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EXTENDED ABSTRACT

Comparatively using the original CAMS and refined dataset of pollution sources for evaluation of a high-resolution application of SILAM model.

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Abstract: The goal of this work is to see, if the CAMS emission inventory can be improved with finer resolution national emissions data, when integrated correctly into the SILAM dispersion model. Main dispersion results show that using the local database improves modelled SO_2, while NO_2 overestimation must be investigated further, along with other adjustments.

Key words: *SILAM model, model validation, nitrogen oxides, ozone, sulfur dioxide, carbon monoxide, particulate matter.*

Introduction

This research is about performance of SILAM model (Sofiev et al., 2008) in Estonian domain (380 by 250 km, horizontal resolution 2 km and 11 layers in vertical up to 6570 m above ground, time step 2 minutes) with different emission inventories. The validation period is one year (01.02.2023 – 01.02.2024), always ECMWF meteorological fields used. The SILAM model was ran inside Airviro modelling system in full chemistry mode for hourly concentrations of NO_x, O_3, SO_2, CO, and calculated $PM_{2.5}$ and PM_{10} as output parameters. Boundary fields originate from permanent European-scale runs by Finnish Meteorological Institute (see https://silam.fmi.fi/).
Firstly, the runs were performed with original CAMS gridded regional anthropogenic emissions inventory, version 5.1 (Denier van der Gon, 2023), grid resolution about 6 km. From there, SILAM uses NO_x, SO_2, CO, NH_3, several VOCs and primary aerosols, in GNFR sectors A-L, with F in subcategories F1-F4 and residential heating being based on heating degree days (Chdd). GNFR sectors span from public power (sector A) to agriculture emissions (sector L), defined in (EEA, 2023).
Secondly, the CAMS emissions of NO_x, SO_2, CO and NH_3 were replaced with data from Estonian national emission inventory database (OSIS, resolution 1 km, initially without temporal variation and based in lowest model layer, without vertical distribution) and keeping the other emissions from CAMS database. As a next step, the sector-based source elevations and temporal variation, that the CAMS data had, were added to OSIS data, making full year re-runs after each bigger addition. Re-runs were necessary to check correct implementation of added variations and the improvement of model performance. The goal of this study is to see, if the finer resolution emission data outperforms CAMS, if integrated correctly into the model.

Validation experiments

Experimental setup

Validation was carried out in the Estonian modelling domain using the "full chemistry" configuration, which accounts for aerosol dynamics as well as both organic and inorganic atmospheric chemistry. This includes the formation of secondary organic aerosols and simplified (linear) chemistry of SO_2 and SO_4. A full-year simulation was performed, with a 4-day spin-up period applied to each re-run. Modelled hourly concentrations for comparison with observations were extracted at the locations of 14 monitoring stations of various types, as detailed in Table 1. They are grouped by the underlined station type, "industrial" counting only large industries in North-East Estonia. Modelled values correspond to a 20 m near-surface layer. Not all stations provided measurements for every pollutant.

Table 1. Monitoring stations used for the validation of the SILAM model and domain corners.

Station	Latitude (deg.)	Longitude (deg.)	Type
Kohtla-Järve	59.4096	27.2787	Urban industrial
Lahemaa	59.5153	25.9282	Rural background
Narva	59.3722	28.2007	Urban industrial
Saarejärve	58.7272	26.5034	Rural background
Tallinn-Liivalaia	59.431	24.7605	Urban street
Tallinn-Õismäe	59.414	24.6497	Urban background
Tallinn-Rahu	59.4451	24.7219	Urban industrial
Tartu	58.3706	26.7348	Urban background
Vilsandi	58.3762	21.8446	Rural maritime
Kiviõli	59.344	27.2218	Urban industrial
Sillamäe	59.4028	27.7502	Urban industrial
Sinimäe	59.3332	27.6274	Industrial
VKG	59.394	27.2449	Urban industrial
Tahkuse	58.2247	25.7116	Rural background
Domain minimum	57.403	21.691	
Domain maximum	59.707	28.411	

Statistical procedure

To evaluate model performance, predicted concentrations (C_p) were compared to observations (C_o) using three statistical metrics recommended by the HARMO initiative (Chang & Hanna, 2004).

