

Fig. 1. Red squares:  $PM_{2.5}$  emissions for January in CMAQ model. Blue raster: distribution of  $PM_{2.5}$  emissions to the IFDM and CALPUFF models (the emission flux is not shown).

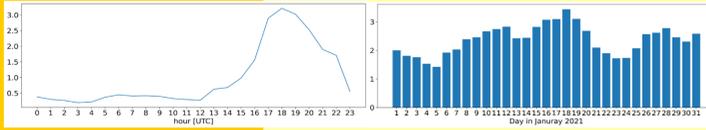


Fig. 5. Emissions profiles for residential heating: diurnal (left) adopted from Guevara et al. (2021) and monthly (right) used in calculations.

Model	CALPUFF	IFDM	CMAQ
Type	Lagrangian puff	Gaussian	Eulerian CTM
Horizontal (terrain) resolution	250 m	no terrain included	2 km
Model output resolution	250 m	10 m	2 km
Number of vertical layers	11	NA	19 (ALA), 40 (WRF)
Top layer height	3 000 m	NA	17 000 m

Table 1. Selected parameters of the models.

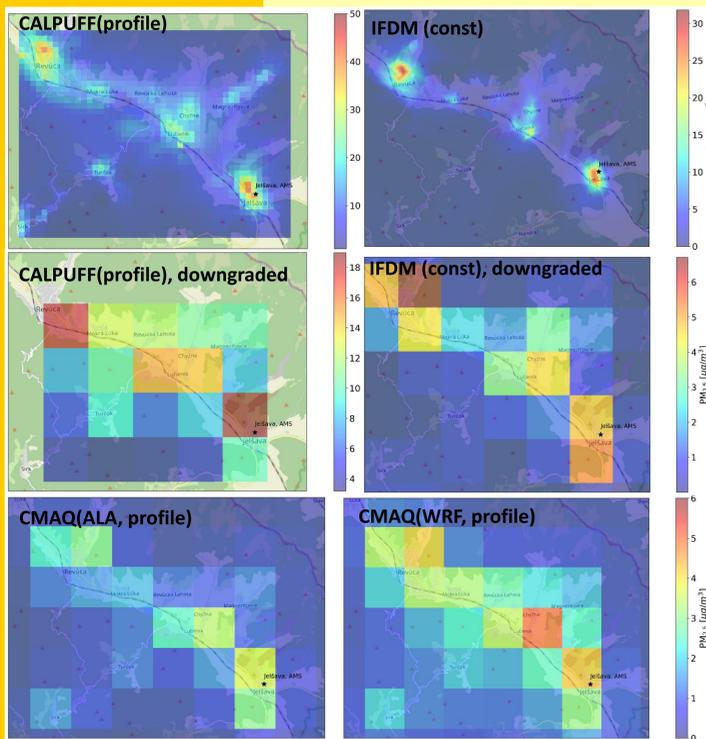


Fig. 6. Mean monthly  $PM_{2.5}$  concentrations.

