Knowledge
for Climate

Coupling of the air quality model LOTOS-EUROS to the climate model RACMO

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Content

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Abstract

Ozone and particulate matter have an adverse impact on the health of humans and other organisms. Their concentrations depend not only on emissions but also on meteorological conditions. In a changing climate, their ambient concentrations are therefore expected to change. However, even the sign of the changes is still highly uncertain, especially for particulate matter. Due to the complex interaction of meteorology and air quality, which is different for different species, and would ideally include feedback mechanisms, the best approach to study the effect of climate change is to use an air quality model that is coupled to a regional climate model, and analyse simulations that are performed with the coupled models.

This study presents the coupling between the regional climate model RACMO and the regional chemistry transport model LOTOS-EUROS and first results for the present-day climate. The coupling consisted of a one-way coupling in which LOTOS-EUROS was able to read the meteorological fields form RACMO, furthermore LOTOS-EUROS was adapted to run in parallel, using openMP and a faster heterogeneous chemisty routine (EQsam instead of Isorropia). First of all, the report shows that the coupled system works and that runs can be performed within an acceptable amount of computation time.

First, a RACMO run was performed for 1989-2009 using ERA-interim fields at the boundaries, and coupled to a LOTOS-EUROS run. A comparison of this meteorology with ECMWF analysis meteorology for 2003-2007 showed that for variables that are important for air quality, namely daily maximum temperature, daily average wind speed and wind direction, the correlation was very good with minor biases. Only for rain, which is also highly important, the correlation was poor, but annual totals were comparable. Rain is notably difficult to model and due to its discrete character small mismatches already lead to poor correlations.

From a comparison of modelled air quality with observed concentrations for LOTOS-EUROS runs with ECMWF meteorology and with the downscaled RACMO-ERA_interim meteorology for the years 2003-2007, it followed that in general, modelled concentrations are correlated with observed concentrations for both runs and that their dependency on wind, temperature and rain is comparable. However, some biases were found in both runs. Ozone is underpredicted for high temperatures. Nitrate and ammonium are too sensitive to high temperatures and not sensitive enough for low temperatures and tend to be underestimated, even more so in the RACMO-LOTOS-EUROS run with EQsam. Sulfate is underestimated but its temperature dependency seems to be represented correctly. For black carbon and sea salt, no validation could be done due to a lack of observations.

Finally, a run with LOTOS-EUROS using RACMO-downscaled ECHAM5r3 A1B climate scenario meteorology was performed for 1970-2060. In the present report, a preliminary analysis was done, restricted to 2003-2007. This already revealed differences in the meteorology: when using the climate scenario there were overall lower daily maximum temperatures, higher wind speeds and more rain, resulting in lower ozone and particulate matter concentrations. The period of 5 years is too short for a solid bias characterization of the meteorology and will be extended to the full ERA-interim period (1989-2009) to get more reliable estimates. This will be done in a follow-up of the present project, together with the analysis of the climate run for our target period: the climate in 2050.

1 Introduction

Background/rationale

North-western Europe, most notably the Netherlands, is one of the most densely populated regions of the world and subject to high levels of air pollution. Anthropogenic activities produce a vast amount of atmospheric emissions of pollutants such as nitrogen oxides, ammonia, sulphur dioxide, volatile organic compounds and particulate matter. In the atmosphere these constituents form secondary products, most importantly ozone and particulate matter. Exposure to air polluted by particles as well as photochemical smog induces adverse health effects, especially for sensitive groups in the population. Ozone may also contribute to crop or vegetation damage in general. Deposition of reduced and of oxidized nitrogen negatively effect biodiversity, when ecosystem critical loads are exceeded.

Meteorology plays an important role in the formation, transport and removal of air pollutants from the atmosphere. Hence, it is common knowledge that there may be a significant impact of climate change on air quality. Under climate change conditions, say in 2040-2050, the concentrations of air pollutants will differ from the current concentrations in 2010, even when present-day anthropogenic emissions are assumed. This implies that the present emission reduction strategies to fulfill air quality criteria may not be sufficient when high temperatures or stagnant conditions would occur more frequently in 2050, which makes a study of the impact of climate change directly relevant for policy making. The increase in temperature, the changing weather patterns including changes in rainfall and its intensity, may impact on air pollutants concentrations like O3, NO2 and PM 2.5. Van Dijk et al. (2010) performed an analysis of the current knowledge on the potential effects of a changing climate to air pollution conditions in the Netherlands. A general finding is that our knowledge basis is still poor. Present-day knowledge indicates that it is likely that ozone concentrations in summer will increase due to higher temperatures, stagnant weather conditions and more frequently occurring heat waves (Vautard et al, 2007, Vautard and Hauglustaine, 2007, Giorgi and Meleux, 2007, Forkel and Knoche, 2007). The extent of the increase is however still highly uncertain. Models show summer mean increases ranging between 1 and 10 ppb, indicating that concentrations during episodes may show a more substantial increase. For PM large uncertainties exist in the effect of climate change, due to the different response of the individual components to changing meteorological conditions. The very few modelling studies do not show consistent results, even on the sign of the effect (Jacob and Winner, 2009). Van Dijk et al (2010) argued that especially the increase in stagnant weather conditions in summer and the decrease in wet deposition efficiency will increase the PM concentrations in a changing climate. Quantification of the effect is not possible at present. Consequently, a dedicated study to assess the first order impact of the predicted climate change on PM and ozone in the Netherlands is necessary.

A further complication is that the concentrations of air pollutants also affect the climate, by acting as a greenhouse gas (ozone) or as cloud condensation nuclei (particles), or by directly scattering or absorbing radiation (particles). The quantification of the full coupled system including feedback of climate and atmospheric chemistry is still under development. First estimates of the difference between climate runs with and without chemistry-climate feedback indicate changes in concentratons up to 40% (Raes et al 2010). Hence, a priority would be the improvement of (the coupling between) the models, since to date important processes are highly parameterized or even missing in models due to uncertainties. . Ideally, climate should be coupled to air chemistry so that continuous feedback can occur.

Strategy for assessment of climate change impact on air quality

Air quality in a future climate should be investigated by using a chemistry transport model (CTM), which is driven by meteorology from a global or regional climate model (GCM or RCM). This is the only methodology to capture the complex interaction between meteorological conditions, natural emissions and the formation and dispersion of air pollutants. As there are many uncertainties associated to climate models and CTMs a stepwise approach is necessary to be able to appreciate the results of a climate change scenario for air quality. One has to take into account that 1) climate models have biases compared to observed climate, 2) CTMs may have biases compared to observed air quality, 3) CTMs may not react well to changing meteorology, and 4) that different global climate models should be used to span the uncertainty in the estimated climate change. To investigate the impact of climate change on air quality using a one-way coupling one needs to perform the following steps:

1. Generate a RCM-CTM interface such that the CTM can be driven with meteorology from the RCM.
2. Verify the results of the air quality model driven by a hindcast simulation of the RCM (verification paired in time) with special attention to the derivative to meteorology and emissions.
3. Use a global climate model to drive the RCM for a regional climate scenario and determine the biases in the meteorology
4. Determine the impact of biases in the meteorology of the RCM on the modelled air quality (verification of air quality PDFs).
5. Perform a number of well defined scenario simulations (current and future) to investigate the impact of climate change on air quality.
6. Perform simulations that account for future developments in important input parameters other than meteorology such as land use and emissions.
7. Evaluate the scenario results to quantify the change with respect to presently observed air quality.

In this study we aim to make progress on this activity list by using the regional CTM LOTOS-EUROS and the regional climate model RACMO. In the Netherlands the CTM LOTOS-EUROS is the community model for air quality.

Within BSIK-CS4 (De Martino et al 2008) a dataflow has been tested and evaluated that enables LOTOS-EUROS to be driven by meteorology from RACMO. Within the BSIK project the RACMO meteorology was generated separately after which LOTOS-EUROS was run for the full period. However, to perform multi-decadal simulations such an off-line approach takes too long in terms of computation time. Moreover, to be able to incorporate the feedback of air pollutants on climate one needs to run the two models simultaneously, with the ability to exchange meteorology and air pollution information. Hence, the first goal of this project was to implement a ‘handshaking’ between the two models and to develop the software enabling running the two models in parallel for multi-decadal simulations. For this purpose, we have pooled resources with the BSIK-CS6 project that envisages determining the climate impact of the aerosol indirect effect.

The improved coupling between RACMO and LOTOS-EUROS is used to test the coupled RACMO-LE system for a multi-year period. For this purpose, RACMO-LE will be forced by ERA-interim boundary conditions, which represent the ‘true’ present-day climate. The second goal of the project is to evaluate the results of RACMO-LE with special attention to the dependency on meteorology (step 2). For this purpose, an evaluation of both modelled meteorological parameters and ozone and PM concentrations will be performed.

Perfect climate models would reproduce observed averages, variability and extremes when running for present-day conditions. Unfortunately, all climate models have significant biases in parameters such as temperature, rain fall and frequency of occurrence of synoptic situations. The third goal of this study is to assess the impact of these biases in the meteorology on modelled air quality parameters (Step 3). Here, we will use results from the GCM ECHAM to force RACMO-LE. In contrast to the original planning, a transient simulation was set up for the period 1970-2060 to obtain results for present day and future climate in a single simulation. We will use the results to investigate the biases in present-day climate simulations compared to hindcast simulations.

Further analysis and quantification of the climate change impact on air quality based on two transient simulations is performed in a follow-up project that is currently running. As such, the present report can be considered a status report.

Set-up of the report

First, a brief introduction in the models used will be given (Section 2) with special attention to the developments needed to be able to run the two models in parallel for long integrations, followed by an outline of the approach (Section 3) Then the performance of the coupled models is demonstrated in terms of representing the analysed meteorology (Section 4) and observed air quality (Section 5). After that, results from the coupled models running in climate mode, representing the present-day climate, are shown and discussed (Section 6). In the present report only the time window 2003-2007 will be analysed. Finally the results and their implications for the next steps in the investigation of the air quality in a future climate are discussed and the main conclusions are summarised.

2 Description of models and runs

A CTM like LOTOS-EUROS needs meteorological fields to drive the transport of species, removal processes (dry and wet deposition), generation of sea salt (wind), and reaction constants and photolysis rates. To model air quality in a changing climate, the regional CTM LOTOS-EUROS was coupled to the RCM RACMO which produces the required meteorological fields. Both models are regional models which need input at the boundary. In the present study, RACMO was driven by ERA climate reanalysis fields, so that it represents the true climate, but also by a free-running global climate model (ECHAM5, in this case). In the present study, LOTOS-EUROS has present-day emissions and present-day climatological boundary conditions of species concentrations. Below, details of the models and their coupling are given.

2.1 RACMO

RACMO is the regional atmospheric climate model of the KNMI. The RACMO 2.2 version used for this project consists of the 31r1 cycle of the ECMWF physics package embedded in the semi-Lagrangian dynamical kernel of the numerical weather prediction model HIRLAM and a few routines to link the dynamics and physics parts.

RACMO uses a rotated longitude-latitude grid to ensure that the distance between neighbouring grid points is more or less the some in the entire domain. For the simulations in this project the RLOTOS50 domain was defined, which encompasses the entire LOTOS-EUROS domain described in the next section. It has a horizontal resolution of 0.44° and runs from 25.04°W to 24.68°E (114 points) longitude and from 11.78°S to 31.78°N (100 points) latitude in the rotated grid. The South Pole is rotated -47° in latitudinal direction and 15° in the longitude. In the vertical, 40 pressure levels were used.

The model runs done for this project used a model time step of 15 minutes and output for coupling with LOTOS-EUROS was generated every three hours. The output fields are shown in Table 1, in which the fields used by LOTOS-EUROS are indicated. For more information on RACMO see Lenderink et al (2003) and Van Meijgaard et al (2008). Friction velocity and u* could be taken from RACMO but are calculated internally in LOTOS-EUROS for consistency with the grid size and land use in LOTOS-EUROS.

Table 1. RACMO meteorological output fields. Fields that are not used by LOTOS-EUROS are in italic.

2D fields:	3D fields:
sensible heat flux [W m-2]	density [kg m-3]
cloud base height [m]	height of layer top from surface [m]
cloud top height [m]	*liquid water path [kg m-2]*
boundary layer height [m]	*mass flux updrafts [kg m-2 s-1]*
low cloud cover [0-1]	relative humidity [0-1]
1 / Monin-Obukhov length [m-1]	temperature [K]
precipitation [kg m-2 3hr-1]	zonal wind speed [m s-1]
2 meter relative humidity [0-1]	meridional wind speed [m s-1]
Snow cover fraction [0-1]	
incoming surface short wave radiation [W m-2]	
skin temperature [K]	
2 meter temperature [K]	
friction velocity (u) [m s-1]*	
10 meter wind speed [m s-1]	

2.2 LOTOS-EUROS

LOTOS-EUROS is a Eulerian chemistry transport model. In this project version 1.6.1 is used on the European domain, from 10°W-40°E, 35-70°N on a 0.5x0.25° longitude-latitude grid with 5 dynamical vertical layers, including a surface and a mixing layer and reservoir layers.

Modelled species are ozone, nitrogen oxides, ammonia, primary PM2.5 and black carbon, primary PM10 (excluding PM2.5 and black carbon), sulfate, nitrate, ammonium and sea salt and species relevant as precursors or reservoir (peroxy-acetylnitrate, volatile organic carbon). For (photo)chemical gas reactions the CBM IV scheme is used, for secondary inorganic aerosol (heterogeneous chemistry) Isorropia or EQSAM is used.

The regional model uses climatological boundary conditions for most species. In the present set-up, these boundary conditions will not change in a changing climate. Anthropogenic emissions are kept constant during all runs and consist of the MACC 2005 emissions (Denier van der Gon et al (2010a, b). In the present study, fixed anthropogenic emissions will be used for the whole period to isolate the effect of climate change. Natural emissions of sea salt and isoprene emissions by trees are calculated on line, they depend on wind speed (sea salt, parameterization of Monahan et al. 1986) and temperature (isoprene). Dust emissions, forest fire emissions and secondary organic aerosols are not included since they are either too uncertain (secondary organic aerosols), mainly fall outside the domain (dust) or cannot be modelled in a realistic way in a climate run (fire emissions). For the removal of species the EMEP parameterization for wet deposition and the depac scheme for dry deposition (Erisman, 1994) is used. A land use database (CORINE/Smiatek) is used for the information needed for the parameterization of natural emissions of isoprene and for the dry deposition.

A full model step represents one hour. Model output consists of concentrations at ground level and model levels, and daily output of cumulative dry and wet deposition of SOx, NOx, NHy. See Schaap et al. (2008) for a more extensive description and validation of the model.

2.3 Coupling

The coupling of RACMO and LOTOS-EUROS required a number of technical adaptations.

First of all, LOTOS-EUROS had to be able to read the meteorological data from RACMO. This means that the RACMO meteorological fields on a rotated grid with 40 pressure levels have to be interpolated to the LOTOS-EUROS 0.5x0.25° lon-lat grid with 5 geometric vertical layers. The development of interpolation routines was part of the BSIK CS-4 project. The air quality results were evaluated against observations for a 5 year period and found to be in line with the results using other meteorological sources.

Although for the 1-way coupling of the present run the LOTOS-EUROS model can be started after the RACMO run has finished, we chose a different approach to facilitate a future 2-way coupling ('handshaking') of the models. RACMO has been adapted to be able to read in concentrations fields of LOTOS-EUROS in BSIK CS-04 (De Martino et al 2008). In a fully coupled version, after each RACMO 3-hourly model step, the corresponding 3 hours are modelled in LOTOS-EUROS and a feed-back is then possible. This implies that the models have to wait for each others output. At present, an intermediate set-up is used, in which RACMO and LOTOS-EUROS are run independently for one-month periods.

LOTOS-EUROS was too slow for a long simulation period. Therefore, as part of the present project the code was adapted to run the model in parallel. This was done by using OpenMP, which resulted in a considerable speed-up of a factor 3.3 when using 8 processors as compared to a standard sequential run. The

Isorropia routine of LOTOS-EUROS was however not parallelized, as an alternative the EQsam parameterization was used, which is faster and can be used in parallel. A further issue is the amount of output. To reduce this, concentrations were only stored every 3 hours, which is also the minimum for a future two-way coupling. For PM, only daily averages are studied, but for ozone this temporal reduction would remove the modelled daily ozone maximum from the output. As a solution, the daily ozone maximum was also written to file.

2.4 Model runs

In the present project, three model runs were performed:

- Model run with ECMWF analysis fields 1-1-2003—31-12-2007 using Isorropia,
- Model run, coupling with RACMO, forced by ERA-interim boundaries, 1-1-1989—31-12-2009, using Eqsam
- Model run coupling with RACMO, forced by ECHAM5r3 A1B scenario boundaries, 1-1-1970—31-12-2059, using EQsam.

The first run can be used as a baseline, illustrating the model's ability to model air quality. The second run gives insight in the ability of the model to be run in climate mode. It illustrates both the technical functioning of the coupled models and gives insight in differences caused by the use of RACMO and the different SIA chemistry, when compared to observations. The third run is the climate run that is the ultimate goal of the project.

3 Observations and variables for bias characterization

In this section, the approach is outlined and the measures and observations which are used in the analysis of the model runs are introduced. Climate change will have an impact on all meteorological variables, but not all of them will have a direct and clear impact on air quality. Therefore, the most relevant and accessible meteorological variables will be identified first. Also the difference between the used meteorologies (ECMWF analysis, and ERA-interim, ECHAM5r3 and their downscaling with RACMO) is briefly addressed. The relevance of the selected parameters is demonstrated in section 5.

To assess the system of coupled models, the modelled concentrations of pollutants will be compared to observations for the runs with ECMWF analysis and downscaled ERA-interim fields. The observations used in this report are briefly introduced at the end of the present section. Then, in section 5, observations and model results are combined with meteorology to assess biases in the model. Not only the averages, but also the variability and maxima of modelled concentrations are presented and compared with observations to investigate overall effect of biases in RACMO and LOTOS-EUROS together. The approach will give an estimate of the biases of the coupled system ECHAM-RACMO-LOTOS-EUROS.

3.1 Meteorology sets

Three different sets of meteorological fields are used in this study. The first set is the ECMWF analysed meteorology, which is nowadays the default for LOTOS-EUROS. This meteorology is based on the ECMWF global meteorological model, in which available observations have been taken into account. This does not imply that the meteorology is identical to the observations at a given location, but that the modelled fields are close to the observations and at the same time consistent with the large scale flow. In the present study, these analysed fields are considered as the baseline for comparison.

The second and third set consists of meteorological fields derived from RACMO. The second set is a downscaling by RACMO for ERA-interim climate analysis fields. Since these ERA-interim fields are analysed fields, the resulting downscaling should closely resemble the first set of meteorological fields. However, since RACMO is not fed by observations, differences will occur, which are illustrated in this study as far as they are relevant for air quality. The third set is a RACMO downscaling of the ECHAM5r3 global climate model run, in the IPCC A1B transient scenario. In the present report, only the ability to represent the current climate is investigated for this scenario.