Correlation R quantifies linear relationship between modelled and observed values:

$$R = \frac{\overline{(C_p - \overline{C_p})(C_o - \overline{C_o})}}{\sigma_{C_p}\sigma_{C_o}},$$

Fractional bias FB provides a symmetric measure of model under- or overestimation:

$$FB = \frac{\text{bias of mean values}}{\text{mean of mean values}} = \frac{\overline{C_o} - \overline{C_p}}{0.5\left(\overline{C_o} + \overline{C_p}\right)},$$

Fraction in factor two *FAC2*, defined as the proportion of cases, when the modelled concentration is two times bigger to two times smaller than observed one.

These metrics were applied not only to hourly mean values but also to daily minimum, maximum, and mean concentrations. Additionally, they were used on hourly values with daily cycle removed and on the isolated daily cycle component.
Time series graphs were also compared visually to see yearly, weekly, daily and seasonal variation performance.

Results

Nitrogen dioxide

The initial run with only CAMS inventory showed in general good correlations with mean R=0.62 for the initial hourly values with slight rural (and urban) overestimation and industrial underestimation (mean rural FB -0.42, industrial 0.11). Best performing statistic was daily average with mean correlation being 0.75, while daily minimum was overestimated in all stations (mean FB -0.64).
Adding OSIS data initially made almost all statistics worse with overestimation prevailing, as the data did not have neither proper temporal variation, nor source elevations. Adding both these features, the diurnal variation became better than with CAMS, while urban overestimation (Figure 1) and industrial underestimation increased, now mean urban FB being -0.53 and industrial 0.45. Hourly data correlation dropped slightly (-0.04 on average). Overall, there is potential to outperform CAMS, if the reason for urban overestimation is found. Fixing poor weekend temporal variation would also improve the performance.

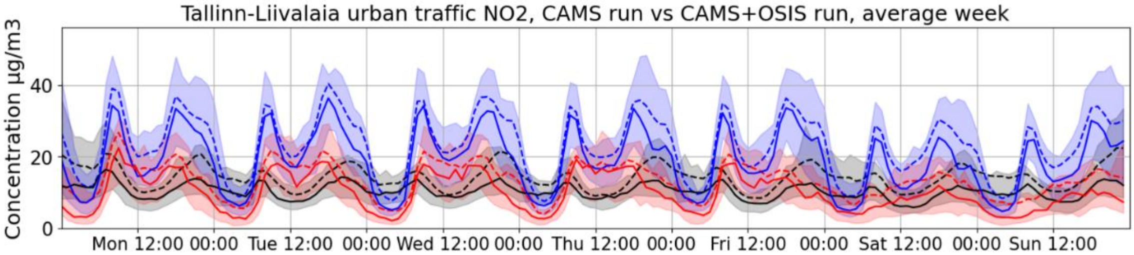

Figure 1. Average week in Tallinn-Liivalaia urban traffic station, hourly values. Model before (black lines, only CAMS) and after (blue lines, CAMS+OSIS). Measured concentration in red. Median, mean and 25th-75th percentile denoted with solid and dotted line and coloured area, respectively. Emerged poor weekend temporal variation is seen.

Nitrogen monoxide

Initial CAMS run shows rather poor correlation for hourly values in industrial and rural stations (mean R = 0.23), with outlier maritime station, R=0.6. Correlation is better in urban stations (mean R = 0.39). Underestimation prevails, mean FB is 1.17 (industrial), 0.66 (rural) and 0.3 (urban).
Adding OSIS data with temporal variation and elevations, correlations remained similar (mean industrial with rural R=0.17, maritime 0.57, urban 0.43). Underestimation increased in industrial (mean FB=1.55) and rural areas (0.73), while urban stations were now largely overestimated (-0.83).

