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SO_2 AND NO_X PEAK CONCENTRATIONS, VERTICAL PROFILES AND MODEL-IDENTIFIED ORIGINS FROM DISTANT SOURCES

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Abstract: Atmospheric chemistry-transport models struggle to predict correct concentrations of the modelled pollutant during ground-based temperature inversions. The aim of this study was to evaluate the performance of SILAM model during such inversions. For evaluation, emissions from known tall stack sources of SO_2 and NO_x were applied; modelled concentrations were compared with mast measurements up to 110 m above surface. In inversion conditions the concentrations related to those elevated point sources were found remarkably higher at higher levels. Results show that in half of the inversion cases SILAM diagnoses the temperature inversion well and in almost all of the latter cases, SILAM predicts the vertical profile of modelled pollutant concentration correctly. In general, SILAM tends to underestimate the inversion strength and near-surface concentrations. The reasons may be, respectively, related to the parametrization of the surface layer and the underestimation of vertical dispersion. In modelled concentrations the usual duration of peaks was shorter than measured. The reason may be underestimation of horizontal dispersion.

Key words: *atmospheric chemistry-transport model; near-surface temperature inversion; model validation*

INTRODUCTION

This practical research grew out of model experiments with the atmospheric chemistry-transport model SILAM (Sofiev et al, 2015). While modelling the dispersion of SO_2 from tall (70-250 m) stacks in North-Eastern Estonia, SILAM showed plumes of higher SO_2 concentration passing over rural monitoring stations far away from the stacks. In these ground-based stations, however, every modelled plume did not bring about a noticeable increase (”peak”) of SO_2 concentration. Assuming that the plume trajectories, driven by HIRLAM short-term forecast, are rather reliable, we hypothesized that thermal inversions formed during such episodes might prevent fast penetration of admixtures from higher above to the surface layer. It is known that atmospheric models are in general less reliable in thermal inversion.

To check our hypothesis, this research was made using a five-year period of SO_2, NO_x and temperature measurements from a rural 130 m tall mast, located 110-150 km from the pollution sources. The mast measurements of vertical profiles of both potential temperature and pollutant concentration were used. This research focuses on cases, when remarkably high (up to 20 times the background level) short-term concentrations (”peaks”, duration up to 12 hours) of SO_2 and/or NO_x occur in a rural area, with specific concentration vertical profile in planetary boundary layer: the concentration above is higher than at lower levels. Such a profile indicates either origin from an elevated point source or possibly ascending air mass at some point on the advection path of these trace gases. The aim of this study was to evaluate the performance of SILAM in modelling SO_2 and NO_x concentrations during these cases of ground-based thermal inversion.

METHODS

The concentration and temperature profiles measured during 2016 – 2020 in a 110 m tall mast at rural (forested) SMEAR Estonia site (Järvselja, Estonia) were used to specify the cases (location: see Figure 1). In total about 20 cases meeting the main criterion (concentration increasing with height) were selected. It appears that all these events have a pronounced temperature inversion between all or some of the 5 mast measurement heights (30, 50, 70, 90 and 110 m), which obviously prevents fast penetration of admixtures from higher above to the surface layer. Both wintertime steady inversions and summer night-time inversions contribute to these events. To confirm the inversion condition at Järvselja was not a fluke,

temperatures measured in 24-meter masts at Aseri, Uulu and Külitse, Estonia (locations: see Figure 1), were used to see the extent of inversions.

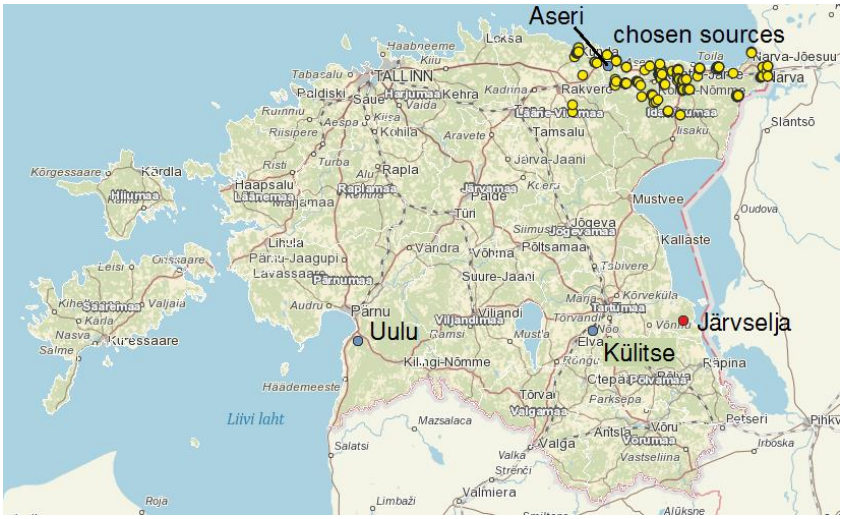

Figure 1. All data sources used in this research: Järvselja mast marked in red, lower masts in blue and initially chosen industrial pollution sources (stacks) in yellow. In the calculations only sources with emission more than 1 g s^{-1} were used.

