

## OPERATIONAL VALIDATION OF SILAM MODEL IN DIFFERENTLY INHABITED AREAS

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**Abstract:** *A pre-operational validation of Estonian application of SILAM model is performed. A denser network of passive sampler measurements of NO<sub>2</sub> and SO<sub>2</sub> and filter measurements of black carbon (BC) is added to the stationary monitoring network. It is found that SILAM tends to “smooth out” the two-week average pollution levels over the country: urban concentrations are underestimated, whereas concentrations at remote sites are represented fairly or slightly overestimated. Thus, this study maintains the results of a former one and gives broader geographical view on these. The modelled and measured values are reasonably correlated in general. Further research on emissions, model features and resolution effects is needed to clarify the reasons of its “smoothing” behaviour and to develop it for more accurate predictions.*

**Key words:** *pre-operational validation, SILAM, nitrogen dioxide, sulphur dioxide, particulate matter, black carbon.*

### INTRODUCTION

This paper is intended to validate the local application of air quality model SILAM running in pre-operational mode in Estonian Institute of Meteorology and Hydrology (<http://meteo.physic.ut.ee/silam>). Regular air quality monitoring network of Estonia is geographically not representative enough. Although 9 stations may seem enough for a country of 45000 km<sup>2</sup> by area, the network is designed for quantifying the public health risks, rather than to understand the geographical spread of pollutants. Three stations are located in capital city Tallinn and three in next largest towns. Two of three rural stations are in North-Eastern part of the country to detect the pollution from oil-shale-driven industrial complex. A maritime station is in far west on a small islet Vilsandi near island Saaremaa. Thus, most of stations are concentrated in North Estonia and no background stations are located in the southern part (Figure 1). In this study the data from monitoring network are combined with passive samplers of NO<sub>2</sub> and SO<sub>2</sub>, and black carbon (BC) filter measurements in small towns and villages. These measurements were carried out by students of secondary schools and basic schools under supervision of their teachers and quality control by University of Tartu, within the GLOBE Estonia environmental measurements' programme (<http://www.globe.ee/>).

### METHODS

#### Measurements

The map of measurement sites is given in Figure 1. In total, the passive samplers and filter measurements of BC were used at 26 sites. The sites were chosen to represent the territory different inhabitation types. The classification of inhabitation types is somewhat subjective. In Estonia, the small villages have well below thousand inhabitants, boroughs about a thousand, small towns a few thousands to about 20 thousands. Medium-sized towns Narva, Kohtla-Järve and Pärnu fit between 40000 and 70000, Tallinn (400000) and Tartu (100000) are classified as cities. For comparison with sampling sites, the monitoring stations are classified as following: one of stations in Tallinn (the Liivalaia street station) and Tartu station in category 1 (city), two stations in Tallinn (Rahu and Õismäe), Kohtla-Järve and Narva in category 2 (city outskirts or medium-sized town), rural station Saarejärve in category 5 (small village) and background stations Lahemaa and Vilsandi in category 6 (uninhabited).

The samples of NO<sub>2</sub> and SO<sub>2</sub> (passive samplers) and BC were taken during four two-week campaigns - 13.02 – 26.02, 14.05 – 27.05, 27.08 – 09.09 and 19.11 – 03.12 in 2012. The February and November – December campaigns belong to intense heating season, whereas the May and August-September campaigns took place in warm season, when no substantial domestic heating is expected. One set of passive NO<sub>2</sub> and SO<sub>2</sub> samplers was exposed during each campaign, thus the average concentration are measured only. The samplers were manufactured and analysed in internationally certified laboratory of Estonian Environment Research Centre (EERC) in Tartu. The filters of BC were changed, depending on

their darkening, but typically each day. The concentration estimates were based on darkening of the filter, measured with photometer. Thus, we have a more detailed set of BC measurements, resolving the changes during each campaign. On the other hand, the NO<sub>2</sub> and SO<sub>2</sub> sampling results are directly comparable with two-week averages from monitoring stations and output of SILAM, whereas BC is directly neither forecasted by SILAM, nor measured in monitoring stations – both these rely on standard aerosol fractions PM<sub>2.5</sub> and PM<sub>10</sub>. However, we expect that often, related with combustion of fuels, the BC is correlated with total mass of particulate matter.

