Dear Dr. Nicholas Henry Savage,

We appreciate valuable comments, which have helped improve the paper. We revised the text according to the suggested corrections and would like to thank you for the thorough reading of the paper. Below we provide our point-by-point replies, where for clarity the comments are displayed in bold italics.

Title. As per the instruction of GMD, please include the version of WRF-Chem in the title.

Thank you for this reminder, model version is now included in the title: »Evaluation of the high resolution WRF-Chem (v3.4.1) air quality forecast and its comparison with statistical ozone predictions«.

Abstract. Please specify the resolution of the model configuration applied (high resolution is rather a relative term)

We included the information about model resolution in the first sentence of the abstract, which is now: »An integrated modelling system based on the regional on-line coupled meteorology-atmospheric chemistry WRF-Chem model configured with two nested domains with horizontal resolution 11.1 km and 3.7 km has been applied for numerical weather prediction and for air quality forecast in Slovenia.«

2.1 WRF-Chem forecast system. Please state the height of the model top.

The height of the model top is 50 hPa, this information is now included in the paper in the following sentence: » The vertical structure of the atmosphere is resolved with 42 vertical levels extending up to 50 hPa, with the highest resolution of ~25 m near the ground.«

Please provide a reference (even if it is only a report) for the emissions inventory.

We added the reference to the project presentation at Slovenian Environment Agency (report is not yet available).

2.2 Statistical ozone daily maximum forecast. Please provide references for the statistical model.

We added the reference to the final report about statistical model (also available online).

2.3 Evaluation methodology. What is the height of the lowest model level, and how does that compare to a typical inlet height?

We added this information to the paper the following way: »In the case of air pollutants, the instantaneous lowest model level mixing ratios (with grid point center about 12 m above model orography - an exception is KRV station as explained below) are compared to the hourly averaged concentrations measured at monitoring stations (which have a typical inlet height of 3 m) from the national network and some other environmental information systems in Slovenia. Figure 3 shows locations of these AQ monitoring stations, and Tab. 1 lists the basic characteristics, including comparison of the station altitude, the height of model orography, model analysis height, and pollutants with higher than 75% availability of valid data during the analyzed time period for each of the AQ monitoring site«

Have you considered using data from above level 1 - in very mountainous terrain, an observation site can be well above the model orography at the relevant grid point and it is more appropriate to use data from level 2 or above.

Thank you for this question. In the case of AQ variables we usually use results from a higher model level for the KRV station. The altitude of this station is well above model topography (model height: 1272 m, model grid point at the lowest level: 1284 m, station altitude: 1740 m). In the present paper we originally included results for all stations (also KRV) at the lowest model level, because the correlation coefficient at the lowest model level is highest (CORR decreases with increasing the model level), showing that in spite of the negative bias due to too low model topography, the near surface processes still play an important role in ozone dynamics. In the review process we reconsidered this and decided to use model data from the 5th model level for KRV (model grid point center: 1414 m), but stay with the lowest model level for all other stations. For KRV the 5th model level is still well below the station altitude, but this reduces the bias for KRV from -12 μgm^{-3} to -2 μgm^{-3} for ozone hourly values, and from -16 μgm^{-3} to -7 μgm^{-3} for ozone daily maxima (which lowers the impact of KRV bias on overall model performance). Unfortunately also CORR then decreases from 0.76 to 0.74 for ozone daily maxima (which has a negligible impact on overall model performance). For other stations the differences between model height and station altitude are smaller. Also for some of the stations model height is too low (e.g. VNA, model height: 468 m, station altitude: 630 m), but for other stations the model height is too high (e.g. HRA, model height: 540 m, station altitude: 290 m), related to very complex topography in sub-alpine region of Slovenia. Consequently, by increasing the model levels we could reduce the negative bias for stations of the first group (with too low model orography), but cannot decrease the positive bias for the stations of the second group with too high model orography. This makes an approach of using higher model levels for stations with too low model orography questionable, also in the light that also CORR decreases with increasing model levels. We thus support the approach of using the data on the lowest model level and make a posterior bias correction, which does not impact the ozone dynamics and can be applied for all stations. We only made an exception for KRV station, for which the height in the model was significantly underestimated, as well as the station is known to be influenced by the conditions of the free troposphere (except during hot summer daytime conditions), which is not the case for other stations.

For meteorological variables we did not explore the impact of using results from higher model levels. This would be far beyond the scope of this study, focused on ozone prediction, also because the impact of using the higher layer data depends on meteorological variable, as well as the set of meteorological stations is not the same as in the case of AQ stations.

In the paper due to using results for KRV on the 5th level we corrected all of the AQ statistics and also the text throughout the paper accordingly. We included the following text:

»In the case of the elevated alpine KRV station, AQ variables are evaluated for the 5th model layer instead of the first model layer. We made this exception for KRV, since the height of the model topography was significantly underestimated there (Tab. 1), as well as the station is known to be strongly influenced by the conditions of the free troposphere. The selection of the 5th model layer for KRV station is based on analyses performed for different model layers (results not shown) and was found to reduce the negative bias for O_3 due to too low WRF-Chem topography at this location.

Although even for this model layer the location of the grid point representing KRV station (1414 m) is still well below the true station altitude (1740 m), the O_3 bias for KRV station is significantly smaller than for the first layer, while the correlation coefficient between the measured and simulated O_3 levels remains similar in both cases (the 5th or the lowest model layer). Taking results from higher model layers would further decrease the negative model bias, but would also worsen the correlation coefficient for O_3 at this station due to decreased impact of surface processes.«

Later in text also:

Instead of: »The elevated alpine KRV station is the only one with negative bias (-12 μgm^{-3}) in forecasted 1-hour O_3 concentrations, which can be explained by the too low altitude of the KRV station in model topography, since the mean O_3 concentration increases with height.«

We added: » In Fig. 4a the elevated alpine KRV station is the only one with high negative bias (-12 μgm^{-3}) in forecasted 1-hour O_3 concentrations at the lowest model layer, which can be explained by the too low altitude of the KRV station in model topography. The high negative bias for hourly O_3 concentrations at KRV station is reduced to a value of only -2 μgm^{-3} by using the 5th model layer concentrations as explained in chapter 2.3. The 5th model level predictions will be used for KRV in all analyses that follow.

We added also: » For sites with highest positive bias in 1-hour O_3 concentrations (TRB, ZAG, HRA and ISK, with bias of 36 μgm^{-3}, 31 μgm^{-3}, 26 μgm^{-3} and 32 μgm^{-3}, respectively), this can also be partly explained by too high altitude of the stations in model orography (Tab. 1), since the mean O_3 concentration increases with height.«

Later in text we deleted: »or Alpine stations (KRV)« and added: » Here we recall that high negative bias in WRF-Chem forecast for alpine KRV site due to too low altitude of the station in model topography was compensated by taking prediction from the 5th model level.«

Also the values of statistics in text and figures are changed throughout the paper.

3.1 Evaluation of meteorological variables. There is a large decrease in the precipitation bias from day 1 to day 2 - is this a model spin up issue? If so would a different initialisation improve this error?

We agree. Additional circumstance here is also that in the 3.4.1 model version it was not possible to include the information about hydrometeors at the boundaries of the nested domain (in the applied 1-way nesting procedure). Since the intensity of (relatively rare) summertime precipitation events was expected to have a less significant impact on ozone concentrations, we considered this issue less problematic (in our study focused on ozone). We added the following text: "It must also be taken into account that the 3.4.1 model version does not allow to include the information about hydrometeors at the boundaries of the nested domain (in the applied 1-way nesting procedure), which contributes to the negative simulated bias of precipitation. A large decrease in the precipitation bias from day 1 to day 2 suggests that different initialization methodology (e.g. using 1 day spin-up for meteorology) could improve the prediction of precipitation events."

Please provide some evidence for the statement "the main precipitation events were well predicted and simulated" or remove this statement.

Although we performed analyses and produced some plots we think that including additional material here is beyond the scope of the paper. We thus decided to remove this statement.

3.3 Evaluation and comparison of different methods for O3 daily maximum predictions. Please correct the statement "ideal forecast would lie in the right-bottom corner". It fact the ideal model would have correlation coefficient of 1 and a standard deviation equal to the observations, i.e. it would be co-located with the black dot which indicates the model. The black dot is not always in the bottom right corner on these plots.

Thank you, we corrected this statement. The statement that is now included is: » The ideal model would have a correlation coefficient of 1 and a standard deviation equal to the observations, which means that it would be co-located with the black dot on the diagram. «

In the section on the evaluation of the model's ability to predict episodes, too much weight is given to accuracy. For example, the statement "Accuracy ... increases with threshold level" is misleading. A model which always forecasts "no event" will have an increasing accuracy as the number of events decreases. To compare skill at different thresholds you need to use a differnt metric e.g. Critical Success Index or Equitable Threat Score. These would be better choices in general than accuracy in this section. There is no harm in including accuracy in the tables, but it should not be the primary criterion for judging forecast skill.

In the revised paper we replaced Accuracy (A) measure by Equitable Thread score (ETS), we also changed the order of categorical statistics in Tab. 5, so that ETS is shown in the first column, followed by CSI, B, FAR and POD. We corrected the text, to give most weight to the ETS and briefly mention the rest of them. The text that we now have in the paper regarding the categorical evaluations is the following: »Equitable Threat Score (ETS) measures the fraction of observed and/or correctly predicted events, adjusted for the frequency of hits that would be expected to occur by random chance. Although this score takes into account the climatology it is not truly equitable. It ranges from -1/3 to 1, where the minimum value depends on climatology (it is near 0 for rare events). Looking at Tab. 5 ETS shows equal skill for WRF-Chem and statistical forecast, higher than persistence for the 120 μgm^{-3} threshold (1-day and 2-day forecast). ETS decreases with increasing the threshold for both WRF-Chem and statistical forecast, indicating the challenge that both models have to accurately predict the extremes. In the case of 140 μgm^{-3} threshold, WRF-Chem has the same ETS as persistence, higher than the statistical model for 1-day forecast, while for 2-day forecast WRF-Chem outperforms the statistical model, followed by persistence. In the case of 160 μgm^{-3} threshold persistance has the highest ETS for a 1-day forecast, followed by statistical model and WRF-Chem, while in the case of 2-day predictions, statistical model shows the highest skill and WRF-Chem the lowest. Another measure, the critical success index (CSI), is similar to ETS, except that it does not take into account the climatology of the events and thus gives poorer scores for rarer events. It measures the percentage of cases that are correctly forecasted out of those either forecasted or observed, and ranges from 0 to 1 (1 indicating the perfect forecast). Similar as ETS, CSI gives higher scores for persistence in the case of 1-day forecast for the higher two thresholds, while on the second day WRF-Chem or the statistical model already performs better. Bias (B) determines whether the same fraction of events are both forecasted and observed. A tendency of the statistical model and of WRF-Chem to under-predict O_3 threshold exceedances shows as a B below 1 for these two models. The false alarm ratio (FAR) that measures the percentage of forecast high O_3 events that turn out to be false alarms, gives highest skill for WRF-Chem, followed by statistical model and persistence. The probability of detection (POD) is a measure of how often a high threshold occurrence is actually predicted to occur, and is relatively low for WRF-Chem with respect to other models. «

Also why were these specific three thesholds chosen?

There was no specific reason for these certain three thresholds. We also performed the calculations for different thresholds, e.g. 130 μgm^{-3} or 150 μgm^{-3}, distinguishing between higher and lower ozone maxima, and the conclusions were similar. We included some thresholds which present an elevated ozone levels and pose a greater risk to human health, and decided to exclude the statistics for a higher threshold (180 μgm^{-3}, a legislation limit value) due to a very low number of exceedances for this threshold. In the paper we extended the following sentence: »Table 5 summarizes the categorical evaluation results for three different thresholds (120, 140, 160 μgm^{-3}) of elevated ozone levels, which pose a greater risk to human health.«

Grammatical and other minor corrections.
p1030 line 22, "The first RT-AQF systems.."
p1030 line 25, delete "existing"
p1032 line 13, "during summertime conditions"
p1032 line 21, "a one-way"
p1032 line 22, "evaluated a forecast"
p1033 line 2, "based on WRF-Chem are implemented worldwide"
p1033 line 4, "over the topographically complex"
p1033 line 6, "with a statistical model"
p1033 line 6, "at the Slovenian"
p1036 line 19, "a southwestern"
p1036 line 24, "shows a mean O3 daily mean"
p1037 line 27, "is a mountainous station"
p1037 line 27, "As well as the elevated station KRV, the ISK, OTL and VNA stations area are also influenced by regional transport of pollutants.
p1038 line 7, "information about the AQ forecast can also be gained by the evaluation of meteorological forecasts for these stations."
p1038 line 16, "index of agreement"
p1041 line 3, "with a range of 0.64 to 0.90 for 1 day forecasts"
p1041 line 7, "On average"
p1042 line 8, "3 month accumulations by"
p1042 line 3, "has problems simulating the"
p1043 line 1, "the model over-predicts"
p1043 line 5, "explained by model error in"
p1043, line 16, "poorly reproduced meteorological"
p1043, line 26, "Also interesting to discuss are the results"
p1045, line 3, "In this section we want to answer the question: 'how accurate is the 1 h O3 daily maximum WRF-Chem forecast in comparison to the statistical model prediction or to persistence?'."

p1045, line 8 "which is, along with their computational efficiency, "
p1045, line 9 "Among the strengths of the deterministic models are that they give"
p1045 line 12, "Furthermore, they also allow forecasts for"
p1045 line 14, "descriptions of"
p1045 line 27, "because a statistical"
p1046 line 1, "with an available"
p1046 line 5, "already beats persistence"
p1046 line 12, "than the statistical forecast"
p1046 line 25, "MNBE in Fig. 8 has very similar results to ME."
p1047 line 13, "also contingency-table-based statistics are an important metric of"
p1047 line 15, "It is important to take into account"
p1048 line 9, "were to be applied to"
p1049 line 7, "local emissions result in model underestimations of NO2"
p1049 line 12, "show good WRF-Chem model performance"

We revised the text according to the suggested corrections and would like to thank again for the thorough reading of the paper.

Dear Dr. Georg A. Grell,

We appreciate and would like to thank you for all the comments and raised questions, which have helped to improve the quality of the paper. Below we provide our point-by-point replies, where for clarity the comments are displayed in bold italics.