The biggest known sources of SO_2 and NO_x in Estonia are tall stacks of thermal power plants and chemical industries in North-Eastern Estonia, about 110 – 150 km away from the measurement site. To initially (vaguely) confirm that measured concentration peaks (present during selected 20 cases) originate from these stacks, NOAA HYSPLIT model (Stein et al, 2015) was used for backward air mass trajectory tracing. If HYSPLIT showed the air mass trajectories going over North-Eastern Estonia, more detailed studies were done in the domain covering Estonia using SILAM model at resolution of 0.025°of latitude and 0.05°of longitude, which is about 2.8 km. Firstly, the model was run in inverse (adjoint, receptor-oriented) mode to more accurately confirm that the measured peaks originated from this industrial area, not from other (more distant) sources. SO_2 concentrations measured at 110 m mast height were used as model input, as on this height, usually the highest concentrations were measured. The output, sensitivity distribution in time, was visualized with the OpenGrADS software. To see if outputs differ, a sensitivity study was conducted: same adjoint runs were made with sensitivity source (measured concentration) emitted from a column at heights 0-50 m. For all SILAM runs, the necessary weather data originated from the HIRLAM NWP model (Estonian Weather Service).

After thoroughly confirming the origin of the concentration peaks to be North-Eastern Estonia, SILAM was run in the direct (forward, source-oriented) mode. For that, NO_x and SO_2 emission data for the industrial stacks (tons per year) was known. As the daily, weekly and monthly variation of the emissions was unknown, it had to be estimated taking into account the type of the source (chemical industry or power plant). Using the emission data (now in g s^{-1}), dispersion of the pollutants was calculated. Only sources emitting more than 1 g s^{-1} were used in the run, in total 20-30 sources were used (Figure 1). Modelled time series and vertical profiles of concentration and (potential) temperature were compared with data measured at the Järvselja mast location.

Regarding the time series, measured and modelled average concentration at peak maximum were compared, as well as peak duration. Using all peak maxima, fraction in factor two index (FA2) was calculated for both SO_2 and NO_x. Furthermore, modelled and measured inversion strengths (in this study, temperature difference between 30 m and 70 m mast height) at peak maximum were compared. Vertical profiles of potential temperature and pollutant concentration (taken from peak maxima) were compared qualitatively. It was considered that SILAM modelled the profiles well, if the shape of profiles was visibly similar.

RESULTS

From the 20 selected inversion cases, HYSPLIT showed the contribution of North-East Estonian industrial stacks for 12 cases, all confirmed by SILAM adjoint runs. Other events are likely induced by more distant sources outside of the country. Due to other limitations, it was possible to completely study 6 cases, containing in total 17 pollutant concentration peaks. During almost all of these cases, lower masts (Aseri, Uulu, Külitse) showed inversions as well (temperature difference between 22 m and 8 m mast heights 0.2...4.5 °C), which means the inversions were extensive, possibly ranging all over continental Estonia.

The sensitivity study showed sensitivity distributions similar to the adjoint run most of the time. However, five modelled cases out of seven still showed some level of difference for at least two hours at a time. The difference was sensitivity distribution converging closer to the monitoring station in the sensitivity study. The distribution was lower further away (Figure 2). These differences were too small to prevent the North-Eastern Estonian sources to contribute to a particular peak.

