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CALCULATION OF THE TRANSBOUNDARY POLLUTION BY CMAQ CHEMICAL TRANSPORT MODEL AND THE ASSESSMENT OF THE NON-LINEARITY EFFECT.

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Abstract: The comparison of two methods in calculating transboundary concentrations of NO_2 and PM_{10} using CMAQ chemical transport model was presented in Štefánik et al. (2020). There, the transboundary pollution was divided to the pollution unambiguously attributed to foreign sources and that which cannot be unambiguously attributed to the foreign sources. It was proposed that the latter is caused by the non-linear processes present in the atmosphere. It can be expressed by the interaction term. In the present paper, the discussion how the interaction term introduced in Štefánik et al. (2020) is connected to the non-linear interaction term discussed in Thunis et al. (2019) is presented. The main goal of this paper is harmonizing some of the concepts proposed in these two works, which is in accordance with the main purpose of HARMO conferences. The main results of Štefánik et al. (2020) for transboundary concentrations of NO_2 and PM_{10} are also shortly presented.

Key words: *Transboundary pollution, non-linearity, source apportionment, NO_2, PM_{10}*

INTRODUCTION

In the first part of this paper, a short description of the two concepts of the interaction term is presented: the one proposed in Štefánik et al. (2020) and the non-linear interaction term introduced by Thunis et al. (2019), followed by the discussion of their mutual relation. In the second part, the calculation of the transboundary pollution by the two methods introduced in Štefánik et al. (2020) and the estimation of the non-linearity effects is discussed.

INTERACTION TERM AND NON-LINEARITY EFFECT

The problem of non-linear relationship between emission and concentrations changes in the source apportionment is well established. Let us consider a source which contribution to the concentration we would like to assess. The real concentration[1] C_R of given pollutant at given point can by in principle divided to three parts as

$$C_R = C_{ext} + C_{source} + I_{ext,source}, \quad (1)$$

where concentrations C_{ext} *is* coming from external emissions originated outside the source, concentration C_{source} *is* coming from the source emissions, and $I_{ext,source}$ represents the concentration coming from the interaction between the source and the external emissions. $I_{ext,source}$ represents the part of the pollution which can be attributed unambiguously neither to the source nor the external sources. The strength of the interaction term depends on the pollutant, meteorological conditions and assumed external and source emissions. The values of $I_{ext,source}$ can vary from 0 – in this case the source apportionment is straightforward, up to C_R – in this case the origin of the concentration is solely secondary due to the interaction of other pollutants coming from the source with external sources.

[1] In this paper, the term *the real concentrations* means the actual concentrations which can be measured with some degree of uncertainty.

Thunis et al. (2019) suggest that even with non-zero $I_{ext,source}$ in case of low emission reduction the sources could be apportioned unambiguously to a certain degree. Let us slightly reannotate their equations into a more general form. The impact of α reduction of source emissions on the concentrations is defined as

$$[\,sourceIM_{\alpha}]_{\%} = \frac{\Delta C_{source(\alpha)}}{\alpha C_R} \times 100\ \%, \tag{2}$$

and, similarly, the impact of α reduction of external emissions on the concentrations can be written as

$$[\,extIM_{\alpha}]_{\%} = \frac{\Delta C_{ext(\alpha)}}{\alpha C_R} \times 100\ \%\ , \tag{3}$$

where the $\Delta C_{source(\alpha)}$ and $\Delta C_{ext(\alpha)}$ represent the concentration changes resulting from α × 100 % percentage reduction of the source and external emissions, respectively. The sum of the two impacts obeys (Štefánik, 2020)

$$[\,sourceIM_{\alpha}]_{\%} + \ [\,extIM_{\alpha}]_{\%} = \left(1 - \frac{\hat{I}_{ext,source(\alpha)}}{\alpha C_R}\right) \times 100\ \%, \tag{4}$$

where the non-linear[2] interaction term $\hat{I}_{ext,source(\alpha)}$ is defined in Eq. (1) of Thunis et al. (2019) paper. Thunis et al. (2019) demonstrates that the absolute value of $\hat{I}_{ext,source(\alpha)}$ should be lower with lower value of α. For $\alpha \leq \alpha_t$, the interaction term should be zero, which implies that with emission reductions lower than α_t the sources can be apportioned unambiguously. What does it actually mean? It means that lowering only source (external) emissions by α causes a concentration reduction by percentage of $[\,sourceIM_{\alpha}]_{\%}$ ($[\,extIM_{\alpha}]_{\%}$) of the concentration reductions achieved in case of α-reduction of both source and external emissions. The sum of both impacts is equal to 100 % in this case. However, in case of emission reduction more than α_t, this sum exceeds 100 % and, therefore, the interpretation of the impacts is not straightforward. In Štefánik et al. (2020), it is shown that, in the special case of α =1, the interaction terms in equations (1) and (2) can by related as