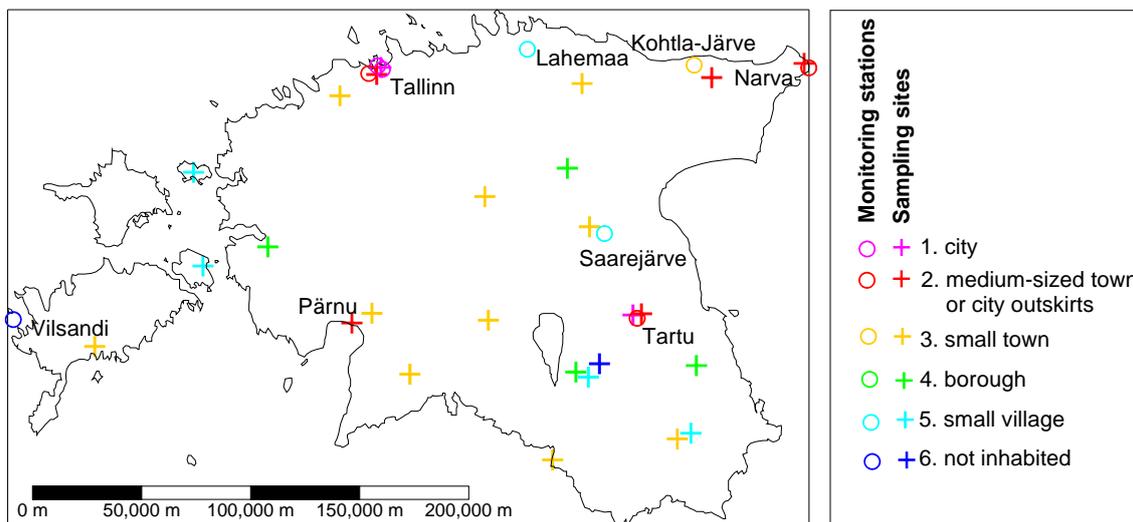


Figure 1. Locations of monitoring stations and sampling sites.

### Modelling

The SILAM model (Sofiev et al., 2008) is running in pre-operational mode in Estonian Institute of Meteorology and Hydrology for Eastern Baltic domain (Estonia, Latvia, Southern Finland, adjacent areas of Russia, Lithuania and Baltic Sea) with horizontal grid resolution 3.3 km. The applied version 5.2 includes basic acid chemistry and aerosol formation. The pre-operational runs apply a detail database of pollution sources within in Estonia that originates from Estonian Centre of Environment Research (see Ots et al., 2013) and is driven by ETB-HIRLAM meteo from EMHI. For surrounding areas the TNO MACC database (resolution 7 km) is applied. The air pollution boundary fields originate from European SILAM runs in Finnish Meteorological Institute. Since this pre-operational configuration applies from July 2012 only, the simulations for first two campaigns, February and May, are made with same grid resolution in the domain of Estonia, boundary fields in North Europe generated by SILAM driven by ETA\_HIRLAM, setup details see (Ots et al., 2013). Differences in setup from the pre-operational one are expected not substantially affect the results.

## RESULTS

### Black carbon and particulate matter

As it was expected, the modelled PM<sub>2.5</sub> concentrations are much higher than measured BC concentrations. Typical differences are 5 – 10 times, but in summertime in rural sites these values are often in same size order. However, it appears that SILAM, in comparison with monitoring stations, underestimates PM<sub>2.5</sub> typically by a few times. Thus, comparing the BC measurements with close monitoring sites, the fraction of BC is typically 2 – 5%. Despite discrepancies in absolute values, the correlations between modelled and measured values are remarkable, referring to rather adequate description of origins and conditions for PM pollution episodes, Figure 2. The measured and modelled values are better correlated during the heating period. Lower correlations in summer are expected, as the combustion emissions, responsible for most of BC, are lower. Somewhat surprisingly, the BC measurements are somewhat better correlated with model estimations than PM<sub>2.5</sub> measurements. The

reason may be in database of emissions: no wind-blown dust neither from agricultural areas, nor from roads is included. Thus, combustion emissions are dominating.

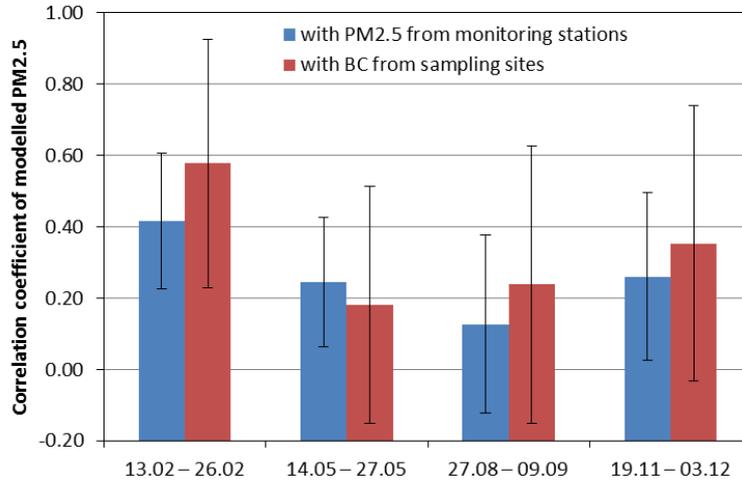


Figure 2. Linear correlations (averages and standard deviations) of PM<sub>2.5</sub> concentrations predicted by SILAM with (i) PM<sub>2.5</sub> measured in monitoring sites and (ii) BC measured in sampling network. In total 7 valid monitoring stations and 22 – 25 BC sampling sites are applied.

