



# Improvement of chemical initialization in the air quality forecast system in North Macedonia, based on WRF-Chem model

Vlado Spiridonov<sup>1</sup> · Nenad Ancev<sup>1</sup> · Boro Jakimovski<sup>1</sup> · Goran Velinov<sup>1</sup>

Received: 26 June 2020 / Accepted: 1 September 2020 / Published online: 16 September 2020  
© The Author(s) 2020

## Abstract

Urban air quality is determined by a complex interaction of factors associated with anthropogenic emissions, atmospheric circulation, and geographic factors. Most of the urban-present pollution aerosols and trace gases are toxic to human health and responsible for damage of flora, fauna, and materials. The air quality prediction system based on state-of-the-art Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) has been configured and designed for North Macedonia. An extensive set of experiments have been performed with different model settings to forecast simultaneously the weather and air quality over the country. The initial results and the finding from other similar studies suggest that chemical initialization plays a significant role in a more accurate, both qualitative and quantitative forecast and assessment of urban air pollution. The main objective of the present research is to develop and test for a novel chemical initialization input in the air quality forecast system in North Macedonia. It is performed using ensemble technique in respect to treatment of the mobile emissions data using scaling factors. The WRF-Chem prediction has shown a high sensitivity to different scaling methods. While scaling of the overall mobile annual emissions tends to produce some discrepancies regarding the PM<sub>10</sub> concentration level (overestimation during summer and underestimation during winter), an improved approach that utilizes scaling, in a wider range, only the mobile emissions originated from household heating offers the possibility of more detailed parameter fitting. The verification results indicate that the best accuracy across all scores for the winter months was achieved when scaling up the baseline pollutant input using a higher factor, while in the other seasons, the best results were achieved when scaling down the baseline pollutant emissions by a significant factor. Taking all into account, we can conclude that the seasonal variation in the pollutant input to the atmosphere is a significant factor in simulating the pollution in this region. Therefore, these seasonal variations must be taken into account when fitting the pollutant emission input to any model.

**Keywords** WRF-Chem · Air quality modeling · Extreme pollution episode · PM<sub>10</sub> · Urban area

## Introduction

High urban air pollution has become one of the main concerns of citizens of Macedonian cities due to the elevated levels of pollutants, especially during the winter season, thus having a negative impact on human health (Mindell and Joffe 2004). The problem associated with the urban air pollution is very

complex, as many factors contribute to increased concentrations of the main pollutants (e.g. urbanization, specific topography, emissions sources, environmental factors, meteorology). The main pollutants emitted into the atmosphere in urban areas such as Skopje are sulfur oxides (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO), and particulate matter (PM/aerosols), mostly consisting of black carbon, sulfates, nitrates, and organic matter. For these reasons, it is necessary to carry out advanced research based on observational, theoretical, and modeling studies in evaluation and estimation of the impact of different emissions (anthropogenic, biogenic, and especially mobile emissions) on air pollutant concentrations in urban areas, since the urban sources of variety of gaseous and particulate matters have the largest contribution to anthropogenic emission (Seinfeld and Pandis 1998). These chemical pollutants include primary PM<sub>10</sub> and PM<sub>2.5</sub>, which are directly emitted in the atmosphere from various sources (e.g., road traffic,

---

**Electronic supplementary material** The online version of this article (<https://doi.org/10.1007/s11869-020-00933-4>) contains supplementary material, which is available to authorized users.

---

✉ Vlado Spiridonov  
vlado.spiridonov@univie.ac.at

<sup>1</sup> Faculty of Computer Science Engineering, Ss. Cyril and Methodius University, Skopje, Republic of North Macedonia

construction sites, soil dust, fires, fuel consumption), and secondary PM, which is formed in the atmosphere via chemical reactions in gas and aqueous phases, leading to the oxidation of precursors such as sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>). The recent economic developments in North Macedonia and the increasing urbanization will potentially cause wide-ranging consequences in terms of environmental and health problems in the urban areas, especially in the larger cities like Skopje.

Real-time forecast of urban air quality is a relatively new discipline in atmospheric science and highly important to the public as an advanced information for both air quality and safety assessment. The ever-increasing computational power of modern computers over the past years allowed the use of advanced offline deterministic 3-D chemical transport models (CTM), leading to more accurate air-quality forecast, replacing the previous empirical and statistical approach with its limitations of adequately representing the physical and chemical processes of pollutants. Nevertheless, the use of increased computational resources of these new models is justified as they show significant advances and are capable of simulating the complex physical-chemical processes (Zhang et al. 2012; Spiridonov et al. 2019a). The details describing the original approach of developing the air quality forecast and alert system for North Macedonia based on the WRF-Chem model are given by Spiridonov et al. (2019b).

The goal of the research described in this paper is to find the optimal pollutant input parameters for air pollution prediction that uses a WRF-Chem weather model that combines various sources of air pollution emission repositories. We will start with a brief description of the novel approach and the evaluation method with comparison of results and verification. At the end, we will conclude our findings with the optimal pollution source inputs that produce the best results.