3.2 Relevant meteorological variables

Climate changes will not only influence the average temperature, but also affect the preferent flow patterns and the frequency, length and intensity of blocking episodes or depressions and the hydrological cycle. All meteorological variables are connected, so a bias in one variable will not be independent of another variable. A full analysis is beyond the scope of this report. Here, the bias characterisation is restricted to a number of meteorological variables, which are known to be related to air quality (Fig. 1, see for example Manders et al 2009 for PM10 in the Netherlands, Tai et al 2010 for PM2.5 in the US, Anderson, 2009 for ozone in Europe). In the present study, we will use daily maximum temperature, daily average wind speed, wind direction and daily amount of rain. Cold days are days with maximum temperatures lower than 5°C or lower, warm days are days with maximum temperatures higher than 25°C. Wet days are days with more than 0.5 mm of rain. Low wind speeds are used here as an indicator for stagnant conditions, although strictly speaking one should look at a set of variables, including pressure. A daily average wind speed of less than 2 m/s is counted as a low wind speed day. Below, their role in air quality processes is discussed in more detail.

Daily maximum temperature

Statistical analysis has shown that temperature is the most important factor for ozone concentrations (e.g. Anderson, 2009). High ozone concentrations are related to high temperatures (and low cloud cover), as ozone is photochemically formed from precursor gases. Also the emission of biogenic precursor gases is higher for high temperatures. For PM relations are less clear. High temperature may cause semi-volatile species to be mainly in the gas phase, lowering the PM concentrations, but may also enhance reaction rates. Cold weather will make semi-volatile species less volatile. But both very warm and very cold conditions may be related to stagnant conditions with little ventilation and in winter additionally a shallow boundary layer. These conditions by themselves favour high concentrations of pollutants. Furthermore, temperature does not only affect the chemical reaction rates, but in an indirect way also the deposition. When the temperature is above a threshold value, vegetation closes their stomata, thereby decreasing the deposition velocity, and temperature has an impact on the wetness of the surface, also an important factor for the deposition velocity.

Wind direction

The wind direction determines the transport of pollutants and precursors from areas. For the Netherlands for example, southeasterly winds bring more pollutants from densely populated and industrial areas in Belgium and Germany (Ruhr area) to the country, whereas northwesterly winds bring relatively clean air although there may be an impact of shipping emissions. For other countries the wind directions associated with less clean air may be different, which reduces the use as a universal parameter.

Wind speed

Wind speed determines not only the transport but also the exchange, both horizontally and vertically. Low wind speeds are related to stagnant conditions, with a low boundary layer in winter. In such a shallow boundary layer, emitted pollutants can reach rather high concentrations. In contrast, high wind speeds are mostly related to low PM concentrations, except for sea salt aerosol, because sea salt aerosol is directly generated by wind over the sea surface.

Rain

Rain is a very efficient removal mechanism for particulate matter through direct rainout or washout. Also, the dry deposition of species is affected by the wetness of the surface. In reverse, a dry surface will favor resuspension of particulate matter and will be more liable to wind erosion. We will not go into the details of soil moisture and evaporation in this study. It is not only rain duration and intensity which are important here, but also the period of drought. Since rain is often associated with the passage of a frontal system, it is in many cases correlated with the entrance of air with a different (often cleaner) origin.

Flow patterns

Other possibilities are to look at pressure differences or the blocking index. The Lamb weather classification is based on the ground level pressure, and changes in the pressure or pressure differences between two locations up to a few 100 km apart and describes the major flow pattern. The blocking index is a similar measure, indicating a specific flow pattern higher up in the atmosphere. The abovementioned variables are not fully independent, as indicated before.

Both are the same kind of measures, but less easy to interpret in relation to air quality than temperature, wind and rain. On the other hand, the number of blockings or passage of strong depressions can be a suitable summary for a year and a measure of climate change.

Cluster analysis and empirical orthogonal functions (EOFs) can be a further integration of meteorological variables. Since the formulation of clusters and cluster analysis are a study on its own, they are beyond the scope of the present study.

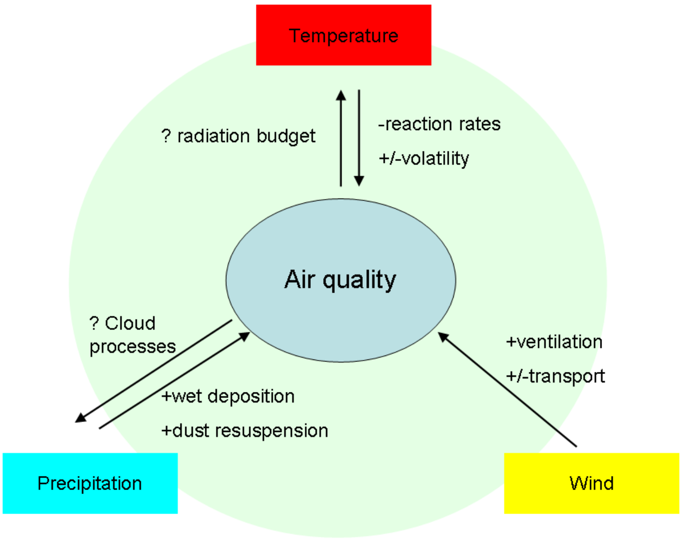

Figure 1. Simplified representation of interaction between air quality and meteorology.

3.3 Observed concentrations

To verify the model results and to characterize biases, the modelled concentrations are compared to measurements in the EMEP database. Countries can contribute observations from their national network to the EMEP database, provided that the observations meet certain standards. Not all national observations are submitted to the EMEP dataset, and not all chemical species or constituents are monitored at all observation locations. Some concentrations are monitored on an hourly basis, these are mainly the gases (ozone, nitrogen oxides) which exhibit strong daily variations due to photochemistry. Particulate matter concentrations can only be measured with reasonable accuracy by weighing filters on which the daily accumulated particulate matter is attached when the air is sucked through it. The total mass is measured at many locations, but, analysis of the several PM10 constituents is limited to a few locations, even then there may be a non-daily coverage.

To verify the output from RACMO and LOTOS-EUROS a number of locations is selected (Fig. 2). These are a sub-selection of the EMEP stations. In the present study, implications of climate change on air quality in the Netherlands are the main focus. Therefore, we concentrate on the Dutch stations, but a few other stations are included to verify the dependency of concentrations of ozone and PM under different climatic circumstances.

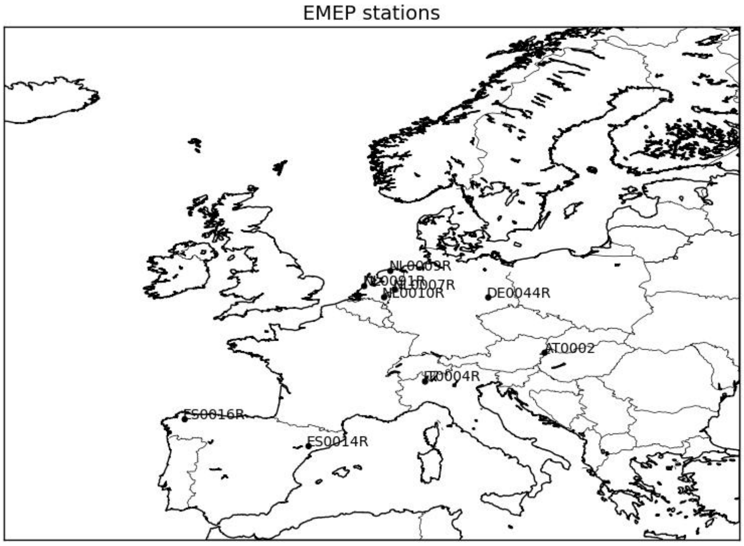

Figure 2. EMEP measurement locations used in the present study

Table 2 Name and code of EMEP stations that are used for more detailed analysis, see Figure 2 for their location

station	station code
Kollumerwaard	NL0009R
Vredepeel	NL0010R
De Zilk	NL0091R
Eibergen	NL0007R
Melpitz	DE0044R
Illmitz	AT0002
Ispra	IT00004R
Els Torms	ES0014R
O Savinao	ES0016R

3.4 Chemical species

In the present study, only a few species are addressed. First of all, tropospheric ozone will be investigated. PM is a container of several subspecies, which may react differently on climate changes (differences in volatility, chemical interactions). Furthermore, not all species that are part of the observed PM are present in the model. Especially secondary organic aerosols, which may be very important and very climate sensitive due to their semi-volatility and chemical equilibria, are not yet understood well enough to be modelled realistically in a CTM like LOTOS-EUROS. In the present study, total PM10 is studied as the sum of sea salt, primary PM2.5 and black carbon, nitrate, sulfate, ammonium and primary coarse PM10 (excluding PM2.5).

4 Meteorology

To test the performance of RACMO, the meteorological fields produced by RACMO, forced by ERA-interim fields, was compared with ECMWF analysis fields. Their features are illustrated for 2003-2007 for the Dutch station Vredepeel (Fig. 3-5). This is an EMEP station for which most observations are available and the station is representative for rural inland conditions. The other Dutch EMEP station Kollumerwaard is close to the Wadden Sea and therefore often has a slightly different behaviour which is less representative for the rest of the Netherlands. The wind direction is taken with 0° as northerly wind, 90° is easterly, -90° westerly and +/- 180° southerly. For the EMEP stations Melpitz and Ispra the behaviour is shown in the appendix and serves as additional illustrations.

First, the correlation between the two sets was investigated for the individual variables. For Vredepeel, daily maximum temperature, wind speed and wind direction of the two meteorologies turned out to be well correlated (Fig. 3). RACMO has slightly higher extreme summer temperatures, and slightly higher wind speeds. For rain however, the correlation is poor, which is at least in part caused by the local and discrete nature of rain. Convective and large scale precipitation were not compared separately. Also the probability density functions of the individual variables were studied. Overall, they are very similar (Fig. 4) for RACMO and ECMWF analysis fields.

As a next step, the correlation between meteorological variables was investigated for both meteorologies. To this end, for each station the five years of modelled daily maximum temperature, wind speed, wind direction and rain were used, results were sorted along the leading variable (x-axis, Fig. 5) and the results (both x and y) were averaged in bins of 50 data points. One should note that the scatter for individual days is large but the approach is used to illustrate the general tendencies. Wind direction is a cyclic variable, so averaging may introduce cancellations of positive and negative values, therefore when it appears on the y-axis it should be interpreted with great care. Also for temperature, averaging summer and winter temperatures may reduce the true signature. To avoid these cancellations, these two variables were put on the x-axis when possible. For Vredepeel, daily maximum temperatures are separated in a few patterns: moderate temperatures (5-20° C) are related to westerly winds (wind from the sea) and both high and low daily maximum temperature are related to easterly (continental) winds although this is not so clear due to the averaging out of wind directions with positive (easterly) and negative (westerly) sign. These higher and lower values also coincide with relatively low wind speeds, whereas temperatures of about 10° C typically occur for higher wind speeds. Winds from the (south)west occur most frequently and have the highest wind speed, whereas northerly and southerly winds have the lowest speed. Rain is clearly related to westerly winds, with a considerable spread. This is consistent with the general pattern of depressions with rain coming from the west, although there are a few occasions with rain coinciding with easterly flow. Rain and wind speed are not clearly correlated. Very cold or very warm days are in general dry, rain occurs at moderate temperatures. This fits in with the moderate temperatures for westerly winds, which are associated with the depressions. The number of consecutive dry days is also related to high and low temperatures, but not so much to wind direction, although for southwesterly winds the number of consecutive dry days is clearly smaller than for other wind sectors.

For Melpitz, a more continental station, the behaviour is roughly similar to that of Vredepeel, with slightly lower wind speeds for southerly winds and more scatter in the rain, in particular for the RACMO fields, and a good general correspondence between the two meteorologies. For Ispra however, at the border between the Alps and the Po valley, the difference between the two meteorologies is larger, mainly in the behaviour of the wind. Due to the presence of the mountains, local topographical effects become important. For this station, high wind speeds are not associated with winds from the sea but winds from the Alps. Due to the cancellation effect described above, the dependency of wind direction and temperature becomes blurred, but the association of high wind speeds to low temperatures, cold winds from the mountains in winter can be deduced, as can be expected. Note also that the wind speeds for Ispra are in general lower than for Vredepeel and Melpitz.

At the EMEP observation locations, the occurrences of cold (daily maximum temperature < 5ºC) and warm days (daily maximum temperature >25 ºC) , calm days (average wind speed < 2m/s) and wet days (more than 0.5 mm rain) were calculated (Table 3a-c, for a selection of stations), as well as overall averages of maximum temperature, wind speed and rain. Quantities were calculated per year and per station, and then averaged over the years. One should keep in mind that the number of warm, cold, wet and dry days is quite sensitive to the threshold value. The results confirm the above findings, with some regional signatures. Total amounts of rain are of the same order of magnitude in both meteorologies, but RACMO tends to be a bit dryer and has at the same time more days with rain, so less rain per event on average. RACMO is also slightly more extreme, regarding the number of warm and cold days, slightly higher maximum temperatures in summer and lower temperatures in winter. Averages are however very close to ECMWF analysis fields. Differences are largest for stations near the coast or in mountaineous areas, where local effects are important.

It has been demonstrated that overall, weather patterns of the two meteorologies are consistent for various locations, showing the same local relationships between the wind, rain and temperature, which implies that RACMO can be used to represent the current climate. Furthermore, the correlation between the two meteorologies is very good except for rain, which means that when forced by ERA-interim fields at the boundaries RACMO indeed reproduced the observed weather.

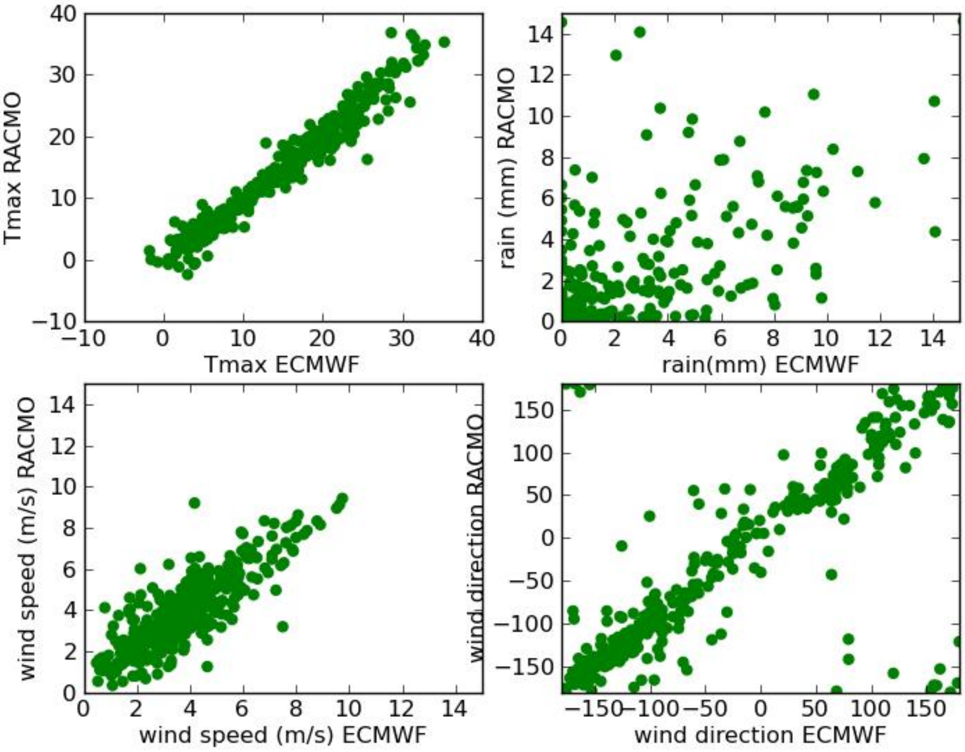

Figure 3. Correlation plot of meteorological variables for Vredepeel, RACMO versus ECMWF, 2006.

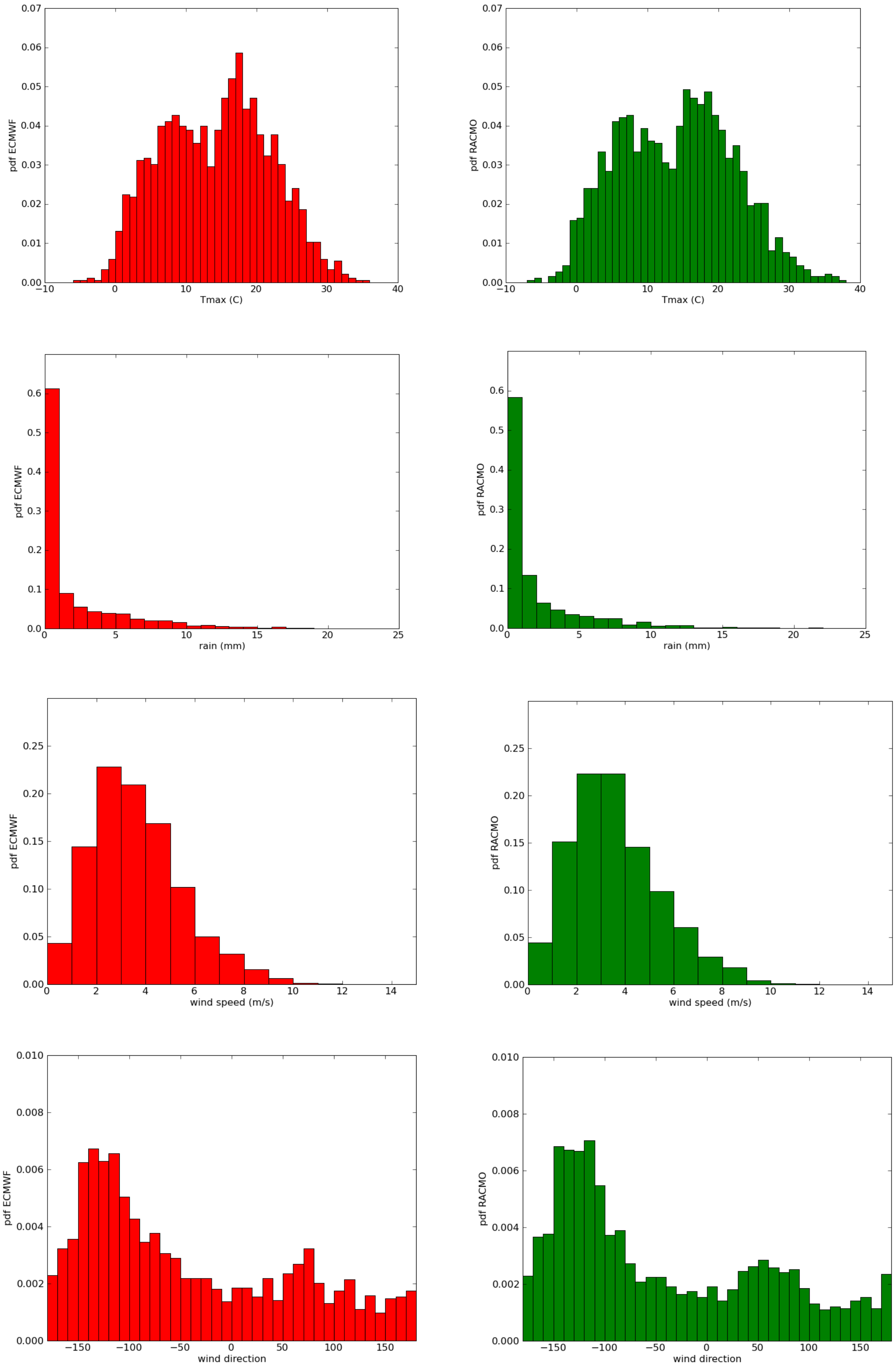

Figure 4. Probability density plots for 2003-2007, ECMWF and RACMO meteorology at Vredepeel.