This paper describes the use of the community version of WRF-Chem for real-time ozone and aerosol predictions. The authors perform statistical evaluations over a 3 month period, comparing the model forecasts with observations as well as statistical forecast methods. In general his paper is well written and should be published in GMD. This can be done with only minor modifications.

We thank for this comment.

Although the authors provide much information on model set-up there are a few details that I was looking for and couldn't find. Is this 2-way nesting or 1-way nesting? If it is 1-way nesting, how was it applied?

It is a 1-way nesting applied by two consecutive simulations (using ndown). We added this information the following way (section 2.1): »A 1-way nesting is applied by two separate consecutive simulations, where outputs from the coarse grid integration are processed to provide boundary conditions for the nested run every 15 minutes.«

Is the choice of physics parameterization the same on both domains?

Yes, schemes are the same on both domains. To include this information in the paper we changed in Section 2.1: »We decided to apply the same schemes as were used…« to »In both domains we decided to apply the same schemes as were used…«.

Which photolysis model have you been using?

Fast-J photolysis scheme (Wild et al., 2000), this information is now added in section 2.1.

All evaluations I am assuming are done on the high resolution domain.

Yes. We included this information in the first sentence of Section 2.3: »We evaluate the 1-day and 2-day WRF-Chem meteorological and AQ forecasts on the high resolution domain during a 3-month period (June - August 2013).«

Also, the color choice for figures 5, 6, and 7 is unfortunate. The two blue colors are almost impossible to separate – at least with my aging eyes. Why not a different color? Figure 5 is even more difficult to read, a bit too small for me.

We replotted these figures with two different colors. Still it is hard to distinguish between 1-day and 2-day forecast (Fig. 5-7), but the purpose of these figures is more to separate model forecast from observations. 1-day and 2-day forecast are more easily distinguished by the use of statistics. Figure 5 is now divided into two parts.

Some other questions I have:

(1) There is a negative temperature bias, but a positive short wave bias? Since you are using the interaction flag for convection/radiation the SW bias could be interpreted as not enough cloud cover, which could give you a low bias at night, but at day? Are you cycling

soil temperature and soil moisture or is that always a new initialization with coarse resolution GFS data?

All meteorological variables, including soil temperature and soil moisture are always initialized with GFS data, which is now mentioned in the paper. This explains higher negative bias for T2m during the first day of simulation (not valid for daily maxima, where bias is the same on the first and the second day of simulation). For all hourly values T2m bias decreases from -2.1 C to 0.8 C due to reduced bias for nighttime temperatures on the second day of simulation. Looking at results station by station the link between T2m and SW bias is not straightforward (they appear not to be directly correlated). On the first day of simulation higher SW is due to less cloudy conditions (more cloud cover on the second day).

(2) The statistics I assume are always over domain 2. The fact that the precipitation under-forecast is a lot less on day 2 may indicate some spin-up issues, especially also when taking into consideration the coarse initial conditions (did you use .5 degree data from GFS?)

Yes, we used the 0.5 degree data from GFS, this information is now added in section 2.1 as »…with meteorological initial (ICs) and lateral boundary conditions (BCs) taken from the 0.5° data from the Global Forecast System (GFS)…«. We also agree that under-prediction of precipitation indicates some spin-up problem, where it must also be taken into account that in 3.4.1 model version ndown procedure does not allow to include the information about hydrometeors at boundaries of the nested domain. Since the intensity of (rare) summertime precipitation events was expected to have a less significant impact on ozone concentrations, we considered this issue less problematic in our study focused on ozone. But we agree that applying a different initialization methodology should reduce the precipitation error. The following text was added: »It must be mentioned that the 3.4.1 model version does not allow to include the information about hydrometeors at the boundaries of the nested domain (in the applied 1-way nesting procedure), which contributes to the negative simulated bias of precipitation. A large decrease in the precipitation bias from day 1 to day 2 suggests that different initialization methodology (e.g. using 1 day spin-up for meteorology) could improve the prediction of precipitation events.«

(3) On page 1047, line 22 you talk about WRF-Chem under-predicting Ozone maxima, while before you had a positive bias. Do you mean under-predict excedences?

We replaced »ozone maxima« to »threshold exceedances«.

(4) In the summary and conclusions you should mention again (you have that hidden somewhere in section 2.1, pg 1034) that different choice of physical or chemical parameterization will influence and possibly change outcomes. However I think your choices are good choices, since they are well documented in other real-time applications.

We added the following sentence to the conclusions: »Since the selection of physical or chemical parameterization schemes influences and possibly changes the outcomes, we decided to apply the schemes that are well documented and have previously been used in other applications (e.g. AQMEII).«

(5) Pg. 1031, line 7: The MM5 reference should be 1994, not 1995 – if I remember correctly

This error is now corrected.

(6) Pg 1032, line 11: 2011 should not be a reference for WRF-Chem. Just 2005 is good enough.

We deleted the 2011 reference.

(7) Pg. 1049, last line: If you want you could add the recent Pagowski et al publication in GMD (also WRF-Chem special issue) as an example of chemical data assimilation

The following sentence was added: »For WRF-Chem model a technical note on the implementation of the aerosol assimilation and a guidance for prospective users has been recently published by Pagowski et al. (2014).«

Evaluation of the high resolution WRF-Chem (v3.4.1) air quality forecast and its comparison with statistical ozone predictions

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Abstract

An integrated ~~high resolution~~ modelling system based on the regional on-line coupled meteorology-atmospheric chemistry WRF-Chem model configured with two nested domains with horizontal resolution 11.1 km and 3.7 km has been applied for numerical weather prediction and for air quality forecast in Slovenia. In the study an evaluation of the air quality forecasting system has been performed for summer 2013. In the case of ozone (O_3) daily maxima, the first ~~day~~ and second day model predictions have been also compared to the operational statistical O_3 forecast and to the persistence. Results of discrete and categorical evaluations show that the WRF-Chem based forecasting system is able to produce reliable forecasts, which depending on monitoring site and the evaluation measure applied can outperform the statistical model. For example, the correlation coefficient shows the highest skill for WRF-Chem model O_3 predictions, confirming the significance of the non-linear processes taken into account in an on-line coupled Eulerian model. For some stations and areas biases were relatively high due to highly complex terrain and unresolved local meteorological and emission dynamics, which contributed to somewhat lower WRF-Chem

skill obtained in categorical model evaluations. Applying a bias-correction could further improve WRF-Chem model forecasting skill in these cases.

Key words: Air quality, forecast, ozone, WRF-Chem, online-coupled model, statistical model

1 Introduction

Real-time air quality forecasting (RT-AQF) is a relatively new discipline in atmospheric sciences, which has evolved as a response to societal and economic needs, reflecting the progress in scientific understanding of physical processes and numerical and computational technologies (Zhang et al., 2012a). The ~~F~~first RT-AQF systems, developed for forecasting air pollution in exposed urban regions, were either empirical methods based on persistence, climatology, human expertise and meteorological forecast (e.g. Wolff and Lioy, 1978), or statistical models taking advantage of ~~existing~~ links between pollutant concentrations, meteorological variables (wind speed and direction, temperature, cloudiness, moisture etc.) and physical (emissions) parameters (e.g. McCollister and Wilson, 1975; Cobourn, 2007; Vlachogianni et al., 2011). The next step in evolution of RT-AQF systems was the use of sophisticated chemical transport models that represent all major processes (meteorological and chemical) that lead to the formation and accumulation of air pollutants. Many of these RT-AQF systems consist of an offline coupled meteorological model and a chemical-transport model, where the meteorological model (e.g., ALADIN, ALADIN International Team, 1997; MM5, Grell et al., 1994~~5~~; WRF, Skamarock et al., 2008) provides meteorological input for the chemical-transport model (e.g., EMEP, van Loon et al., 2004; CMAQ, Byun and Schere, 2006; CAMx, ENVIRON, 2011; CHIMERE, Menut et al., 2013) with an output time interval typically around 1 hour. Examples are the EURAD (http://db.eurad.uni-koeln.de/index_e.html), SILAM (http://silam.fmi.fi/), ForeChem (http://atmoforum.aquila.infn.it/forechem/), CALIOPE (http://www.bsc.es/caliope/) forecast systems and others. The new generation of an online coupled models (e.g., MCCM, Grell et al., 2000; GATOR-GCMM, Jacobson 2001; Meso-NH-C, Tulet et al. 2003; WRF-Chem, Grell et al., 2005; Enviro-HIRLAM, Baklanov et al., 2008; GEM-AQ, Kaminski et al. 2008; COSMO-ART, Vogel et al., 2009; WRF-Chem-MADRID, Zhang et al., 2010a) presents an alternative approach with one unified modelling system, in which meteorological and air quality variables are simulated together within the same model. The online approach permits the simulation of two-way interactions between different atmospheric processes including emissions, chemistry, clouds and radiation, and a better response of the simulated pollutant transport to changes of the wind field (Grell et al., 2004), and can thus provide a more realistic representation of the atmosphere. The use of online coupled models can be particularly important in regions with high aerosol loadings and cloud coverage (Otte et al., 2005; Eder et al., 2006), where physical processes in the atmosphere may be modified by the aerosol direct effect on radiation or by aerosol cloud interactions. Several reviews summarized the strengths and limitations of offline and online coupled models (e.g. Zhang 2008; Klein, 2012; Baklanov et al., 2014).There is an increasing awareness that an integrated online approach is needed not only for assessment, forecasting and communication of air quality, but also for weather forecasting (e.g. Baklanov, 2010; Grell and Baklanov, 2011; Klein et al., 2012; Zhang et al., 2012b; Baklanov et al., 2014). Nevertheless, there are several issues regarding the inclusion of chemistry into numerical weather prediction models. More evidence is required whether an integrated model can produce a good climatology of the most important chemical species, and if such a model is, considering many uncertainties, able to beat persistence forecasts of these species (Grell and Baklanov, 2011). These questions are calling for further research and studies exploring the performance of the models with an online coupled chemistry.

In recent years extensive efforts have been devoted to develop air quality (AQ) forecasting systems for Slovenia. In this study we explore the use of the state-of-the-science WRF-Chem model (Grell et al., 2005~~, 2011~~) with coupled meteorological, microphysical, chemical, and radiative processes for forecasting AQ in Slovenia during~~ the~~ summertime conditions. In last decade WRF-Chem has been increasingly applied to many areas worldwide (e.g., Misenis and Zhang, 2010; Fast et al., 2009; Zhang et al., 2010a, 2010b; Li et al., 2011; Tie et al., 2009; Hu et al., 2012; Forkel et al., 2012, Žabkar et al., 2011a, 2013). In most of these studies WRF-Chem model has been successfully used to simulate historical poor AQ conditions in hindcast approach. To our knowledge, only a few studies focused on using WRF-Chem for forecasting AQ, most of these have applied WRF-Chem forecast before and during field campaigns (McKeen et al., 2005, 2007, 2009; Yang et al., 2011). Takigawa et al. (2007) evaluated O_3 forecast for a 1 month time period from a~~n~~ one-way nested global-regional RT-AQF system with full chemistry based on the global CHASER (Sudo et al. 2002) and regional WRF-Chem models, while Saide et al. (2011) evaluated a forecast system based on~~ the~~ WRF-Chem model for simulating carbon monoxide (CO) as a PM10/PM2.5 surrogate over Santiago de Chile for wintertime conditions. WRF-Chem-MADRID (Zhang et al., 2010a) with two additional gas-phase mechanisms, sectional representation for particle size distribution and more advanced model treatments compared to WRF-Chem, was applied by Chuang et al. (2011) and by Yahya et al. (2014) for forecasting AQ over the Southeastern U.S.. In spite of a limited number of evaluation studies published in the literature, an increasing number of real-time weather and air quality forecasting systems based on WRF-Chem ~~is performed~~are implemented worldwide (http://ruc.noaa.gov/wrf/WG11/Real_time_forecasts.htm).

In our study we explore the forecasting skill of WRF-Chem model over the topographically complex and geographically diverse area of Slovenia for three summer months (June - August 2013). Furthermore, in the case of O_3 we compare WRF-Chem predictions with a statistical model for predicting O_3 daily maxima, currently used at the Slovenian Environment Agency (SEA). Both first day (1-day) and second day (2-day) forecasts are considered, while a persistence model, which assumes that pollutant level today and tomorrow will be the same as yesterday, is used as a threshold for useful model prediction. Since the availability of accurate and reliable forecasting system could be useful to the local authorities and could help to advise the public the proper preventive actions, we want to answer the question whether WRF-Chem model outperforms the statistical model or persistence. Namely, considering many uncertainties related to one unified model, it may not be easy for models with online chemistry to be able to perform well enough to meet the required standards, and more research and studies are needed to investigate that (Grell and Baklanov, 2011). Due to the limited number of previous studies focused on online coupled forecasting systems, the aim of our study is also to provide a greater insight into potential that lies in the approach based on an unified model for forecasting weather and air pollution. Finally, identified strengths, limitations and deficiencies of analyzed RT-AQFs, are expected to present the basis for further research.

2 Methodology

2.1 WRF-Chem forecast system

The RT-AQF system for Slovenia based on the WRF-Chem model version 3.4.1 is configured with two nested domains (Fig.1) with horizontal resolution 11.1 km and 3.7 km, and 151×100 and 181×145 grid points, respectively. A 1-way nesting is applied by two separate consecutive simulations, where outputs from the coarse grid integration are processed to provide boundary conditions for the nested run every 15 minutes. The vertical structure of the atmosphere is resolved with 42 vertical levels extending up to 50 hPa, with the highest ~~near ground~~ resolution of ~25 m near the ground. About 15 levels are located within the lowest 2 km to assure high vertical resolution of the daytime planetary boundary layer (PBL). To produce the 48-hour forecast, the model is run every day, starting at 00 UTC, with meteorological initial (ICs) and lateral boundary conditions (BCs) taken from the 0.5° data from the Global Forecast System (GFS)~~, a global numerical weather prediction system~~ operated by the US National Weather Service (NWS). For chemical BCs forecasts from global MOZART-4/ GEOS-5 (Emmons et al., 2010) RT-AQF system with temporal availability of 6 h are used. The instantaneous outputs at the 24th hour of the previous day forecast are used to initialize next day's forecasting simulation. An exception is the very first day of the first 48-hour forecasting cycle, when global MOZART-4/ GEOS-5 fields were used also to initialize chemistry. A three day spin-up ahead of the first analyzed forecast day is then taken into account to allow pollutants to accumulate in the air masses.