## Air quality forecast system of North Macedonia

The air quality forecast system has been developed and extensively tested to simulate the atmospheric physical and chemical transport processes over North Macedonia. The main design of the system is based on using the Weather Research Forecast, a non-hydrostatic meso-scale model WRF-ARW (see Skamarock et al. 2008), and chemical module WRF-Chem developed by Grell et al. (2005). An integral part of the system are global data from emissions from various sources (e.g. Stockwell et al. 1990; Ackermann et al. 1998; Schell et al. 2001). For simulation of the urban air quality, WRF-Chem utilizes a set of inventories that are processed with PREP-CHEM-SRC algorithm created by Freitas et al. (2017). The global anthropogenic emissions utilize the Emission Database for Global Atmospheric Research (EDGAR) (e.g. Olivier et al. 2005; Muntean et al. 2014;

Crippa et al. 2016a, b) and upgraded data set, the EDGAR-HTAP, with resolution of 0.1° × 0.1° according to Janssens-Maenhout et al. (2015). The WRF-Chem model also incorporates the RETRO, a 1960–2000 global anthropogenic emission base, and GOCART background emission data. A more detailed information about the system can be found in Emmons et al. (2010) and Spiridonov et al. (2019b).

## Improving the chemical initialization of the WRF-Chem model

We started by using the same configuration of the WRF-Chem model as described in our previous research (Spiridonov et al. 2019b), with a modified approach in sourcing the urban mobile emissions. Our hypothesis is that the pollution emissions in the atmosphere strongly depend on seasonal variations due to the household heating emissions being concentrated in winter. Given that the majority of the households in North Macedonia use heating wood or other household combustion stoves<sup>1</sup>, we expect that these sources would have significant environmental effects and contribution to increased air pollution levels during the winter period. The high intensity and scale of the measured pollution values during winter further supports our hypothesis that the household heating emissions form a significant component to the total air pollution. Therefore, any model designed for our region should put special emphasis in correctly incorporating these emissions. Our main contribution is to perform a thorough model evaluation with different pollutant emission levels in different seasons. Thus, we would be able to find the optimal mobile pollution sources for each season that should be incorporated into the model chemistry input. This novel approach will significantly improve the model performance and its precision for air quality forecast in urban areas.

## Emission sources

In addition to the standard sources integrated in the PREP-CHEM-SRC preprocessor, we use additional data composed by the inventory of gas emissions updated by the Ministry of Environmental and Physical Planning (MOEPP). This inventory represents the total yearly emissions of gases by different GNFR defined over the entire area of the country in geographic rectangles (grid boxes) with size of 0.1 × 0.1 lat/lon deg. For each of these rectangles, we have the total emission of some significant chemical compounds (e.g. nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO), and non-methane volatile organic compounds (NVOC)). Each of these pollutants is classified (grouped) by source within the following categories (GNFR sectors): public

<sup>1</sup> This report has been prepared by the Environmental Information Center of North Macedonia, a department within the Ministry of Environment and Physical Planning (MOEPP 2017). More details can be found on the following link: [http://air.moepp.gov.mk/wp-content/uploads/2017/05/0302\\_IIR\\_Macedonia\\_2017\\_FINAL.pdf](http://air.moepp.gov.mk/wp-content/uploads/2017/05/0302_IIR_Macedonia_2017_FINAL.pdf)

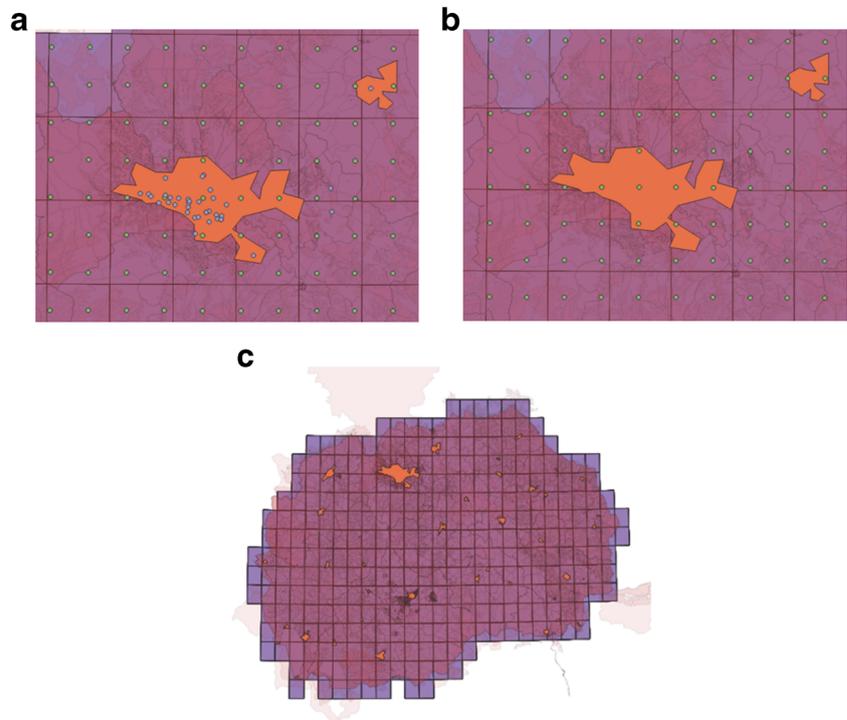
power, industry, other stationary combined, fugitive, solvents, road transport, shipping, aviation, off road, waste, agricultural livestock, other agriculture, and others. The emissions are given as the total annual emissions in (kiloton/year) for each category and each rectangle on the map. These emissions are later added to the default inventories of the Prep-Chem-Src preprocessor and are compiled and used as initial chemical conditions in the WRF-Chem simulation. As Prep-Chem-Src requires us to overlay the emissions to cities, with their rural and urban areas given, we took a rather different adaptation of this approach. We took the urban areas of North Macedonia and overlapped them with the mobile emission data defined in a grid boxes. We have arbitrary added an additional 1.0 km<sup>2</sup> of urban area to each rectangle without an urban area overlap or given the overlapping area of the city with that rectangle if an overlap exists (Fig. 1). The rural area was calculated as the remainder of the area of the square. These areas, together with the emissions (scaled by the factors described in the next chapter) were inserted into Prep-Chem-Src for preprocessing.