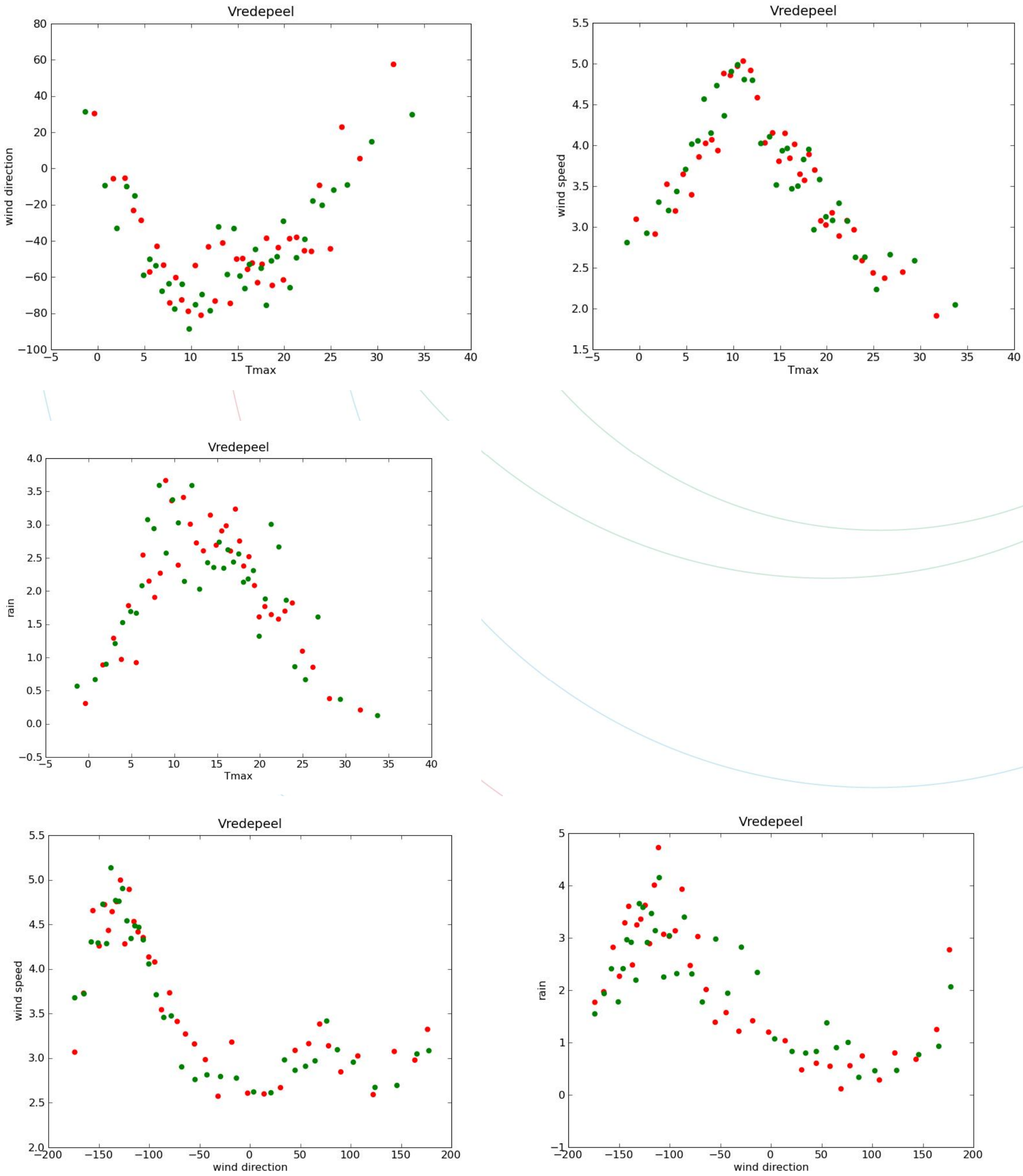

Figure 5. Scatter plots of sorted and binned meteorological data for 2003-2007, ECMWF (red) and RACMO (green).

Table 3a: ECMWF and RACMO-ERA_INTERIM, averages 2003-2007

station	ECMWF				RACMO-ERA_INTERIM			
	# T>25 C	# T<5 C	av Tmax	av std	# T>25 C	# T<5 C	av Tmax	av std
Vredepeel	30.2	48.4	14.3	7.5	32.8	55.8	14.0	8.0
Kollumerwaard	8.4	42.8	12.8	6.3	15.4	49.8	13.0	6.9
De Zilk	10.2	36.8	13.4	6.3	18.0	42.2	13.6	7.0
Eibergen	29.8	54.6	13.9	7.7	28.8	56.0	13.7	7.8
Illmitz AT002	0.2	7.6	14.6	9.5	0.2	91.4	14.1	10.3
Melpitz DE44	42	74.8	13.9	8.8	45.6	89.4	13.4	9.4
Ispra IT004	46.2	53.4	14.6	8.3	61.8	65.6	14.6	9.2
ES0014	110.4	3.8	19.5	7.8	137.8	1.6	21.6	8.8
ES0016	30.5	5.6	16.0	6.1	60.2	2.2	17.0	6.7

Table 3b Averages rain 2003-2007. Wet days are days with more than 0.5 mm rain.

station	ECMWF			RACMO-ERA_INTERIM		
	#wet days	average yearly rain (mm)	av. stdev daily rain (mm)	# wet days	average yearly rain (mm)	av. stdev daily rain (mm)
Vredepeel	163.6	760	3.5	193.8	750	3.5
Kollumerwaard	177.6	882	3.9	206.8	835	3.6
De Zilk	173.4	882	4.0	196.0	757	3.3
Eibergen	166.0	785	3.6	202.8	830	3.7
Illmitz AT002	116.6	513	3.4	133.2	551	3.7
Melpitz DE44	147.8	587	3.0	166.6	522	3.0
Ispra IT004	122.8	959	6.9	107.8	938	7.9
ES0014	77.6	426	3.9	68.2	322	3.1
ES0016	152.6	941	4.8	196.0	965	4.8

Table 3c Averages wind speed 2003-2007

station	ECMWF			RACMO-ERA_INTERIM		
	#v<2m/s	average v	av stdev v	#v<2m/s	average v	av stdev v
Vredepeel	68.6	3.6	1.8	71.4	3.6	1.8
Kollumerwaard	37.8	4.9	2.4	25.6	5.4	2.5
De Zilk	46.2	4.5	2.3	30.2	5.3	2.5
Eibergen	67.8	3.7	1.8	59.6	3.8	1.8
Illmitz AT002	116.4	3.1	1.8	145.8	2.5	1.3
Melpitz DE44	82.8	3.4	1.7	71.4	3.6	1.9
Ispra IT004	335.4	0.8	0.7	285.0	1.3	0.9
ES0014	183.8	2.3	1.5	151.1	2.6	1.6
ES0016	92.8	4.0	2.4	124.0	3.1	1.9

5 Air quality modelling

To investigate the performance of the coupled system RACMO-LOTOS-EUROS, model results are compared with both observations and LOTOS-EUROS results driven by ECMWF analysis. First the general behaviour of the model is demonstrated, then the dependency of the modelled and observed concentrations on wind, temperature and rain is investigated per species.

5.1 General behaviour

To illustrate the general functioning of the coupled system, model runs with ECMWF analysis meteorology using Isorropia (run1) and with RACMO ERA meteorology using EQsam (run2) were compared with observations (Fig. 6) for ozone (Vredepeel) and total modelled PM10 (Melpitz, no observed total PM10 available for Vredepeel). For ozone, the correlation is quite good, for both meteorologies. For run1, the variability in time is slightly better than in run2, which will be caused by the use of analysed meteorology which will represent the true meteorology slightly better. However, high concentrations tend to be underestimated by LOTOS-EUROS, in particular in run 1. This is probably the effect of the somewhat higher temperatures for the warmes days when using RACMO, and not related to the difference in heterogeneous chemistry. For PM10, the model seriously underestimates the observed concentrations, as expected, since not all PM10 components are present in the model. Below, the behaviour of the individual components is illustrated. Here, we just show that the total modelled PM10 of both model runs is comparable, with a quite good correlation and slightly larger high concentrations when using EQsam. These high concentrations often correspond with observed higher concentrations.

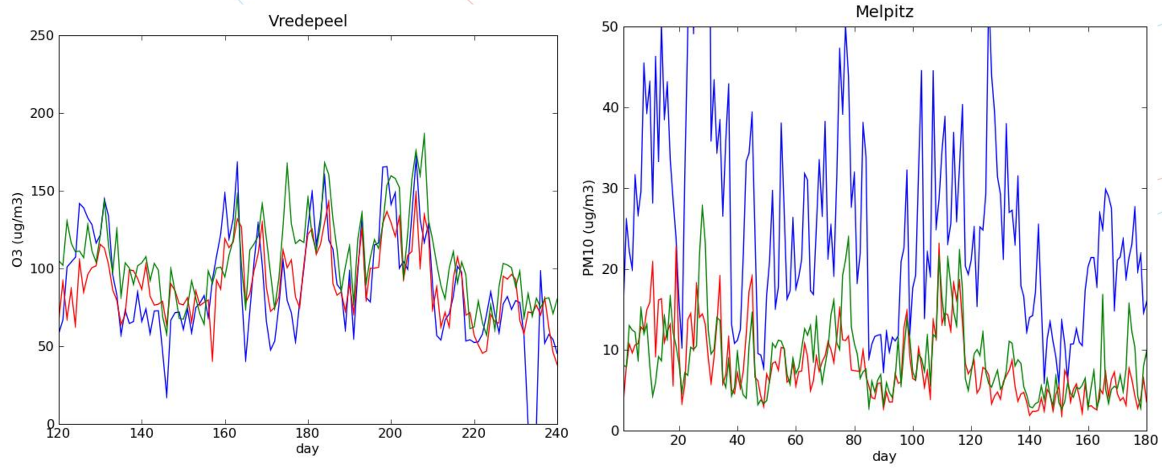

Figure 6. Time series of observed(blue) and LOTOS-EUROS modelled (red, ECMWF, green: ERA-interim) concentrations, Left: ozone daily maxima, Vredepeel 2006. Right: daily average PM10, Melpitz, 2006.

Scatter plots of modelled versus observed concentrations for Vredepeel (Fig. 7) confirm this general picture. For run2 using RACMO, the spread is larger than for run1. For ozone, the tendencies to overestimate ozone concentrations in the moderate range (50-100 $\mu g/m^3$) and to underestimate them for the high concentrations are clearly visible. For NO3 and NH4 both model runs overestimate the concentration for this station, whereas SO4 concentrations are underestimated. For this station, it should be mentioned that the PM3 fraction is sampled, not PM10. Since SO4 is mostly in the finer fraction of PM10 it is expected that the observed SO4 concentrations are representative for SO4 in PM10. Observed SO4a concentrations have discrete values, resulting from the method of analysis.

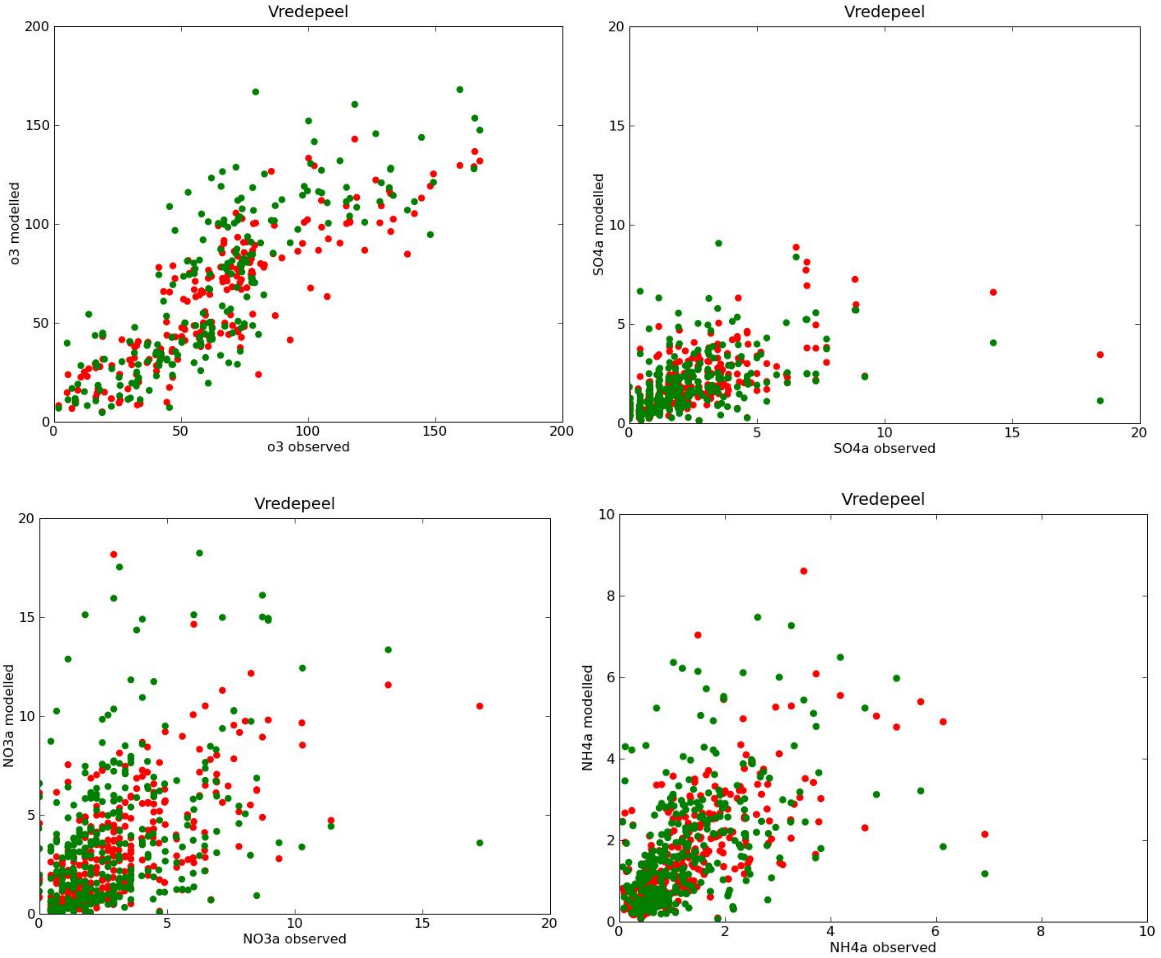

Figure 7. Modelled versus observed ozone daily maxima and daily averages of sulfate, nitrate and ammonium, 2006, Vredepeel, concentrations in μg/m^3*.*

The fairly good correlation makes it possible to compare two snapshots of the model fields for an instant of high ozone concentrations in the Netherlands (Fig. 8). General patterns in the two runs are comparable, with the elongated structures with high ozone concentrations from southwestern France to northern Finland, albeit with some shift in the run using RACMO.

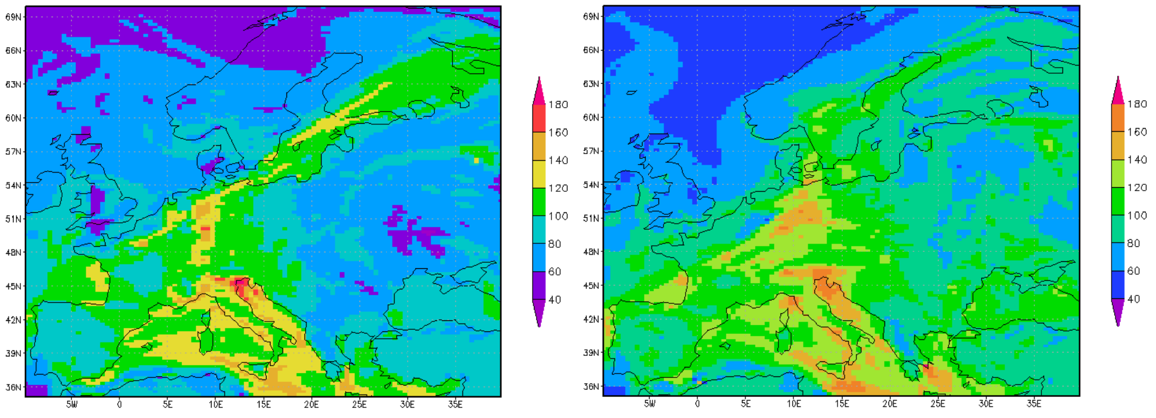

Figure 8. Modelled event with high ozone concentrations, 14 June 2006, 15h. Left: run1, right run2 (RACMO).

For PM10, 5-year averages of total modelled PM10 of the two runs were compared (Fig. 9), as well as the averages of the modelled species (Fig 10 and 11).

Run2 has higher concentrations of secondary inorganic aerosols. This is mainly caused by using EQsam instead of Isorropia, which results in higher production or stability of NO3, NH4. Sodium concentrations have increased above the North Sea but decreased above the Mediterranean, which will be caused by a change in wind speed: larger wind speeds in RACMO above the North Sea, lower wind speeds above the Mediterranean. For primary PM2.5 and PM10, mainly the major cities stand out as locations with differences, with higher concentrations when using RACMO.

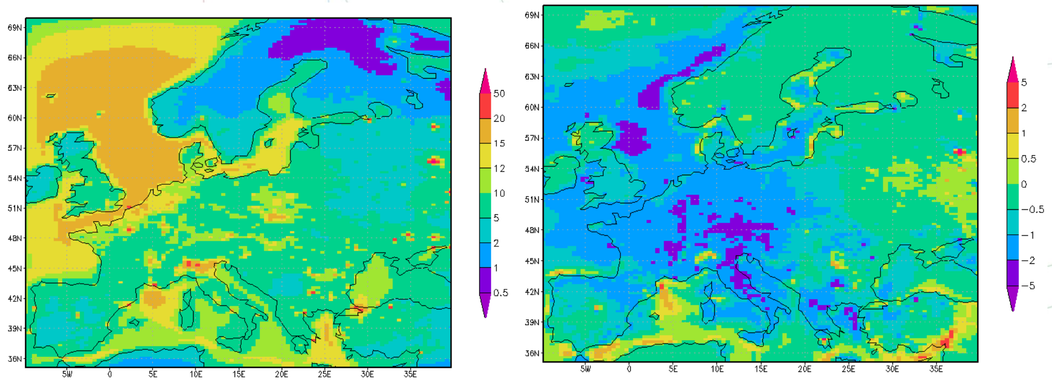

Figure 9. Average modelled PM10 concentration, 2003-2007.Left:: run2, right: concentration difference, run1-run2.