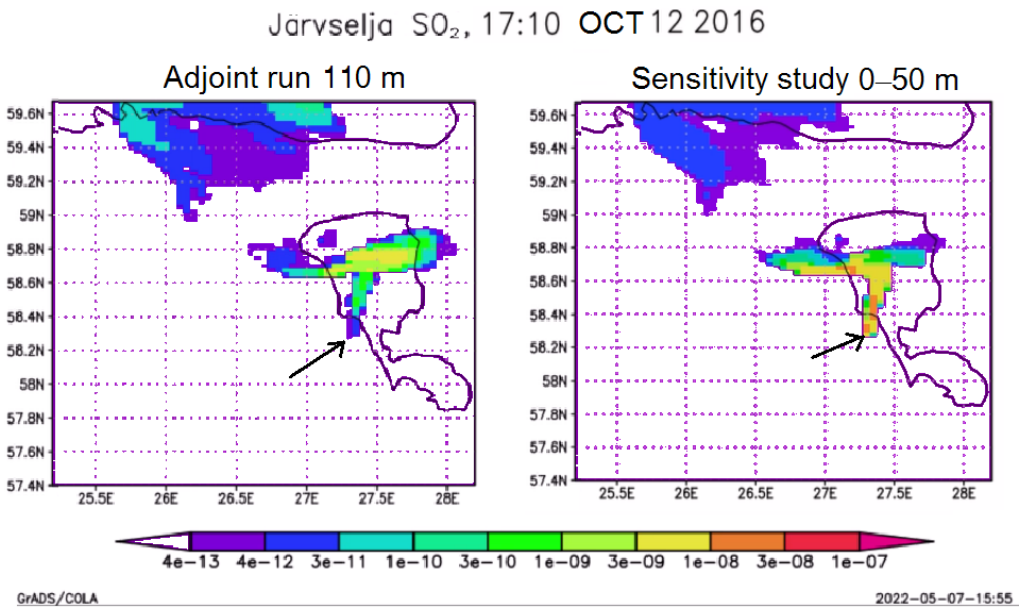

Figure 2. Comparison between an adjoint run at 110 m and sensitivity study at 0-50 m for an SO_2 peak. The sensitivity study shows higher sensitivity distribution close to Järvselja (marked with an arrow) and lower distribution further away. The colour scale is in arbitrary, non-normalized units.

After direct (source-oriented) runs were made, comparison of time series at mast location showed that, on average, SILAM-modelled peaks lasted three hours less than measured peaks. The fraction in factor two (FA2) results for SO_2 and NO_x were, respectively 5/9 and 1/8, which means that (e.g. for SO_2) the average modelled concentration at peak maximum did not differ more than two times from the measured value (5 cases out of 9). SILAM predicts the peak values better during a weaker inversion (measured temperature difference between 30 and 70 m less than 1 °C). Inversion strength at concentration peak maximum was up to 3.1 °C underestimated by SILAM. Potential temperature profiles were properly reproduced by SILAM in four cases out of eight. If temperature profile was reproduced, three times out of four the concentration profile fitted as well. In total, 3/9 and 5/8 concentration profiles for respectively SO_2 and NO_x were reproduced. For detailed results see Table 1. Examples of vertical profiles are given on Figure 3.

Table 1. A concise table of most important results.

Event	Measured inversion strength 30-70 m (°C)	Modelled inversion strength 30-70 m (°C)	Inversion strength underestimated (°C)	Potential temperature profile correct?	Concentration profile correct?		Peak estimated by SILAM	
					SO_2	NO_x	SO_2	NO_x
13.10.2016 1	gaps in data	2.1	-	-	no	yes	under	under
13.10.2016 2	gaps in data	0.8	-	-	yes	no	under	under
13.10.2016 3	gaps in data	-0.2	-	-	no	yes	under	under
16.05.2017	3.4	0.4	3.1	no	no	no	under	under
24.07.2017	1.5	-0.1	1.6	yes	no	no	well	over
27.08.2017	2.9	1.6	1.3	yes	yes	yes	well	over
04.03.2018 1	0.4	-0.4	0.8	yes	-	yes	-	under
04.03.2018 2	0.3	-0.4	0.7	no	no	-	well	-
04.03.2018 3	0.7	-0.2	0.9	yes	yes	-	well	-
24.01.2019	0.3	-0.3	0.6	no	no	yes	well	well
						FA2:	5/9	1/8