With both NO and NO_2, for the mean daily course in a year, OSIS data gave better correlation, especially in urban stations, yet worse FAC2 results, as seen in Table 2.

Table 2. Mean statistics for mean daily course in a year.

Mean daily course	NO, CAMS	NO, CAMS+OSIS	NO_2, CAMS	NO_2, CAMS+OSIS
Urban mean R	0.53	0.73	0.46	0.93
Urban mean FAC2	0.96	0.46	0.97	0.71

Sulfur dioxide

In CAMS run, SO_2 was the worst-performing pollutant with the mean R for hourly values being 0.04 (industry), 0.36 (urban) and 0.19 (rural). Overestimation prevailed with mean FB being -0.32 (industry), -1.12 (urban) and -0.50 (rural).

Having added temporal variation and source elevations, hourly values mean R improved for industry (0.21) and stayed the same for urban (0.34) and rural (0.22) stations. Biases were reduced a lot with mean FB being now -0.24 (industry), -0.38 (urban) and -0.37 (rural stations). Concerning daily averages, minimum, maximum and mean daily course in a year, the CAMS-OSIS hybrid database outperformed regular CAMS.

CAMS v5.1 has data for the year 2018, OSIS has for the year 2019 and simulation period is 2023-2024. It is possible that CAMS v6.1 (for 2019 and 2020, Denier van der Gon, 2023) would yield much better results especially for SO_2, as there is 13% less SO_2 emissions for 2019 and 24% less emissions for 2020.

Ozone

Generally, the CAMS run O_3 performed quite well, as it is generated by the model and depends less on emissions. Hourly values had mean correlation of 0.79, mean FB=0.02. Daily course had near-perfect correlation, except in the traffic station, with R=0.38.

OSIS data did not change rural or industrial O_3 statistics, but urban FB and correlation suffered due to the interaction with overestimated NO_2. Mean urban R=0.74, FB=0.16.

Carbon monoxide

CAMS run underestimates CO hourly values, mean FB=0.23, and moderate correlation, R=0.62. Daily course has mean FAC2=1, mean R=0.65 with outlier traffic station, R=0.32.

Results with OSIS data go both ways in small magnitude. Urban daily course was improved most, with mean R=0.55 (before) and 0.75 (after). Concurrently, urban correlation slightly worsened, FB slightly improved. Narva industrial station performed slightly worse in all observed statistics (hourly values, daily course, hourly values without daily course and daily maximum, minimum and average).

Particulate matter $PM_{2.5}$ and PM_{10}

As there was no usable primary $PM_{2.5}$ or PM_{10} components available in OSIS database, only results with CAMS database are described. OSIS run did not change the results in any meaningful way. $PM_{2.5}$ had moderate correlation with mean R=0.51. Slight overestimation prevailed with mean FB=-0.27 while urban mean FB was -0.46. Although daily course mean FAC2=0.95, there was no correlation with mean R=0.05, while Sinimäe industrial station R=0.86, Lahemaa rural station had R=-0.86.

PM_{10} was poorly correlated: mean hourly R=0.28, and underestimated: mean FB=0.20. Similarly to $PM_{2.5}$, daily course had no correlation with mean R=-0.05, but mean FAC2=1. Outlier was Sillamäe industrial station with R=0.67. PM_{10} had no rural stations.

Conclusions

Combining the CAMS data on vertical distribution and variations in time with high-resolution national database OSIS, we can achieve better agreement with measurements than any of these emission data on its own. For SO_2, the hybrid database is already better, while NO_2 and NO have potential to outperform CAMS, improving the O_3 prediction in the process. While improving CO must be looked into, temporal variation of particulate matter could be tweaked.
Better performance with SO_2 can be explained with the fact that CAMS data is from 2018, while OSIS is from 2019, when there were less emissions. New CAMS version for the years 2019-2022 may give good results, too. Next steps are planned for understanding the remaining moderate overestimation of NO_2 concentrations, testing better temporal variations for NO_2 and PM (and possibly others), and running a FAIRMODE evaluation on the results.

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