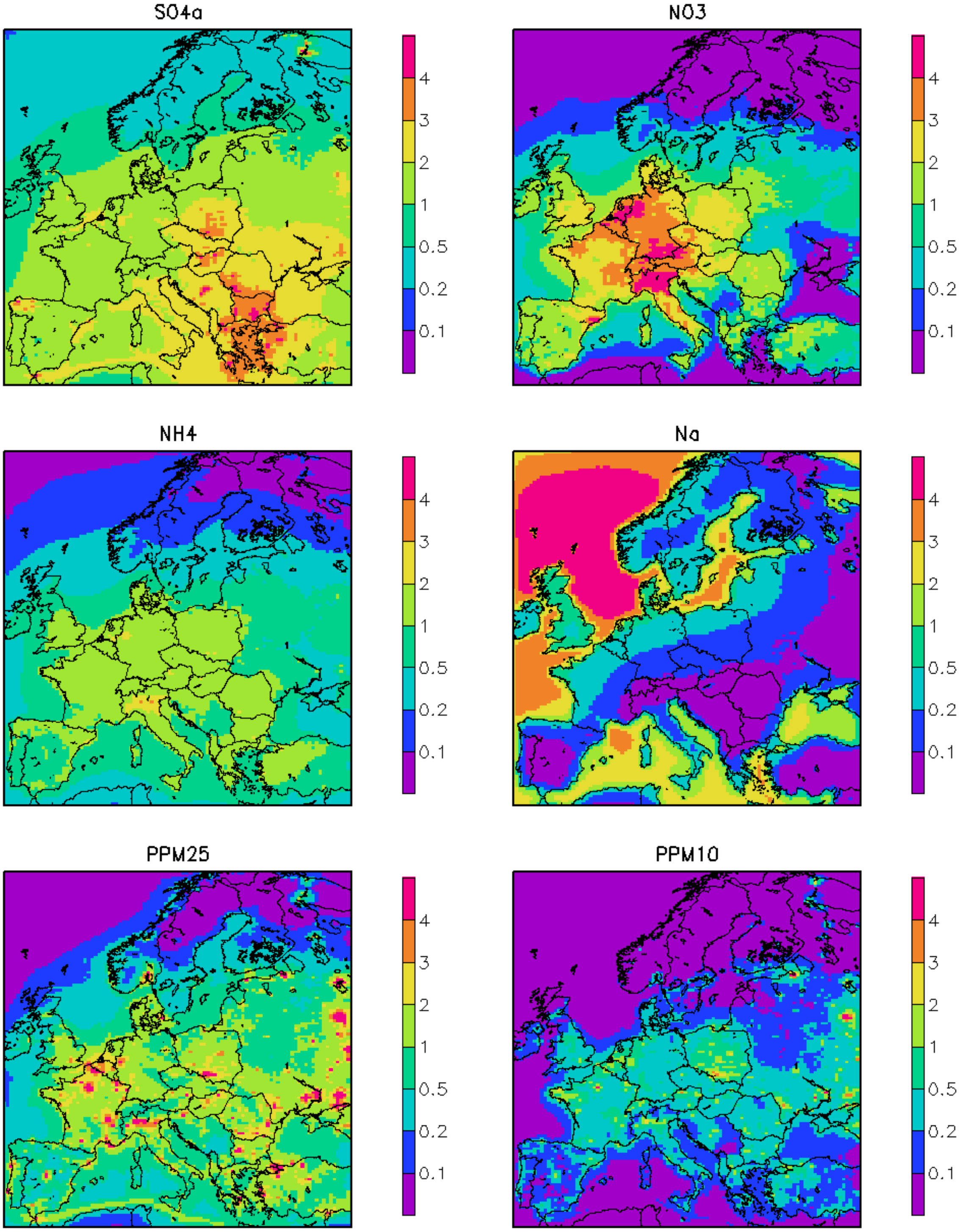

Figure 10. Average PM concentrations ,run2, 2003-2007.

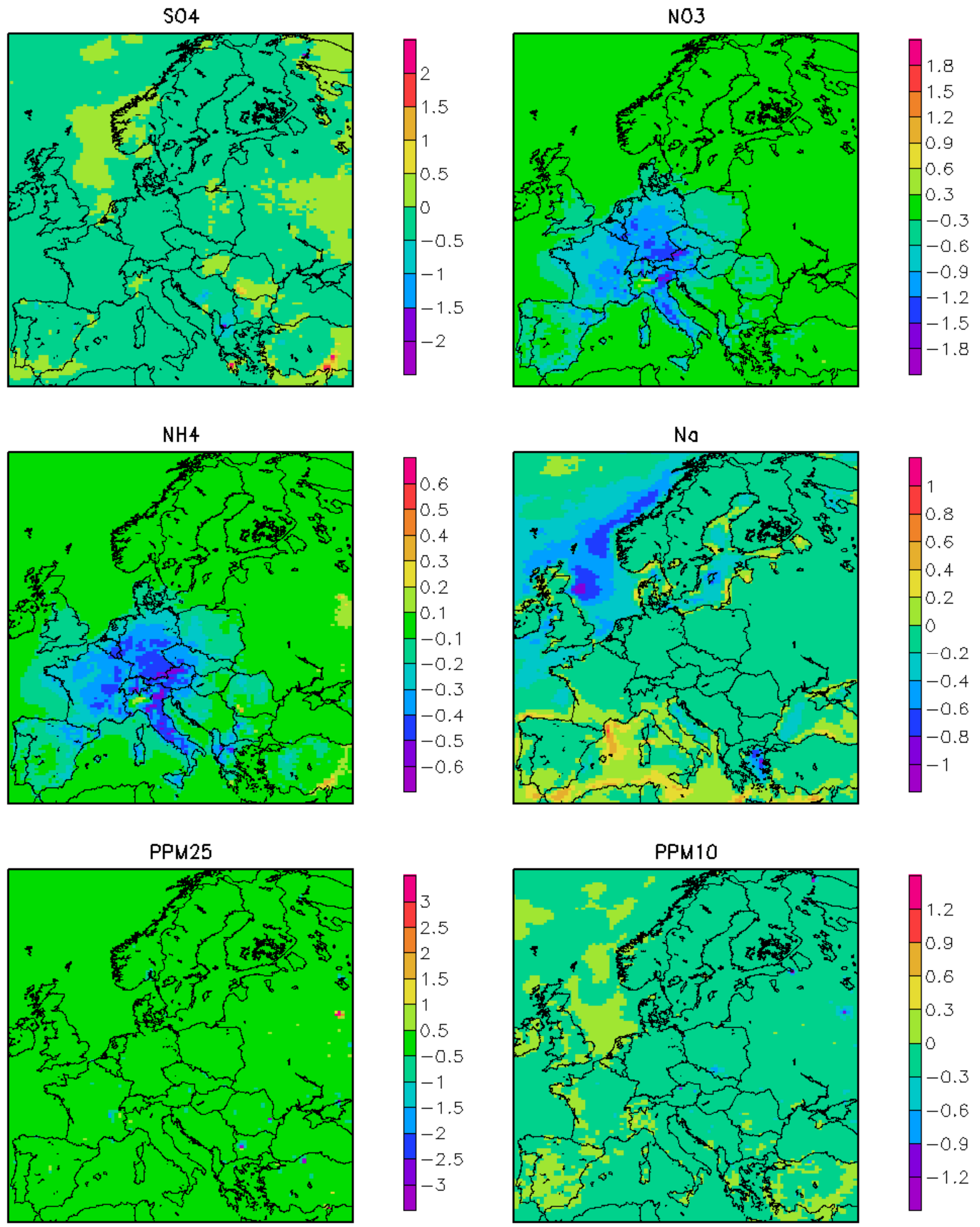

Figure 11. Absolute differences in average PM concentrations, run1-run2.

5.2 Effect of temperature on ozone daily maximum

To study general relationships, daily maximum temperature, wind and rain have again been sorted for 2003-2007. Results were averaged in bins of 25 data points for the summer periods (April-September) except for temperature for which the whole year was taken and 50 data points per bin were taken. For ozone, the maximum temperature is the main criterion for high concentrations both in the model and for observations (Fig. 12). The increase of ozone concentration with temperature is stronger than linear. For maximum temperatures above 20°C, the ozone concentrations increase rapidly. The model tends to underestimate the concentration, both in run1 (red) and in run2 (green), in particular for the higher temperatures. In run2, this underestimation is a little less strong, but there it is overestimated for the moderate temperatures. This effect is probably caused by the difference in heterogeneous chemistry, but can also be caused in part by the slightly higher daily maximum temperatures on the warmest days in RACMO. Also the duration of a warm episode may be relevant. We have investigated the ozone concentration against the number of days with maximum temperature above 25°C (episode length).

For Kollumerwaard there seems to be an increase in ozone concentration with increasing episode length, but due to the closeness to the sea this station is not representative for large areas. Since for the other stations the impact of the length of the episode is less clear, the duration of the episode is not used further in the present study. There is also a general correlation with wind speed and rain, but these have much more scatter and can be ascribed to the general relationship between temperature and these variables. For Ispra and Melpitz (appendix B) the same strong correlation with temperature, including the underestimation for the highest concentrations were found. Again, when using RACMO and EQsam, the modelled concentrations are higher than for ECMWF and Isorropia. In a test using EQsam and ECMWF meteorology, the ozone concentrations were not different from the run using Isorropia, so the differences found here must be ascribed to differences in meteorology. Daily maximum temperatures of RACMO on warm days were up to a few degrees higher than for ECMWF.

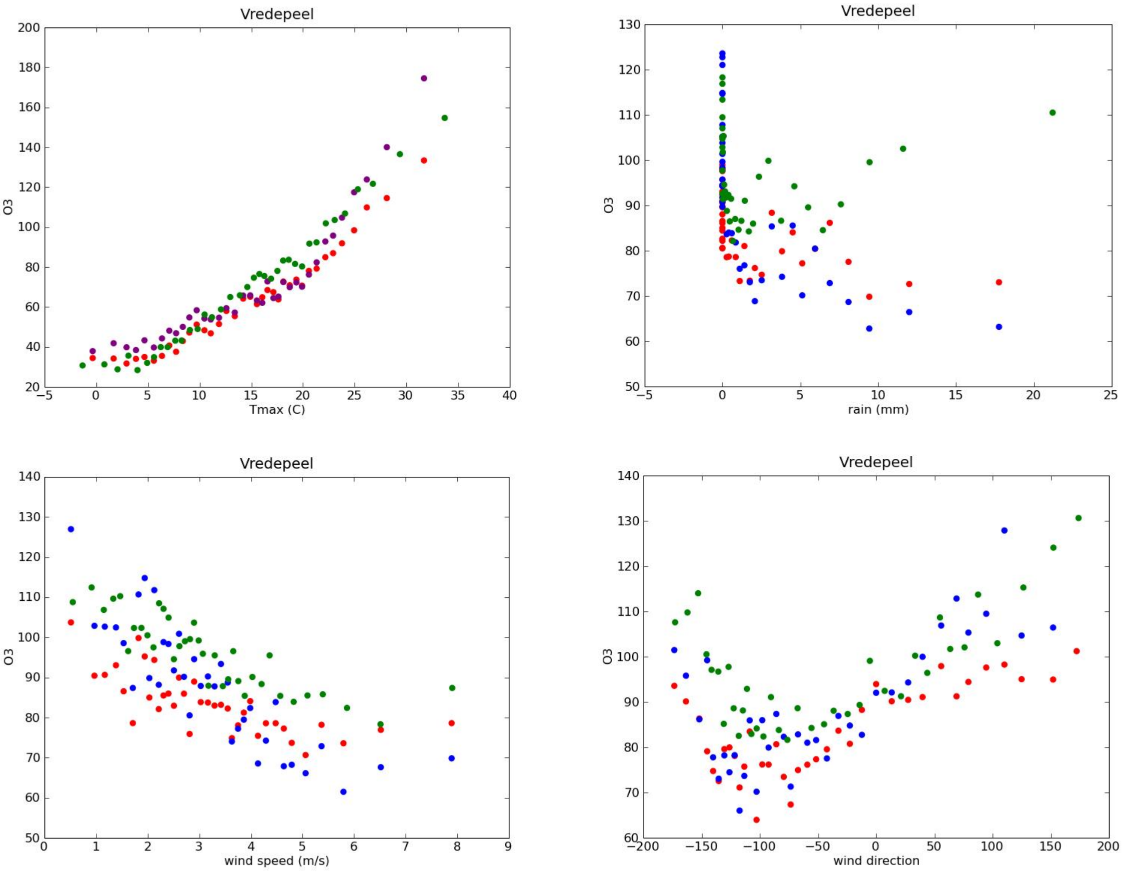

Figure 12. Daily maximum ozone concentrations versus daily maximum temperature, 2003-2007, rain, wind, April-September, 2003-2007 Vredepeel. Blue/purple: observed, red run1, green run2.

5.3 Effect of temperature, wind and rain on daily average PM concentrations

Total PM is always underestimated in models, since some components are not taken into account in the model, like dust and secondary organic aerosol. Below, the behaviour of the modelled components are discussed per component and is compared with observations when possible.

5.3.1 Black carbon

Black carbon concentrations are small, but relevant since black carbon is associated with a negative health effect, and has at the same time an impact on the radiation budget, thus influencing the climate. Black carbon is a passive tracer, which means that it is not affected by chemistry and therefore only depends on emission, transport and deposition. The concentrations decrease with increasing wind speed, with a nearly linear trend (Fig. 13, and appendix B). Winds from the southeast bring most BC to the measurement location in Vredepeel (air from Ruhr Area) but this wind direction is also correlated to low wind speeds, we cannot fully separate the effect. Concentrations are higher for daily maximum temperatures below 7°C. Rain is a very efficient removal process, and concentrations are indeed higher for days with no to little rain than for rainy days, with a sharp decrease in concentrations for wet days, and the lowest concentrations for the wettest days. The relationship with number of consecutive dry days was not so clear, and is therefore not studied in more detail. Results with RACMO are more scattered than with ECMWF, as we have seen for the meteorology itself. Since chemistry plays no role here, this is the pure effect of meteorology differences. For Melpitz the concentrations are much lower than for Vredepeel, for Ispra they are somewhat higher.

Also for these stations the BC concentrations decrease with increasing temperature, but with different patterns than for Vredepeel. Ispra shows no clear relationship with wind direction for ECMWF meteorology, but for RACMO meteorology concentrations are higher for easterly winds. For Melpitz, concentrations are clearly lowest for westerly winds. Again, a nearly linear negative relationship for concentration and wind speed is found.

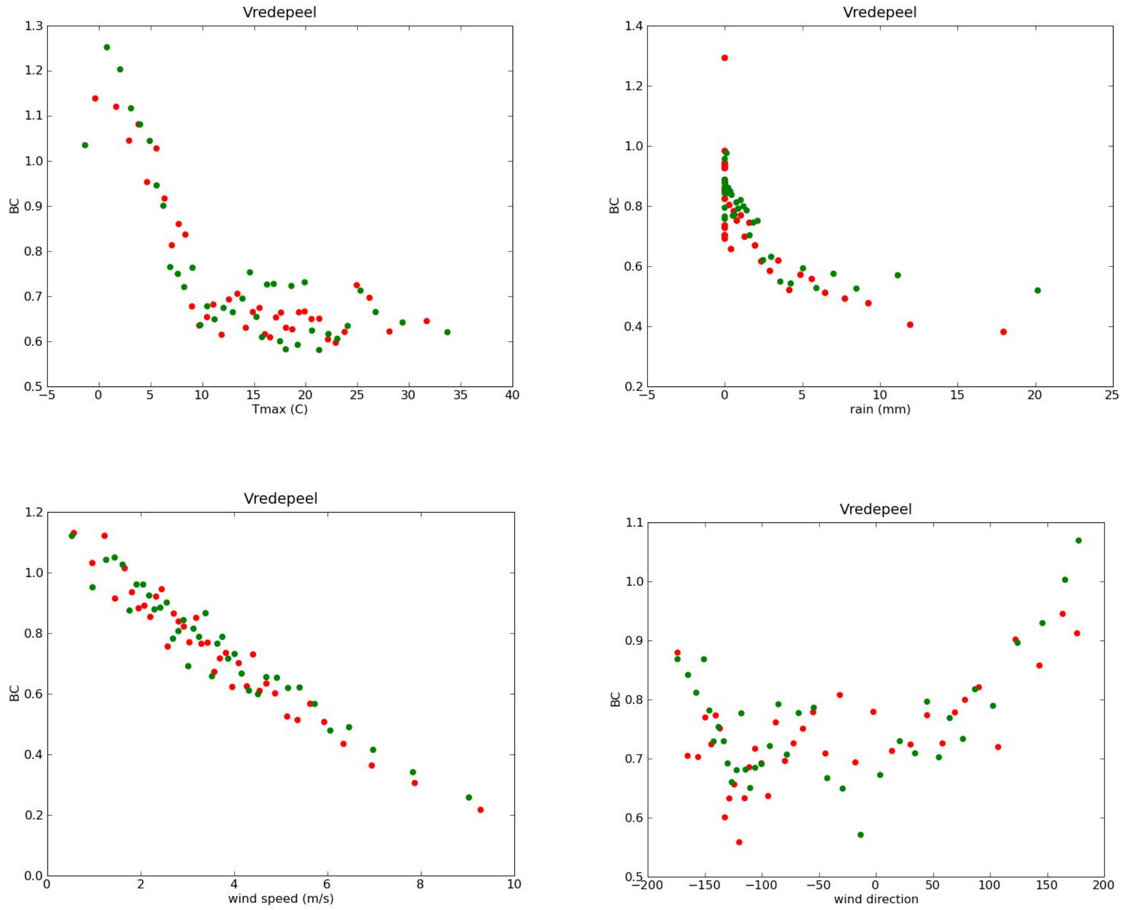

Figure 13. Black carbon concentrations versus meteorological variable, Vredepeel, 2003-2007, binned per 50 data points.

5.3.2 Sea salt aerosol

Sodium concentrations are a tracer for sea salt. They are strongly related to wind speed and direction (Fig.14), since sea salt aerosol is generated by the wind over the sea surface. In the present study LOTOS-EUROS used the parameterization by Monahan et al 1986, but in fact the generation is weakly dependent on sea surface temperature (Mårtensson et al 2003). Modelled concentrations were indeed higher for Kollumerwaard and De Zilk, close to the coast (not shown), than for Vredepeel, and higher for (north) westerly winds and for higher wind speeds than for easterly winds and calm conditions. The temperature dependency in Fig. 14 probably mimics the relationship between the wind direction, wind speed and temperature, rather than a causal relationship between sea salt concentrations and temperature. Sodium concentrations are negatively correlated with rain, as expected, and negatively correlated with the number of consecutive dry days. The latter is probably a reflection of the fact that dry days are often related to continental flows, which contain little sea salt.

Furthermore, sea salt aerosol is relatively coarse so that gravitational settling plays a role, in absence of wet deposition. For sea salt this is far more important than for fine aerosol like black carbon. For 2003-2007 a few observations of sodium were available, indicating a good spatial correlation of annual averages but an overestimation by the model. In a different study it was observed that for the Netherlands, maximum concentrations were reached for southwesterly to westerly winds, rather than for northwesterly winds (Manders et al. 2009). For Ispra and Melpitz sodium concentrations were not studied, since they are far from the sea.

