

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



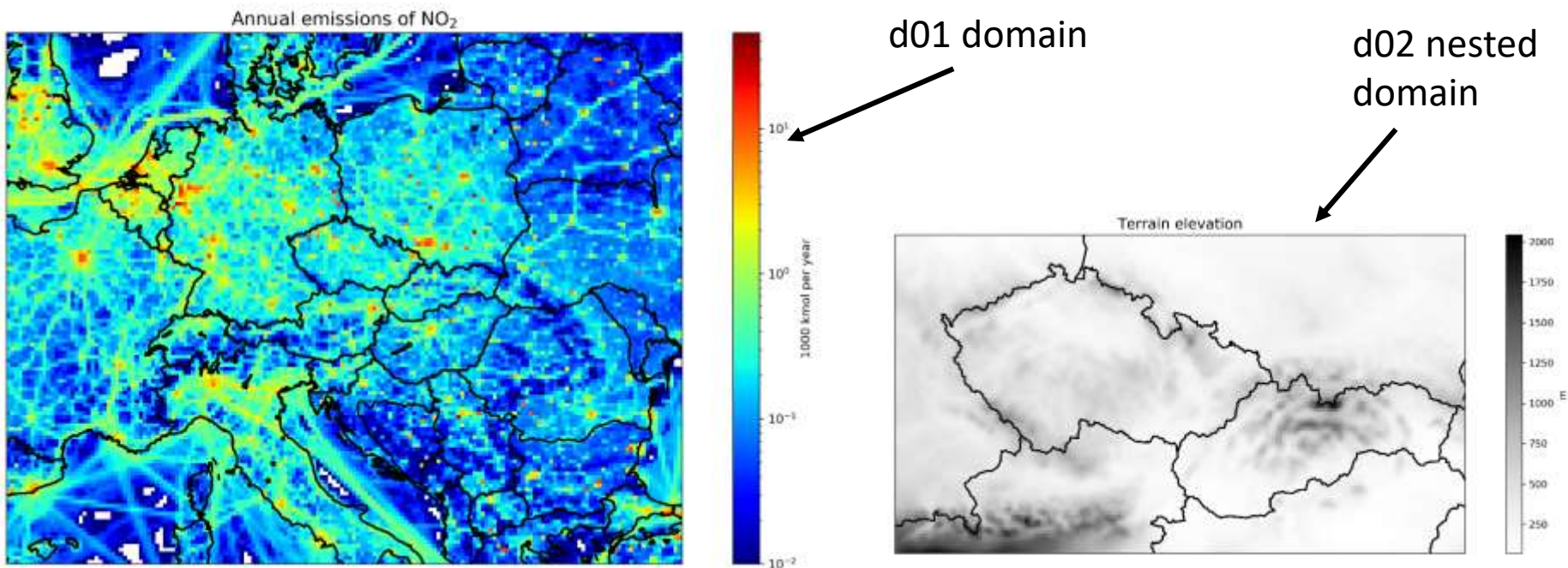
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# Motivations

- 1. Quantify the transboundary pollution over the Slovakia for  $\text{NO}_2$  and  $\text{PM}_{10}$  in 2015**
- 2. Quantify the pollution which cannot be unambiguously attributed to the foreign sources nor the national sources**
- 3. Discussed the interaction term and the non-linearity**

- 0. Model configuration**
- 1. Two methods of calculation of the transboundary pollution**
  - 1.1. Difference between the two methods**
- 2. Estimation of the real transboundary concentrations**
- 3. Results for NO<sub>2</sub> and PM<sub>10</sub> 2015**
- 4. Results for PM<sub>10</sub> from SHERPA model**
- 5. The interaction term and the non-linearity**
- 6. Conclusions**



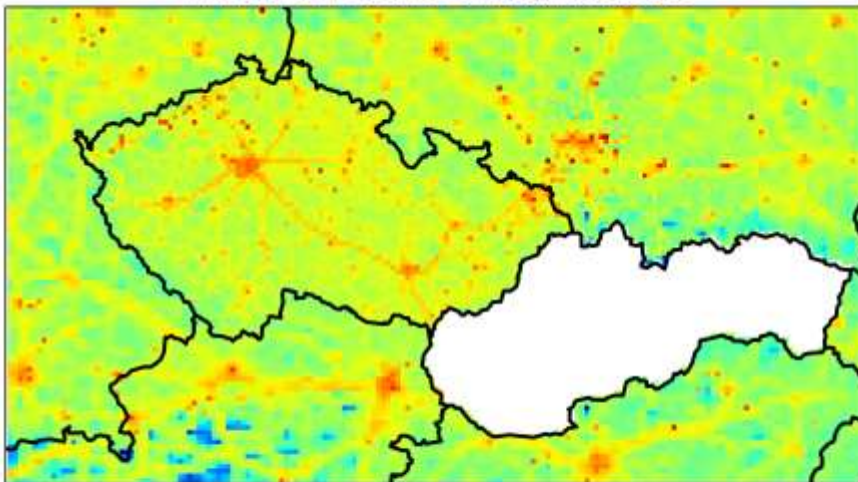
The CMAQ version v4.7.1 (US EPA, 2010; Byun and Schere, 2006) with the cb05cl gas-phase chemistry mechanism (Yarwood et al., 2005) and the AERO4 version of the aerosol module (Binkowski, 2003).

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.