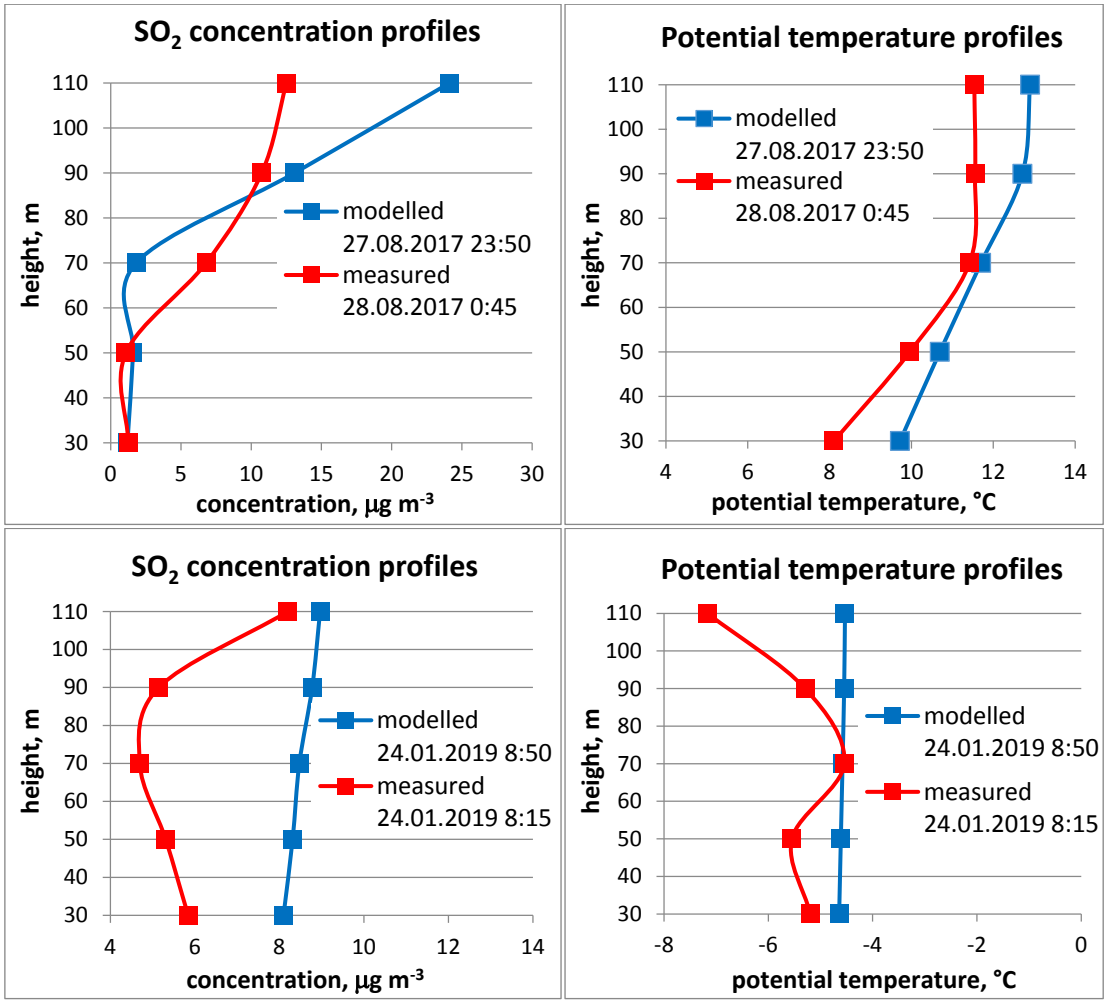

Figure 3. Examples of SO_2 concentration and potential temperature vertical profile for two events. Upper ones were deemed well-assessed, lower ones poorly assessed by SILAM.

DISCUSSION

The sensitivity study shows all the pollution originating roughly from the same area for both 110 m and 0-50 m height. At the same time, sensitivity study does not reflect all the details in reality. Forest canopy reaches up to a height of 30 meters at Järvselja, but SILAM does not consider the in-canopy processes.

Most of modelled concentration peaks (averages of concentrations at peak maximum) are seen to be underestimated. This may mean underestimation of pollutant vertical dispersion in the model. SILAM usually showed highest pollutant concentration above the mast (110-600 m). If pollutant dispersion is underestimated in the model, the resulting peak concentrations are underestimated. However, as variation of emissions in time had to be estimated (having only yearly emissions available), a notable amount of uncertainty was introduced.

Modelled durations of peaks were shorter than measured. One explanation is that horizontal dispersion of pollutant is underestimated, too. If pollutants dispersed horizontally more, the plume would take a longer time to pass over the monitoring station in the model, resulting in a longer duration of the peak.

SILAM in most of the cases underestimates the strength of inversion. Inaccuracies are understandable, as maximum three layers in HIRLAM meteorological fields cover five measurement heights in the mast. If these data points are poorly represented by HIRLAM, it is likely that SILAM assesses the inversion condition poorly as well. In addition, tree canopies can affect the air mass on lower levels. To pinpoint the cause of inversion strength underestimation, it is necessary to look into the parametrization of the surface layer in SILAM.

CONCLUSIONS

The aim of this study was to evaluate the performance of atmospheric chemistry-transport model SILAM during specific conditions of PBL thermal inversion using peak concentrations originating from distant elevated sources. Based on the fraction of factor two (FA2) index, SILAM assesses average peak concentrations fairly less than half of the time. SO_2 peaks are assessed better than NO_x. Most of the time, average peak concentrations are underestimated, which may mean underestimation of pollutant vertical distribution. However, uncertainty originating from emission data is considerable. The horizontal dispersion of the pollutant may be underestimated as well, as modelled peaks are shorter in duration. SILAM tends to underestimate inversion strength (at least at heights 30-70 m). To understand the reasons, it is necessary to look into the parametrization of the surface layer in SILAM.

Acknowledgements

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