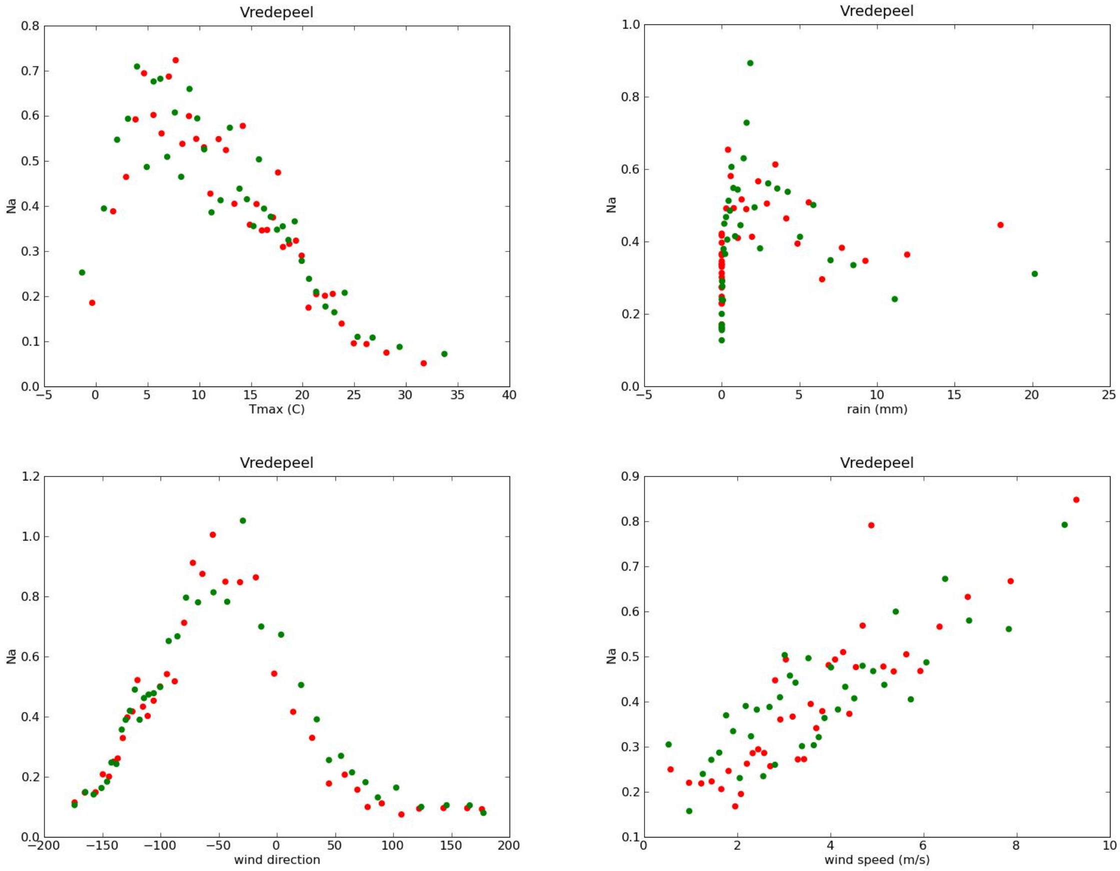

Figure 14. Sodium concentrations versus meteorological variable, Vredepeel, 2003-2007, binned per 50 data points.

5.3.3 Nitrate

Nitrate is a secondary aerosol, resulting from chemical reactions and depending on the presence of other species and meteorology. For Vredepeel, concentrations clearly decrease with wind speed and concentrations are higher for easterly winds (Fig. 15). Differences between modelled and observed concentrations are most clear in the temperature plot. For maximum temperatures of 5-10º C, model and observations agree most. For higher temperatures the modelled concentrations are higher whereas observed concentrations are not, for RACMO this effect is larger, probably due to the use of EQsam instead of Isorropia. For low temperatures, observed concentrations are largest, and the modelled concentrations tend to underestimate the concentration. Concentrations are negatively correlated to rain due to wet deposition, although for run2 the values for large amounts of rain increase again. The poor representation of the temperature dependency of the model is most probably related to the dependency of chemistry on temperature (volatility and reaction rates). Due to the correlation of temperature and wind and of temperature and rain, mismatches are also visible in the scatter plots of wind and rain, but they do not stand out as much. The passive transport and removal processes seemed to be correctly modelled, according to the BC results.

For Melpitz and Ispra the correlation with temperature of the two model runs and observations deviates more strongly than for Vredepeel. Especially for Melpitz there is a strong general underestimation of the model. Melpitz shows a rather constant offset for concentration against wind direction and wind speed, pointing at structural underestimation in the model, and the relationship with temperature is clearly not well represented. For Ispra the relationship with wind speed is clearly nonlinear, the relationship with wind direction is much stronger for the model runs than for the observations.

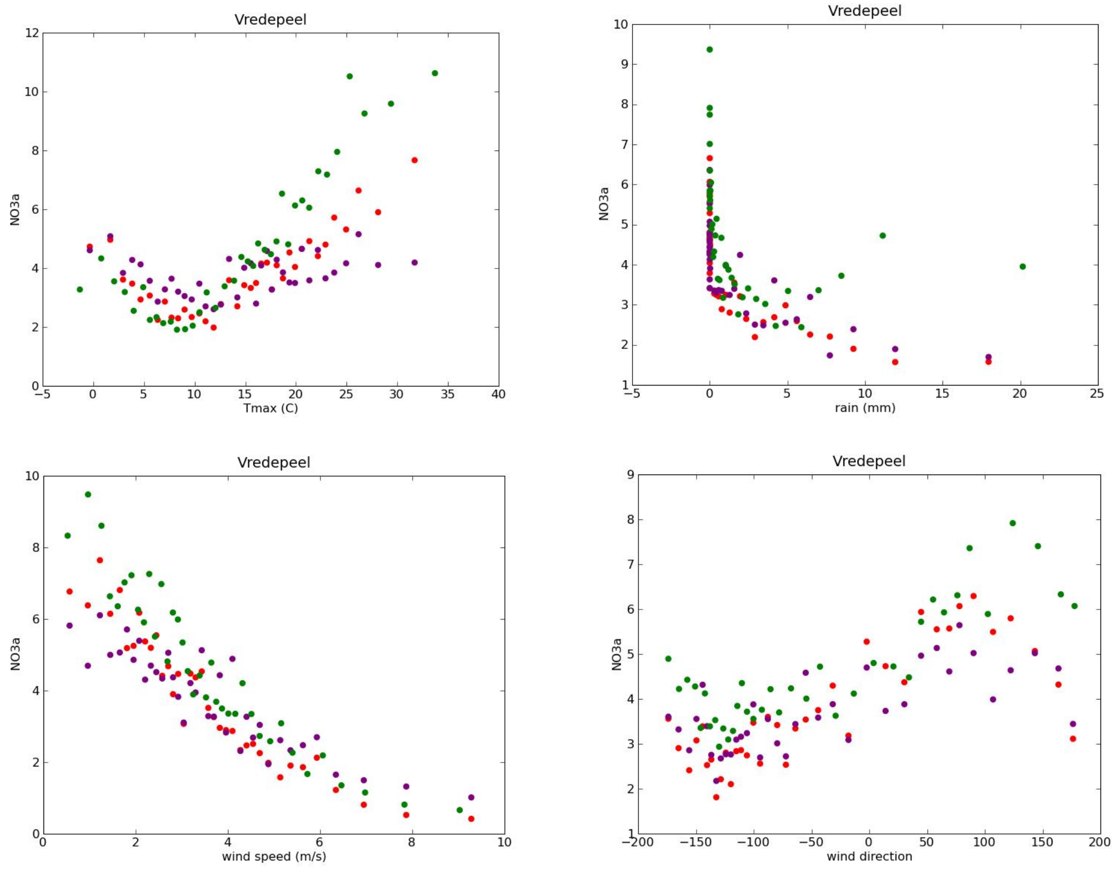

Figure 15. Nitrate concentrations versus meteorological variable, Vredepeel, 2003-2007, binned per 50 data points.

5.3.4 Ammonium

Also ammonium is a secondary aerosol and a clear correlation with temperature can be observed in both model runs and observations (Fig. 16). Again, minimum concentrations are found for temperatures around 10ºC. For lower temperatures, the model runs have the same higher concentrations as the observations, for high temperatures they overestimate the increased concentrations, in particular with EQsam. Concentrations tend to a be a bit higher for (south)easterly winds, as expected since they are associated with both warmer and colder regimes, and are related to lower wind speeds. Concentrations decrease nearly linearly with increasing wind speeds, this decrease levels off for wind speeds larger than 6 m/s. There is no clear relationship with daily maximum temperature, except for very low temperatures for which concentrations are elevated. There is a clear negative correlation with daily rain for the observations and for run1, but less clearly for run2, similar to the behaviour of nitrate. For Melpitz, the correlations of ammonium with wind speed and direction of observations and model runs are similar, but with a general underestimation of the concentrations, stronger for run1 than for run2, probably due to the use of EQsam which gives higher concentrations. There is a linear decrease of concentrations with wind speed, and concentrations are slightly lower for northwesterly winds than for southeasterly winds. But for temperature differences between the runs are large again, with an underestimation of concentrations for low temperatures and an overestimation for high temperatures. For Ispra results are very scattered, with a tendency to mimic the results for nitrate.

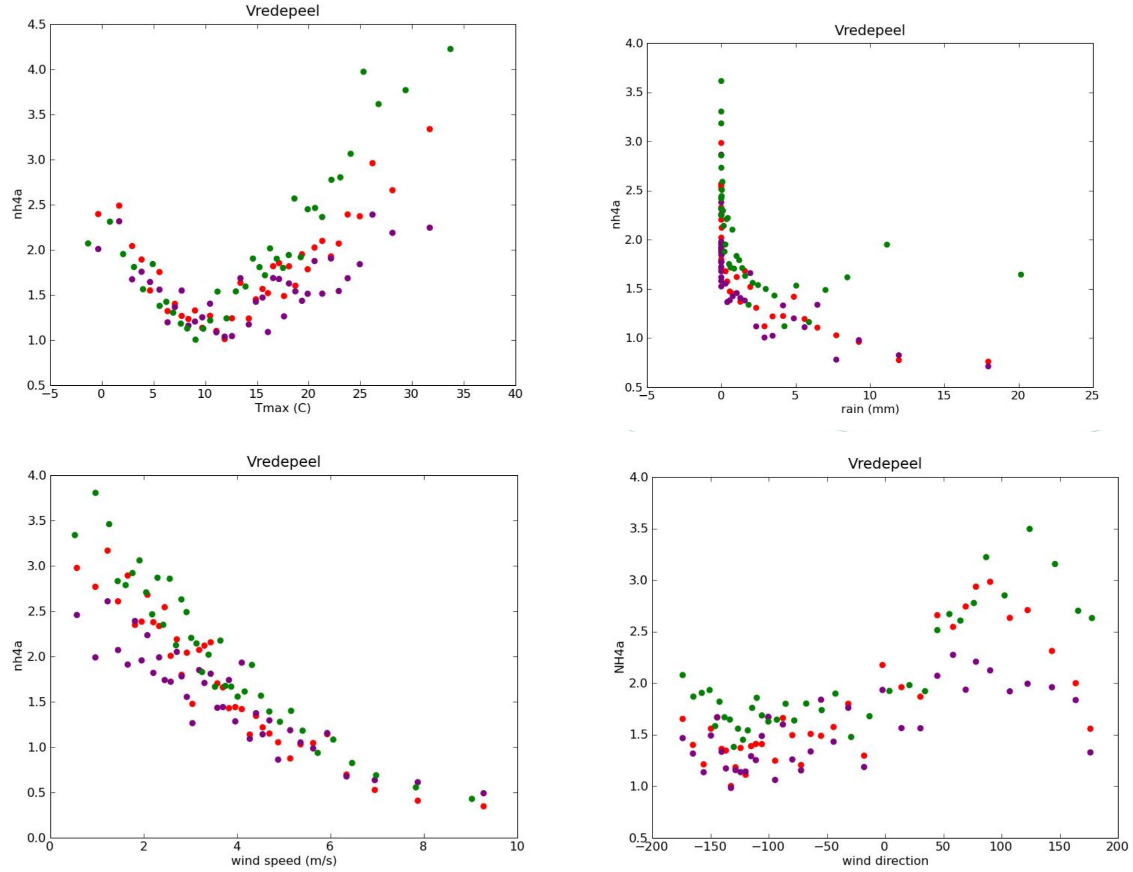

Figure 16. Ammonium concentrations versus meteorological variable, Vredepeel, 2003-2007, binned per 50 data points.

5.3.5 Sulfate

Also sulfate is a secondary aerosol, but for this species the difference between the two model runs is smaller than for nitrate and ammonium. Sulfate concentrations are clearly underestimated by the model for Vredepeel. There is a negative correlation with wind speed in model results and observations, as expected, but the dependency of wind direction is much stronger in the model than in the observations. Concentrations tend to be higher for for easterly winds. This could be the result of many causes, like too strong local sources east of Vredepeel or too weak sources in the west, but also be caused by the relationship between easterly winds and warm and cold days. The correlation between temperature and concentration for run1 and run2 represent the observed concentration better than for ammonium and nitrate, albeit with a structural underestimation of the concentrations. For sulfate, the underestimation in summer is larger than in winter, in contrast to the behaviour of modelled nitrate and ammonium. For Melpitz and Ispra, modelled and observed sulfate show the same dependency on rain and wind and nearly the same dependency on temperature as observations, but absolute values are seriously underestimated.

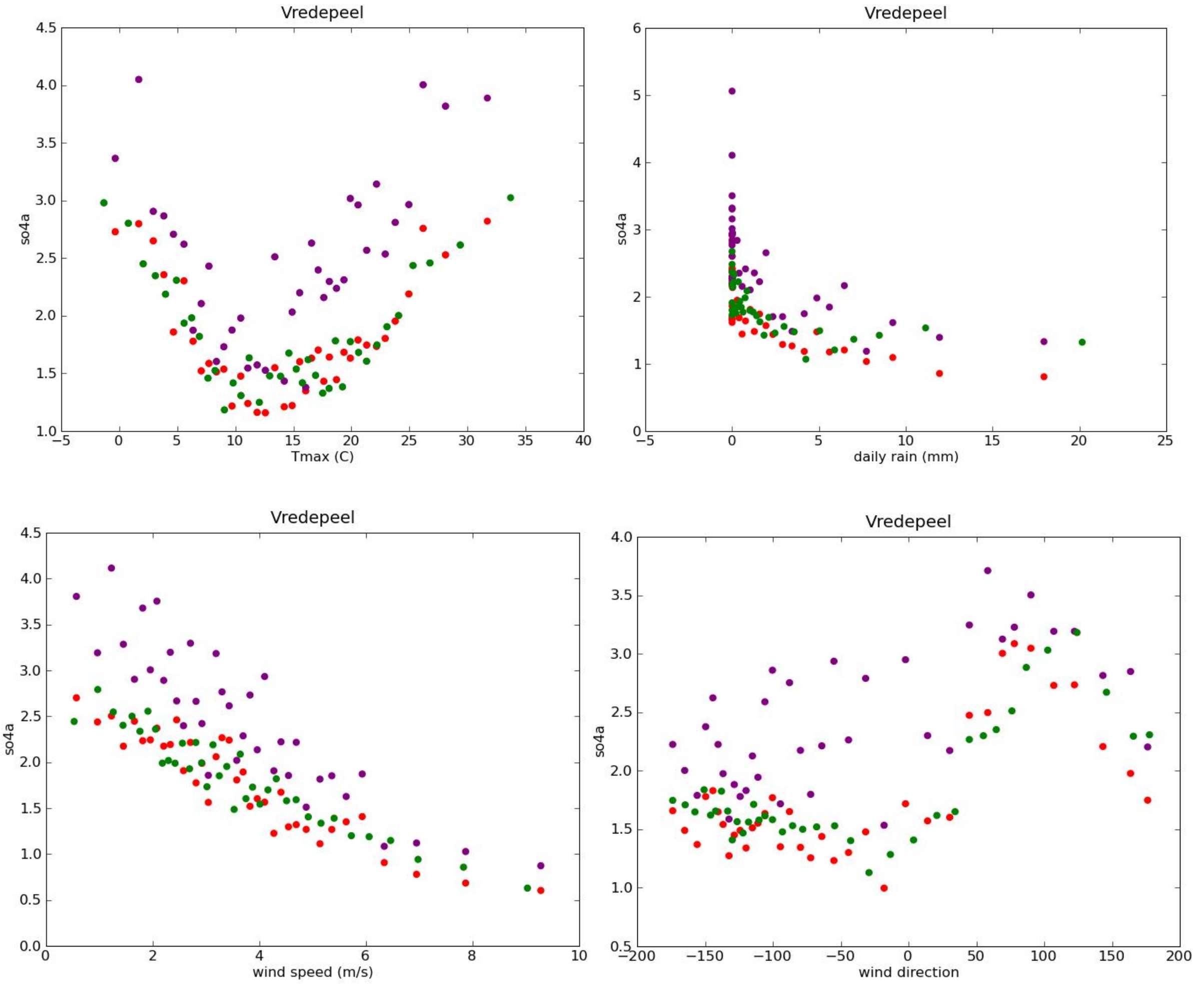

Figure 17. Sulfate concentrations versus meteorological variable, Vredepeel, 2003-2007, binned per 50 data points.

The results above show that LOTOS-EUROS on average represents the main dependencies of concentration on daily maximum temperature, daily average wind direction and wind speed. This means that the meteorological parameters that we have selected are indeed useful, and that the model is sensitive to them. However, several biases were identified:

- an underestimation of ozone for temperatures > 20°C, which is slightly less prominent for RACMO-ERA meteorology with ECMWF meteorology.
- a general tendency to underestimation of the secondary inorganic aerosol concentrations, the mismatch depends on the location.
- the temperature dependency of nitrate and ammonium is not modelled correctly. For low temperatures LOTOS-EUROS tends to underestimate the concentrations, with EQsam the sensitivity to temperature is lower than with Isorropia. For high temperatures, LOTOS-EUROS gives a too high sensitivity to temperature, for EQsam even more than with Isorropia.

For black carbon and sea salt the model could not be validated because of a lack of observations.

6 Results from climate run: 2003-2007

For the climate run, the ECHAM53r A1B transient scenario was used to drive RACMO at the boundaries. This climate model has biases as compared to the observed climate, which add to the biases of RACMO – LOTOS-EUROS forced by ERA-interim. As a first indication, the years 2003-2007 of the RACMO runs forced by ERA-interim and by ECHAM5r3 A1B have been analysed. This period is too short for a solid analysis, due to the interannual variability. Furthermore, the free-running ECHAM5r3 has no correlation with observed climate. This implies that it is possible that the selected period does not contain years with very high temperatures like the true realisation of 2003 and 2006. Therefore the presented results only serve as examples for our approach but they will illustrate the kind of biases that may occur. The two runs are by no means correlated, so the 5 years studied here are not connected and we might as well have chosen to compare a different 5-year period of the ECHAM5r3 run.
First, the RACMO downscaling of this climate model is compared to RACMO forced by ERA-interim. Then the impact on air quality is shown.

6.1 Meteorology

For 2003-2007, clear differences with run2 can be found. The distributions of wind, temperature and rain for Vredepeel (Fig. 18) and the relationship between the meteorological variables (Fig. 19) show that wind speeds for run3 are higher for the southwesterly and northeasterly winds, the wind directions for which the highest wind speeds occur. Furthermore, precipitation has increased for westerly winds and moderate temperatures. There daily maximum temperatures are somewhat lower, with more cold days and less warm days. This gives the picture that in the ECHAM5r3 run there are more episodes with westerly flows, bringing frontal systems with lower temperatures, higher wind speeds and more rain.