### Scaling of the chemical inputs

The model chemistry initialization employs an ensemble technique with two different approaches of scaling the inputs:

- a) The first scaling was done independently of the pollution emission source, *by scaling factor of the total emissions for every grid box in the model domain* in factors ranging from 0 to 200% of the original input with rate of 10%. The scaling was done uniformly across the entire domain.

**Fig. 1** Graphical illustration of the adjustment method of the urban mobile emission data pollutants. **a)** The mobile data emission provided by the Ministry of Environmental and Physical Planning (MOEPP) with a grid resolution of  $0.1^\circ \times 0.1^\circ$  lat/long. The shaded orange area represents the municipality of Skopje with location of all available air quality measuring stations from the state network, and private stations. **b)** Same as Fig. 1a but only for the mobile data emission provided by the (MOEPP). **c)** Same as Fig. 1b, but for the main municipalities of North Macedonia



- b) The second approach was done *by taking into account the pollution source* by scaling only the sources we considered having a seasonal variability. Namely, the column of emission data termed “other stationary combined” sources mostly consist of the pollution emissions from household heating for which the peak occurs in winter and has a significant impact to the urban air quality over that period (see Mirakovski et al. 2018). The justification for this consideration is based on the fact of seasonal variability. For these reasons, it is necessary to delineate emissions and to apply scaling in order to avoid overestimating emission values in summer and underestimation in winter. Furthermore, as these sources are the most significant source of CO and a quite insignificant one for NO, the importance of a separate scaling of these sources is more pronounced.

As for the other pollution sources (traffic, power generation, waste) we do not expect a significant seasonal variation; therefore, these sources were not a subject to scaling in the second approach.

### Numerical experiments and model evaluations

#### WRF-Chem predictions

The best approach for evaluation of any model will be achieved on a wide variety of atmospheric conditions. Given

the large number of ensemble members for simulations (required about 40 per run), the computational time and computer performance needed to simulate each day would be significant. This would require us to limit the simulation span and prioritize certain types of atmospheric conditions that we want to evaluate on. As our goal is basically the best estimation of the input parameters for pollutant emissions, we chose to conduct evaluations on days with stable atmospheric conditions that would yield the most reliable results. This way, by comparing our results with the measured results from the network of air quality measurements stations, we could estimate the pollutant inputs on those specific dates in the area around the measurement stations. Numerical experiments have been conducted for 3 winter cases 3 Jan 2020, 17 Jan 2019, and 27 Jan 2017, two summer cases 20 Sep 2019 and 25 Jun 2017, and one autumn case 17 Oct 2017. WRF-Chem model runs were initialized with GFS 0.25 deg data taken at 00:00 UTC for 4-day advance forecasts. All selected dates are specified for the stable atmospheric conditions. However, we have intentionally chosen evaluation time frames over the different seasons, with widely different measured pollutant concentrations, in order to test our hypotheses that these differences are mostly due to the seasonal distribution of the pollutants inputs to the atmosphere from the local sources.

