Temperature Dependent Initial Chemical Conditions for WRF-Chem Air Pollution Simulation Model

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Abstract. Air pollution is a health hazard that has been brought to public attention in the recent years, due to the widespread networks of air quality measurement stations. The importance of the problem brought the need to develop accurate air prediction models. The coupled meteo-chemical simulation systems have already been demonstrated to correctly predict the episodes of high pollution events. Due to the complexity of these models, which simulate the emissions, interactions and transport of pollutants in the atmosphere, setting up the correct parameters tailored for a specific area is a challenging task. In this paper we present an exhaustive analysis of the historical air pollution measurements, a detailed evaluation of an existing WRF-Chem based predictive model and propose an approach for improvement of that specific model. We use a specific temperature-dependent way of scaling the initial chemical conditions of a WRF-chem simulation, which leads to significant reduction of the bias by the model. We present the analysis that led us into these conclusions, the setup of the model, and the improvements made by using this approach.

Keywords: WRF-Chem · Air pollution · Prediction · Simulation · Initial conditions · PM10 modeling · Temperature dependent

1 Introduction

The problem of air pollution has been in the focus of the public interest in our country in the past few years. The enabling reason for that was the availability of air pollution monitoring stations with publicly available data in real time [1, 2]. The data measured are showing consistently high pollution with PM10 and PM2.5 during winter, often with high pollution events with measurements up to more than 10 times the limit of what is considered a safe concentration [3]. These episodes of extreme air pollution have increased the public interest in monitoring the pollution levels in real time, for the purpose of personal planning of outdoor activities and minimizing exposure to hazardous air.

A step towards in the direction of more effective planning and preparedness for minimizing exposure to hazardous air would be the creation of a reliable model for air pollution prediction. Few such systems have appeared in the recent years, with some of them still in the research phase, while others have been brought to the stage of user-accessible air quality prediction apps. These systems are based on a variety of different models, like WRF-Chem, SILAM, CAMS and others [4-8]. In this paper we

are going to take a more detailed look in the WRF-Chem model and work on its improvement.

Most of these models, while working relatively correctly throughout most of Europe, struggle to correctly predict the pollution concentrations in the area of the Balkans. The discrepancy is mostly due to two factors, the first one is the complex orography, which often causes temperature inversions in the valleys of the larger towns, where localized extreme high concentrations of pollutants occur. The second one is the use of wood and other solid fuels for domestic space heating which is a large non-industrial pollution source in these areas, further amplified by the orographic effect. These domestic emissions are hard to be modeled properly and are endemic to these areas. The pan-European prediction models focusing mostly on industrial and transportation emissions often fail to integrate these domestic emissions properly on a local level. The combination of the two factors often leads to these models making incorrect predictions of the pollution in the larger urban areas in the Balkans.

The solution to the problem caused by orography is increasing the simulation grid density around the urban areas, and thus improve the localized predictions. This is a fairly straightforward task, but due to the significantly increased computational demand of the models, we are not be able to test it in our experiments for this paper.
The second problem is a more challenging one, and we will try to tackle it in our research. Our hypothesis is that the total pollution emissions during the winter have a strong component originating from the domestic heating. As we expect that the cumulative solid fuels combustion intensity (and therefore the pollutants emissions) depends on the air temperature, we propose a temperature dependent scaling of the pollution emissions. We will show that the improved accuracy of estimating the pollutants emissions will cause an overall improvement of the correctness of the model.

The base model we will be improving is a WRF-Chem model, which has al-ready been developed for our country [9] We will present the setup of that model in Section 2, which we will refer to as 'the initial model'. In Section 3 we will analyze the data from air pollution measurements and try to get insight and find patterns that will define our model. In Section 4 we elaborate the improved model and present our contribution, based on the combined findings of the previous two chapters. Section 5 presents the results of the evaluation of the model, with a discussion on the obtained improvements. In the final section, we present our conclusion and further improvements possible in future.

2 WRF-Chem Model

The core meteo-chemical model setup we use is based on the WRF-Chem model. Its core is the Weather Research and Forecasting (WRF) Model, a mesoscale numerical weather prediction system designed for both atmospheric research and operational forecasting applications[10]. The WRF-Chem extension simulates the emission, transport, mixing, and chemical transformation of trace gases and aero-sols simultaneously with the meteorology [11].