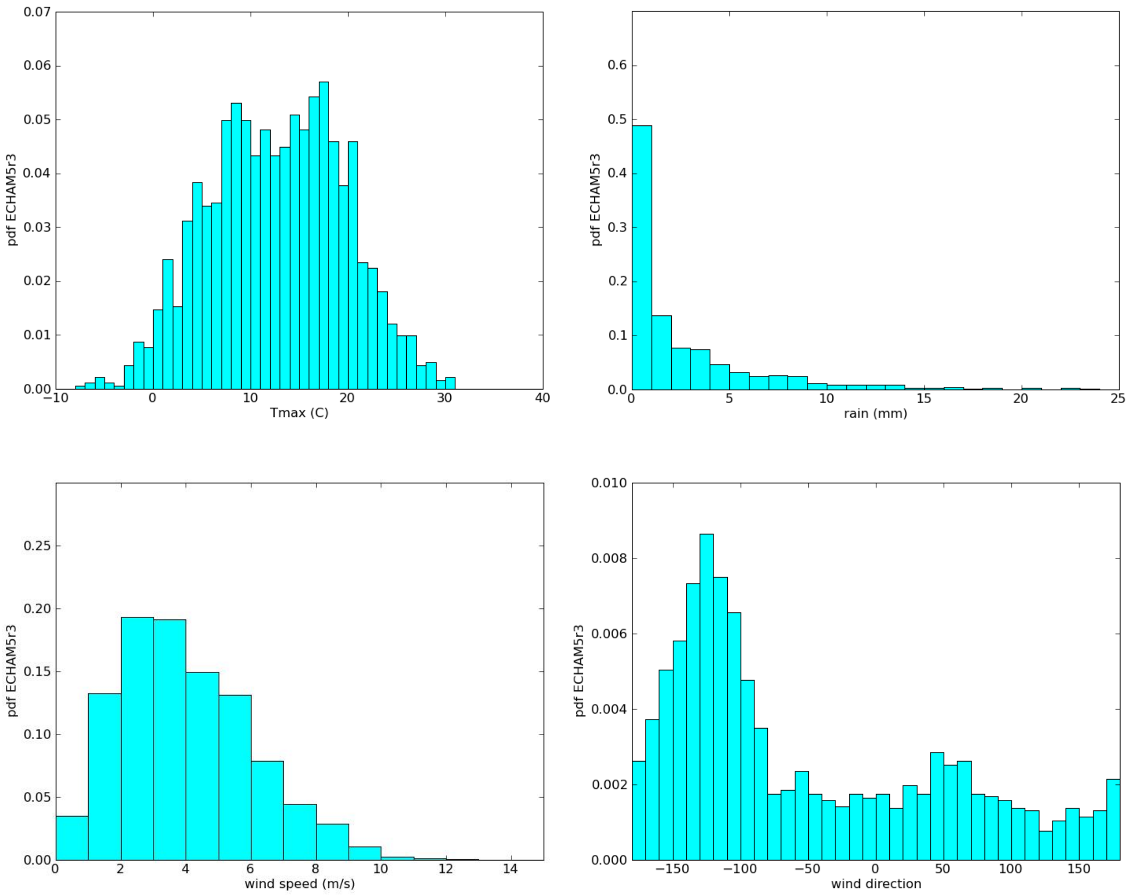

Figure 18. Probablility density plots for Vredepeel, 2003-2007, RACMO with ECHAM5r3 forcing.

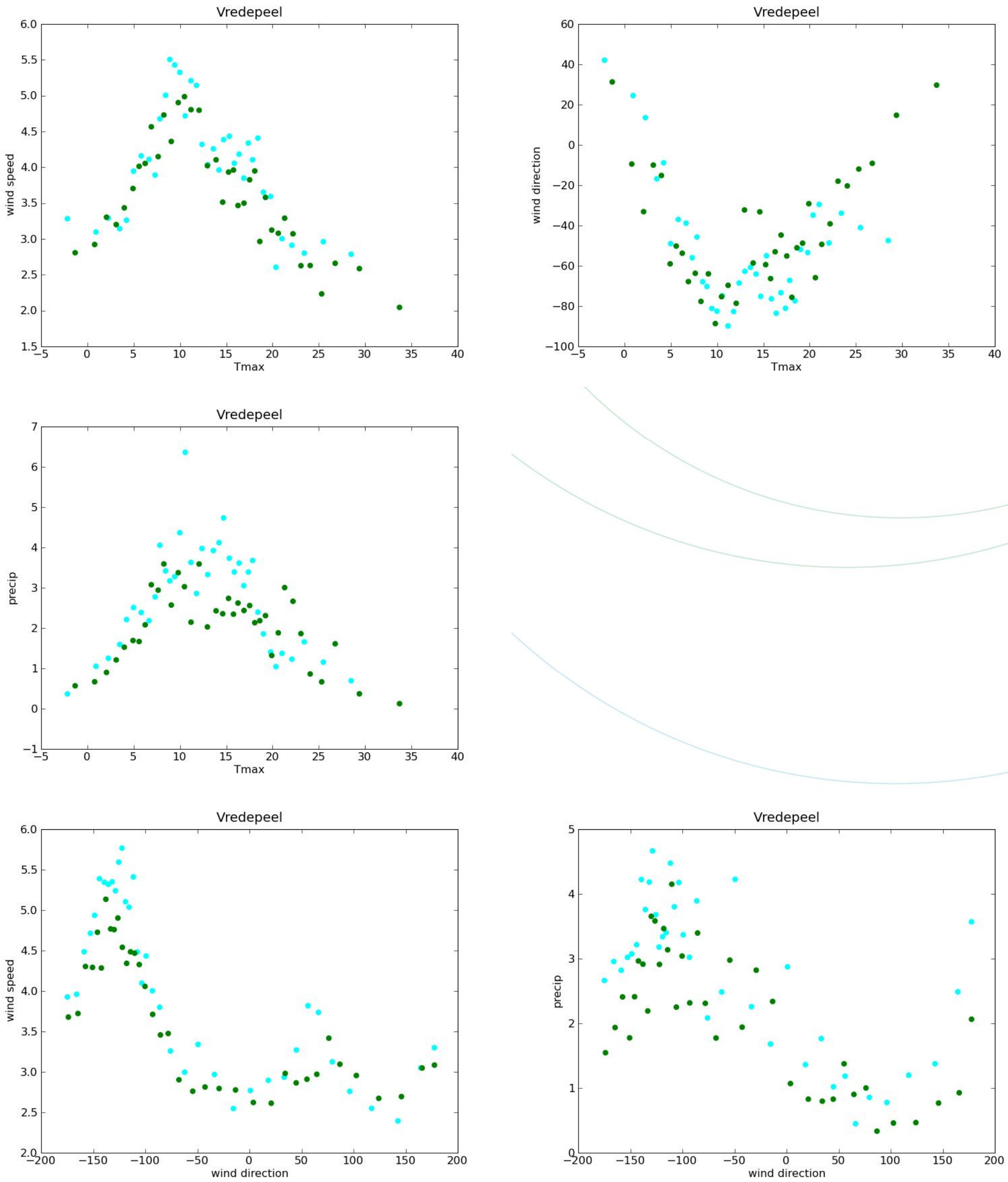

Figure 19. Scatter plots of meteorological data for 2003-2007, RACMO_ERA (green) and RACMO_ECHAM5r3 (cyan), binned per 50 data points.

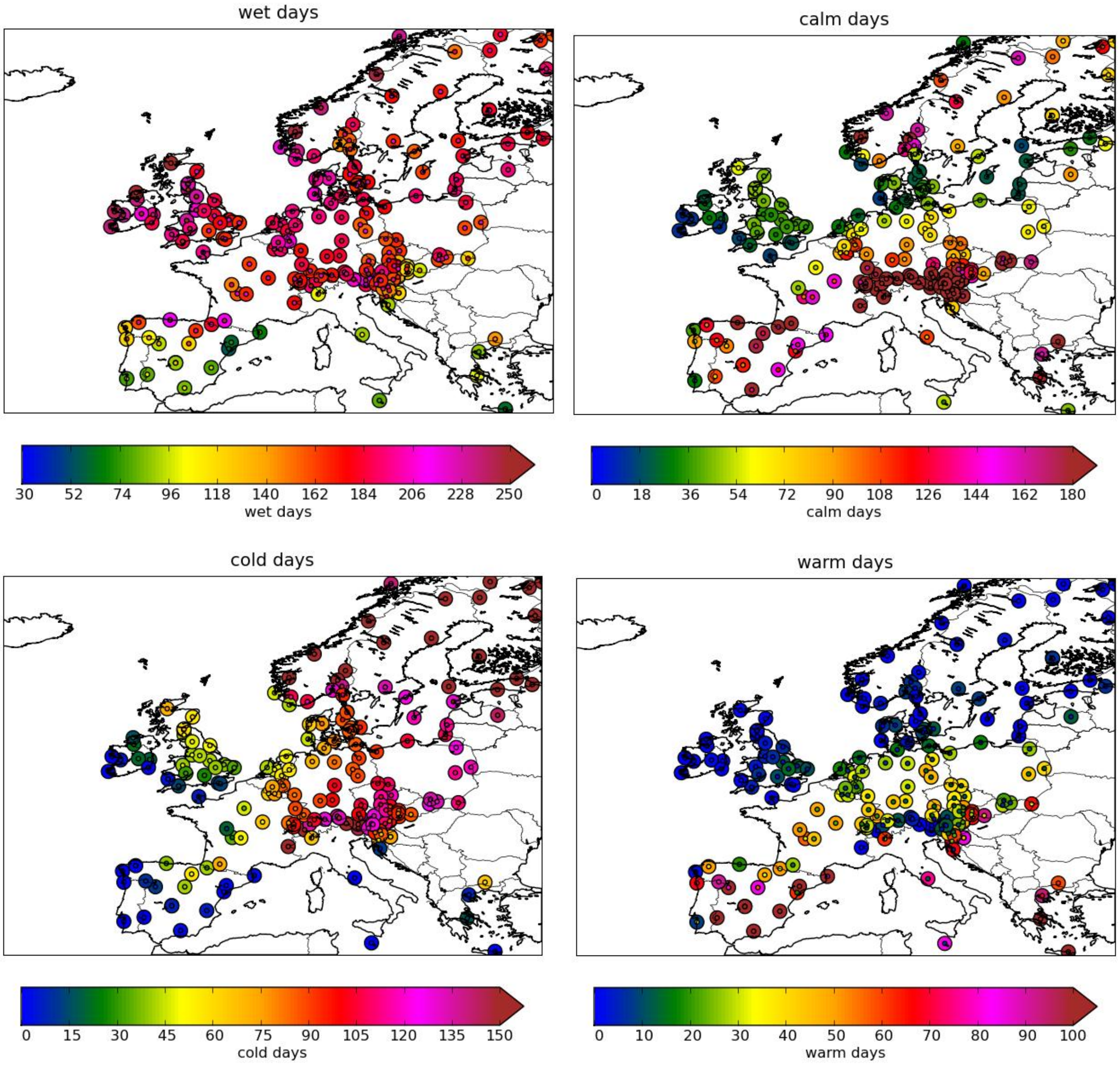

Figure 20. Average number of wet days (upper left), calm days (upper right , cold days (lower left) and warm days (lower right) per year, 2003-2007, ECHAM5r3 forcing (small circles) and ERA-interim forcing (large circles).

Also when looking at other locations (Fig. 20), clear differences can be found. The climate run is wetter, has higher wind speeds, slightly more cold days and less warm days. Note that the exact numbers of warm and cold days are very sensitive to the choice of the threshold value. Also the average maximum temperature, wind speed and rain differ considerably from ECMWF and RACMO-ERA meteorology (Table 4). ECHAM5r3 is colder, wetter and has higher wind speeds for nearly all locations. Differences between RACMO-ERA and RACMO-ECHAM5r3 are larger than between RACMO-ERA and ECMWF. The present analysis indicates that there is a bias current climate as represented by RACMO with ECHAM5r3 boundary conditions as compared to RACMO with ERA-interim boundary conditions. But the analysed period is too short to draw final conclusions.

Table 4 RACMO_ECHAM5, averages for 2003-2007

station	av Tmax ºC	av std Tmax ºC	average yearly rain (mm)	av. stdev daily rain (mm)	average v (m/s)	av. stdev v (m/s)
Vredepeel	12.7	6.8	1007.8	4.0	4.0	2.0
Kollumerwaard	11.7	6.1	996.0	3.8	5.7	2.6
De Zilk	12.3	6.1	1009.7	4.0	5.7	2.7
Eibergen	12.4	6.8	1098.1	4.3	4.2	2.0
Illmitz	13.0	8.7	836.4	4.9	2.7	1.5
Melpitz	12.1	7.9	763.5	3.3	3.9	2.0
Ispra	13.2	8.0	1545.4	11.4	1.5	1.0
Els Torms	21.4	7.9	314.5	3.1	2.9	1.8
O Savinao	16.3	6.5	1768.1	8.1	3.6	2.2

6.2 Air quality

To get a general impression of the changes in air quality, long-time (2003-2007) averages of summer ozone and PM10 for run 2 and run 3 were compared. For summer daily maximum ozone concentrations (April-September, Fig. 21), average concentrations in the free climate run are considerably lower than for run 2 with ERA boundaries for large parts of western Europe. For some locations differences are more than 10%. Only in the southeast of the domain concentrations are higher than in run 2. The concentration differences are most probably the result of the lower daily maximum temperatures in the ECHAM5r3 over most of Europe, since ozone is very sensitive to temperature, as demonstrated in Section 5.

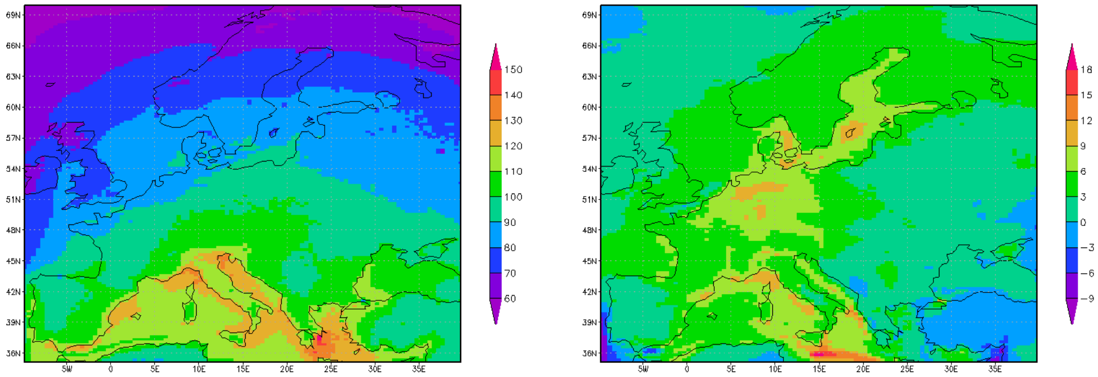

Figure 21. Modelled daily ozone maxima, averaged for April-September , 2003-2007. Left: ozone maxima, run3, right: difference plot of averages summer daily maximum 2003-2007, run2-run3. Concentrations are in µg/m^3.

Also for PM10 (Fig. 22) concentrations in run3 tend to be lower for most of Western Europe for latitudes below 55º. Differences are largest for regions with high PM concentrations: large cities (Paris, Madrid) and the Po valley. Although the absolute difference looks rather uniform, the fractional difference is not: in Spain concentrations are much smaller than in the Netherlands, but the absolute difference is comparable.

The various components of PM10 were investigated, since they do not all react the same on changes in meteorology (Fig. 23). The concentrations of secondary inorganic aerosols has decreased in the climate run, which could be an effect of the colder summers, the warmer winters, enhanced precipitation or increased ventilation. Probably all factors play a role. For sea salt, concentrations have increased slightly above large parts of the Atlantic and North Sea, resulting in a slight increase in coastal areas. Especially above the Mediterranean the increase is large.

This is most probably related to increased wind speeds, which have a direct impact on sea salt emissions. Primary PM2.5, has decreased in the areas of high concentrations (cities, industrial areas), whereas for primary PM10 the difference between the two runs is close to zero.

As a further check, for a few species the relationship with temperature, wind and rain was plotted (Fig 24). The general behaviour closely resembles to the relationships for the run using ERA-interim forcing, with minor differences that must be ascribed to the differences in realized meteorology. These differences are most clearly visible for the passive tracer black carbon.

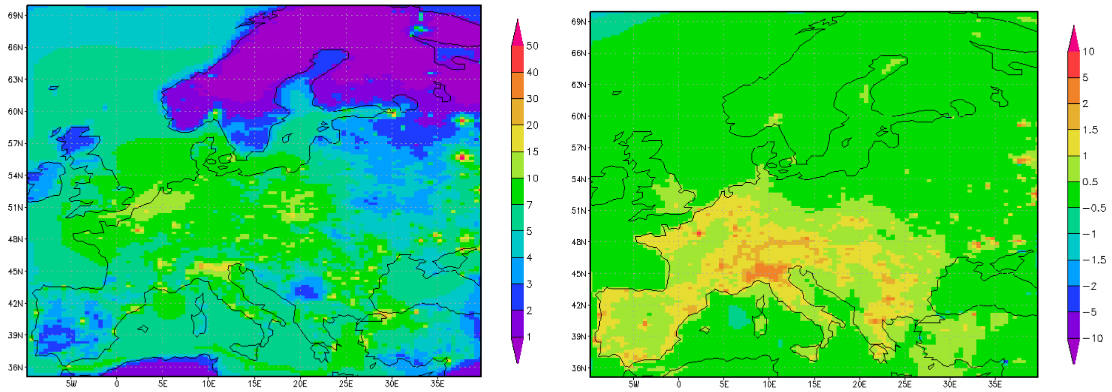

Figure 22. Modelled total PM10. Left: average concentrations for 2003-2007, run3. Right: difference plot of averages 2003-2007, run2-run3. Concentrations are in µg/m^3.