In the WRF-Chem model, several choices for parameterizations of physical and chemical processes are available (Grell et al., 2005; Skamarock et al., 2008; Peckham et al., 2011), and their choice can have a strong impact on the model predictions. In both domains w~~W~~e decided to apply the same schemes as were used in simulation SI1 for Phase-2 of the Air Quality Model Evaluation International Initiative (AQMEII) (e.g., Balzarini et al., 2014, Baró et al., Curci et al., 2014, Forkel et al., 2014, Im et al., 2014a and 2014b, Kong et al., 2014, 2014, San Josè et al., 2014). These include Yonsei University (YSU) PBL scheme (Hong et al., 2006), NOAH land-surface model (Chen and Dudhia, 2001), Rapid Radiative Transfer Method for Global (RRTMG) long-wave and short-wave radiation scheme (Iacono et al. 2008), Grell 3D ensemble cumulus parameterization scheme (Grell and Devenyi, 2002) with radiative feedback, Morrison double-moment cloud microphysics (Morrison et al., 2008), Fast-J photolysis scheme (Wild et al., 2000), RADM2 gas phase chemistry (Stockwell et al., 1990) and the MADE/SORGAM aerosol module (Ackermann et al., 1998, Schell et al., 2001). Current model implementation includes a modified RADM2 gas phase chemistry solver as described in Forkel et al. (2014), which avoids under-representation of nocturnal O_3 titration in areas with high NO emissions. According to Forkel et al. (2014) the modified solver tends to over-estimate the low NO_2 concentration for pristine regions and in the free troposphere, which results in an overestimation of O_3. Due to the focus on polluted regions this deficiency was considered as less important than the advantage of better description of the titration. In addition, the comparatively small modelling domain (D1) ensures that the boundary conditions constrain the high bias of the modified solver for O_3 and NO_2 in the free troposphere. Also according to our sensitivity tests (results not shown) the modified solver showed better performance for O_3 daily maxima and O_3 nighttime minima than the QSSA RADM2 solver supplied originally with WRF-Chem model.

Among feedbacks only the aerosol direct effects on radiation according to Fast et al. (2006) and Chapman et al. (2009) are taken into account. As shown by Kong et al. (2014) for two air pollution episodes, this degree of aerosol-meteorology interactions in 3.4.1 version of the WRF-Chem improved model performance for high aerosol loads, while the representation of the indirect effects needs to be further improved to be able to outperform simulations with direct effects only.

Biogenic emissions are estimated using MEGAN (Model of Emissions of Gases and Aerosols from Nature; Guenther et al., 2006) online model calculations, while dust emissions are modelled according to Shaw et al. (2008) with an adjustment to avoid high dust fluxes from some Dalmatian islands in Croatia. A detailed anthropogenic inventory for pollutants CO, NH_3, NOx, SO_2, and NMVOC, which has been for the purpose of AQ forecasting constructed for year 2009 by SEA (SEA, 2014), is used to estimate anthropogenic emissions in Slovenia. For areas outside Slovenia the recently updated anthropogenic emissions for the year 2009 based on the TNO-MACC-II (Netherlands Organization for Applied Scientific Research, Monitoring Atmospheric Composition and Climate – Interim Implementation), the same as prepared for phase-2 of the AQMEII exercise (Pouliot et al., 2014), are being used. Daily updates of the WRF-Chem based experimental AQ forecast are provided at http://meteo.fmf.uni-lj.si/onesnazenje.

2.2 Statistical ozone daily maximum forecast

The statistical O_3 model (Žabkar, 2011b), currently used at SEA for forecasting O_3 daily maxima at 8 measuring sites in Slovenia (Fig.3), is a multivariate regression tool combined with clustering algorithms to take into account measured data, weather forecast data, as well as the predicted backward trajectories of each monitoring site. As regards measurements, yesterday (at 12, 15, 18 and 21 local time, daily maximum, daily minimum, daily average) and today early morning (7 local time) meteorological (pressure, relative humidity, direct and diffusive solar radiation, wind speed) and AQ data (O_3, NO_x, NO_2, CO, PM_{10}, SO_2) are used. For meteorological predictions the 24-h ECMWF forecast variables at 12 UTC of the forecast day at different vertical levels (1000 hPa, 925 hPa, 850 hPa, 500 hPa, 300 hPa) above the measuring sites are taken into account. Among all these variables by the use of stepwise technique, based on the F-statistic only significant variables were selected to be included in multivariate regression equations for different monitoring sites (from 15 to 26 variables, depending on monitoring site).

The important part of the statistical forecast is calculation of 24-h backward trajectories on meteorological fields of ALADIN/SI forecast. The inclusion of 24-h predicted trajectories into statistical model is based on the study (Žabkar et al., 2008) which showed, that the highest O_3 daily maxima at monitoring sites in Slovenia are in general associated with short (slow-moving) backward trajectories with a~~the~~ southwestern origin, while the lowest measured daily maximum O_3 values for all the stations are associated with the clusters of long northwestern trajectories. Clusters of similar trajectories were for the purpose of statistical forecast calculated by *k*-means clustering algorithms (Moody and Galloway, 1988; Žabkar et al., 2008) on 6 years (2004-2010) of data (ALADIN/SI trajectories). As an example, Fig. 2 shows a mean ~~is showing mean~~ O_3 daily maxima for clusters of similar trajectories for one of the monitoring sites. The same 6-year time period of training data was used in the stepwise multiple regression procedure to determine the multiple regression prognostic equations associated with monitoring sites and trajectory clusters, from measurements, ECMWF forecast data, average cluster O_3 daily maximum, and day-of-the-year variable.

The first step of the statistical O_3 prediction is the calculation of trajectories approaching the monitoring stations at 12 UTC of the forecast day. In the next step these backward trajectories of each monitoring site are associated to the nearest pre-calculated cluster of similar trajectories. Finally, the multiple regression equation of the associated group of trajectories is used to calculate the O_3 daily maximum prediction. It must also be noted, that the decision on declaring O_3 episodes is only partially based on the results from this statistical model; it also involves a decision made by AQ forecasters.

2.3 Evaluation methodology

We evaluate the 1-day and 2-day WRF-Chem meteorological and AQ forecasts on the high resolution domain during a 3-month period (June - August 2013).The main focus is on O_3 predictions. In the case of air pollutants, the instantaneous lowest model level mixing ratios (with grid point center about 12 m above model orography - an exception is KRV station as explained below) are compared to the hourly averaged concentrations measured at monitoring stations (which have a typical inlet height of 3 m) from the national network and some other environmental information systems in Slovenia. Figure 3 shows locations of these AQ monitoring stations, and Tab. 1 lists the basic characteristics, including comparison of the station altitude, the height of model orography, model analysis height, and pollutants with higher than 75% availability of valid data during the analyzed time period for each of the AQ monitoring site. In the case of the elevated alpine KRV station, AQ variables are evaluated for the 5th model layer instead of the first model layer. We made this exception for KRV, since the height of the model topography was significantly underestimated there (Tab. 1), as well as the station is known to be strongly influenced by the conditions of the free troposphere. The selection of the 5th model layer for KRV station is based on analyses performed for different model layers (results not shown) and was found to reduce the negative bias for O_3 due to too low WRF-Chem topography at this location. Although even for this model layer the location of the grid point representing KRV station (1414 m) is still well below the true station altitude (1740 m), the O_3 bias for KRV station is significantly smaller than for the first layer, while the correlation coefficient between the measured and simulated O_3 levels remains similar in both cases (the 5th or the lowest model layer). Taking results from higher model layers would further decrease the negative model bias, but would also worsen the correlation coefficient for O_3 at this station due to decreased impact of surface processes.

All AQ stations are background, 7 of them are measuring urban background, 1 suburban and 9 rural conditions. Valid O_3 measurements are for the analyzed time period available for 13 AQ stations. When studying the general model performance, data from additional 4 stations for two other pollutants (NO_2, PM10) are also analyzed to get a better picture of model behavior over the domain, known for its large topographical and climate diversity. The coverage of three climate zones in Slovenia (Mediterranean, sub-alpine and mountainous) with monitoring stations is the following: NG, KOP and OTL are Mediterranean sites, KRV is a mountainous station, and the remaining stations are sub-alpine. As well as the~~Besides~~ elevated station KRV, the ~~also~~ ISK, OTL and VNA stations are also influenced by ~~measuring~~ regional transport of pollutants.

For evaluation of predicted meteorological variables, data from SEA meteorological stations (MET, Fig. 3) for 2m temperature (T2m), 10 m wind speed (W10m), relative humidity (RH), incoming shortwave radiation (SR) and precipitation (RR) are used. It must be noted, that

MET stations with lower spatial representativeness (e.g. alpine stations) were not a priori excluded from the analyses, which needs to be taken into account when looking at evaluation results. The reason for not excluding these stations was that some~~ interesting~~ information about the~~for~~ AQ forecast can also be gained ~~also ~~by the evaluation of meteorological forecast for these stations.

Basic statistical measures (correlation coefficient (CORR), mean error (ME), mean absolute error (MAE) and root mean square error (RMSE)) are used for evaluating model's forecasting skills of meteorological and AQ variables. In the case of O_3, correlation coefficients are presented also by Taylor diagrams (Taylor, 2001), which graphically summarize the similarity between model forecasts and observations not only in terms of their correlation, but also with their centered root-mean-square difference and the amplitude of their variations, represented by their standard deviations. Furthermore, some additional discrete statistical measures, including index ~~i~~of agreement (IOA), the mean normalized bias error (MNBE), and the mean normalized gross error (MNGE) are calculated for O_3 daily maximum concentrations predicted by the different models. Finally, to evaluate the model's ability to predict exceedances and non-exceedances also several categorical indices including Equitable Threat Score~~accuracy~~ (ETS~~A~~), Critical Success Index (CSI), ~~b~~Bias (B), F~~f~~alse ~~a~~Alarm ~~r~~Ratio (FAR) and~~,~~ P~~p~~robability O~~o~~f D~~d~~etection (POD) ~~and critical success index (CSI) ~~are calculated for different thresholds. Definitions of statistical measures are shown in Appendix A.

2.4 Meteorology and air quality of June-August 2013

The analyzed period was marked by three heat wave events, which contributed to the summer characterized by high temperatures, sunny weather and lack of precipitation in Slovenia. The first heat wave event with measured temperature daily maxima up to 35 °C occurred after a rather cold beginning of the month and lasted from June 15 – 21. The event was terminated by a cold front passage and followed by the pronounced cold episode during the end of June and the beginning of July. Another heat wave event with temperatures above 35 ºC observed in the lowland, started on July 26 and was briefly interrupted on July 29, when thunderstorms related to frontal passage were accompanied by exceptionally strong wind gusts. The most remarkable of three extraordinary hot episodes was recorded from August 1 – 8. On the last day of this episode, August 8, temperatures reached 40 °C at some measuring sites in Slovenia, and many of them observed their highest temperature ever recorded.

As expected for summertime conditions, measured concentrations of most air pollutants, including PM10, were in general low during the analyzed time period. The only exception was O_3 with exceedances of 8-hour target value (120 μgm^{-3}) measured at all AQ monitoring stations during the three heat wave events, which is the reason why the main focus of the present study is on this pollutant. During the second two events (in July and August) also threshold exceedances of 1-hour daily maxima were recorded for O_3. In spite of the hot and sunny conditions during the first heat wave event in June 2013, measured daily O_3 maxima at the Slovenian stations did not exceed the 1-hour information threshold value (1h ITV; 180 μgm^{-3}), but reached 171 μgm^{-3} at the Mediterranean OTL and the elevated alpine KRV stations. During the second heat wave event 1-hour daily maxima exceeded 180 μgm^{-3} at KRV, OTL, NG and KP (July 23 – 28), while the highest number of 1-hour exceedances (20) has been in July measured at OTL station. Similarly, during the August heat wave event O_3 concentrations exceeded the 1h ITV at LJ, MB, OTL, NG and KP from August 2 – 7. To summarize, the Mediterranean stations (NG, OTL, KP) due to very high O_3 concentrations measured during the heat wave events (especially the second two events) exhibited the poorest AQ in Slovenia during the analyzed time period, while the legislation limit values have been exceeded only occasionally for the sub-alpine stations.

3 Results and discussion

3.1 Evaluation of meteorological variables

Table 2 shows conventional statistical scores evaluating the 1-day WRF-Chem forecast for the basic meteorological variables, 2m temperature (T2m; for hourly values and daily maxima), 10 m wind speed (W10m), relative humidity (RH) and incoming solar radiation (SR). Results for three selected measuring sites (LJ, NG, MS) and overall result for all 24 MET monitoring sites (shown in Fig. 3) are presented separately.

Incoming solar radiation is the main energy source that drives all atmospheric processes, including PBL processes, and has a critical role also in atmospheric chemistry. For almost all sites the mean SR was overestimated by the model, with an overall ME of 16 W/m^2 and 11 W/m^2 for 1-day and 2-day forecast, respectively. CORR was higher for 1-day (0.77) than for 2-day (0.71) forecast, with a range of 0.64 to 0.90 for 1-day forecasts at different stations. The larger positive bias during the first day than for the second day can be attributed to less cloudy conditions during the first day of simulation.