We use the setup of the simulation as proposed for a similar model [9], which we are going to describe in this section thoroughly. The initial and boundary meteorological conditions are prepared by WPS (WRF Preprocessing System) and the initial chemical conditions are calculated by PREP-CHEM-SRC[12]. A crucial part of their setup is the addition of user-defined data, where they provide the grid of yearly emissions on the entire territory of the Republic of North Macedonia, aggregated by economic sectors and sources. This grid of sector aggregated pollution sources is compiled by the MOEPP [13, 14], and with the addition of these data, they are able to correctly predict the episodes of high pollution in their evaluation domain.

The meteorological module of the model is based on the Advanced Research WRF (ARW) core developed by the NCAR [15]. ARW is a non-hydrostatic mesoscale model with a compressible equation on C-grid staggering, using a terrain following hydrostatic pressure-vertical coordinate. It conserves mass, momentum, dry entropy, and scalars using a flux-conserving form for all prognostic equations. The numerical methods utilize third-order Runge-Kutta split-explicit time differencing, together with higher-order advection.

The Weather Research and Forecasting model ARW is coupled with chemistry (WRF-Chem), as an efficient and flexible system for weather and air quality forecast. WRF-Chem was developed at NOAA/ESRL (National Oceanic and Atmospheric Administration/Earth System Research Laboratory) [16] and updated by incorporating complex gas-phase chemistry, aerosol treatments, and photolysis scheme [17]. The air quality component of WRF-Chem is fully consistent with the meteorological component; both components use the same transport scheme (mass and scalar preserving), the same horizontal and vertical grids, the same physical schemes for subgrid scale transport, and the same time step for transport and vertical mixing.

In the initial model, WRF-Chem v.4.0 released at NCEP in June 2018 is employed as a basis for the chemical transport forecast. The Regional Acid Deposition Model version 2 (RADM2) chemical mechanism for gas-phase chemistry schemes [18] is without kinetic pre-processor (KPP). In addition, the air quality modeling system includes also the Modal Aerosol Dynamics Model for Europe (MADE) [19], coupled with the SORGAM (Secondary Organic Aerosol Model) parameterization [20] for PM10 simulations.

The urban emissions are derived from daily inventories built in the emission preprocessor PREP-CHEM-SRC. The PREP-CHEM-SRC is a tool developed to estimate the emission fields of aerosols and trace gases from biomass burning (by satellite observations and inventories), biogenic, urban-industrial, biofuel use, and volcanic and agricultural waste burning sources for regional and global transport models based on available inventories and products [21]. The main objective of PREP-CHEM-SRC is to estimate the emission fields of the main trace gases and aerosols for use in atmospheric-chemistry transport models, such as WRF-CHEM.

In this work, we are using the global anthropogenic emission data for gaseous species (CO2, CO, NOx = NO + NO2, SO2, NH3) compiled and distributed by the Emission Database for Global Atmospheric Research (EDGAR) system (http://www.mnp.nl/edgar) [22]. The EDGAR-HTAP project compiled a global emission data set with annual inventories for CH4, NMVOC, CO, SO2, NOx, NH3,

PM10, PM2.5, BC, and OC and covering the period 2000–2005 for 10 aggregated sectors and on a global 0.1° × 0.1° resolution. The global emission data comes from the Reanalysis of the Tropospheric (RETRO) (http://retro.enes.org) (0.50 × 0.50) monthly 1960–2000 emission base and GOCART background emission data. The model uses anthropogenic emissions from a number of global and regional inventories, biomass burning emissions from the Global Fire Emission Database, and biogenic emissions from Model of Emissions of Gases and Aerosols from Nature (MEGAN) [23]. This data set is inserted in PREP-CHEM-SRC to estimate the emission fields over the user-specified simulation domain.