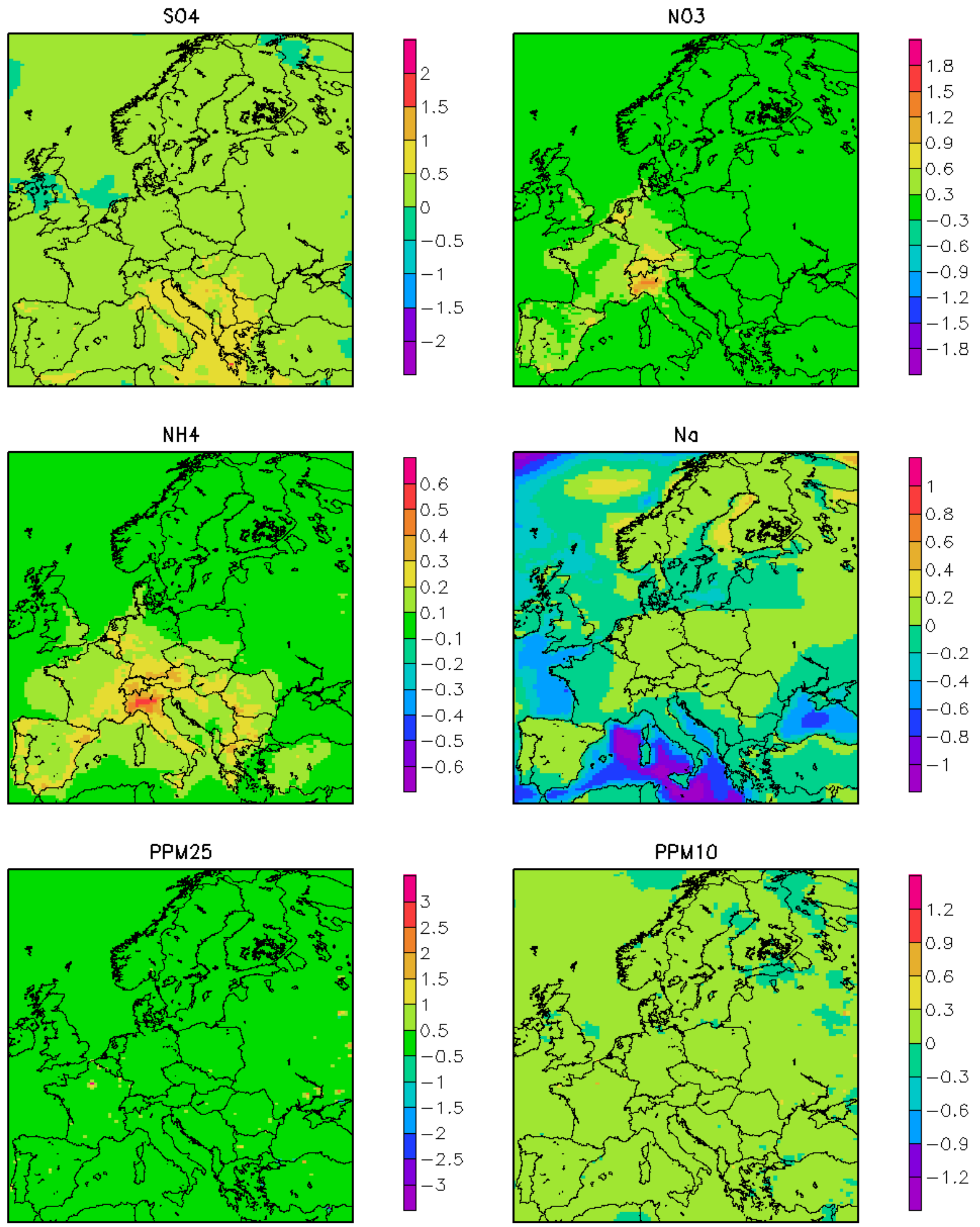

Figure 23. Differences between averages in species belonging to PM10 2003-2007, run2-run3. Concentrations are in µg/m^3.

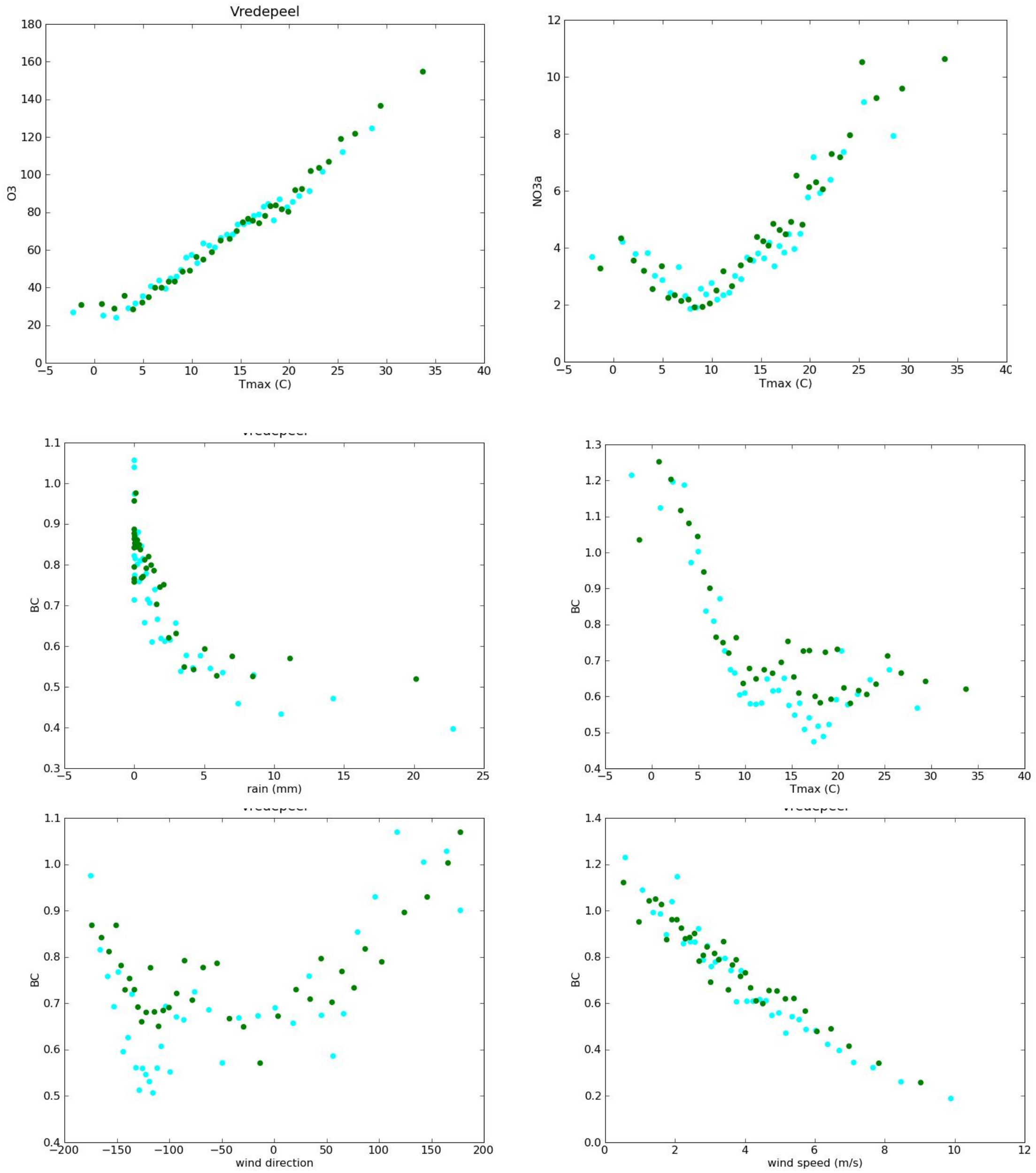

Figure 24. Modelled results for station Vredepeel, 2003-2007. Green: run2, cyan: run 2. Dependency of ozone, nitrate and black carbon on temperature, and the dependenc of black carbon on rain, wind direction and wind speed.

7 Discussion

First of all, it is clear that the coupled system RACMO-LOTOS-EUROS works. The report shows that it is feasible to use the system with a one-way coupling in a parallellized version for a long climate integration. The full run of 1970-2060 of RACMO and LOTOS-EUROS took about 45 days on 32 processors at the ECMWF computing facility. This is acceptable for present standards for the desired application.
The RACMO meteorology was compared with ECMWF analysis fields instead of comparing with observations. This has the disadvantage that biases in the ECMWF analysis and their effect on the air quality modelling cannot be taken into account. But on the other hand, including all available meteorological data from different sources, which may be contradictory at some point, is far beyond the scope of the present research. ECMWF analysis fields are thoroughly validated and may be considered a solid basis. The runs were compared in terms of daily maximum temperature, daily rain, daily average wind speed and daily average wind direction. These variables were chosen for their relevance for air quality. Indeed, average relationships between observed concentrations and these meteorological variables were found, although with a considerable scatter. The episode length (warm period, dry period) did not give a clear relationship with air quality and was not used further. Convective and large-scale precipitation can be separated, but this was not done at this stage, since for air quality only total precipitation is relevant, although for the pure comparison of the difference in meteorology it would be interesting. Also boundary layer height is important, but since it is related to temperature and wind it was not treated separately, although it would be useful to verify whether RACMO reproduces the ECMWF analysis boundary layer height.

From the present comparison for 2003-2007 it follows that for a run with ERA-interim boundary forcing, the correlation of daily maximum temperature and wind with ECMWF analysed temperature and wind are very good. Slightly higher maximum temperatures were found, resulting in somewhat more warm days. Only for rain the correlation was poor, with comparable annual totals of rain but more days with rain in RACMO. This poor correlation was to be expected, since rain is known to be difficult to predict and a rather localized event which is discrete in time, so that small mismatches lead to a poor correlation. Overall, one must conclude that RACMO reproduces the ECMWF meteorology rather well and can therefore be used with confidence for the downscaling of a global model for the purpose of driving an air quality model. In a next study, the analysis can be further refined by separating the different seasons.

The next step was to drive LOTOS-EUROS with the RACMO meteorology. Results were compared with a LOTOS-EUROS run with ECMWF meteorology and with observations from the EMEP network. Differences between the model runs are partly caused by the small differences in meteorology, leading to differences ozone concentrations on warm days, but to a larger extent by differences in the heterogeneous chemistry (EQsam versus Isorropia), leading to higher nitrate and ammonium concentrations for high temperatures, most notably in areas which are already characterised by high nitrate and ammonium concentrations. This becomes also clear when comparing the concentrations with observations, separating the effects of temperature, wind and rain. This comparison reveals clear biases, with underestimations of ozone for warm conditions (T>20ºC), underestimates of nitrate and ammonium for cold weather and overestimations for warm weather. For the secondary aerosols a structural tendency to underestimate is found when taking other stations into account (Melpitz). For primary PM, it was difficult to validate the results with observations. When the relationship of concentrations with meteorological variables is identical to observations but with a constant bias, this implies that the emissions are too low. For other cases it is less clear. There are strong indications that the main biases are caused by the impact of temperature on the chemistry (ozone, nitrate, ammonium) which is not modelled well for warm and cold days, and in part also by underestimations in emissions (SO2-sulfate).

Overall, LOTOS-EUROS underestimated the PM10 concentrations. Apart from the points raised above, there are sources that are not part of the model, notably dust and secondary organic aerosol, and forest fires, which are more event-driven. These are highly relevant for climate studies, since hotter and dryer conditions will lead to more dust and fire emissions, whereas for SOA the sign of the change is totally uncertain. Despite the biases and uncertainties, we feel that the model represents the sensitivity of air quality to meteorology on average good enough to use LOTOS-EUROS for the study of the impact of climate change on air quality. However, the biases must be taken into account when the climate run for 2050 is analysed, since in particular for warm and cold days a poor air quality is found.

When RACMO is driven by ECHAM5, scenario A1B, one of the more realistic IPCC scenarios, additional biases occur in the meteorology, which have their impact on the modelled air quality. These biases must stem mainly from the biases in the global model ECHAM5, since we have seen that RACMO reproduces the ECWMF analysis rather closely when forced by ERA-interim. On the positive side, the biases in the downscaled ECHAM meteorology imply that RACMO can indeed be used to downscale a climate scenario run. For the five-year averages of 2003-2007, it has been illustrated that there are less hot days, less calm days and more days with rain, so that air pollutant concentrations were on average lower than for the run representing the real climate. Because of the interannual variability, these five years, which are not physically linked to the true climate 2003-2007, may be not representative and a longer period is needed for intercomparison. The model simulations are there and a period of 20 years will be analysed in a follow-up of the present project. When biases are found for this longer period, they will be taken into account in the analysis of the future climate.

We have only compared with observations for the period 2003-2007 for several reasons. One reason is that this study focuses on modelling the effect of changes in meteorology. This implies that the emissions should be taken constant. When it comes to model uncertainly, emissions are formally not part of the model, but uncertainties in the emissions, both anthropogenic and biogenic, will contribute to uncertainties of the modelled concentrations. Over the period 2003-2007 the emissions did not change drastically (MACC project, emissions 2003-2007 available) although some changes are present which contribute to differences in observed and modelled concentrations. When comparing with observed concentrations, the emissions should be more or less correct to identify biases in the model itself. Even when the annual totals are correct, still the emissions in the model are not perfect since the annual totals are translated to hourly emissions by generalised relationships for time of the day and week and temperature, leading to small mismatches, but this is the best one can get for the present purpose. When going back further in time, the emissions for 2005 will become less and less representative, due to differences in technology, changes in locations of industry and plants etcetera. The other reason to compare with 2003-2007 is that not all EMEP stations have been using the same monitors and locations further back in time and have sampled the same species, so that comparison with observations becomes problematic. Also changes in land use may play a role.

In a follow-up of the present study (KvK 2nd Tranche), the climate run with ECHAM A1B scenario boundary forcing will be analysed for 1989-2009, to identify biases in meteorology, and for 2039-20 59 to study the effect of climate change. In the whole run, emissies are kept constant. Runs with different emission scenarios, both anthropogenic and natural (additional NOx due to flashes, additional mixing in of stratospheric ozone), and land use may follow. In addition, it is intended to establish a two-way coupling between RACMO and LOTOS-EUROS, so that RACMO can use aerosol fields modelled by LOTOS-EUROS to model their effect on cloud formation and radiation. Ultimately, the interaction between air quality and climate is taken into account, although there is still a long way to go (e.g. Korsholm, 2009). First steps towards a two-way coupling of LOTOS-EUROS and RACMO have been taken.

8 Conclusions and outlook

The one-way coupling between RACMO and LOTOS-EUROS works and after parallelization of LOTOS-EUROS and technical implementation of scripts to run the two models together, a climate run of the coupled system was feasible. At the ECMWF computing facility, this run was done for the period 1970 to 2060. The run took about 45 days on 32 processors which is reasonabe for the present purpose.

In general, RACMO seems to be fit to downscale global meteorological fields for the purpose of air quality modelling. A comparison with ECMWF analysis fields for 2003-2007 revealed that RACMO forced by ERA-interim reproduces the ECMWF analysis fields well except for rain. For daily average wind speed and direction and for daily maximum temperature good correlations were found, and RACMO was up to few degrees warmer on the warmest days. The global circulation model ECHAM5, scenario A1B, has biases which are downscaled by RACMO, implying that these biases are fed into LOTOS-EUROS and have an impact on the modelled air quality. For the investigated period, the modelled present-day climate was less warm, had less calm days and more precipitation. However, these conclusions are preliminary, since a five-year period is too short to form a solid basis to quantify biases.

LOTOS-EUROS itself models the air quality fairly well in the set-up with ECMWF and RACMO-ERA-interim, with slightly higher ozone, ammonium and nitrate concentrations when RACMO and EQsam were used. For 2003-2007, the modelled concentrations were compared with EMEP observations. Ozone, ammonium and nitrate are strongly dependent on temperature. For very warm days, ozone is underestimated. In contrast, ammonium and nitrate tend to be overestimated for warm days, whereas they are underestimated for cold days. Biases may originate in part by uncertainties in the emissions, but are probably to a large extent caused by the parameterization of the chemistry in the model.

In general, the sensitivity of the model to changes in meteorology is good enough to use the model for a study on the impact of climate change to air quality, although great care should be taken to account for biases. When LOTOS-EUROS is driven by RACMO with the ECHAM5r3 A1B scenario, the biases in temperature, rain and wind, lower concentrations of ozone and PM were found, which is consistent with the identified relationships between modelled (and observed) concentrations and the meteorological variables. The present analysis will be extended to both 1989-2009, to identify biases on a more solid basis, and to 2039-2059, to study the impact of climate change. This will be part of a follow-up of the project (KvK 2nd tranche).

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Knowledge
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Appendix A Meteorology for Ispra and Melpitz

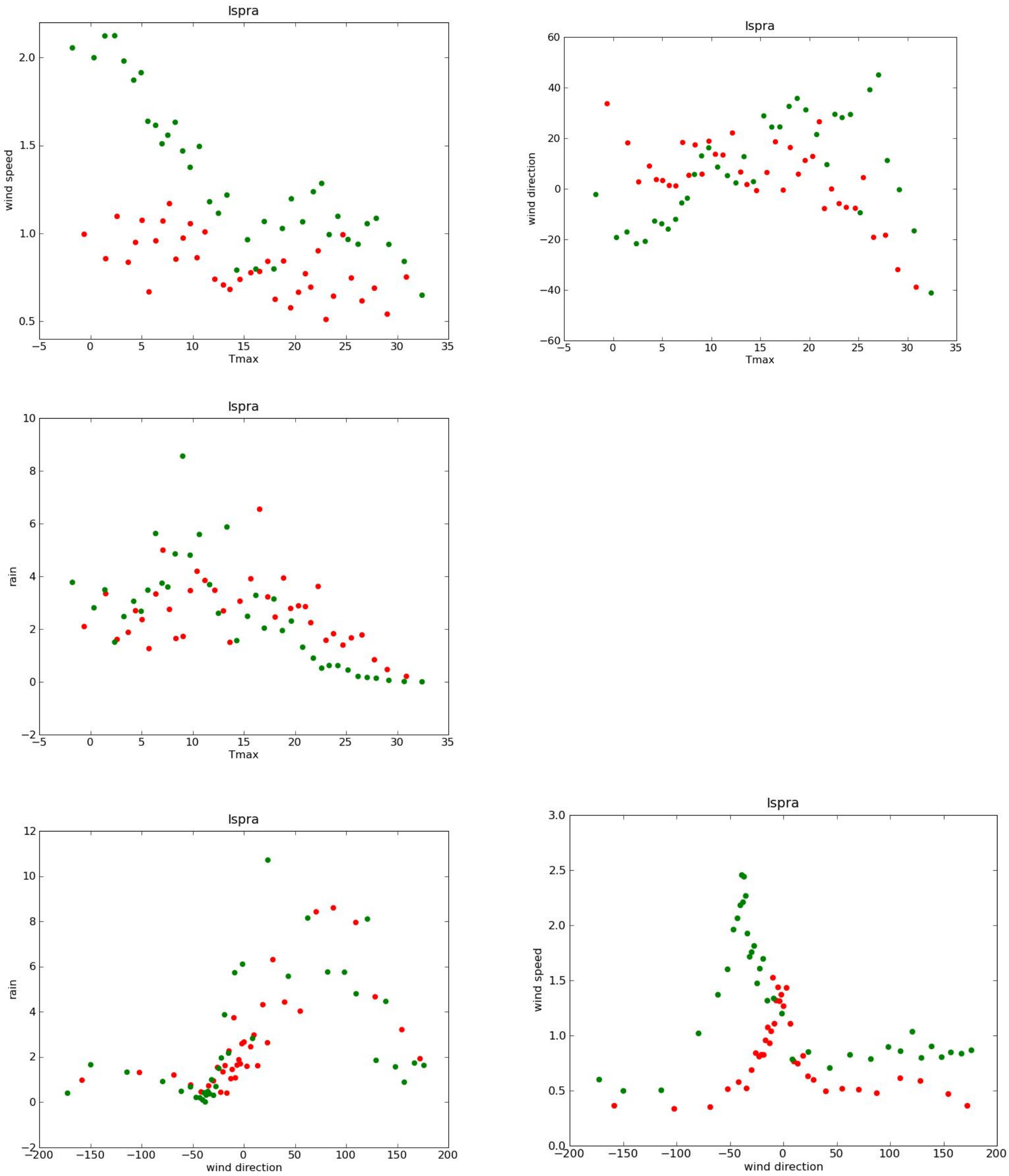

Figure A1 Scatter plots of sorted and binned meteorological data for 2003-2007, ECMWF (red) and RACMO (green) for Ispra, an Italian station in between the Alps and the Po valley