In the case of T2m 1-day (2-day) WRF-Chem meteorological forecast showed an overall correlation with measurements of 0.93 (0.94) for all 1-hour values and 0.97 (0.96) for 1-hour daily maxima. With an exception of three alpine stations with higher simulated positive bias, daily T2m maxima were simulated with ME between -3.9 °C and -0.6 °C, depending on station spatial representativeness. All meteorological variables, including soil temperature and soil moisture, are always initialized with GFS data. This explains higher negative bias for T2m during the first day of simulation in spite of the overestimated of solar radiation. ~~A while a~~An average systematic underestimation of T2m daily maxima was -2.1 °C both for 1-day and 2-day forecast. Nighttime T2m minima showed lower systematic bias for 2-day forecast, which resulted in overall bias for all hourly T2m values of -1.3 °C for 1-day and -0.8 °C for 2-day forecast. Predominant weak wind conditions with variable direction at stations located in complex topography were challenging to simulate. The general model tendency was to overestimate W10m with overall ME of 0.8 m/s for 1-day and 2-day forecast, where for some stations bias can be very low (e.g. LJ; Tab. 2) and much higher for some other stations due to their local positioning in complex topography (e.g. HRA located in valley with ME of 1.9 m/s). For hourly values the correlation is lower (Tab. 2), but for mean daily W10m values Pearson correlation coefficient between 0.4 and 0.9 has been simulated, depending on monitoring site. Relative humidity shows slightly better results for 1-day than for 2-day forecast with CORR of 0.77 and low overall ME of 2 % for 1-day forecast, which for particular stations can be positive (e.g. KRV) or negative (e.g. LJ, NG; Tab. 2).

~~Incoming solar radiation is the main energy source that drives all atmospheric processes, including PBL processes, and has a critical role also in atmospheric chemistry. For almost all sites the mean SR was overestimated by the model, with an overall ME of 16 W/m^2 and 11 W/m^2 for 1-day and 2-day forecast, respectively. CORR was higher for 1-day (0.77) than for 2-day (0.71) forecast, with span from 0.64 to 0.90 for 1-day forecast at different stations.~~

Precipitation (RR) has an important role in cleansing of the atmosphere by wet deposition and scavenging. O~~I~~n average, the predicted precipitation underestimated the measured 3-month accumulations by~~for~~ -55 mm (1-day) or -8 mm (2-day forecast), where the station averaged predicted 3-month precipitation was 145 mm for 1-day, and 194 mm for 2-day forecast (results not shown). It must also be taken into account that the 3.4.1 model version does not allow to include the information about hydrometeors at the boundaries of the nested domain (in the applied 1-way nesting procedure), which contributes to the negative simulated bias of precipitation. A large decrease in the precipitation bias from day 1 to day 2 suggests that different initialization methodology (e.g. using 1 day spin-up for meteorology) could improve the prediction of precipitation events. ~~Although the WRF-Chem simulations sometimes failed to correctly predict the true amount and location of the more randomly spread summertime convective precipitation, the main precipitation events (e.g. those terminating three heat wave events) were well predicted and simulated.~~

3.2 Evaluation of air quality variables

In this section we evaluate WRF-Chem predictions for O_3, NO_2 and PM10, as three of the most problematic pollutants in terms of harm to human health and compliance with EU limit values (EEA, 2012). Table 3 shows the domain wide performance statistics for 1-day and 2-day forecasts of these pollutants, where in the case of O_3 1-hour and 8-hour averages and daily maxima are analyzed separately. The comparison of 1-day and 2-day forecasts shows that concentrations of air pollutants were somewhat better forecasted 1-day than 2-days ahead by means of almost all of statistics shown in Tab. 3, with higher impact on O_3 predictions. Although the 2-day prediction was generally not worse for the majority of meteorological variables, the reason for better 1-day prediction in the case of O_3 could be somewhat stronger simulated winds on the second day of simulation. Stronger winds impact the transport and dispersion of pollutants, and have the greatest consequence for secondary pollutants (like O_3) which need time to be formed.

As shown in Tab. 3 the WRF-Chem simulations tend to overestimate the 1-hour and 8-hour O_3 values with ME of 14.5~~3.7~~ μgm^{-3} and 14.6~~3.8~~ μgm^{-3}, respectively. Looking at MAE, RMSE and CORR statistics, agreement with measurements is better for 8-hour (22.6~~9~~ μgm^{-3}, 28.1~~5~~ μgm^{-3} and 0.69) than for 1-hour O_3 values (25.1~~5~~ μgm^{-3}, 32.1~~5~~ μgm^{-3} and 0.65), which is in line with results of previous studies (e.g. Tong and Mauzerall, 2013) and suggests that the current modeling system has problems simulating~~to simulate~~ the small-scale fluctuations of O_3. On the other hand evaluations of predicted 8-hour and daily O_3 maxima, which are of most concern, show a nice model performance (ME, MAE RMSE and CORR of -2.7~~3.4~~ μgm^{-3}, 13.3~~7~~ μgm^{-3}, 16.7~~7.1~~ μgm^{-3} and 0.81 for daily maxima, respectively), in line or even better than obtained in some previous studies (e.g. Tong and Mauzerall, 2006; Chuang et al., 2011; Yahya et al., 2014), which could be to some extent related to higher model resolution.

To understand results of the domain wide statistics (in Tab. 3) we further analyze spatial and temporal characteristics of model O_3 predictions. Figure 4 shows a spatial pattern of average simulated 1-day predictions for O_3, NO_2 and PM10 overlaid with measured averages, where in the case of O_3 results for all hourly values and for daily maxima are shown separately. Examples of forecasted and measured time series for O_3 at different stations are shown in Fig. 5. In Fig. 4a ~~T~~the elevated alpine KRV station is the only one with high negative bias (-12 μgm^{-3}) in forecasted 1-hour O_3 concentrations at the lowest model layer, which can be explained by the too low altitude of the KRV station in model topography~~,~~. The high negative bias for hourly O_3 concentrations at KRV station is reduced to a value of only -2 μgm^{-3} by using the 5th model layer concentrations as explained in chapter 2.3. The 5th model ~~-~~level predictions will be used for KRV in all analyses that follow.~~since the mean O_3 concentration increases with height.~~ Besides KRV also the Mediterranean KOP and OTL stations, as well as the rural ZAV site, are stations with comparatively high measured nighttime O_3 levels, which results in low overall bias for all hourly O_3 values for these stations (from -2~~3~~ to -7 μgm^{-3}). Namely, WRF-Chem model cannot capture well the profound nighttime O_3 reductions (shown also by Žabkar et al, 2013; Im et al., 2014a), which contributes to the overall over-prediction of hourly O_3 concentrations (from 10 to 36 μgm^{-3}) for stations with very low measured nighttime O_3 concentrations. For sites with highest positive bias in 1-hour O_3 concentrations (TRB, ZAG, HRA and ISK, with bias of 36 μgm^{-3}, 31 μgm^{-3}, 26 μgm^{-3} and 32 μgm^{-3}, respectively), this can also be partly explained by too high altitude of the stations in model orography (Tab. 1), since the mean O_3 concentration increases with height.

Looking at O_3 daily maxima (Fig. 4b), the under-predictions occur at alpine KRV (-16 μgm^{-3} for the lowest model level shown in Fig.4) and at three Mediterranean stations (OTL, NG, KOP; from -14 to -11 μgm^{-3}). For Mediterranean stations the underestimations of daily maxima are most probably due to inaccurate representation of costal processes in model, which are crucial for PBL height evolution and accumulation of pollution in the near ground air layers. For TRB station located in narrow valley of the very complex terrain that cannot be appropriately resolved in the current model topography, the model over-predicts O_3 daily maxima for 14 μgm^{-3}. For other sub-alpine stations the bias of O_3 daily maxima predictions is lower.

To some extent the previously mentioned model over-predictions of nighttime O_3 minima could be explained by model ~~incongruity~~ error in predicted NO_2 levels. When evaluating the primary pollutants one must be aware that in the model the instantaneous emissions are spread over an entire grid box, which results in underestimated emissions and concentrations close to the source regions and overestimated emissions and concentrations at rural locations adjacent to the source regions, and can thus cause a combined effect of negative and positive biases at urban and rural sites. Comparisons of WRF-Chem predicted NO_2 levels with measurements show that in spite of the high spatial resolution the concentrations of the small urban areas are insufficiently represented by the model (Fig. 4c). In Slovenia many towns are located in basins or very narrow valleys, usually poorly or even not resolved in model topography. Smoothed local emissions for these towns show significant underestimations of NO_2 concentrations (e.g. ZAG in Fig. 6). In combination with poorly~~deficiently~~ reproduced meteorological processes (calm and stable nighttime conditions in valleys and basins) this results in an underestimation of the O_3 loss by titration. This can explain the positive nighttime bias of O_3 found at these sites. The situation is better for bigger cities, located in wider basins, like LJ or CE (LJ; Fig. 6), while at rural sites NO_2 is either well simulated (e.g. MOH; Fig. 6), or slightly over-predicted due to increased emissions from adjacent urban area (e.g. ZAD; Fig. 6). The overall agreement of hourly NO_2 predictions with measurements was good for rural sites, while urban sites experienced under-predictions, which were highest for small cities, especially for NG (ME of -13 μgm^{-3}) and ZAG (ME of -14 μgm^{-3}).

Also ~~I~~interesting to discuss are the~~also~~ results for predicted PM10 concentrations (Tab. 3 and Fig. 4d), showing slight over-prediction of daily PM10 levels at all stations which is somewhat surprising due to the fact that nearly all current off-line and on-line coupled chemical transport models show large systematic PM10 underestimations. For example, within AQMEII exercise, where seventeen modeling groups from Europe and North America were brought together, running eight operational online-coupled air quality models over Europe and North America, the rural PM10 concentrations over Europe were underestimated by all models (model configurations) by up to 66% while for the urban PM10 concentrations the underestimations were even much larger (up to 75%) (Im et al., 2014b). The reason for slight over-prediction of PM10 levels could be to some extent attributed to the high model spatial resolution used in our study. Further, CORR for daily PM10 concentrations is rather low (0.34 and 0.37 for 1-day and 2-day forecasts, respectively; Tab. 3), which is partly due to the low temporal dynamics of measured daily PM10 concentrations during the analyzed time period (no recorded PM10 exceeding~~exceedances~~), and partly due to the simulated PM10 overestimations during the heat wave events. These over-predictions contributed also to the overall positive bias of predicted PM10 levels. As shown in Fig. 7 for two monitoring sites, there was a significant PM10 over-prediction simulated on June 10 (day 8 in Fig. 7), related to the pre-frontal advection of polluted air-masses coming from the north-western part of the domain D2 (coming from domain D1). The next significant PM10 over-prediction occurred during the first heat wave episode (June 17-22), when during the hot and low wind conditions (after June 17) the PM10 levels started to build up in the PBL over entire domain D2 (and over southwestern parts of domain D1), and reached the maximum concentrations in Slovenia again with prefrontal advection of polluted air masses. Both over-predictions contributed to an overall positive bias in forecasted PM10 concentrations. Detailed analyses showed that high concentrations in domain D1 originated from boundary conditions, and appear to be a consequence of overestimated advection of Saharan dust in MOZART model predictions. The increase in PM10 concentrations over Slovenia was also simulated during the prefrontal advection related to the cold front which terminated the next two heat wave events in July and August (days 56-57 and days 67-68 in Fig. 7), but during these days predicted PM10 levels were close to the measured PM10 concentrations.

3.3 Evaluation and comparison of different methods for O3 daily maximum predictions

In this section we want to answer the question: “~~–~~how accurate is the 1-hour O_3 daily maximum WRF-Chem forecast in comparison ~~with~~ to the statistical model prediction or ~~with~~ to persistence?”. According to Zhang et al. (2012a) statistical models are known to be generally more suitable for complex site-specific relations between concentrations of air pollutants and predictors. With appropriate and accurate predictors they have a higher accuracy as compared to deterministic models, which is, along with their ~~–beside the~~ computational efficiency their main advantage (Zhang et al., 2012a). Among the strengths of the deterministic models are~~is~~ that they give prognostic time- and spatially-resolved concentrations under typical and atypical scenarios, and can give scientific insights into pollutant formation processes (Zhang et al., 2012a). Furthermore, they also allow forecasts ~~also~~ for locations which are not monitored due to their complete spatial coverage. In spite of simplified descriptions of physical and chemical processes in the deterministic models and inaccuracies and uncertainties in model inputs (in particular the emissions), some previous studies already suggested that deterministic models can also have skills close to statistical forecasting tools (e.g. Manders et al., 2009). In addition to evaluation and comparison of O_3 daily maxima predictions with WRF-Chem and the statistical model, we decided to add a persistence model as a threshold for useful model prediction. Persistence works well under stationary conditions, but because it cannot handle changes in weather and emissions, fails at the beginning and at the end of the episodes (Zhang et al., 2010a). Regarding the extremes, models of all types are known to have problem to accurately predict them, while persistence predicts extremes with a 1-day (2-day) time lag.

Figure 8 compares discrete statistics site by site for 1-day and 2-day model predictions of 1-hour O_3 daily maxima. Similarly, Tab. 4 shows these statistics for all data with different thresholds applied (only for WRF-Chem and persistence, because a statistical forecast is not available for all stations), and separately for different types of stations (sub-alpine urban, rural, Mediterranean urban) with an available statistical forecast. Looking at ME persistence gives results close to zero as long as no threshold is applied, while with threshold of 140 μgm^{-3} (Tab. 4) ME of 1-day persistence (-10.2 μgm^{-3}) is very close to the WRF-Chem model for 1-day predictions (-11.2~~9~~ μgm^{-3}), and for 2-day predictions WRF-Chem (-13.8~~4.6~~ μgm^{-3}) already beats ~~the~~ persistence (-19.4 μgm^{-3}). Site-by-site comparison (Fig. 8) shows that for most stations the statistical forecast has a lower ME than WRF-Chem forecast, but there are also stations (ISK, HRA, LJ, KRV) with lower or equal ME for WRF-Chem than for statistical model, indicating the possible occurrence of atypical conditions not resolved by the statistical model. Looking at MAE and RMSE, at all stations except those with highest ME (~~KRV,~~ TRB, KOP) WRF-Chem outperforms the persistence already in the 1-day forecast. Among sites with available statistical forecast there are only two (OTL~~KRV~~, KOP) with WRF-Chem performing worse than the statistical forecast. CORR is one of the parameters that suggest how much the model is able to follow the true nature of processes regardless the possible bias. For almost all stations WRF-Chem shows higher CORR than persistence for 1-day and 2-day forecasts. Only at the KRV station the 1-day statistical forecast (CORR=0.80) somewhat outperforms WRF-Chem (0.74~~6~~), and at NG and KOP CORR for WRF-Chem and statistical model is very similar.