$$\hat{I}_{ext,source(\alpha=1)} = -\,I_{ext,source}. \tag{5}$$

It is easy to show that the values of non-linear interaction term from (Thunis et al. 2019) are in following range

$$\begin{aligned} &\hat{I}_{ext,source(\alpha)} = 0 && \text{for} && \alpha \leq \alpha_t \\ &\hat{I}_{ext,source(\alpha)} \in \langle -\,I_{ext,source},\, 0) && \text{for} && \alpha > \alpha_t \quad . \end{aligned} \tag{6}$$

Note that the threshold value α_t is different in various situations and several simulations of the chemical-transport model (CTM) need to be performed in order to estimate it.

The knowledge of the interaction term $I_{ext,source}$ gives us an insight into the limitations of source apportionment. From the equations (4) and (6) we can see that when its value is negligible in comparison with C_R, the source can be apportioned in any value of emission reduction level α. The value of the interaction term $I_{ext,source}$ can be obtained by running the chemical transport model in three different configurations. The first configuration represents the simulation in which all emissions in the domain are taken into account. By performing the full run simulation[3] one can obtain the values of C_M

$$C_M = C^M_{ext} + C^M_{source} + I^M_{ext,source}. \tag{7}$$

[2] The term *non-linear* appears here because due to the presence of interaction term $I_{ext,source}$, concentrations change $\Delta C_{source(\alpha)}$ and $\Delta C_{ext(\alpha)}$ are not linear with respect to α.

[3] *full run of the model* refers to the simulation taking into account all emissions in the domain.

This equation is identical with equation (1), except for the index *M*, which is introduced in order to distinguish modelled values from the real ones. The second configuration represents running the model without the targeted source; it results in concentrations C_{ext}^{M}, while from the last configuration including emissions solely from the targeted source, C_{source}^{M} are obtained. The desired term $I_{ext,source}^{M}$ is computed by subtracting the last two simulations from the full run simulation as: $I_{ext,source}^{M} = C_M - C_{ext}^{M} - C_{source}^{M}$. The difference between $I_{ext,source}^{M}$ and the real value $I_{ext,source}$ depends on the quality of the chemical and physical mechanisms included in the model. This difference can be potentially used as a test of the quality of chemical transport models in in some field experiments.

TWO METHODS OF CALCULATION OF TRANSBOUNDARY POLLUTION

In order to determine the interaction term defined in equation (1) in case of the transboundary pollution estimation in Slovakia, two different methods denoted as Method1 and Method2 were proposed as

$$\begin{aligned} T_M(Method1) &= C_{ext}^{M} \\ T_M(Method2) &= C_M - C_{SK}^{M} \\ with \quad C_M &= C_{ext}^{M} + C_{SK}^{M} + I_{ext,SK}^{M}, \end{aligned} \tag{8}$$

where modelled transboundary concentrations by Method1: $T_M(Method1)$ are obtained by switching off all emissions from the territory of Slovakia, while emissions outside the boundaries remain the same as in the full model run. The modelled transboundary concentrations by Method2: $T_M(Method2)$ are calculated as the difference between the full model run concentrations and concentrations obtained by the model run including only Slovak emissions while the boundary conditions for all pollutants except for O_3 are set to zero. From Eq. (8) one can see that the difference between $T_M(Method2)$ and $T_M(Method1)$ is just the interaction term $I_{ext,SK}^{M}$.

In this paper, the transboundary concentrations are estimated using $T_M(Method1)$ which is unambiguously attributed to foreign sources. The pollution which cannot by unambiguously attributed to the foreign sources is identified as $I_{ext,SK}^{M}$. Both portions of the contribution - unambiguously and ambiguously attributed to the foreign sources - are de-biased using bias values between observed and modelled concentrations by method described in paper (Štefánik et al., 2020).