Additionally, a mobile emission inventory in the urban areas in Macedonia is added, with special emphasis on the city of Skopje. The mobile emission implemented in the system represents an emission inventory updated by the Ministry of Environmental and Physical Planning (MOEPP). The data emissions distributed by the GNFR sectors with a grid resolution of 0.1° × 0.1° lat/long are part of the Central Data Repository of European Environment Information (EIONET) and Observing Network of Long-Range Transport and Pollution Convention (CLRTAP). The inventory data set represents emissions in kt (kilotons) per year for each grid cell by chemical species (including CO and NOx). The emission rates and the coordinates are positioned in the central point of each given grid. The emissions are provided using the gridded mobile inventory data sourced by the MOEPP with a resolution of 0.1° × 0.1° lat/long with a surface area of about 11.10 km × 8.539 km or 94.78 km2 approximately which corresponds at 40° latitude. The domain-averaged emission rates of CO and NOx for the central point of each grid box are then calculated by the given emission rates from the four adjacent points, using a bilinear interpolation method.

The system employed four single model configurations defined on a Lambert projection. The basic numerical integration is performed with 5-km horizontal grid resolution centered at 41.55° N, 21.45° E and covers North Macedonia, with parts of Serbia, Bulgaria, Albania, and Greece. The grid network contains 70 × 70 grid points in both the east-west and north-south directions. The vertical grid in the model is composed of 35 levels from the surface to about 30 km with 10 levels within 1 km above the model surface.

The model is initialized by the real boundary conditions using NCAR-NCEP's final analysis (FNL) data [24] having a spatial resolution of 0.25° × 0.25° (~ 27.7 km × 27.7 km) and a 6-h temporal resolution or NCEP GFS data with the same spatial resolution.

The described setup is what we will later refer to as the initial model and we will use this as the referent model that we will further improve [19]. Although it is a relatively good model for predicting the peaks of urban air pollution in winter, the model as described shows a temperature dependent bias (as shown in Fig. 4). The cause for this bias will be presented in the next section, following with our proposed improvements.

3 Analysis of Measured Pollution Data

In order to understand the distribution of the air pollution and find patterns of correlation, we analyzed the publicly available pollution measurements data. The sources we used are a combination from the pollution monitoring stations operated by the MOEPP and the ones from the various other networks (crowdsourced sensors, experimental and research sensors). The available data was measured over a time period of few years and it is publicly available [25].

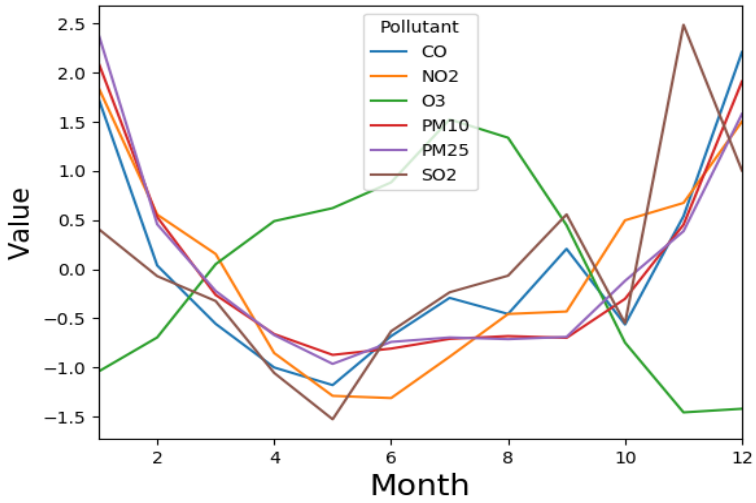

Fig. 1. Normalized monthly distribution of various pollutants

The data we were mostly interested in were the PM_{10} measurements. Most of the measurement stations have sensors that measure this parameter, and the fact that it is a pollutant that often gets the public attention due to the extreme measured values in winter, makes it the most interesting parameter to analyze and predict. The preprocessing of the data we did was minimal: removal of negative values and removal of values above 1300 ug/m^3 for the PM_{10} and $PM_{2.5}$. The upper limit was set at this value, as it is slightly above the official confirmed record high measured value, for which the initial model has been specifically evaluated for [9]. Therefore, the values above were considered outliers. We should mention here that we should be very careful with the filtering of outliers, as most of the standard outlier filters would likely fail by removing the rare extremely high values of air pollution, and might label them as outliers.