The Taylor diagrams in Fig. 9 show CORR together with the centered root-mean-square difference (RMSD) between model forecasts and observations, and the amplitude of their variations (standard deviation)~~,~~. The ideal ~~where ideal~~ model~~forecast~~ would ~~lie in the right~~

~~bottom corner~~have a correlation coefficient of 1 and a standard deviation equal to the observations, which means that it would be co-located with the black dot on the diagram. WRF-Chem gives higher CORR and lower RMSD for all types of stations, while standard deviation of WRF-Chem O_3 daily maxima predictions is underestimated and lower than for other model forecasts. The latter shows that the variability in WRF-Chem model predictions is not as large as that in observed values. MNBE in Fig. 8 has ~~a course~~ very similar results to ME. For all forecasts except WRF-Chem for the TRB site (with MNBE of 16%) which is located in a narrow valley that is not resolved in the current model resolution, MNBE is below the ±10-15%, which is the U.S. EPA (US EPA, 1991) recommended threshold for the models used for regulatory applications. For MNGE the U.S. EPA recommendation below 30-35% for O_3 applications is met by all forecasts, even in the case of 2-day persistence model. With exception of the MS and KOP sites MNGE is lower for WRF-Chem than for statistical forecast, while for KOP and KRV sites ~~with highest negative bias in WRF-Chem predictions,~~ 1-day persistence gives best results, followed by the statistical forecast ~~and~~ or WRF-Chem. Very similar are results for IOA with the range of 0-1, and score 1 indicating perfect model agreement with the observations. We can conclude that for most stations the WRF-Chem predictions are in line or even outperform the statistical model. With the exception of the stations with high bias due to very complex local topography (TRB) or~~,~~ unresolved coastal processes (KOP) ~~or alpine stations (KRV)~~, the WRF-Chem forecasts are more accurate than persistence. Here we recall that high negative bias in WRF-Chem forecast for alpine KRV site due to too low altitude of the station in model topography was compensated by taking prediction from the 5th model level.

The key requirement for a forecast system is to be able to predict O_3 concentration levels greater than a given threshold. Thus, in addition to the discrete evaluation just presented, also the contingency-table-based statistics are an important metric~~s~~ of forecast performance. Table 5 summarizes the categorical evaluation results for three different thresholds (120, 140, 160 μgm^{-3}) of elevated O_3 levels, which pose a greater risk to human health. Namely, it is important to take ~~should be taken~~ into account that results of categorical statistics are very sensitive to the threshold chosen, as well as to the overall pollution levels during the analyzed months. Equitable Threat Score (ETS) measures the fraction of observed and/or correctly predicted events, adjusted for the frequency of hits that would be expected to occur by random chance. Although this score takes into account the climatology it is not truly equitable. It ranges from -1/3 to 1, where the minimum value depends on climatology (it is near 0 for rare events). Looking at Tab. 5 ETS shows equal skill for WRF-Chem and statistical forecast, higher than persistence for the 120 μgm^{-3} threshold (1-day and 2-day forecast). ETS decreases with increasing the threshold for both WRF-Chem and statistical forecast, indicating the challenge that both models have to accurately predict the extremes. In the case of 140 μgm^{-3} threshold, WRF-Chem has the same ETS as persistence, higher than the statistical model for 1-day forecast, while for 2-day forecast WRF-Chem outperforms the statistical model, followed by persistence. In the case of 160 μgm^{-3} threshold persistance has the highest ETS for a 1-day forecast, followed by statistical model and WRF-Chem, while in the case of 2-day predictions, statistical model shows the highest skill and WRF-Chem the lowest. ~~Accuracy (A), which measures how often the forecasts are correct either above or below the threshold, increases with threshold level. Looking at 1-day forecast A is highest for statistical forecast at 120 μgm^{-3} threshold, for WRF-Chem forecast at 140 μgm^{-3} threshold, and in the case of 160 μgm^{-3} threshold applied, for persistence.~~ Another measure, the critical success index (CSI), is similar to ETS, except that it does not take into account the climatology of the events and thus gives poorer scores for rarer events. It measures the percentage of cases that are correctly forecasted out of those either forecasted or observed, and ranges from 0 to 1 (1 indicating the perfect forecast). Similar as ETS, CSI gives higher scores for persistence in the case of 1-day forecast for the higher two thresholds, while on the second day WRF-Chem or the statistical model already performs better. ~~There is~~ Bias (B) determines whether the same fraction of events are both forecasted and observed. A tendency of the statistical model and of WRF-Chem to under-predict O_3 threshold exceedances shows as a B below 1 for these two models. ~~a tendency of the statistical model and of WRF-Chem to under-predict O_3 daily maxima. This shows as a bias (B) below 1 for these two models, where B determines whether the same fraction of events are both forecasted and observed.~~ The false alarm ratio (FAR) that measures the percentage of forecast high O_3 events that turn out to be false alarms, gives highest skill for WRF-Chem, followed by statistical model and persistence. The probability of detection (POD) is a measure of how often a high threshold occurrence is actually predicted to occur, and is relatively low for WRF-Chem with respect to other models. ~~Another useful measure, the critical success index (CSI), measures the percentage of cases that are correctly forecasted out of those either forecasted or observed, and is for higher two thresholds best for persistence in the case of 1-day forecast, while on the second day WRF-Chem or the statistical model already perform better.~~

It must be noted, that in categorical evaluations systematic biases like those obtained with WRF-Chem for some stations (e.g. KOP~~, KRV~~), significantly impact the model performance. For example, if KOP ~~and KRV~~ station~~s~~ was~~were~~ excluded from categorical evaluations, WRF-Chem performance improved by means of all statistical measures (results not shown). If correction techniques, based on observations and the previous day's forecast (e.g., McKeen et al., 2005, 2007; Kang et al., 2008) were to~~would~~ be applied to correct the systematic biases, WRF-Chem forecasts might outperform the other two models even in categorical evaluations.

4 Summary and conclusion

A high resolution modelling system based on an on-line coupled WRF-Chem has been applied for numerical weather prediction and for forecasting air quality in Slovenia. In the study the evaluation of the forecasting system has been conducted for three summer months. Since the selection of physical or chemical parameterization schemes influences and possibly changes the outcomes, we decided to apply schemes which are well documented and have previously been used in other applications (e.g. AQMEII). Both 1-day and 2-day predictions of meteorological and air quality variables have been analyzed. The focus has been on O_3 as the only pollutant with recorded exceedances of legislation limit values during the three heat wave events in June, July and August 2013. WRF-Chem daily O_3 maximum predictions have also been compared to the operational statistical model and persistence forecasts to answer the question how skillful are the WRF-Chem model predictions compared to these two models.

1-day and 2-day WRF-Chem PM10 forecasts showed a very low bias. Exceptions were two events with significantly over-predicted PM10 levels due to prefrontal advection of polluted air masses from neighboring regions. Knowing that majority of the current chemical transport models show large negative biases in simulated PM10 concentrations, these results present a good starting point for studying the importance of aerosol feedbacks with realistic model aerosol concentrations, left for future research.

The overall agreement of WRF-Chem NO_2 forecast with measurements was good for rural sites, while urban sites experienced model under-predictions, which were highest for small towns. One important reason is that many small towns are located in basins or very narrow valleys, usually poorly presented in model topography. Smoothed local emissions result ~~show~~in ~~as~~ model underestimations of NO_2 concentrations for these towns. This in combination with insufficiently reproduced calm meteorological conditions in basins and valleys during the nighttime hours explains also WRF-Chem over-predictions of nighttime O_3 concentrations.

Evaluations of predicted 1-hour and 8-hour daily O_3 maxima, which are in the case of this pollutant of the highest interest, show good~~a nice~~ WRF-Chem model performance. Nevertheless, there are also stations which experience high over- or under-predictions of O_3 daily maximum levels. For Mediterranean sites the under-predictions of the daily maxima are most probably due to inaccurate representation of costal processes in model, which are crucial for the PBL height evolution and accumulation of pollution in the near ground air layers. For some sub-alpine stations the reason for the higher bias in O_3 daily maximum predictions is their location either at elevated mountainous or coastal regions, or in narrow valleys which cannot be appropriately resolved in the current model resolution - that impacts how accurately model simulates the local processes responsible for the level of local pollution. Comparisons of WRF-Chem O_3 daily maximum forecasts with persistence and with statistical model predictions show that with respect to some statistical parameters the deterministic WRF-Chem forecast can outperform the other two for both 1-day and 2-day predictions. For example, correlation coefficient shows highest skill for WRF-Chem model, confirming the importance of complex processes as taken into account in an on-line coupled Eulerian model. Further improvement of WRF-Chem forecasting skill could be obtained by applying one of the bias-correction methods in order to account for unresolved topographical and coastal effects, as well as emission patterns. Chemical data assimilation, although currently still in its infancy for online coupled meteorology-chemistry models (Bocquet et al., 2014), could in future also be used as an efficient method for improving prediction of chemical concentration fields. For WRF-Chem model a technical note on the implementation of the aerosol assimilation and a guidance for prospective users has been recently published by Pagowski et al. (2014).

Appendix A: Statistical measures

For i-th observed (O_i) and the corresponding modelled (M_i) value of variable, discrete statistical measures are calculated as follows:

Mean error:

$$ME = \frac{1}{N}\sum_{i=1}^{N}\left(M_i - O_i\right)$$

Mean absolute error:

$$MAE = \frac{1}{N}\sum_{i=1}^{N}\left|M_i - O_i\right|$$

Root mean square error:

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(M_i - O_i\right)^2}$$

Correlation coefficient:

$$r = \frac{\sum_{i=1}^{N}\left(M_i - \overline{M}\right)\left(O_i - \overline{O}\right)}{\sqrt{\sum_{i=1}^{N}\left(M_i - \overline{M}\right)^2\left(O_i - \overline{O}\right)^2}}$$

Index of agreement:

$$IOA = 1 - \frac{\sum_{i=1}^{N}\left(M_i - O_i\right)^2}{\sum_{i=1}^{N}\left(\left|M_i - \overline{O}\right| + \left|O_i - \overline{O}\right|\right)^2}$$

Mean normalized bias error:

$$MNBE = \frac{1}{N}\sum_{i=1}^{N}\frac{M_i - O_i}{O_i}\times 100$$

Mean normalized gross error:

$$MNGE = \frac{1}{N}\sum_{i=1}^{N}\frac{\left|M_i - O_i\right|}{O_i}\times 100$$

For categorical evaluation all model predictions are first classified into four groups (*a*, *b*, *c* and *d*):

a prediction is above, but observation is below the threshold
b prediction and observation are above the threshold
c prediction and observation are below the threshold
d prediction is below, but observation is above the threshold

Categorical statistics are calculated as follows:

Equitable threat score: $ETS = \frac{b - a_r}{a + b + d - a_r}$, where $a_r = \frac{(a+b)(b+d)}{a+b+c+d}$

Critical success index: $CSI = \frac{b}{a+b+d}$

Bias: $B = \frac{a+b}{b+d}$

False alarm ratio: $FAR = \frac{a}{a+b}$

Probability of detection: $POD = \frac{b}{b+d}$

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Table 1: *AQ monitoring sites.*

Monitoring site	Abbreviation	Type of zone	Altitude (m)	Model orography (m)	Model analysis height (m)	Pollutants
Celje	CE	Urban	240	300	313	O_3, PM10, NO_2
Hrastnik	HRA	Urban	290	540	552	O_3, SO_2
Iskrba	ISK	Rural	540	579	591	O_3, NO_2
Koper	KOP	Urban	56	72	85	O_3, PM10
Kovk	KOV	Rural	608	516	528	NO_2
Krvavec	KRV	Rural	1740	1272	1414	O_3
Ljubljana	LJ	Urban	299	287	300	O_3, PM10, NO_2,
Murska Sobota	MS	Rural	188	189	202	O_3, PM10, NO_2
Nova Gorica	NG	Urban	113	150	163	O_3, PM10, NO_2
Otlica	OTL	Rural	918	874	886	O_3
Sv. Mohor	MOH	Rural	394	254	266	NO_2
Trbovlje	TRB	Suburban	250	459	471	O_3, PM10, NO_2
Velenje	VEL	Urban	389	461	474	O_3, SO_2
Vnajnarje	VNA	Rural	630	468	480	NO_2
Zadobrova	ZAD	Rural	280	275	287	PM10, NO_2
Zagorje	ZAG	Urban	241	431	443	O_3, PM10, NO_2
Zavodnje	ZAV	Rural	765	678	690	O_3, NO_2

Table 2: S*tatistical scores for 1-hour values of 2m temperature (T2m), 10 m wind speed (W10m) and relative humidity (RH), and for daily average incoming solar radiation (SR). Shown are results for 1-day forecast, calculated separately for three measuring sites (LJ, NG, MS) and for 24 MET monitoring stations (ALL) during the 3-month period. In the case of temperature results for daily maxima are also shown.*

Variable	Station	NoCases	Mean	ME	MAE	RMSE	CORR
T2m 1h (°C)	LJ	2129	20.3	-1.6	2.3	2.9	0.91
	NG	2184	21.8	-1.1	2.1	2.5	0.94
	MS	2184	19.2	-2	2.3	2.8	0.95
	ALL	47836	18.7	-1.3	2.3	2.9	0.93
T2m max (°C)	LJ	89	26.5	-1.6	1.8	2.1	0.98
	NG	90	26.8	-3	3	3.3	0.96
	MS	90	26.2	-1.7	1.8	2	0.98
	ALL	1976	24.2	-2.1	2.7	3.2	0.97
W10m (m/s)	LJ	2129	1.5	0	0.7	1	0.58
	NG	2183	2.7	1	1.4	1.9	0.35
	MS	2184	2.3	0.4	1.1	1.4	0.53
	ALL	43378	2.4	0.8	1.4	1.9	0.36
RH (%)	LJ	2066	62	-2	8	10	0.85
	NG	2121	62	-1	12	15	0.75
	MS	2121	69	3	8	11	0.88
	ALL	48556	68	2	11	14	0.77
SR (W/m2)	LJ	90	276	19	31	43	0.84
	NG	90	278	4	32	43	0.77
	MS	90	273	15	26	37	0.9
	ALL	1710	273	16	35	49	0.77

Table 3: *Domain wide performance statistics for 1-day and 2-day forecast in* μgm^{-3}. *For different pollutants statistics for all hourly (hour), 8-hour averages (8h), 8-hour daily maximum (8h max), daily maximum (max) or daily average (day) concentrations are shown.*