### Gaseous admixtures

The best overall fit of modelled *vs.* measured concentrations of nitrogen dioxide is found weakly non-linear: there is a tendency of stronger underestimation of high concentrations (Figure 3). Comparing the measurement campaigns, the data are obviously inhomogeneous: in cold season the concentrations are higher and somewhat better represented by model than in warmer one. Despite general underestimation, there is a positive intercept in quadratic fit – SILAM gives a steady background about 1 – 1.5  $\mu\text{g m}^{-3}$  that is not always the case in warm season measurements.

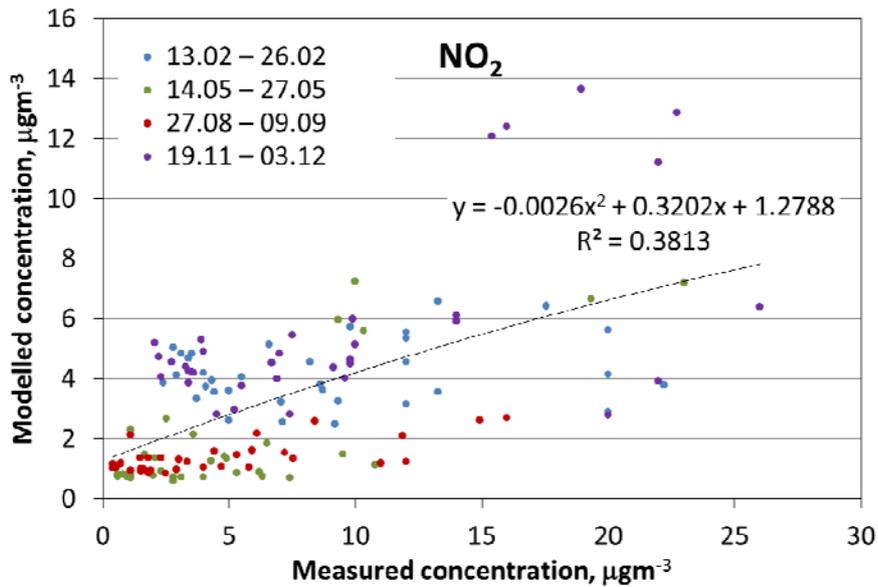


Figure 3. Scatterplot of modelled and measured concentrations of NO<sub>2</sub>. Each dot represents a two-week average concentration (either from passive sampling or monitoring station). Four campaigns are distinguished by colours.

The sulphur dioxide modelling-measurement comparison (Figure 4) is so heavily scattered that no meaningful regression is available. However, the main feature is similar to nitrogen dioxide: higher concentrations in cold season, both measured and predicted. The minimal levels are nearly  $0.2 \mu\text{gm}^{-1}$  in both measured and modelled data. In a few cases rather high concentrations are measured, whereas model doesn't predict anything substantially different from usual background. These cases are identified as impact of local point sources (boiler houses, industrial enterprises) that by no means can be predicted by a regional-scale model with 3.3 km resolution.