The rest of the pollutants were filtered with the threshold of 5 sigma values of the data. This is larger than the standard of 3 sigmas, due to the fact that the distribution here is not Gaussian: it is significantly skewed, with a cutoff below 0. Another reason we were not very interested in a more thorough outlier removal is that we only intend to use the data in an aggregated form, by taking monthly and hourly averages over the entire available data. The eventual outliers would not seriously affect the conclusions

that we would draw from the aggregated data, therefore we consider that the effort to remove the outliers in this case would be unnecessary for our purpose.

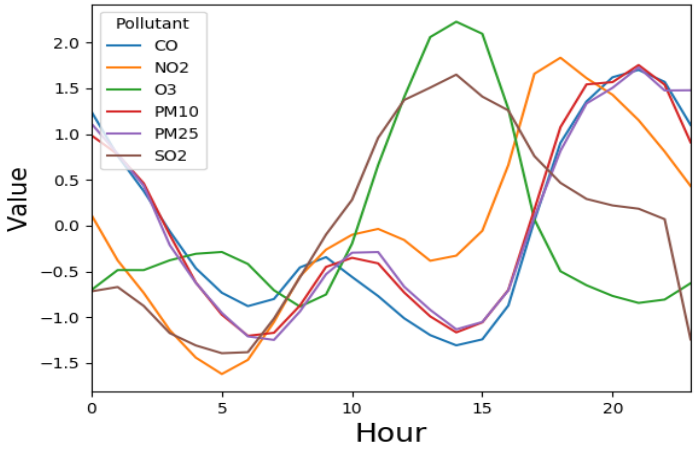

Fig. 2. Normalized hourly distributions of various pollutants in January

The plots that are crucial in improving the model can be seen in Fig. 1 and Fig. 2. Figure 1 shows the average monthly distribution of various pollutants. As we can see, for almost all of the pollutants, the peak concentrations occur in winter, while the measured pollution in summer was relatively low. The only exception here are the tropospheric ozone concentrations, which show a peak in summer. This is related to the increased solar radiation, and the atmospheric processes that create the ozone out of the oxygen. We can confirm that hypothesis in Fig. 2 and Fig. 3, where the ozone concentrations are the highest around noon.

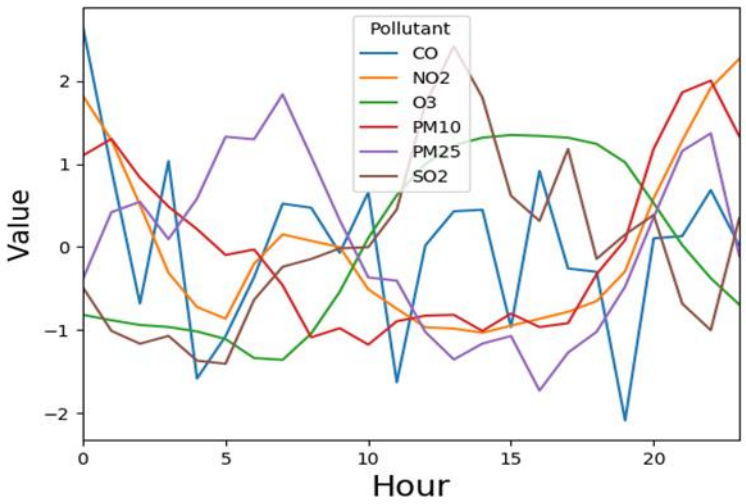

Fig. 3. Normalized hourly distributions of various pollutants in July

Figure 2 shows a bimodal distribution of the rest of the pollutants, except for SO2, for which we determined that the data quality is low and therefore we would not analyze it. The bi-modal distribution of PM10, PM2.5, CO and NO2 has two peaks: a lower one in the morning hours, corresponding with the early commute, and a higher one in the late afternoon, starting with the evening commute and continuing with the sharp increase, peaking before midnight. We conclude that the morning peak is caused by the traffic pollution and partly due to the home heating. The process of pollution caused by home heating is more pronounced in the late evening: at these hours, the traffic calms down from the evening commute, and most of the population is at home. Given that a majority of the population uses solid fuels for space heating [26], this is a plausible hypothesis [27].

Another hint to this hypothesis is the plot in Figure 3. Here we present the data from July in a similar way as in Fig. 2. However, we do not see such a pronounced bimodal distributions with large peaks over night. All of the other pollution sources, like traffic and industry are present in July, however, the pollution from domestic heating is lacking. This supports the hypothesis that the combustion of solid fuels for space heating is a significant cause for the increased pollution at winter. In the next chapters, we will propose a modification of the setup for the WRF-Chem model presented in the previous chapter, and will experimentally test a model that takes modified initial chemical conditions (pollutants emissions) based on the temperature.