		NoCases	Mean	ME	MAE	RMSE	CORR
O_3 (hour)	1 day	28391	94.8	14.5~~3.7~~	25.1~~5~~	32.~~5~~1	0.65
	2 day	28391	95.0~~4.2~~	14.5~~3.8~~	25.5~~8~~	32.5~~9~~	0.64
O_3 (8h)	1 day	28072	94.8~~1~~	14.6~~3.8~~	22.6~~9~~	28.1~~5~~	0.69
	2 day	28072	95.0~~4.2~~	14.6~~3.8~~	23.0~~3~~	28.5~~9~~	0.68
O_3 (8h max)	1 day	1157	111.5~~0.7~~	-0.1~~7~~	13.2~~6~~	16.5~~7~~	0.77
	2 day	1157	111.6~~0.9~~	-0.2~~1~~	13.7~~4.1~~	17.0~~4~~	0.75
O_3 (max)	1 day	1170	116.5~~5.8~~	-2.7~~3.4~~	13.3~~7~~	16.7~~7.1~~	0.81
	2 day	1170	116.6~~5.8~~	-3.1~~9~~	14.0~~4~~	17.5~~9~~	0.78~~9~~
NO_2 (hour)	1 day	26178	7.3	-5.1	7.5	10.8	0.3
	2 day	26178	7.5	-4.9	7.6	10.8	0.3
PM10 (day)	1 day	718	29.0	7.1	12.0	18.8	0.34
	2 day	718	29.1	7.2	12.0	19.1	0.37

Table 4: *Discrete evaluation of 1-hour daily maximum ozone predictions.*

Stations	Threshold, NoCases	Forecast	Mean (µgm^{-3})	ME (µgm^{-3})	MAE (µgm^{-3})	RMSE (µgm^{-3})	CORR	MNBE (%)	MNGE (%)	IOA
All	> 0 1170	F 1day	11~~5~~6.~~8~~5	-~~3~~2.6~~.4~~	13.3~~7~~	16.7~~7.1~~	0.81	-0.05	11.7~~2.0~~	0.86
		F 2day	116~~5~~.6~~8~~	-3.~~9~~1	14.0~~4~~	17.5~~9~~	0.78~~9~~	-0.~~7~~1	12.3~~6~~	0.84
		PER 1day	119.5	-0.4	15.8	21.1	0.65	1.6	14.5	0.81
		PER 2day	119.8	-0.4	21.7	27.7	0.39	2.8	19.6	0.65
	> 140 1102	F 1day	144~~3~~.1~~3~~	-11.2~~9~~	15.2~~7~~	17~~8~~.9~~4~~	0.52	-6.8~~7.4~~	9.5~~9~~	0.57
		F 2day	141~~0~~.4~~6~~	-13~~4~~.8~~6~~	16~~7~~.5~~1~~	19~~20~~.4~~0~~	0.42~~1~~	-8.6~~9.1~~	10.5~~8~~	0.48~~7~~
		PER 1day	145.0	-10.2	15.6	19.6	0.41	-6.5	10.0	0.52
		PER 2day	135.8	-19.4	24.76	29.2	0.31	-12.4	15.9	0.38
Sub-alpine urban with SF (LJ, HRA)	> 0 180	F 1day	115.3	1.1	10.7	14.0	0.84	3.4	11.1	0.91
		F 2day	115.4	0.8	12.0	15.2	0.80	3.5	12.2	0.88
		PER 1day	114.3	-0.3	16.7	21.7	0.64	2.2	16.5	0.80
		PER 2day	114.6	-0.3	21.9	27.8	0.41	3.9	21.6	0.65
		SF 1day	114.0	-0.5	11.9	15.7	0.81	1.6	11.2	0.88
		SF 2day	116.2	0.6	13.4	17.1	0.75	3.2	12.7	0.84
Rural with SF (MS, ISK, KRV, OTL)	> 0 360	F 1day	117.6~~5.2~~	-5.6~~8.1~~	13.3~~4.6~~	16.3~~7.6~~	0.80	-3~~5~~.0	10.8~~1.7~~	0.86~~5~~
		F 2day	117.4~~4.8~~	-6.4~~8.8~~	14.2~~5.5~~	17.4~~8.9~~	0.76~~7~~	-3~~5~~.4	11.4~~2.4~~	0.84~~1~~
		PER 1day	123.6	-0.3	15.0	20.7	0.65	1.4	13.1	0.81
		PER 2day	124.1	-0.4	21.6	27.8	0.37	2.4	18.5	0.64
		SF 1day	121.5	-2.9	15.0	19.4	0.74	-0.7	12.2	0.83
		SF 2day	122.9	-1.8	15.8	20.5	0.67	0.5	13.2	0.79
Mediterranean urban with SF (KOP, NG)	> 0 179	F 1day	123.5	-11.8	17.4	22.5	0.76	-6.9	12.5	0.80
		F 2day	124.5	-11.2	17.2	21.8	0.77	-6.5	12.4	0.82
		PER 1day	135.9	-0.5	17.4	23.0	0.68	1.2	13.8	0.83
		PER 2day	136.0	-0.2	25.2	31.5	0.41	2.8	19.7	0.66
		SF 1day	129.3	-7.0	15.9	20.7	0.75	-3.6	11.6	0.83
		SF 2day	131.6	-4.5	15.6	20.4	0.74	-1.6	11.6	0.84

Table 5: *Categorical evaluation of 1-hour daily maximum ozone predictions for different thresholds, calculated for 8 monitoring sites with available statistical forecast.*

Threshold	Forecast	ETS~~A~~	CSI	B	FAR	POD	a	b	c	d
> 120	F 1day	0.42~~78~~	0.63~~1~~	0.81~~76~~	0.13	0.70~~66~~	39~~5~~	253~~39~~	313~~7~~	107~~21~~
	F 2day	0.39~~77~~	0.61~~59~~	0.79~~75~~	0.14	0.68~~4~~	41~~37~~	245~~33~~	303~~7~~	115~~27~~
	PER 1day	0.31~~74~~	0.59	0.99	0.25	0.74	91	267	249	93
	PER 2day	0.17~~64~~	0.49	1.00	0.34	0.65	123	235	209	124
	SF 1day	0.42~~80~~	0.67	1.02	0.21	0.81	67	257	243	61
	SF 2day	0.38~~77~~	0.65	1.03	0.23	0.80	77	264	225	66
> 140	F 1day	0.40~~84~~	0.50~~47~~	0.64~~59~~	0.15~~4~~	0.551	19~~7~~	111~~03~~	490~~2~~	92~~100~~
	F 2day	0.37~~82~~	0.47~~4~~	0.66~~0~~	0.19~~8~~	0.53~~49~~	25~~2~~	108~~99~~	476~~9~~	95~~104~~
	PER 1day	0.40~~82~~	0.53	1.00	0.31	0.69	62	141	435	62
	PER 2day	0.19~~72~~	0.35	1.00	0.48	0.52	97	106	391	97
	SF 1day	0.30~~79~~	0.43	0.73	0.29	0.52	40	99	398	91
	SF 2day	0.30~~79~~	0.43	0.70	0.27	0.51	37	98	403	94
> 160	F 1day	0.19~~91~~	0.22	0.38~~7~~	0.34~~2~~	0.25	~~9~~10	19	626~~7~~	57
	F 2day	0.17~~91~~	0.20~~1~~	0.34~~0~~	0.35~~26~~	0.22	9~~6~~	17	619~~22~~	59
	PER 1day	0.40~~92~~	0.45	1.00	0.38	0.62	29	47	595	29
	PER 2day	0.22~~88~~	0.28	1.00	0.56	0.43	43	33	572	43
	SF 1day	0.23~~90~~	0.27	0.49	0.35	0.32	13	24	539	52
	SF 2day	0.25~~90~~	0.29	0.63	0.41	0.37	19	27	540	46

1

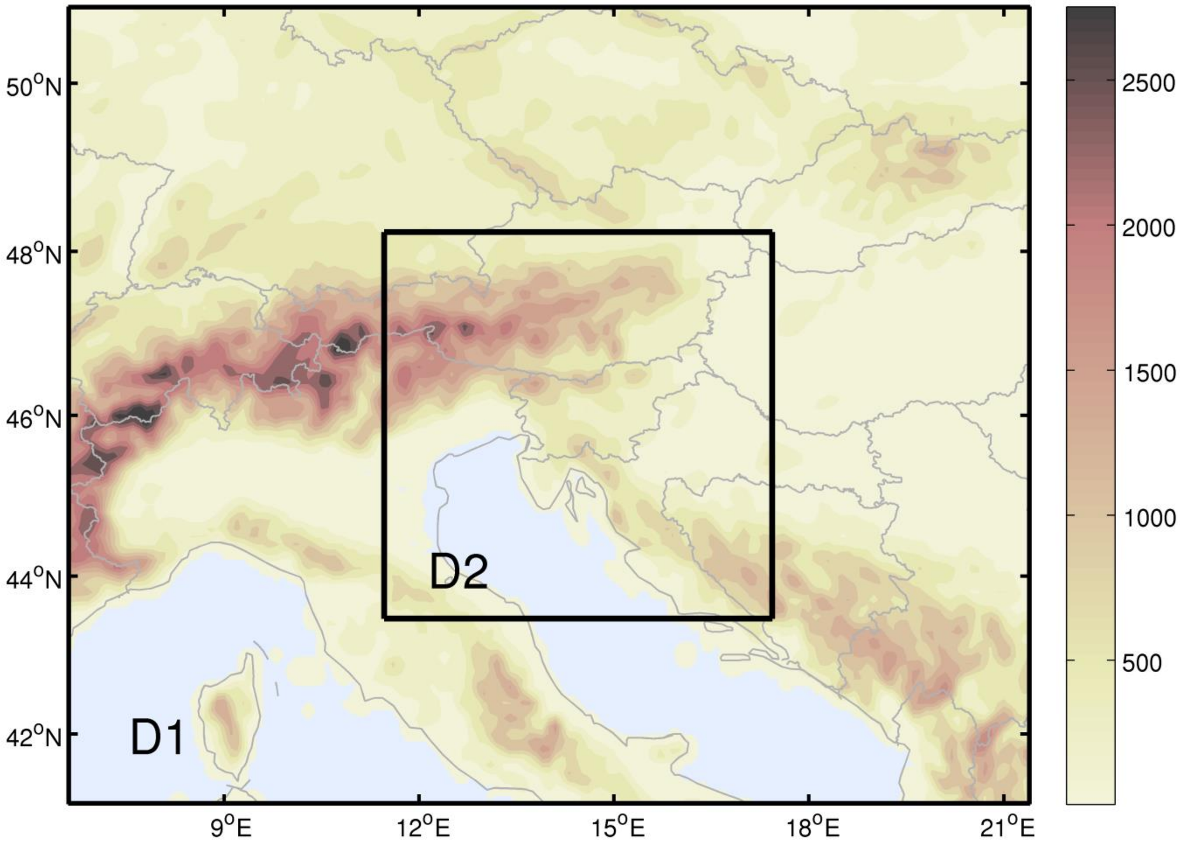

Figure 1: Modelling domains (D1, D2) used in WRF-Chem RT-AQF system. Orography (in meters) is shown in resolution of D1 domain (11.1 km).

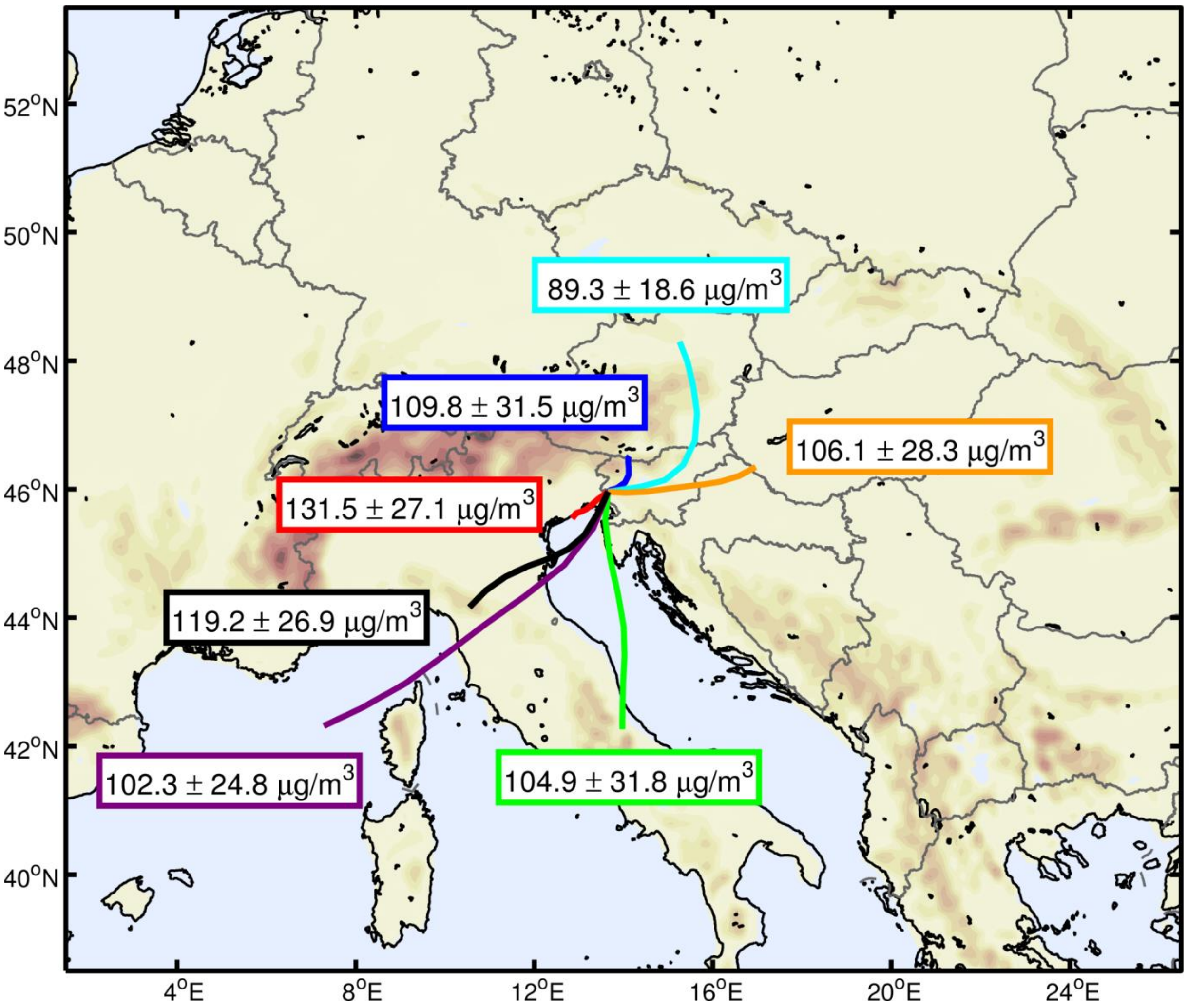

Figure 2: Example of ozone analysis for the Nova Gorica (NG) monitoring site (average daily maximum ± standard deviation) for 7 clusters of similar trajectories, as used in the statistical ozone daily maximum forecast for the NG station.