The Community Multiscale Air Quality modelling system CMAQ developed by EPA National Exposure Research Laboratory in Research Triangle Park, NC (EPA, 2008) was used for the simulations. The simulation was performed on the computational model domain with 103×184 grid cells of 4.7 km resolution for the year 2015. This domain was nested in a mother domain with 169×134 grid cells and spatial resolution of 14.1 km. Hourly meteorological fields were generated by the Weather Research Forecasting (WRF) model version 3.9.1 (Skamarock et al.,2008) using data from the European Centre for Medium-Range Weather Forecast (ECMWF) reanalysis as the boundary and initial conditions. The gridded speciated hourly-resolved emissions were configured as in the LIFE-IP Malopolska project (Ondřej Vlček et al., 2019). More details about model configuration are in Štefánik et al. (2020).

Results for NO_2

These results are a brief summary of those published in Štefánik et al. (2020). The estimated annual mean transboundary NO_2 concentrations unambiguously attributed to foreign sources T_R and its uncertainty δ are plotted in Figure 1 the annual mean of the interaction term $I_{ext,SK}^{R,ann}$ with its uncertainty δ is plotted on the right. The estimated values of the interaction term in Slovakia are $I_{ext,SK}^{R,ann}$= -0.0006 ± 0.0509 μg/m^3 with maximum and minimum values 0.7 and -0.1 μg/m^3 respectively. The uncertainty associated with the model and its input data (lower left panel in Figure 1) are δ =1.5±0.6 μg/m^3 with maximum and minimum values 4.1 and 0.5 μg/m^3, respectively. Therefore, the uncertainty of calculated NO_2 transboundary annual mean concentrations which comes from the presence of the non-linear interaction term is small in comparison

with the uncertainty which comes from the model itself and its input. However, in Štefánik et. al (2020), it was shown that the interaction term expressed in hourly concentrations can be very large in some episodes. Mean measured concentrations of NO_2, estimated transboundary concentrations T_R and concentrations which cannot be attributed to national nor foreign sources $I_{ext,SK}^{R,ann}$ at the places of monitoring sites for year 2015 are plotted in Figure 2.

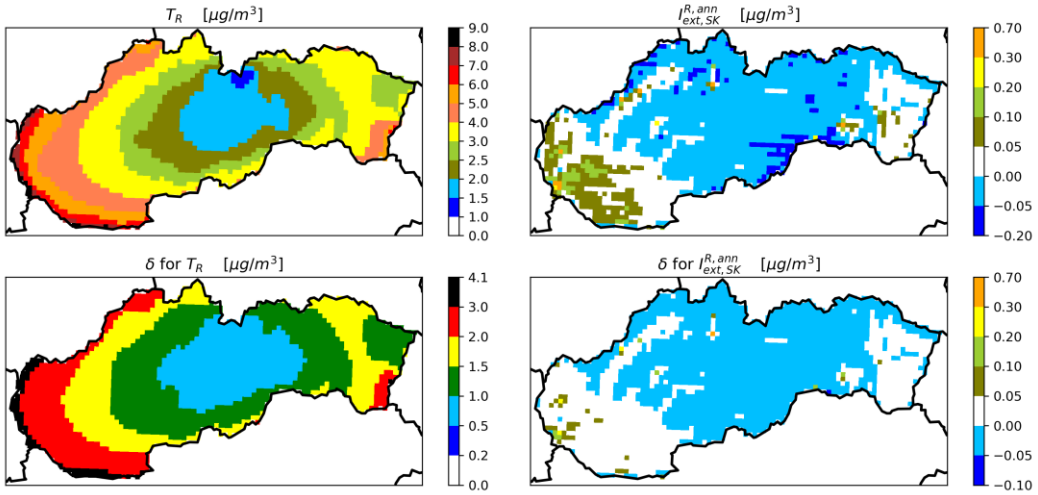

Figure 1. Left panel: Estimated mean transboundary NO_2 concentrations unambiguously attributed to foreign sources T_R (upper panel) and its uncertainty δ (lower panel). Right panel: Estimated mean NO_2 concentrations which cannot be attributed unambiguously to national nor foreign sources $I_{ext,SK}^{R,ann}$ (upper panel) and its uncertainty δ (lower panel).

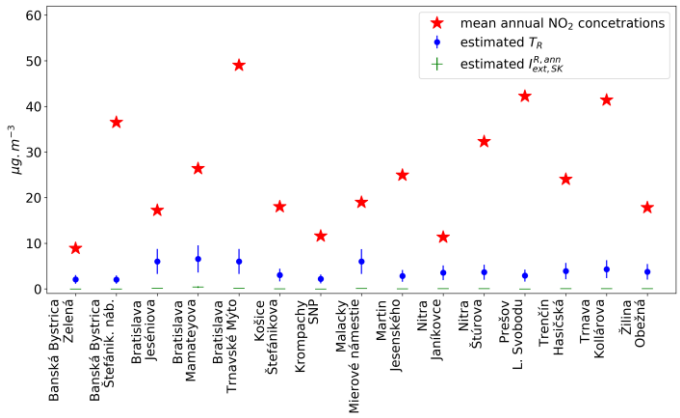

Figure 2. Mean measured concentrations of NO_2, estimated transboundary concentrations T_R and concentrations which cannot be attributed to national nor foreign sources $I_{ext,SK}^{R,ann}$, at the monitoring sites.