4 Temperature Dependent Model

All the data we have seen so far points towards the need of a temperature dependent model for the initial chemical conditions. Both the behavior of the measured air pollution, and the temperature-dependent bias of the model are strong indicators for that. We propose that by scaling the initial chemical conditions by the predicted temperatures over our simulation domain, we would expect improvements in the predictions. A concrete proposal for a temperature dependent model will be presented in this chapter.

If we take the average value of PM_{10} at each degree of Celsius predicted by our model and subtract the average measured value at the stations for the same temperature, we will get a measure that represents the average temperature dependent bias of the model. This is shown in Figure 4, where we present the measured and simulated data for the first half of 2019 over all of the measurement points in the country (X-temperature in Celsius, Y- Bias). The simulated data is sourced by the initial mode, running online for day to day predictions [9]. We can notice that the initial setup of the model is highly temperature biased, namely, there is a large underestimation of the pollution at low temperatures. The temperature dependent bias seems to have a negative linear dependence by the temperature, but only at temperatures below 15 degrees Celsius. In temperatures above that point, the model does not show a temperature dependent bias.

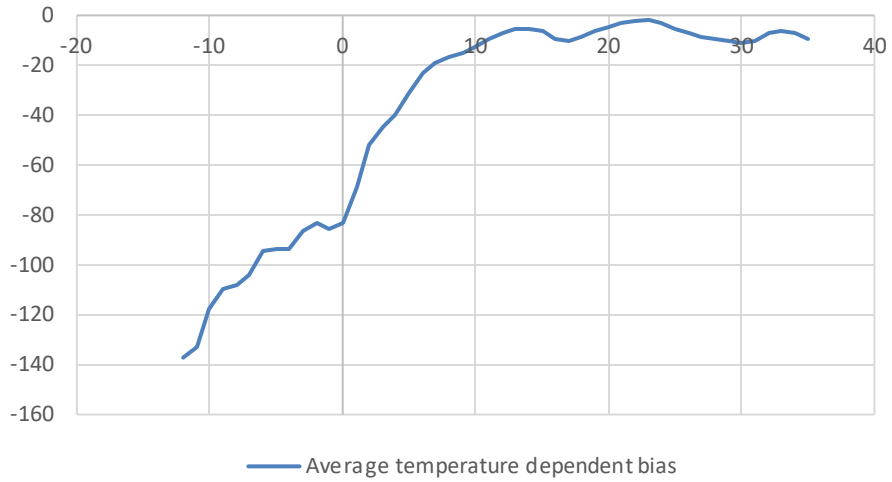

Fig. 4. Average temperature-dependent bias of the initial WRF-Chem setup over all measurement points

The authors of [9] describe that the user defined emissions were defined on a total yearly basis, and were fed to the model indiscriminately of the season. This deviates from what we have presented in Section 3, where a seasonal variation was shown. As we have explained, we expect that the seasonal variation is due to the hypothesis we postulated for domestic space heating emissions. Namely, these emissions occur almost exclusively in winter, and are expected to be temperature dependent. The negative temperature dependence may be explained by the fact that the heating intensity, and therefore the combustion of solid fuels is higher at lower temperatures.

In order to improve the temperature dependent bias of the model seen in Figure 4, we will do a modification in the initial chemical conditions to include temperature dependent scaling. The scaling is done only in the data provided by the MOEPP for urban mobile emissions. More specifically, we only scale the data from the sector of "other stationary combined" emissions, which contains, and mostly consists of the emissions coming from domestic heating sources.

The scaling of the emissions is done in the following way: for each emission cell of the grid that contains an urban area, we multiply the "other stationary combined" source by a factor F. The factor depends on the average predicted temperature for that cell box for the period we intend to run the simulation. The average temperature for the cell box is calculated by averaging the predicted temperatures for all of the simulation nodes within that cell, over the entire period we intend to do our simulation run. The visualization of the cell boxes where the pollution is given, and our simulation nodes from which we get the temperatures are given in Figure 5.