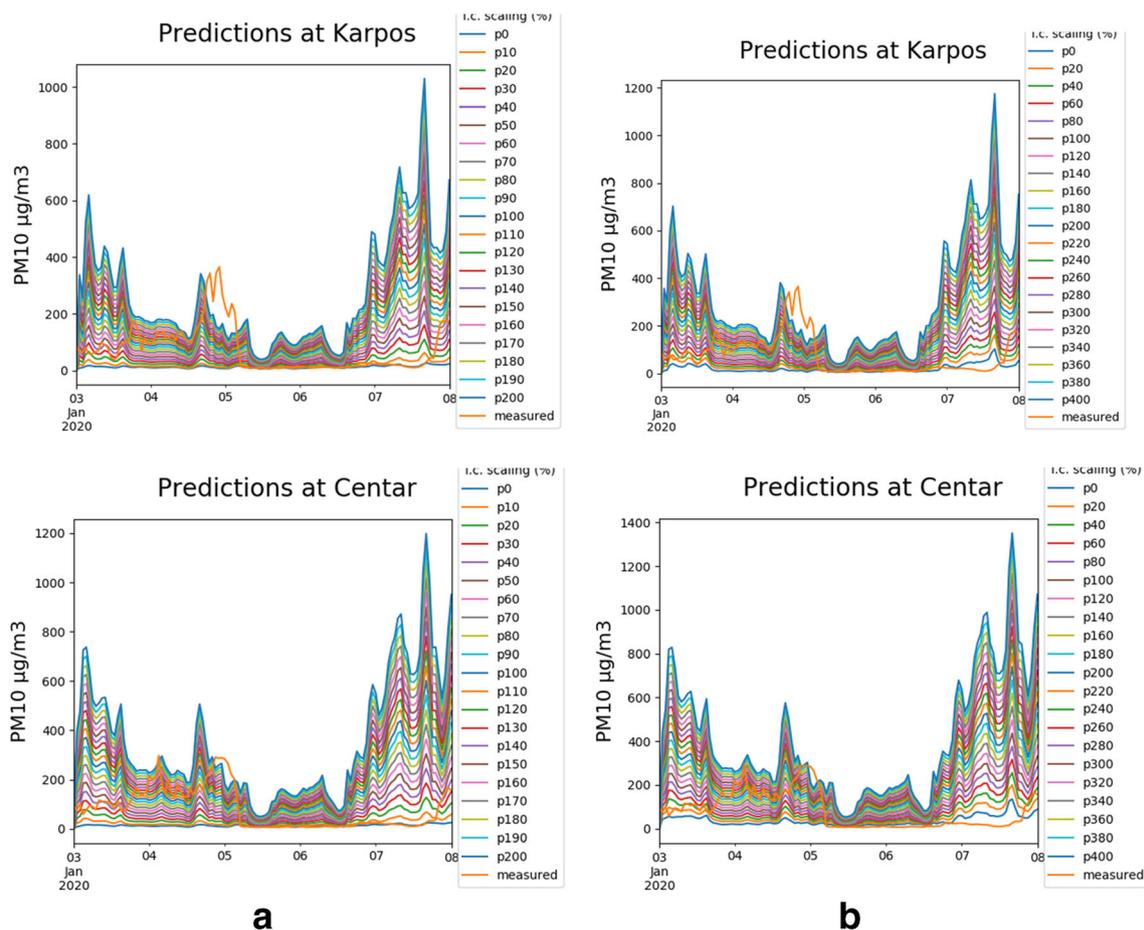
### GIS processing of the data and point-based evaluation

The evaluation of the model is done using the publicly available data from a network of measurement stations. This network consists of a combination of government-operated stations by the MOEPP, plus a supplementary network of home installed stations from the Pulse project. We expect that the MOEPP stations are producing better accuracy than that by the home-based stations, as the latter ones are usually non-certified cheap, household devices, without any guarantee on their measurement precision and calibration and the correctness of their placement and installation. Therefore, we think that evaluation results against each of these stations should be taken with caution and would be chosen only to evaluate against the reliable measurements of the MOEPP stations. The only feasible way to successfully evaluate on these home-installed measurements would be to average and use them as a stack of sensor evaluation, which would hopefully smooth out the high-frequency noise. In order to evaluate the simulation against the in situ measurements by the MOEPP stations, we need to transform the model predictions for the specific geographic point of the sensor locations. For that, we first calculated the interpolated simulated data at each of the stations. The geographic interpolation is performed using a GIS database extension for Postgres, named PostGIS. It was done in such way that for each of the measuring points of the location, we took all the simulation sampling points in a radius

of 5 km around the measurement point. Then, we calculate weighted average of the sampled simulation points within this radius, where the weighted factor is calculated to be proportional to the inverse distance between the measured point and the simulation sampling point. Finally, the measured and the simulated results for each point were compared with the statistical verification scores explained in the next paragraph.

### Comparison with measurements

WRF-Chem predictions have been performed for each of the selected cases using this novel ensemble technique regarding to the mobile emissions data sets. We used two different evaluation strategies regarding these measures: first, we evaluated for each separate day, and then we evaluated the data for the entire simulation period of 4 days ahead for each previously defined intervals of the simulation. The model initialization has been performed using the two scaling methods: The first one utilizes an overall scaling of total annual mobile emissions regardless of the source. The second approach is based on scaling in a wider percentage range only on the total mobile emissions arising from household heating. In addition, it should be emphasized that in several cases, there are stations that were turned off at some time and they either do not provide any data or are constantly zero. Due to the automated way of generating graphical representations, it was more difficult to omit them. Hence, if the measured value is a constant zero, or a time series graph is missing, that means we don't have measured data for that station at that time. Figure 2 a shows a time series of PM<sub>10</sub> predictions using overall scaling factor regardless the emission source, while Fig. 2b shows WRF-Chem predictions initialized with scaling only of the mobile emissions, which were generated from heating and other non-industrial pollutants. The results using two different approaches are compared with corresponding measurements obtained at three urban stations. Looking closely at the results, we found that for a winter case study on 3 Jan 2020, WRF-Chem initialization with scaling only mobile emissions caused by heating, provides a better agreement and better fit with measurement series compared with the first model chemistry initialization approach. It is more evident in evaluating the results shown on Supplemental Material Fig. S1, for other winter case (left panels) and summer event (right panels). One sees that WRF-Chem predictions of PM<sub>10</sub> concentration for all measured stations are more sensitive to the mobile emissions during a winter time when home heating plays significant role in pollution accumulation in urban areas under stable, high-pressure anticyclonic weather conditions. The findings confirmed our hypothesis that there is a strong seasonal irregular distribution of atmospheric pollutant emissions. It suggests that the seasonal variation of mobile emission input should be considered in the chemical model initialization.