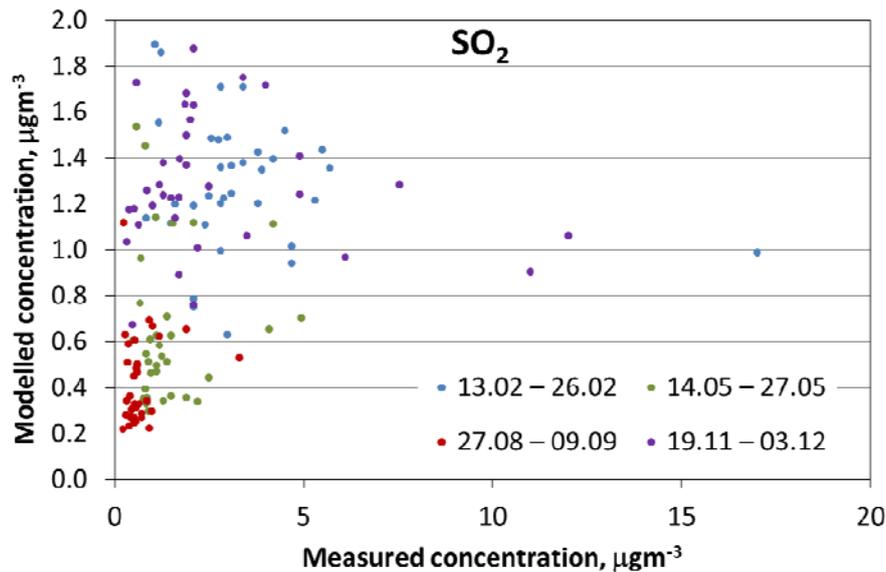


Figure 4. Scatterplot of modelled and measured concentrations of  $\text{SO}_2$ . Each dot represents a two-week average concentration (either from passive sampling or monitoring station). Four campaigns are distinguished by colours.

Despite worse representation of absolute values, the model gives meaningful site-wise linear correlations with measured concentrations in warm season (Table 1). In cold season the correlations do not exist in fact, except for  $\text{NO}_2$  in late autumn. Splitting the samples by measurement site types (see Figure 1) and averaging over both sampling campaigns and sites within each type, a clear trend for  $\text{NO}_2$  appears: substantial underestimation for heavily urbanised areas and slight overestimation for background sites.  $\text{SO}_2$  is slightly underestimated everywhere, not counting the severely underestimated “borough” type 3, where the reason is identified as sharp impact of a local boiler house in one of sites out of four, and the remote background sites (one passive sampling and two monitoring stations), where the averages fit nearly perfectly. However, all the two-week average concentrations of  $\text{SO}_2$  are rather low, including the highest one in mentioned site,  $17 \mu\text{gm}^{-3}$ .

Table 1. Correlation coefficients between modelled and measured site-wise values in passive sampling campaigns.

| Campaign, 2012 | $\text{NO}_2$ | $\text{SO}_2$ |
|----------------|---------------|---------------|
| 13.02 – 26.02  | 0.09          | -0.07         |
| 14.05 – 27.05  | 0.78          | 0.39          |
| 27.08 – 09.09  | 0.68          | 0.53          |
| 19.11 – 03.12  | 0.46          | -0.10         |

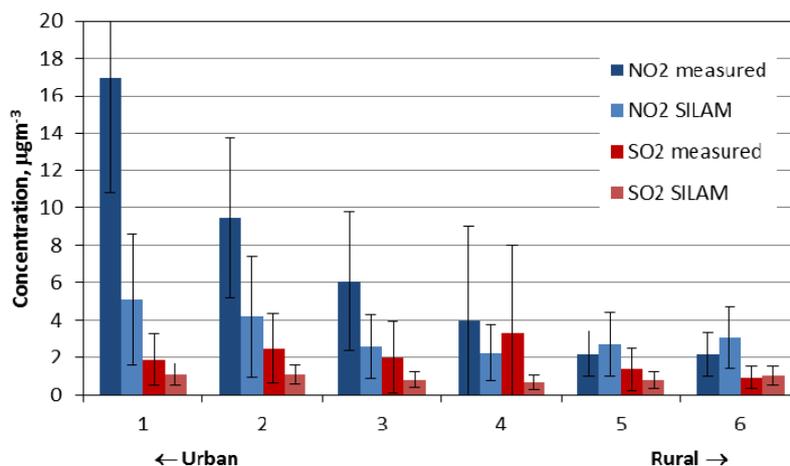


Figure 5. Average concentrations and site-wise standard deviations of measured and modelled NO<sub>2</sub> and SO<sub>2</sub> concentrations (passive sampling sites, plus monitoring stations) in differently inhabited areas (see Figure 1 for classification).

## CONCLUSIONS

In terms of long-term average concentrations, the Estonian application of SILAM tends to “smooth out” the urban-rural differences, producing a rather uniform geographical distribution of pollutants. Although in most urbanised sites the concentrations are underestimated by factor 2 or 3, the low levels of NO<sub>2</sub> and SO<sub>2</sub> in rural and remote sites are reproduced nearly perfectly. This study is maintaining the results of an earlier one based on monitoring sites only: the average modelled values in cities were found too low, however the peak levels during stagnant atmospheric conditions were reproduced fairly (Ots et al., 2013).

The reasons of “smoothing behaviour” of Estonian SILAM application are not completely understood yet. Grid cell size cannot be the single reason of that: central part of Tallinn is big enough to be resolved by 3.3 km resolution, except one street station. In small towns the grid resolution effect must be more decisive. Critical revision of urban emission data is needed – considering that detail emission inventory of city traffic is an extremely complicated task, the emissions may be underestimated in current database. On the other hand, formerly tested peak levels are too high in respect to average values, which is most likely a feature of the model.

## ACKNOWLEDGEMENTS

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