Results for PM_{10}

From Figure 3 we can see that, contrary to the NO_2 case, it can be concluded that in the calculation of annual mean transboundary PM_{10} concentration the uncertainty associated with the model and its input data can be comparable to that coming from the non-linearity of the model. Indeed, for PM_{10} the non-linearity is important even in case of annual mean calculations of transboundary pollution and can reach 2.7 $\mu g/m^3$ and up to 25% of the calculated transboundary pollution. The results of calculated transboundary pollution at the monitoring sites in 2015 are plotted in Figure 4 together with measured annual mean concentrations.

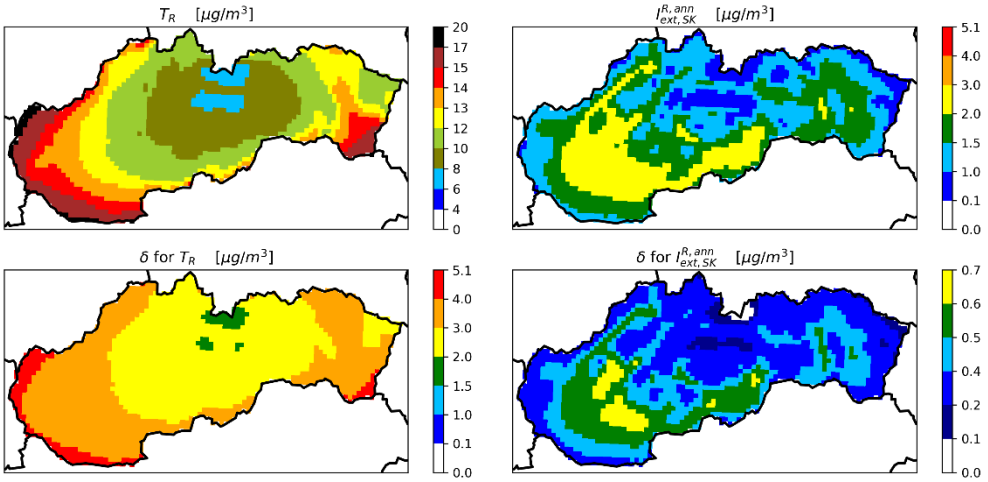

Figure 3. Left panel: Estimated mean transboundary PM_{10} concentrations unambiguously attributed to foreign sources T_R (upper panel) and its uncertainty δ (lower panel). Right panel: Estimated mean PM_{10} concentrations which cannot be attributed unambiguously to national nor foreign sources $I^{R,ann}_{ext,SK}$ (upper panel) and its uncertainty δ (lower panel).

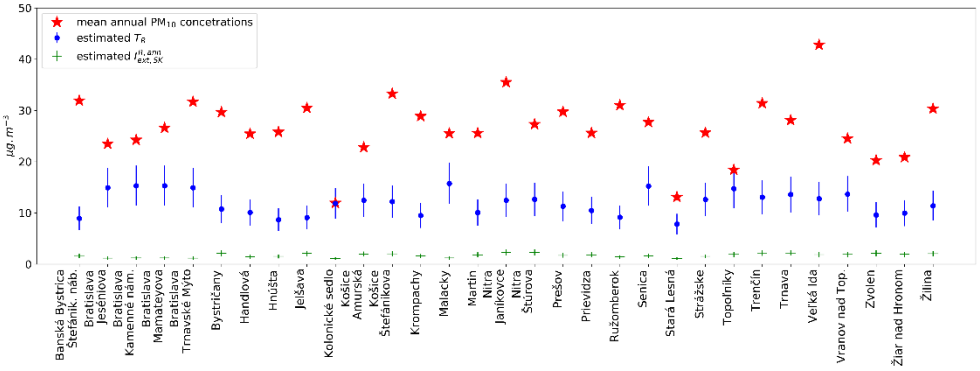

Figure 4. Mean measured concentrations of PM_{10}, estimated transboundary concentrations T_R and concentrations which cannot be attributed to national nor foreign sources $I^{R,ann}_{ext,SK}$, at the monitoring sites.

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