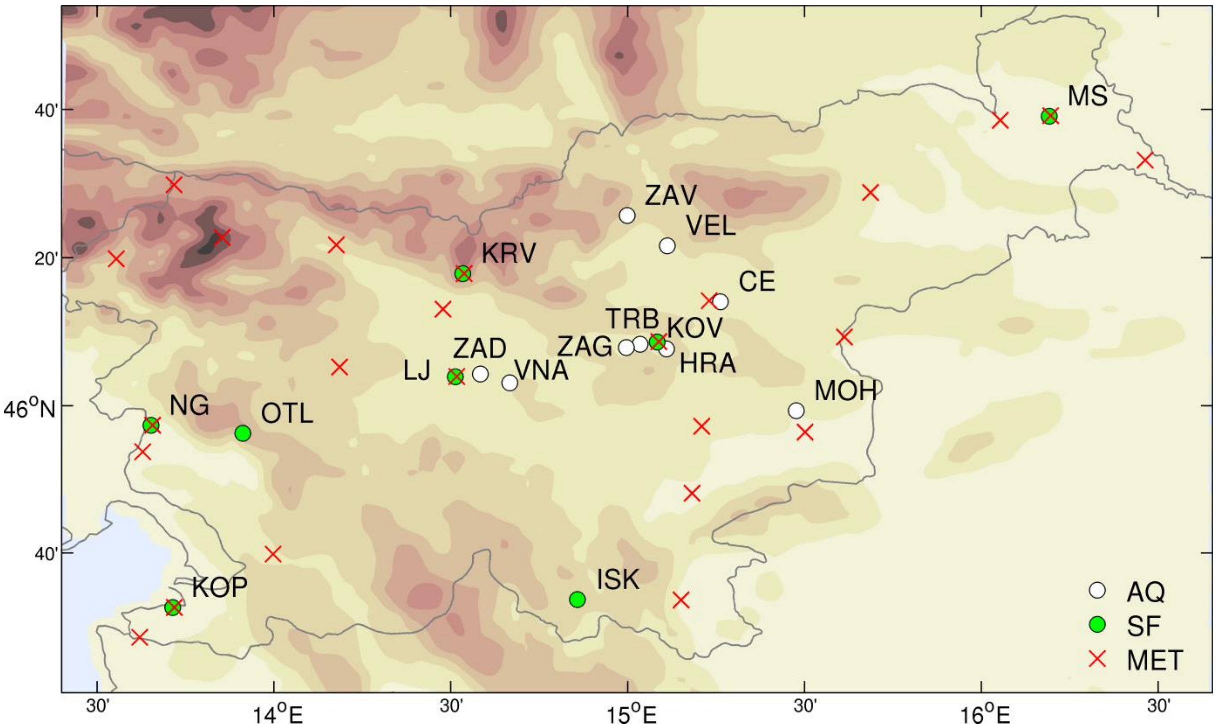

Figure 3: Locations of monitoring stations used in evaluation of air quality variables (AQ stations; shown are also station abbreviations) and meteorological variables (MET stations). Green dots indicate measuring sites with available ozone daily maximum statistical forecast (SF). For the meaning of abbreviations of AQ sites see Tab. 1.

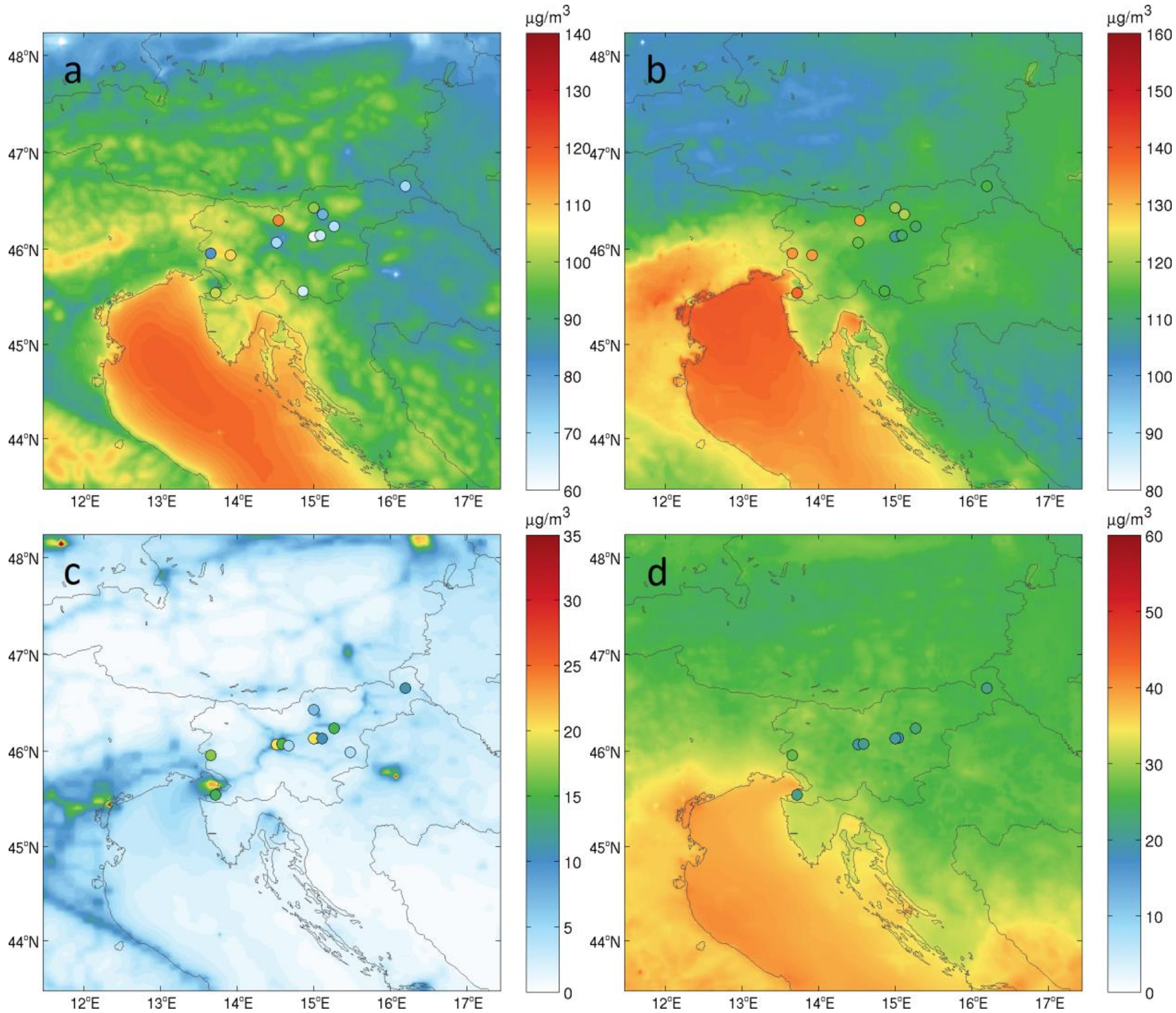

Figure 4: 3-month average 1-day predictions of a) hourly O_3, b) O_3 daily maximum, c) hourly NO_2, and d) daily PM10concentrations for the first model layer, overlaid with measurements.

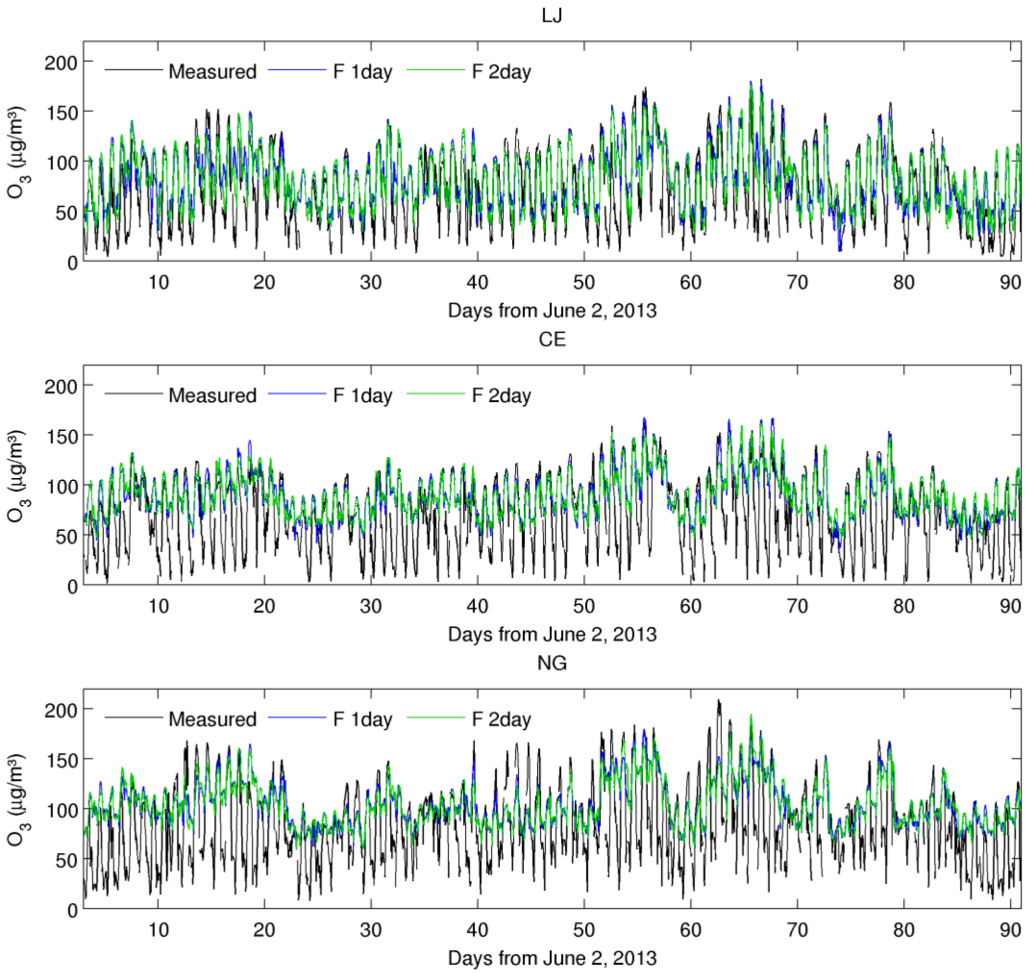

Figure 5: Time evolution of hourly ozone concentrations for 1-day (F 1day) and 2-day (F 2day) WRF-Chem predictions and measurements for some stations during the 3-month period. (continued)

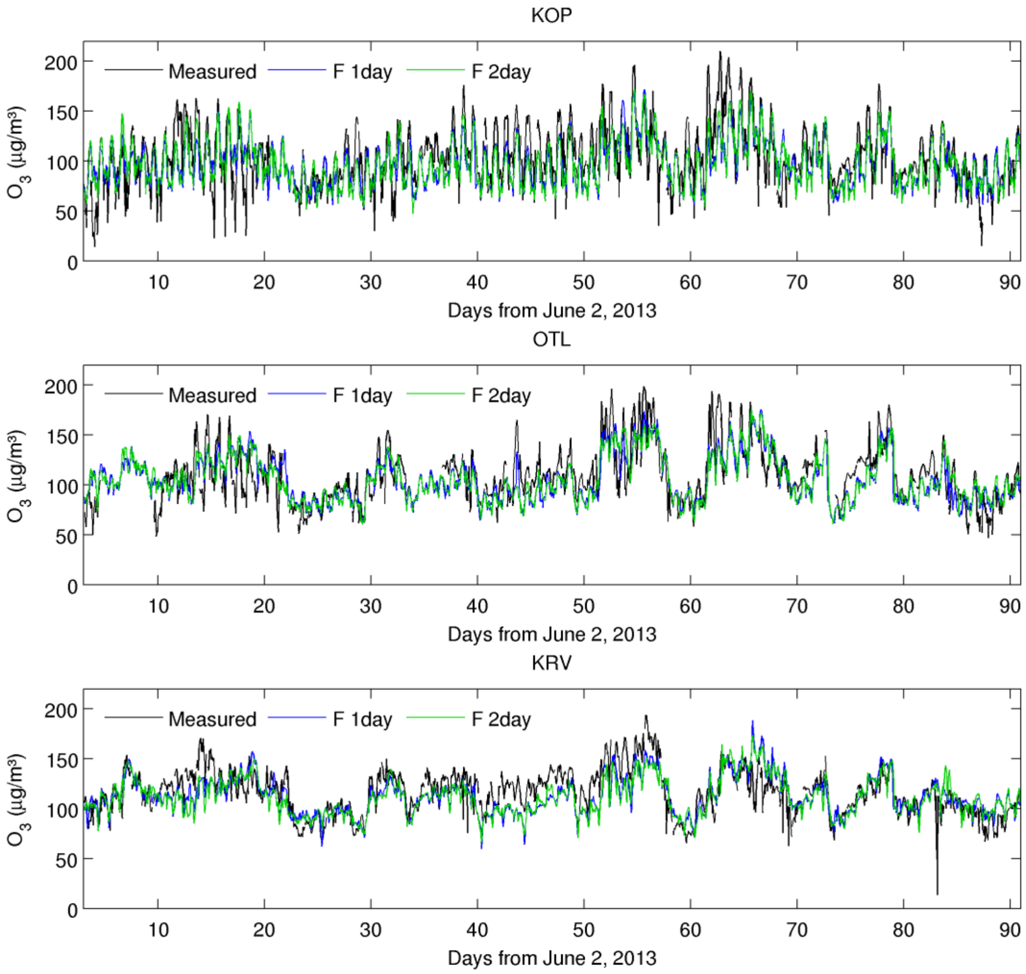

Figure 5: (continued)