Once the average temperature is calculated, we need to calculate the scaling factor F. For average temperatures above 15 degrees Celsius, F=0.1, and for temperatures

below that point, we increase F by 0.15 for the drop of each degree Celsius below 15 degrees Celsius. The formula for scaling is the following one:

$$F=0.1,\ T>=15C; \tag{1}$$

$$F=0.1+(0.15*\ (15C-T)),\ T<15 \tag{2}$$

These modified user defined emissions are fed into the PREP-CHEM-SRC program, and the rest of the simulation workflow is unchanged from the one presented in Chapter 2. With this approach, we only change the emission that come from the domestic heating sources, by making them follow a temperature-dependent formula which is hypothesized to match the reality. The reasoning behind this is that the domestic heating is intensified when the temperatures drop, and is almost non-existent when the average temperatures are above 15 degrees celsius.

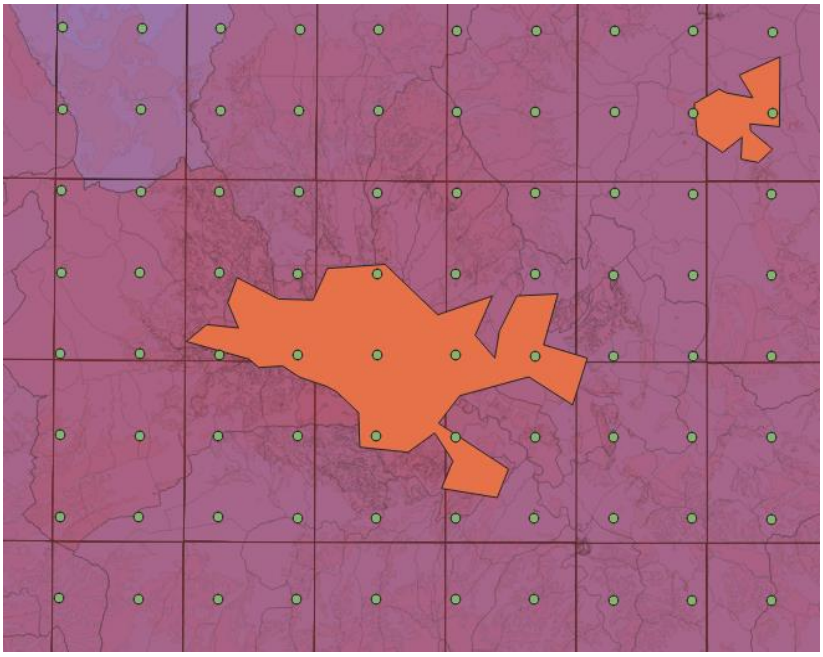

Fig. 5. A map of the wider Skopje area with the GIS data we use in our system

We should also point that our initial conditions are static, i.e. we do not modify these conditions during the simulation run. Feeding dynamic initial conditions into the WRF-Chem model is a challenging task. However, when using the model for day-to-day predictions, we would rarely use simulation domain larger than 3 days, over which the average temperatures are very unlikely to change. Thus, we expect that the cost benefit of integrating over dynamic initial conditions would be small.

5 Evaluation

We have evaluated the proposed model by doing 24 simulation runs for intervals of five days each. The simulation periods were distributed evenly across the year, as we have simulated five day intervals starting at the first and 16-th day of every month. In this way, we expect to simulate a variety of different meteorological and meteo-chemical conditions, as well as be able to produce enough data to evaluate the temperature-related bias of the model for each of the measurement point locations separately.