**Fig. 2** Time series of WRF-Chem ensemble forecast of  $PM_{10}(\frac{\mu g}{m^3})$  for three urban air quality monitoring stations. Model is initialized on 3 Jan 2020, with a two different scaling approaches of the initial mobile

data emissions. **a** Overall (bulk) scaling; **b** scaling of pollution emission as the result of heating and other non-industrial pollutants

### WRF-Chem model verification

A number of different verification scores were used for evaluating the predictions. The data from every MOEPP station and for every day of the simulation was used to compute the following statistical tests: mean error (BIAS), mean absolute error (MAE), root mean square error (RMSE), and correlation coefficient (CC) as well as an additional metrics like difference between the predicted and measured maximum for a given period (maximum difference), difference between the predicted and measured minimum for a given period (minimum difference), difference between the predicted and measured average for a given period (difference of averages), and difference between the sum of all predicted values and the sum of all measured values for a given period (difference of sums), which should be proportional to the difference of the areas of the curves for a given period. The first four measures are standard evaluation metrics, while the latter four are additional, more intuitive metrics that were used for evaluation of the model, which in our opinion are more relevant from human perspective, in case the model data would be made

available to the general public. However, due to the sheer volume of data collected, the charts presented here will be focused on the four standard measures: Mean bias error (BIAS), mean absolute error (MAE), root mean square error (RMSE), and the correlation coefficient of the  $PM_{10}$  concentrations. The distribution of all metrics as a function of the scaling factors for the 3 Jan 2020 event using a different chemical initialization is shown in Supplemental Material Fig. S2. The chemical initialization is performed using the overall mobile emissions, regardless of the pollution emission sources, while Supplemental Material Fig. S3 shows metrics dependence on scaling factor when ensemble forecasts utilize chemical initialization with scaling only performed for mobile emissions generated by heating. As it is expected, the best accuracy across all scores during the winter months was achieved when scaling up the baseline pollutants input by a higher factor, while in the summer (see Supplemental Material Fig. S4), late spring, and early autumn (not shown), the best results were achieved when scaling down the baseline pollutant emissions by a significant factor. Additionally, we noticed that the scaling factors are not uniform across the entire

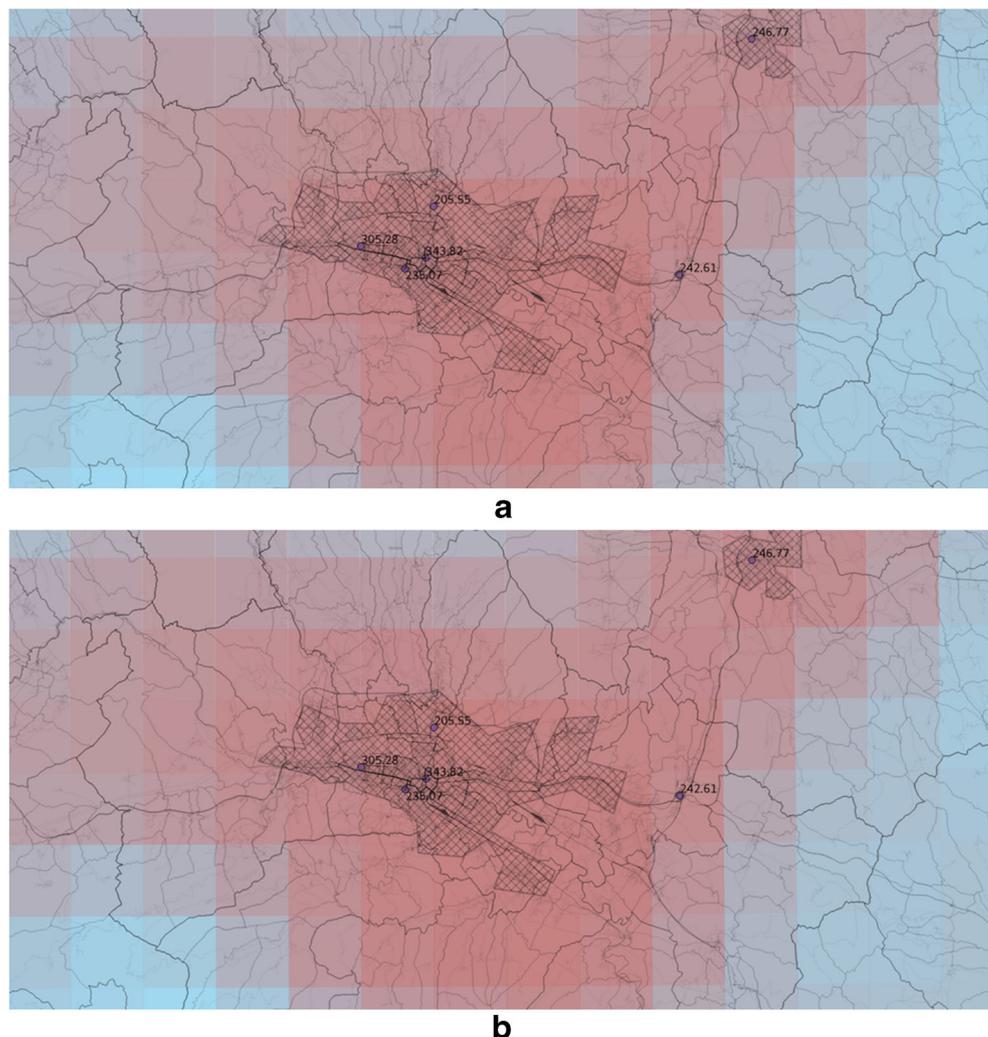
domain, which might come from the incorrectness of some of the pollutant inputs. The spatial distribution of the predicted  $PM_{10}$  concentrations over the Skopje area in winter is given in Fig. 3, where Fig. 3a shows the predictions of the initial model and Fig. 3b shows the predictions with the optimal parameter chosen by our approach. The same information is presented in Fig. 4 for the summer event. By doing the comparison on days with stable atmospheric conditions and weak wind, we tried to eliminate the influence of the atmospheric factors in our experiment.