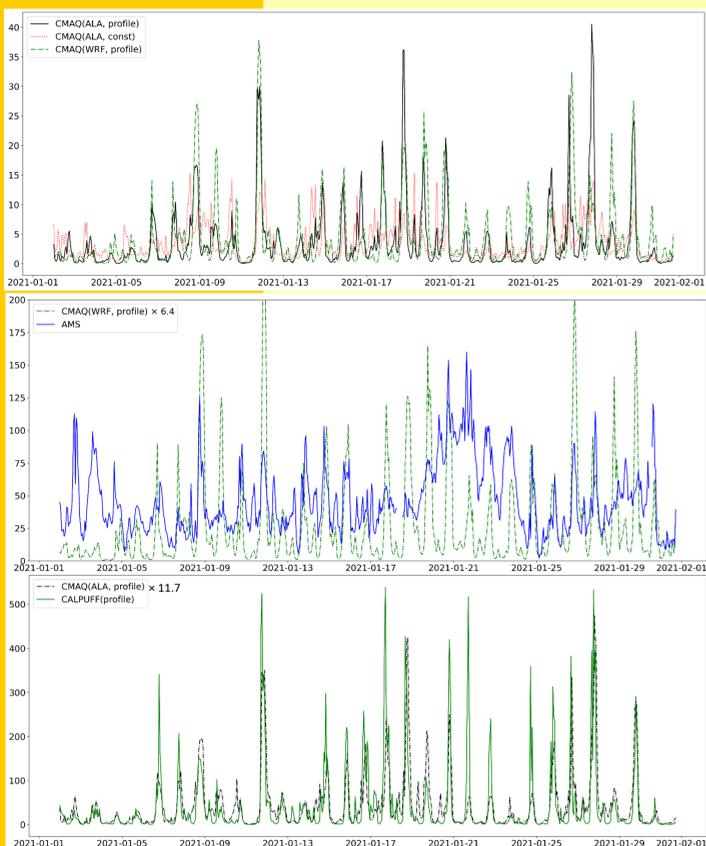


Fig. 7. Comparison between hourly  $PM_{2.5}$  concentrations for different models and models setups and measured concentrations (AMS) at the AMS place.

## Abstract

Comparison of three models differing in resolution and mathematical formulation - CALPUFF Lagrangian puff model, CMAQ Eulerian chemical transport model and IFDM Gaussian dispersion model - is presented. Modelling results for  $PM_{2.5}$  concentrations coming from residential heating emissions over a selected local domain in Slovakia are compared and the differences as well as the usability of the models for local source apportionment is discussed.

## Introduction

Residential heating is the main contributor to the adverse air quality during winter periods in many regions in Central and Eastern Europe. In Slovakia, there are many villages and small towns without connections to central heating systems or natural gas distribution. Therefore, the local heating using solid fuel (mainly wood) is mostly used there. Moreover, the increasing energy prices also contribute to households leaning towards cheaper solid fuel. The consequence of this development is that the annual concentration limits for benzo(a)pyrene and  $PM_{2.5}$ , and the number of daily  $PM_{10}$  exceedances continue to occur at many air quality monitoring sites situated near residential areas. Since the number of air quality monitoring sites is rather limited, there is a need for reliable modelling outputs not only to assess the concentrations at locations without monitoring stations, but also to carry on the source apportionment at monitoring sites. In this paper, we attempt to compare the modelling results for  $PM_{2.5}$  concentrations of several models differing in resolution and mathematical formulation: CALPUFF Lagrangian puff model, CMAQ Eulerian chemical transport model and IFDM Gaussian dispersion model. The local modelling domain includes the town of Jelšava, which according to the measurements is one of the locations with the worst air-quality due to PM in Slovakia, as well as the whole mountain valley NW of Jelšava with the town of Revúca and several smaller villages with solid fuel heating. The simulations are only performed for the residential heating emissions. The results are demonstrated using  $PM_{2.5}$  as it represents almost all of  $PM_{10}$  emissions from residential heating.

## Computational model setup

ALADIN forecasting model (Termonia et al., 2018, Derkova et al., 2017) meteorological data output with the resolution of 4.5 km was used as input to IFDM and CALPUFF. For the CMAQ model a complex set of meteorological 2D and 3D parameters from model Aladin with 2 km resolution were used.

Weather Research Forecasting (WRF) model version 4.3.2 using ECMWF meteorological reanalysis data was also used for CMAQ model.

## CALPUFF

CALPUFF (Scire et al, 2000a) version 7.2.1 was used to model concentrations of  $PM_{2.5}$ . CALPUFF is a Lagrangian puff model which is capable of treating complex terrain, low wind and calm situations which frequently occur in the mountain valleys. CALMET (Scire et al, 2000b) version 6.5.0 meteorological fields was used to process ALADIN meteorological inputs to high resolution grid. CALMET is a diagnostic meteorological model for computation of high resolution terrain-following winds and micrometeorological parameters necessary as inputs for CALPUFF model. The emissions were represented as volume sources corresponding to the emission squares of 50m.