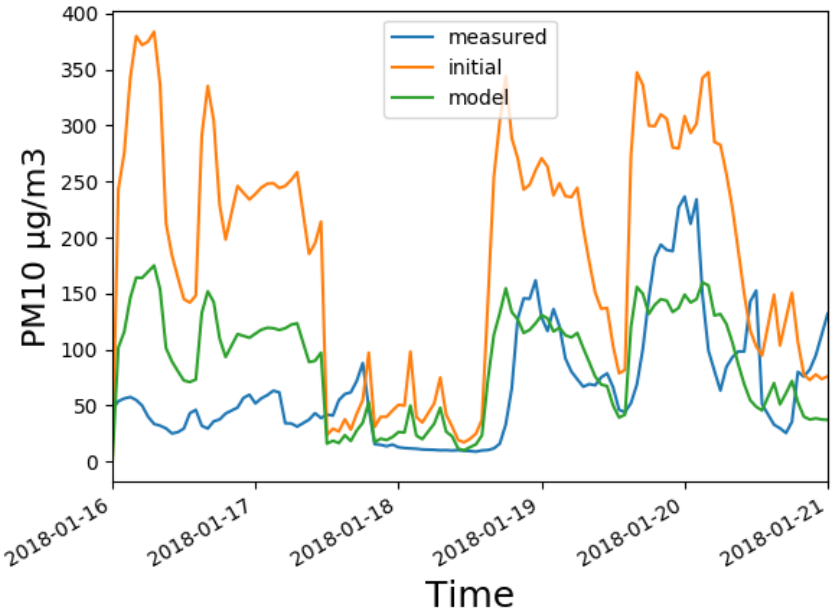

Fig. 6. Measured and predicted values for Karposh in winter

Due to the regional variation and bias of the model, we selected the wider Skopje area for a detailed evaluation, given that the temperature dependent bias, and the general bias was calculated over this area. The model is easily extensible over the other regions as well in the same way as done for this region, but due to the high population density, we decided to focus our attention on the Skopje region. The methodology of evaluation was to calculate the predictions at every point in space where a measurement station exists. As the measurement stations do not match with the nodes in our simulation grid, the predictions at these specific points were calculated by taking the interpolated values of the nearby simulation nodes. We took all the values at the simulation nodes in radius of 5 kilometers around the measurement point, and calculated the interpolated value by taking a weighted average, where the weights are inversely dependent on the distance between the measurement point and the simulation nodes.

Figure 6 shows the measured values for PM10 concentrations at the Karposh measurement station, for a period of 5 days during winter. We can notice that alt-

hough the initial model predicts the pattern of the curve relatively well, it does tend to overestimate the pollution over the entire period. We can also notice that with our proposed improved model, we are able to follow the observed curve with our predictions more tightly, which points to a significant improvement.

A similar pattern, though with a significantly larger differences can be observed for a period of 5 days during the summer. Due to the previously mentioned temperature dependent bias, the initial model tends to significantly overpredict the pollution levels in these conditions. On the other hand, our improved model tends to drastically minimize the discrepancy between the measured and predicted values during summer, when the pollution emissions are much lower than the yearly averages.

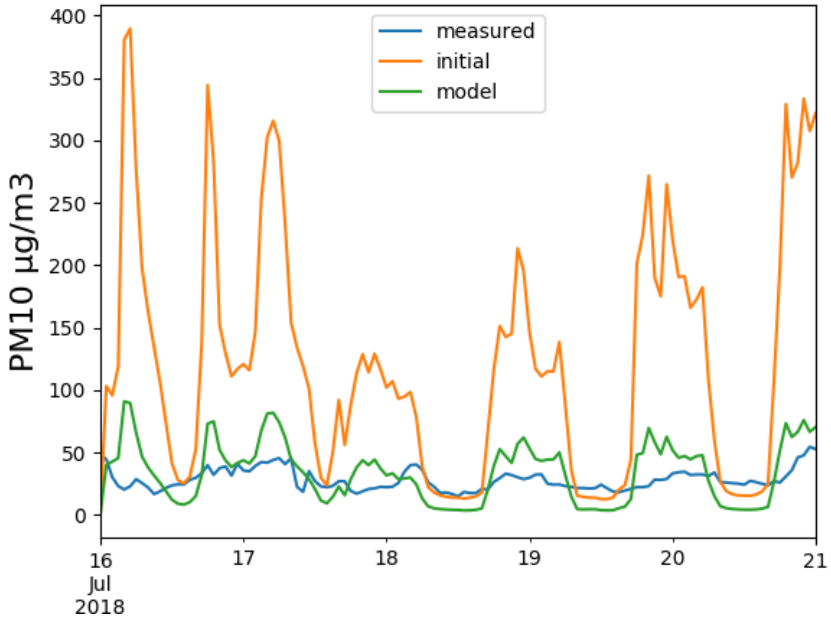

Fig. 7. Measured and predicted values for Karposh in summer.