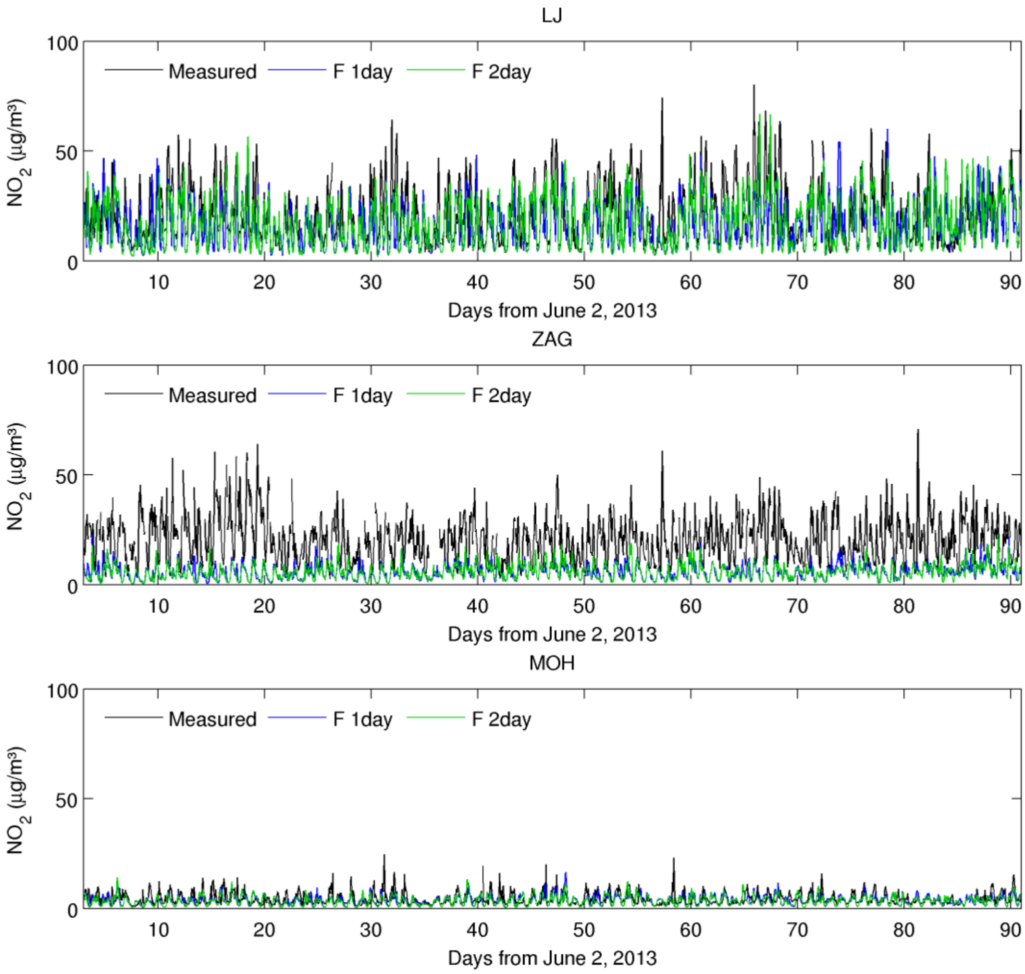

Figure 6: The same as Fig. 5 but for NO_2 at LJ, ZAG and MOH stations.

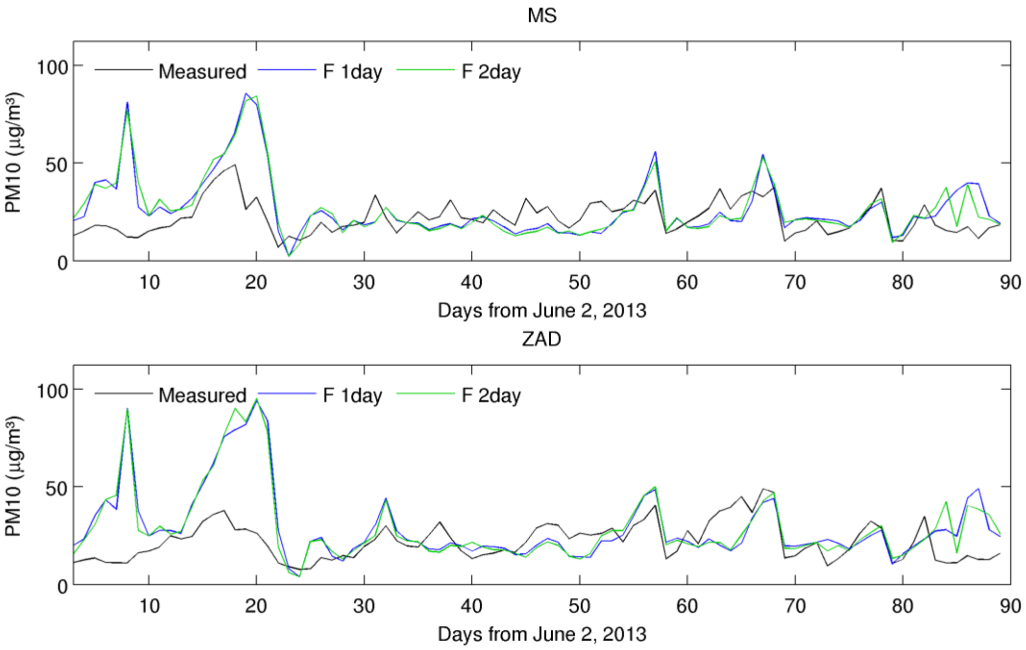

Figure 7: The same as Fig. 5, but for daily PM10 concentrations at MS and ZAD stations.

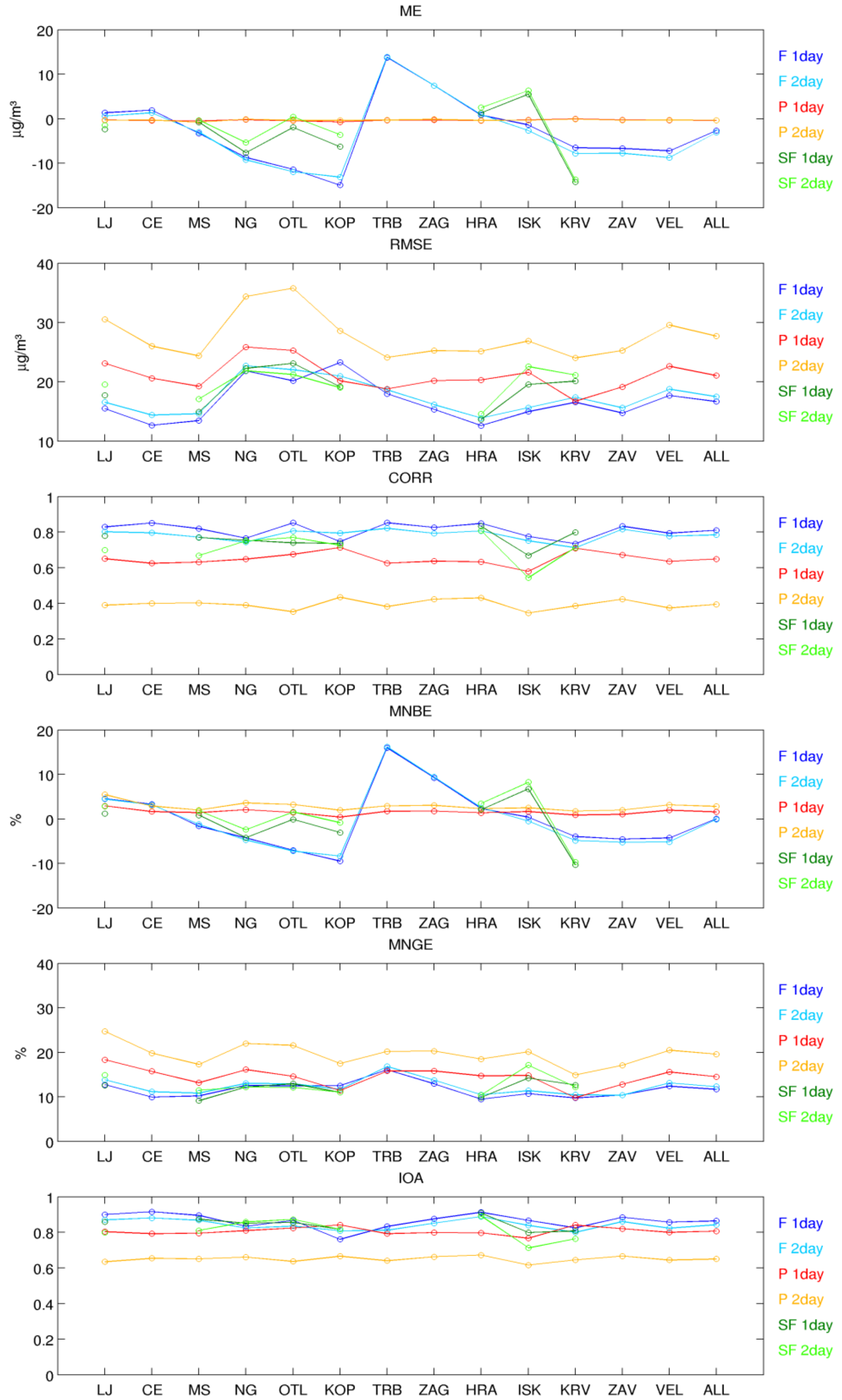

Figure 8: Site-by-site comparison of discrete statistics for 1-day and 2-day WRF-Chem (F 1day, F 2day), statistical (SF 1day, SF 2 day) and persistence model (P 1day, P 2day) predictions of ozone daily maxima during the 3 analyzed summer months.

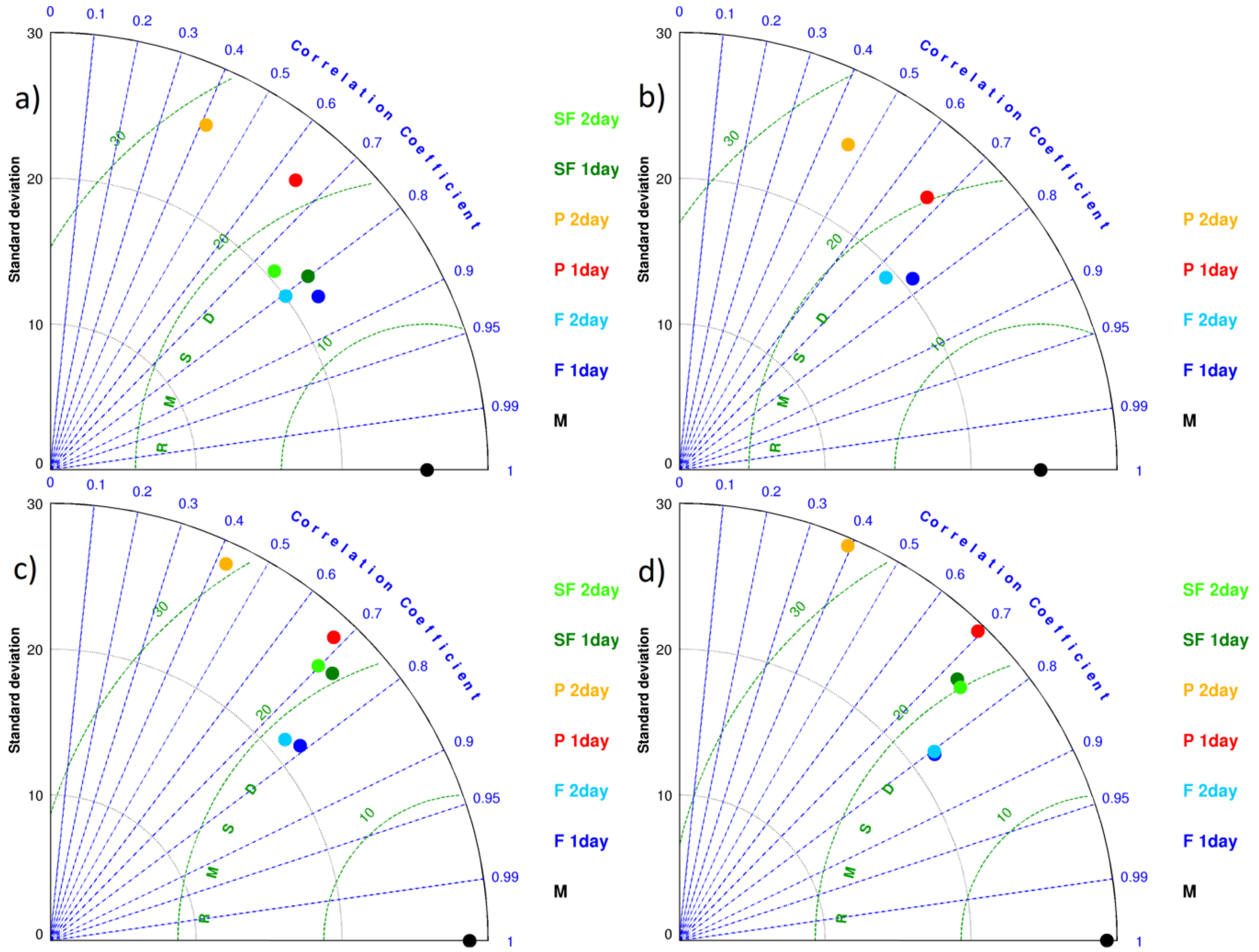

Figure 9: Taylor diagrams comparing 1-day and 2-day ozone daily maximum statistical forecast (SF), persistence (P) and WRF-Chem forecast (F) for a) sub-alpine urban stations with SF (LJ, HRA), b) sub-alpine urban stations without SF (CE, TRB, ZAG, VEL), c) rural stations with SF (MS, ISK, KRV, OTL) and d) Mediterranean urban stations (NG, KOP).