## CMAQ

The Community Multiscale Air Quality (CMAQ) model is a third-generation Eulerian mathematical air quality model (Byun and Schere, 2006). It can be used on various spatial scales from local to hemispheric and for corresponding time scales. It simulates ozone, particulate matter (PM), toxic airborne pollutants, visibility, and acidic and nutrient pollutant species throughout the troposphere. In the simulation, the CMAQ meteorological inputs are taken from the model Aladin, corresponding to the model resolution of 2 km. Boundary conditions are zero except for the ozone. The CMAQ model version 5.3.3 was used (US EPA, 2021). The residential heating emissions are represented as area sources with 2 km resolution.

## IFDM

IFDM (Immission Frequency Distribution Model) is a bi-Gaussian dispersion model developed by VITO to calculate the local dispersion of pollutants in the atmosphere based on meteorological data such as wind speed, wind direction and temperature (Lefebvre et al., 2011a, 2011b). It does not explicitly include the influence of the terrain and is unable to capture calm wind periods. However, as the meteorology for a particular source is always taken from the nearest Aladin gridpoint, a terrain influence is indirectly included through the wind speed and direction from the meteorological model. Emissions gridded in the 50 m squares were represented as point sources at the centres of grid cells. Results were interpolated into the regular grid with 10 m resolution.

## Results

Model results for mean monthly  $PM_{2.5}$  concentrations are shown in Fig.6 for CALPUFF with emission profile from Fig. 5 denoted as CALPUFF(profile), IFDM with constant profile denoted as IFDM(const), CMAQ model with emission profile from Fig. 5 with ALADIN and WRF meteorology denoted as CMAQ(ALA, profile) and CMAQ(WRF, profile), respectively. Both CALPUFF(profile) and IFDM(const) are downgraded to CMAQ 2 km resolution. It can be seen that concentrations of the small hotspots lower substantially as the resolution of model decreases.

The predicted hourly  $PM_{2.5}$  concentrations at the monitoring station location in Jelšava, together with the measured values (AMS) are presented in Fig. 7. At the top of the Fig. 7 the CMAQ results with different meteorology - ALADIN vs. WRF and emission profiles - profile from Fig. 5 vs. constant profile are compared. The importance of the meteorology and used emission profile is seen. In the middle of Fig. 7. the comparison between CMAQ(WRF, profile) and measured concentrations is present. The CMAQ results are multiplied here by factor 6.4 in order to have the same mean monthly value as AMS. The model was able to describe the measured concentrations only with  $R=0.35$ . This can be caused by the meteorological inputs which do not capture the low temperature inversions or by using the unsuitable emission profile, or by the fact that the measured concentrations even in the locked valley during the winter conditions are affected significantly by other sources than residential heating. In the bottom part of Fig. 7 there is comparison between CMAQ and CALPUFF models. Correlations between concentrations time series of different models and measured concentrations at the AMS place are in Fig. 8. It can be seen that the best correlation with the measured values is in the case of CMAQ(WRF, profile). Using constant emission profile gives low correlations in all cases.

## Conclusions

Regional air quality models with resolutions of few km are not capable of reaching measured PM concentrations in small villages or towns. This needs to be considered in the assimilation processes.

Preliminary results show that the diurnal emission profile for PM from the local heating in Fig. 9 gives better correlations with measurements than the CAMS diurnal emission profile.

Meteorological models need to be further tested for cold inversion situations. More realistic emission profile based also on solar heating needs to be developed.

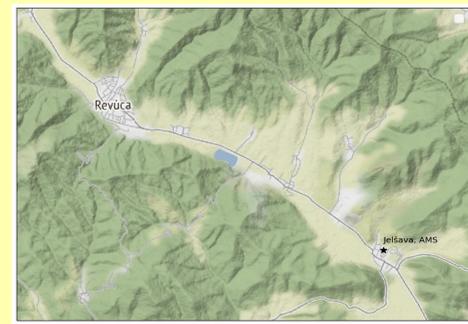


Fig. 2. Orography of the domain



Fig. 3. Typical winter weather in Jelšava. Street near AMS with Coburg castle.