It is worth noting that the improvements proposed in these paper do not change the shape of the curve, and we can still notice the exact same spikes and holes in the curve in both of the charts. This is due to the fact that our work was focused on modeling the initial and boundary chemical conditions in the simulation. These are kept constant over the entire simulation run, so it is reasonable to expect that they might only linearly shift the curve by a certain amplitude factor. In order to be able to change the curve shape, we would need to intervene in the chemical simulation model itself, or simulate with dynamic initial and boundary conditions. This might be interesting for future work, but it is well beyond the scope of our current research.

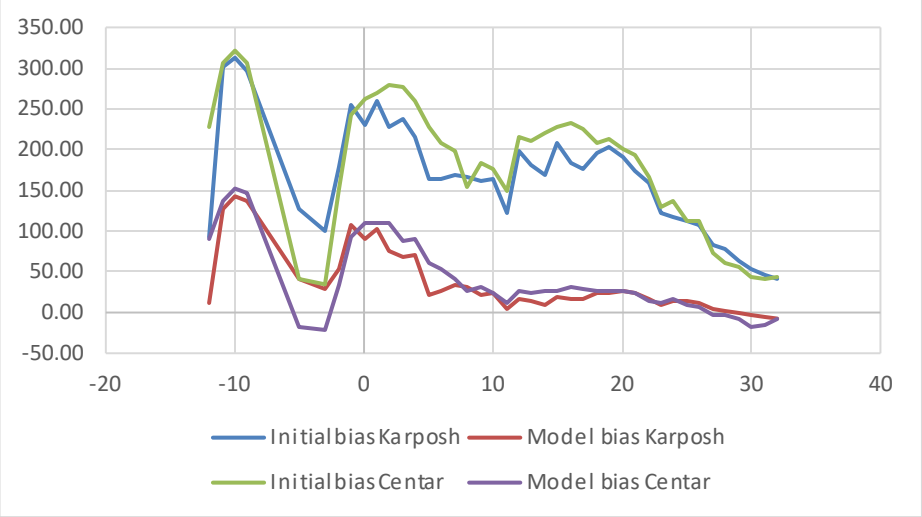

Fig. 8. Temperature dependent bias of the initial model versus our improved model, for the stations Centar and Karposh

Figure 8 (should put a table here instead) shows the bias of the model at two stations, Centar and Karposh. We can note that our model shows a smaller bias in general for both stations, as well as a flatter curve. The flatter curve is a clear indicator that in comparison to the base model, the temperature dependent bias has been decreased by our proposed improvements.

6 Conclusion and Future Work

In this paper, we first presented a general overview of the air pollution prediction models, which are becoming increasingly important in simulating the air pollution in urban areas. Our detailed overview was focused on the WRF-Chem model, where we evaluated a concrete setup of the model, tailored for the conditions of the wider Skopje region. Then we were able to propose a possible improvement of the initial chemical conditions used by the model. Our hypothesis was supported by the data measured from the network of air pollution sensors, which also indicated to a temperature dependent bias of the model. Combining the two findings, we managed to quantitatively measure the deviations of the model from the observed values, and based on that we proposed an improvement for the initialization of the initial chemical conditions.

Our proposed change to the model resulted in measurable improvements in predictions. The total bias of the model, evaluated over the entire simulated data, was decreased from 205.28 to 40.15 for the measurement station in Centar, 179.67 to 32.79 for Karposh, and the charts presented show that we are now able to make predictions that tightly follow the curves of the observed values.

However, the new model still has regional variations, which points out to a regional-dependent bias. In this paper we focused our improvements for the wider Skopje

area, but any future work should address this issue of regional bias, by providing specific scaling factors for every region separately, with the same methods elaborated here that we employed for the Skopje region. This issue of regional variations might also be caused by the relatively coarse simulation grid we use (5km x 5km horizontally). Due to the constraints on computational power, we were not able to test a model with a finer simulation mesh and this could also be an interesting topic for further research. The coarse mesh might also be the cause for the regional variations, which is to be investigated as well.

Finally, our future work may also be directed towards a model that will have a learning property and be able to improve itself dynamically on the base of the calculated bias of its previous predictions. This would be useful in practical application, where the model would be brought to online service for making day to day pollution predictions.

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