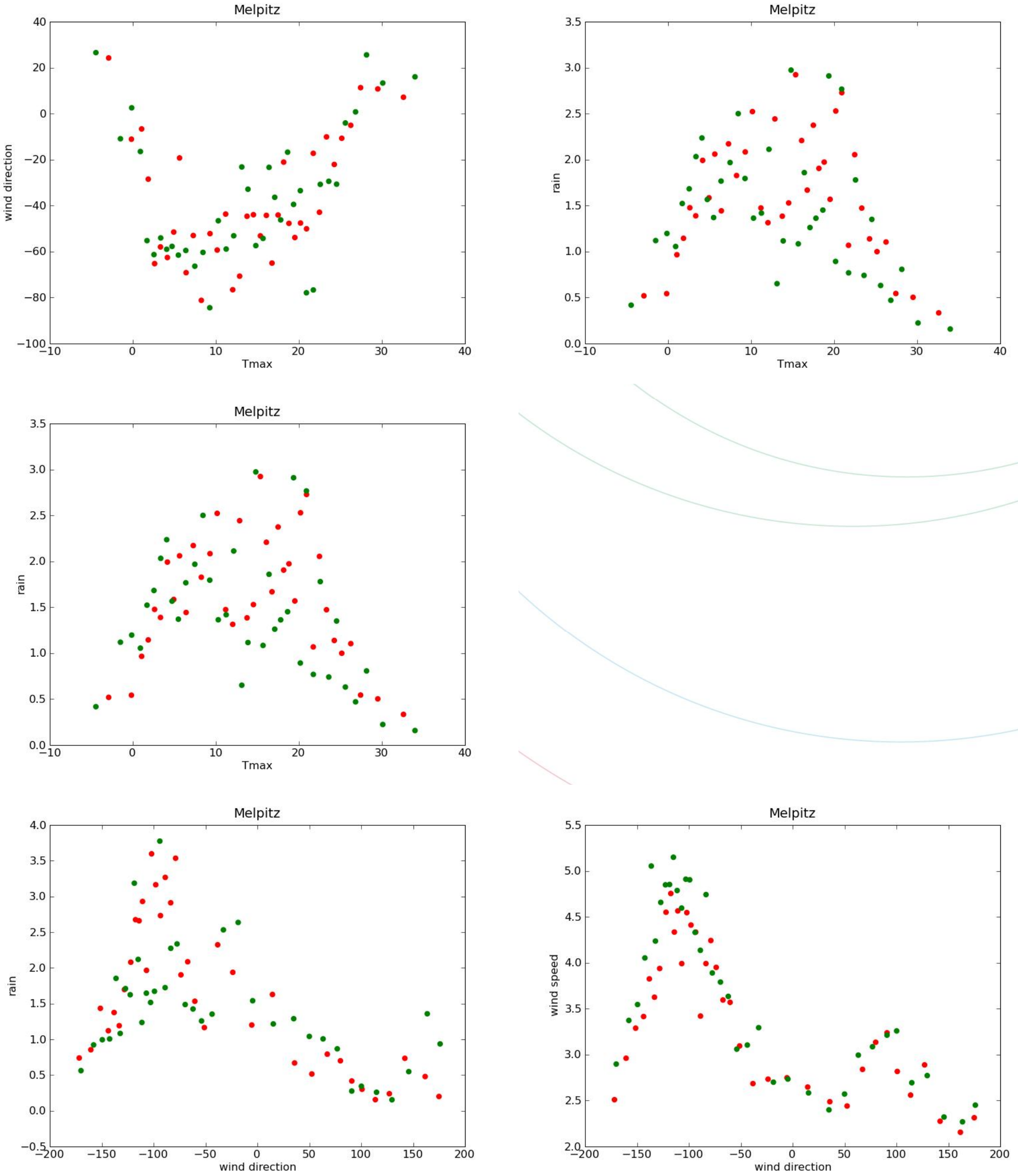

Figure A2 Scatter plots of sorted and binned meteorological data for 2003-2007, ECMWF (red) and RACMO (green) for Melpitz, an inland German station.

Appendix B Modelled concentrations for Ispra and Melpitz

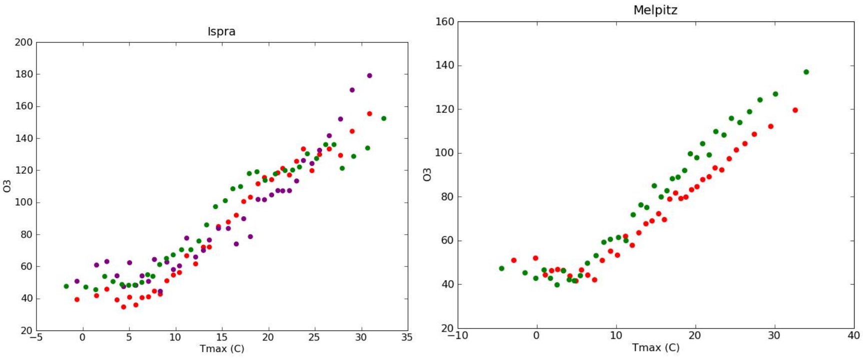

Figure B1. Modelled and observed ozone daily maximum concentrations, 2003-2007, binned per 50 datapoints.

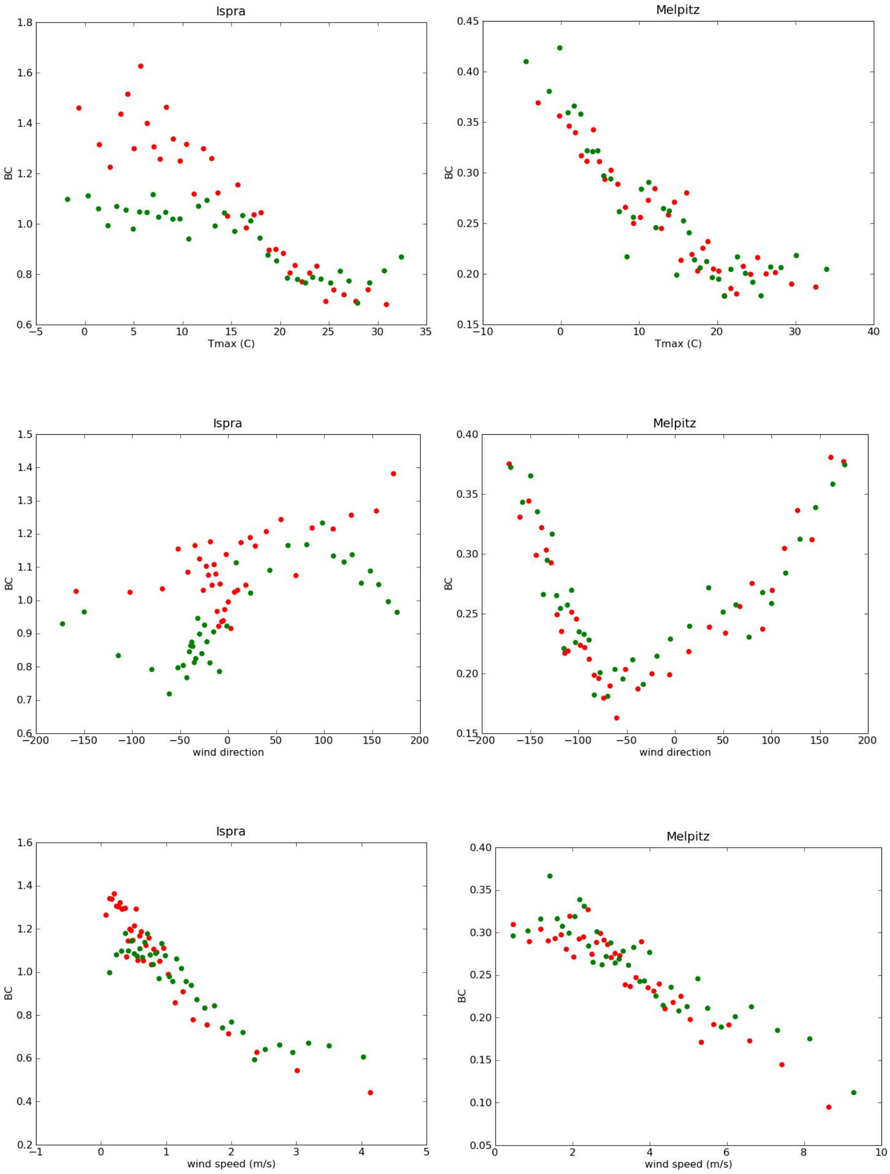

Figure B2. Modelled and observed daily average black carbon concentrations, 2003-2007, binned per 50 datapoints.

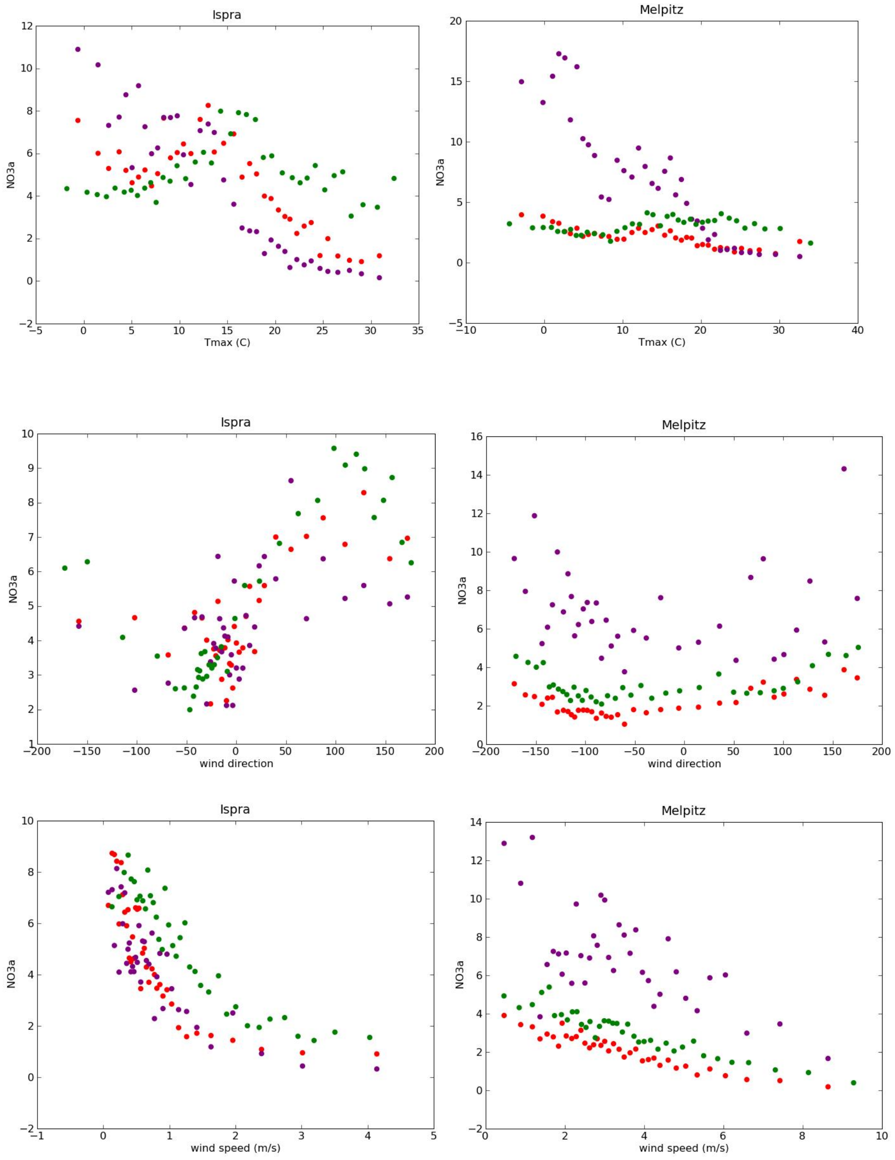

Figure B3. Modelled and observed daily average nitrate concentrations, 2003-2007, binned per 50 datapoints.

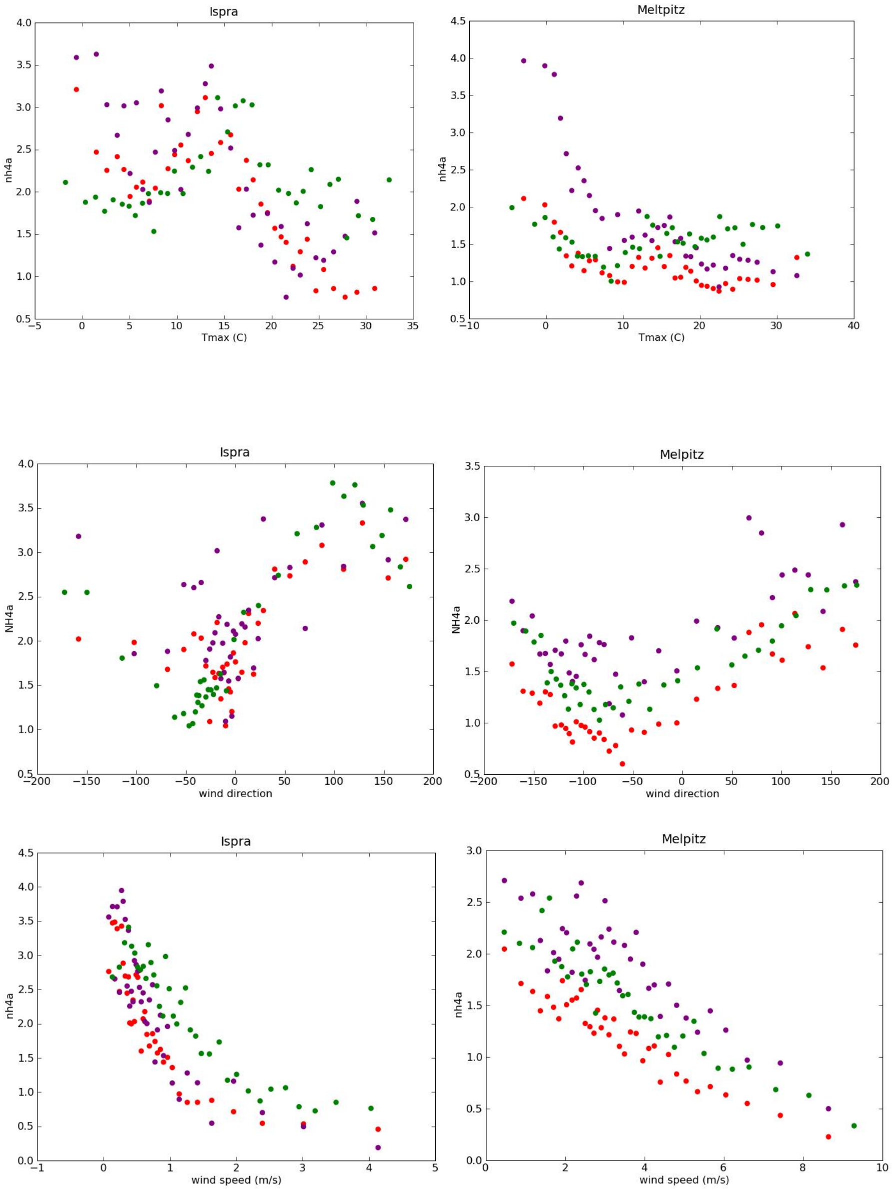

Figure B4. Modelled and observed daily average ammonium concentrations, 2003-2007, binned per 50 datapoints.

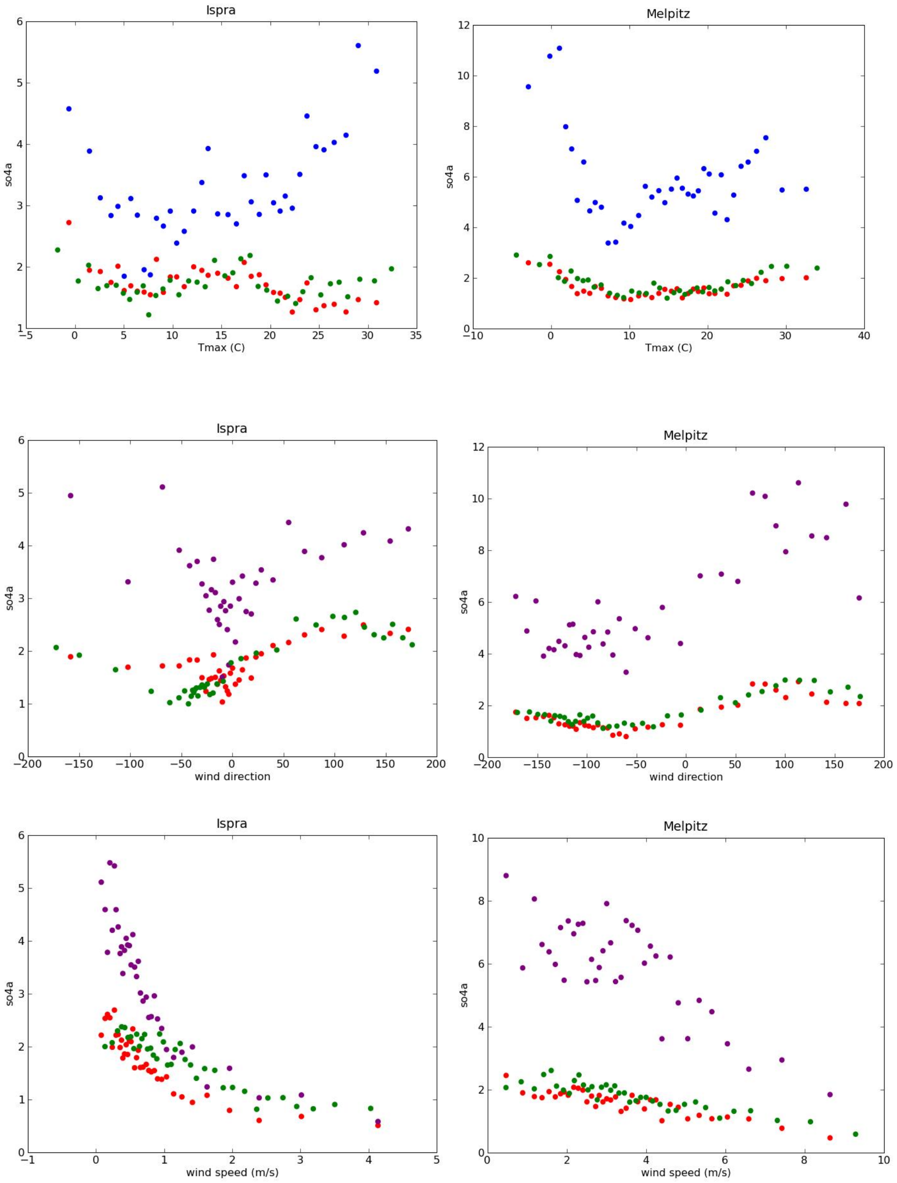

Figure B5. Modelled and observed daily average sulfate concentrations, 2003-2007, binned per 50 datapoints.

To develop the scientific and applied knowledge required for Climate-proofing the Netherlands and to create a sustainable Knowledge infrastructure for managing climate change

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