Fig. 4. AMS station in Jelšava.

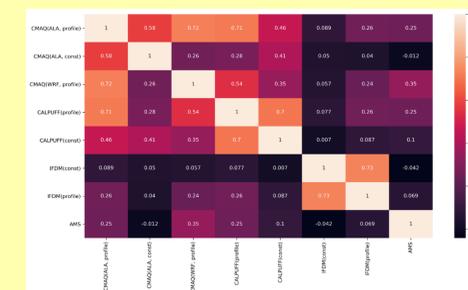


Fig. 8. Correlations between concentrations time series of different models and measured conc. (AMS) at the AMS place.

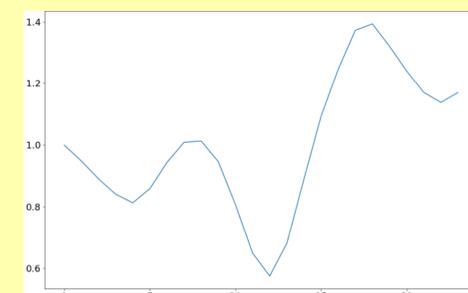


Fig. 9. Preliminary diurnal emission profile for PM from the residential heating obtained in this work

## References

Derková et al., 2017: Recent improvements in the ALADIN/SHMU operational system. *Meteorological Journal*, Vol.20, No. 2, pp 45-52.

Byun, D., Schere, K., 2006. Review of the governing equations, computational algorithms, and other components of the Model-3 Community Multiscale Air Quality (CMAQ) Modeling System. *Appl. Mech. Rev.* 59 (2), 51-77. <https://doi.org/10.1115/1.2128636>.

Guevara, Marc Jorba, Oriol Tena Medina, Carles Denier van der Gon, Hugo Kuenen, Jeroen Elguindi, Nellie Darras, Sabine Granier, Claire Pérez García-Pando, Carlos. (2021). Copernicus Atmosphere Monitoring Service TEMPORAL profiles (CAMS-TEMPO): global and European emission temporal profile maps for atmospheric chemistry modelling. *Earth System Science Data*. 13. 367-404. 10.5194/essd-13-367-2021

Lefebvre, W., Fierens, F., Trimpeneers, E., Janssen, S., Van de Vel, K., Deutsch, F., Viane, P., Vankerkom, J., Dumont, G., Vanpoucke, C., Mensink, C., Peelaerts, W., Vlieghe, J., 2011a. Modeling the effects of a speed limit reduction on traffic-related elemental carbon (EC) concentrations and population exposure to EC. *Atmospheric Environment* 45, 197e207. <http://dx.doi.org/10.1016/j.atmosenv.2010.09.026>.

Lefebvre, W., Vercauteren, J., Schrooten, L., Janssen, S., Degraeuwe, B., Maenhaut, W., de Vlieger, L., Vankerkom, J., Cosemans, G., Mensink, C., Veldema, N., Deutsch, F., Van Looy, S., Peelaerts, W., Lefebvre, F., 2011b. Validation of the MIMOSA-AURORA-IFDM model chain for policy support: modeling concentrations of elemental carbon in Flanders. *Atmospheric Environment* 45/37, 6705e6713. <http://dx.doi.org/10.1016/j.atmosenv.2011.08.033>.

Scire J.S., Robe F.R., Fernald M.E., Yamartino R.J., 2000a: A User's Guide for the CALMET Meteorological Model, Earth Tech, Inc., Concord, MA

Scire, J.S., Strimaitis, D.G. and Yamartino, R.J., 2000a: A User's Guide for the CALPUFF Dispersion Model, Earth Tech, Inc., Concord, MA.

United States Environmental Protection Agency. (2020). CMAQ (Version 5.3.2) [Software]. Available from <https://doi.org/10.5281/zenodo.4081737>

Termonia et al., 2018: The ALADIN System and its Canonical Model Configurations AROME CY41T1 and ALARO CY40T1. *Geosci. Model Dev.*, Vol. 11, pp 257-281.