## Conclusion and the future work

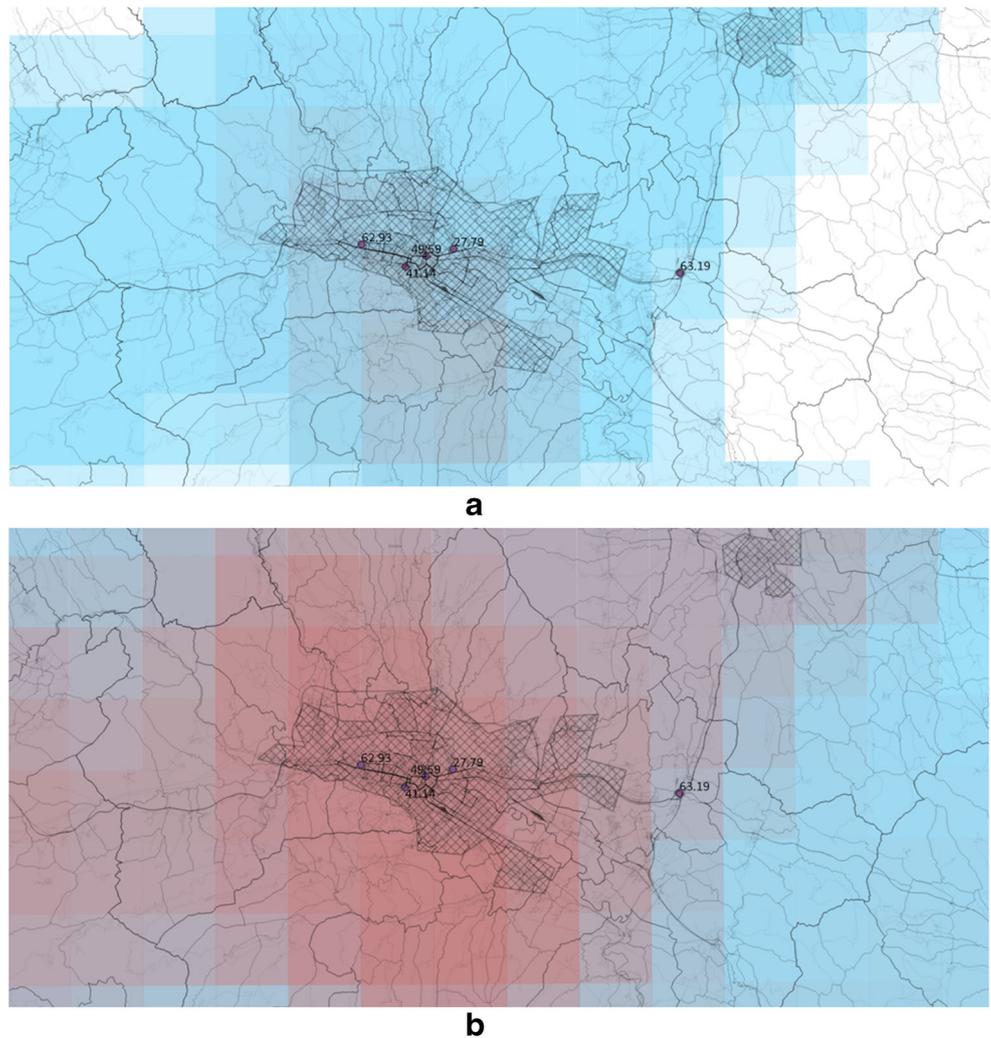
The main goal of the present research is to develop and test a novel chemical initialization input that is being implemented in the air quality forecast system in North Macedonia. It is performed using ensemble approach regarding the treatment of the mobile emissions data and modeling of the appropriate scaling factor. The WRF-Chem model forecasts have shown a

high sensitivity to different chemical initialization scaling methods. While scaling the overall (total) mobile annual emissions, regardless their emissions sources, tends to produce some differences regarding to  $PM_{10}$  concentration level. Our results show some overestimation during summer and underestimation during the winter season. More precisely, the scaling approach that utilizes scaling in a wider range only of the mobile emissions originated from heating for household needs offers a better flexibility for more accurate pinpointing of the optimal parameters, relative to the method that utilizes indiscriminate scaling of the overall mobile emissions. The verification results indicate that the best accuracy across all scores during the winter months was achieved when scaling up the pollutant input by a higher factor, while in the other seasons, the best results were achieved when scaling down the pollutant emissions by a significant factor. Namely, using our evaluation metrics, we can conclude that the best results are obtained if initializing the model for the Skopje region with values around 50–70% of the total urban mobile emissions for the chosen intervals in January (peak of heating season),

**Fig. 3** Spatial distribution of  $PM_{10}$  pollution prediction in the Skopje area for the 17 Jan 2019 event. **a** Forecast of the original model. **b** Forecast of the model with optimal scaling factors



**Fig. 4** Spatial distribution of PM<sub>10</sub> pollution prediction in the Skopje area for the 20 Sep 2019 event. **a** Forecast of the original model. **b** Forecast of the model with optimal scaling factors



with certain days where the optimal range is up to 130% of the original emissions. Values of 10% or less of total urban mobile emissions give the optimal parameters for late summer and early autumn (out of heating season). Taking all into account, we can conclude that the seasonal variation in the pollutant input to the atmosphere is a significant factor in simulating the pollution in this region. Therefore, these seasonal variations must be taken into account when fitting the pollutant emissions input to any model. The seasonal variation of the chemical initialization is modeled even better with selective scaling only of the domestic heating sources. These findings have the potential of future work that may focus on producing a better estimation of the pollutant inputs at any time. A promising area for future work would be investigating the dependence of the temperature and the pollutant inputs, which would be expected due to the higher use of heating fuels with lower temperatures. Our future work will be focused on this area, in producing a better prediction of the emission of pollutants based on the temperature and even coupling the emissions with the temperature predictions of the model.

**Acknowledgments** The authors wish to express their appreciation to Ministry of Environmental and Physical Planning for provision of air quality data from the online monitoring stations of North Macedonia as well as for additional air quality data provided by Ms Martina Spasovska from the Environmental Information Center of North Macedonia. A special thanks to Mr Gorjan Jovanovski for providing the data of the additional measurement stations. We also express our gratitude to the Faculty of Computer Science and Engineering for allowance of using the high-performance computing resources for running and WRF-Chem model and processing.

**Funding** Open access funding provided by University of Vienna.

**Open Access** This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will

need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

## References

- Ackermann IJ, Hass H, Memmsheimer M, Ebel A, Binkowski FS, Shankar U (1998) Modal aerosol dynamics model for Europe: development and first applications. *Atmos Environ* 32:2981–2999
- Emmons LK, Walters S, Hess PG, Lamarque J-F, Pfister GG, Fillmore D, Granier C, Guenther A, Kinnison D, Laepple T, Orlando J, Tie X, Tyndall G, Wiedinmyer C, Baughcum SL, Kloster S (2010) Description and evaluation of the model for ozone and related chemical tracers, version 4 (MOZART-4). *Geosci Model Dev* 3:43–67. <https://doi.org/10.5194/gmd3-43-2010>
- Crippa M, Janssens-Maenhout G, Dentener F, Guizzardi D, Sindelarova K, Muntean M, Van Dingenen R, Granier C (2016a) Forty years of improvements in European air quality: regional policy-industry interactions with global impacts. *Atmos Chem Phys* 16:3825–3841. <https://doi.org/10.5194/acp-16-3825-2016>
- Crippa M, Janssens-Maenhout G, Guizzardi D, Galmarini S (2016b) EU effect: Exporting emission standards for vehicles through the global market economy. *J Environ Manage* 183:959–971. <https://doi.org/10.1016/j.jenvman.2016.09.068>
- Freitas SR, Panetta J, Longo KM, Rodrigues LF, Moreira DS, Rosário NE, Silva Dias PL, Silva Dias MAF, Souza EP, Freitas ED, Longo M, Frassoni A, Fazenda AL, Santos e Silva CM, Pavani CAB, Eiras D, França DA, Massaru D, Silva FB, Santos FC, Pereira G, Camponogara G, Ferrada GA, Campos Velho HF, Menezes I, Freire JL, Alonso MF, Gácita MS, Zarzur M, Fonseca RM, Lima RS, Siqueira RA, Braz R, Tomita S, Oliveira V, Martins LD (2017) The Brazilian developments on the Regional Atmospheric Modeling System (BRAMS 5.2): an integrated environmental model tuned for tropical areas. *Geosci Model Dev* 10:189–222. <https://doi.org/10.5194/gmd-10-189-2017>
- Grell GA, Peckham SE, Schmitz R, McKeen SA, Frost G, Skamarock W, Eder B (2005) Fully coupled “online” chemistry within the WRF model. *Atmos Environ* 39:6957–6975
- Janssens-Maenhout G et al (2015) HTAP\_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution. *Atmos Chem Phys* 5:11411–11432
- Mindell J, Joffe M (2004) Predicted health impacts of urban air quality management. *J Epidemiol Community Health* 58:103–113
- Ministry of Environment and Physical Planning, the Macedonian Environmental Information Center (2017) North Macedonia Informative Inventory Report 1990 – 2015. Submission under the Convention on Long-Range Transboundary Air Pollution (CLRTAP), p 239
- Mirakovski D, Hadzi-Nikolova M, Boev I, Sijakova-Ivanova T, Zendelska A, Doneva N (2018) Sources of urban air pollution in Macedonia – behind high pollution episodes. In: International Scientific Conference GREDIT 2018 – Green Development, Green Infrastructure, Green Technology, 22–25 March 2018, Skopje, North Macedonia
- Muntean M et al (2014) Trend analysis from 1970 to 2008 and model evaluation of EDGARv4 global gridded anthropogenic mercury emissions. *Sci Total Environ* 494–495:337–350
- Olivier JGJ, van Aardenne JA, Dentener FJ, Pagliari V, Ganzeveld LN, Peters JAHW (2005) Recent trends in global greenhouse gas emissions: regional trends 1970–2000 and spatial distribution of key sources in 2000. *Environ Sci* 2(2–3):81–99. <https://doi.org/10.1080/15693430500400345>
- Schell B, Ackermann IJ, Hass H, Binkowski FS, Ebel A (2001) Modeling the formation of secondary organic aerosol within a comprehensive air quality model system. *J Geophys Res* 106:28275–28293
- Seinfeld JH, Pandis SN (1998) Atmospheric chemistry and physics from air pollution to climate change. Wiley, New York
- Skamarock WC, Klemp JB, Dudhia J, Gill DO, Barker DM, Duda MG, Huang XY, Wang W, Powers JG (2008) A description of the advanced research WRF Version 3. NCAR Technical Note, NCAR/TN-475thSTR, 113 pp
- Spiridonov V, Čurić M, Jakimovski B (2019a) Examination of in-cloud sulfate chemistry using a different model initialization. *Air Qual Atmos Health* 12:137–150. <https://doi.org/10.1007/s11869-018-0632-y>
- Spiridonov V, Jakimovski B, Spiridonova I, Pereira G (2019b) Development of air quality forecasting system in Macedonia, based on WRF-Chem model. *Air Qual Atmos Health* 12:825–836. <https://doi.org/10.1007/s11869-019-00698-5>
- Stockwell WR, Middleton P, Chang JS, Tang X (1990) The second-generation regional acid deposition model chemical mechanism for regional air quality modeling. *J Geophys Res* 95:16343–16367
- Zhang Y, Bocquet M, Mallet V, Seigneur C, Baklanov A (2012) Real-time air quality forecasting, part I: history, techniques, and current status. *Atmos Environ* 60:632–655

**Publisher's note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

## Terms and Conditions

Springer Nature journal content, brought to you courtesy of Springer Nature Customer Service Center GmbH (“Springer Nature”).

Springer Nature supports a reasonable amount of sharing of research papers by authors, subscribers and authorised users (“Users”), for small-scale personal, non-commercial use provided that all copyright, trade and service marks and other proprietary notices are maintained. By accessing, sharing, receiving or otherwise using the Springer Nature journal content you agree to these terms of use (“Terms”). For these purposes, Springer Nature considers academic use (by researchers and students) to be non-commercial.

These Terms are supplementary and will apply in addition to any applicable website terms and conditions, a relevant site licence or a personal subscription. These Terms will prevail over any conflict or ambiguity with regards to the relevant terms, a site licence or a personal subscription (to the extent of the conflict or ambiguity only). For Creative Commons-licensed articles, the terms of the Creative Commons license used will apply.

We collect and use personal data to provide access to the Springer Nature journal content. We may also use these personal data internally within ResearchGate and Springer Nature and as agreed share it, in an anonymised way, for purposes of tracking, analysis and reporting. We will not otherwise disclose your personal data outside the ResearchGate or the Springer Nature group of companies unless we have your permission as detailed in the Privacy Policy.

While Users may use the Springer Nature journal content for small scale, personal non-commercial use, it is important to note that Users may not:

1. use such content for the purpose of providing other users with access on a regular or large scale basis or as a means to circumvent access control;
2. use such content where to do so would be considered a criminal or statutory offence in any jurisdiction, or gives rise to civil liability, or is otherwise unlawful;
3. falsely or misleadingly imply or suggest endorsement, approval, sponsorship, or association unless explicitly agreed to by Springer Nature in writing;
4. use bots or other automated methods to access the content or redirect messages
5. override any security feature or exclusionary protocol; or
6. share the content in order to create substitute for Springer Nature products or services or a systematic database of Springer Nature journal content.

In line with the restriction against commercial use, Springer Nature does not permit the creation of a product or service that creates revenue, royalties, rent or income from our content or its inclusion as part of a paid for service or for other commercial gain. Springer Nature journal content cannot be used for inter-library loans and librarians may not upload Springer Nature journal content on a large scale into their, or any other, institutional repository.

These terms of use are reviewed regularly and may be amended at any time. Springer Nature is not obligated to publish any information or content on this website and may remove it or features or functionality at our sole discretion, at any time with or without notice. Springer Nature may revoke this licence to you at any time and remove access to any copies of the Springer Nature journal content which have been saved.

To the fullest extent permitted by law, Springer Nature makes no warranties, representations or guarantees to Users, either express or implied with respect to the Springer nature journal content and all parties disclaim and waive any implied warranties or warranties imposed by law, including merchantability or fitness for any particular purpose.

Please note that these rights do not automatically extend to content, data or other material published by Springer Nature that may be licensed from third parties.

If you would like to use or distribute our Springer Nature journal content to a wider audience or on a regular basis or in any other manner not expressly permitted by these Terms, please contact Springer Nature at

[onlineservice@springernature.com](mailto:onlineservice@springernature.com)