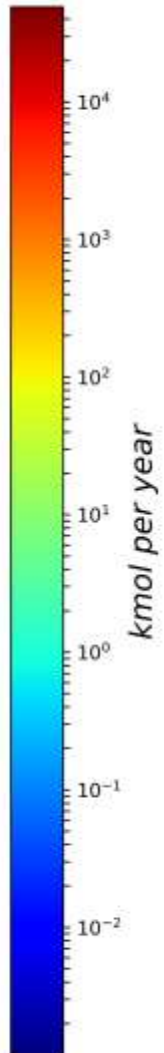
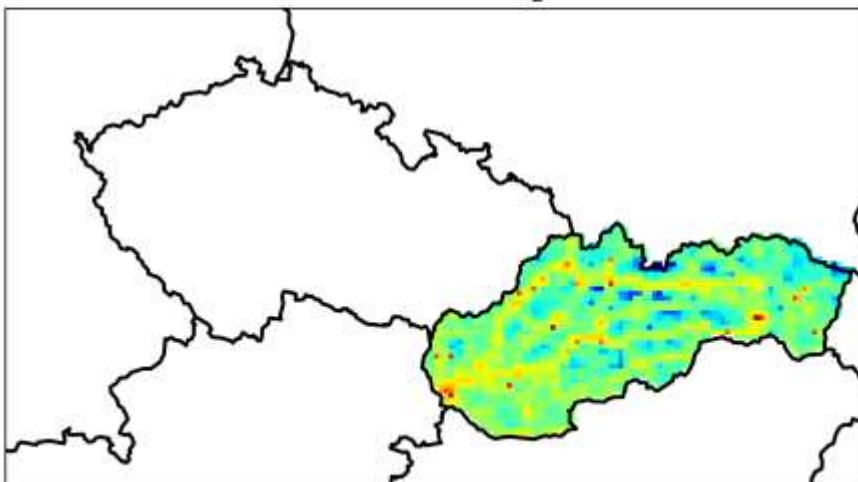
Emissions TNO MAC-III 2015 database (Kuenen et al., 2014), for the SK, CZ and PL from LIFE-IP Malopolska project (Ondřej Vlček et al., 2019).

# 1. Two methods of calculation the transboundary pollution

Annual emissions of NO<sub>2</sub> method1

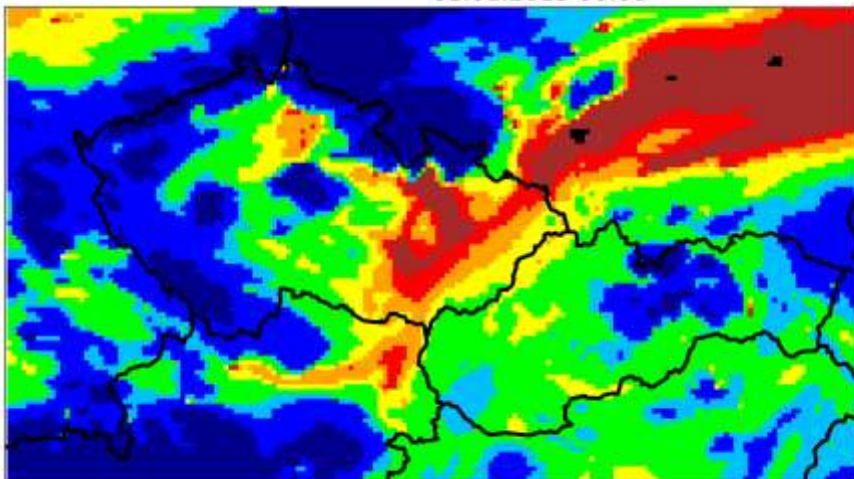


Annual emissions of NO<sub>2</sub> method2

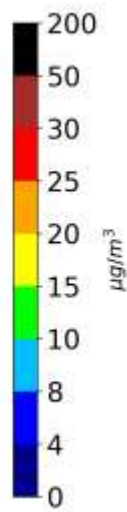
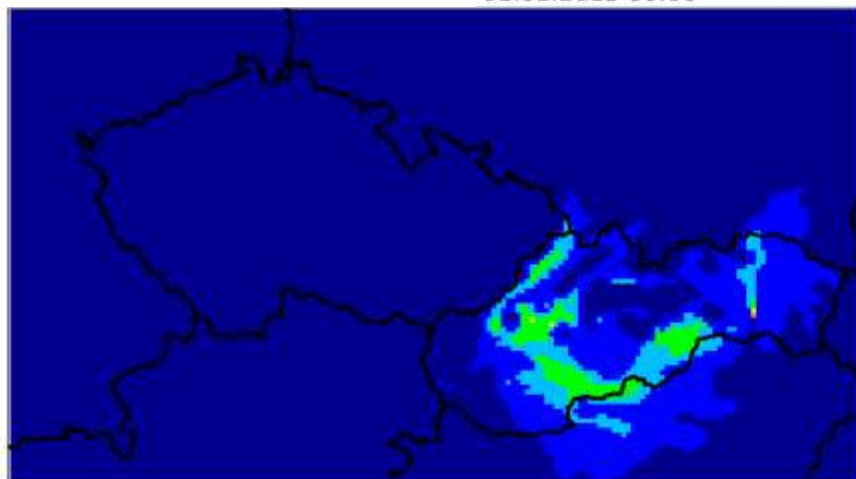


# 1. Two methods of calculation the transboundary pollution: $PM_{10}$ concentrations

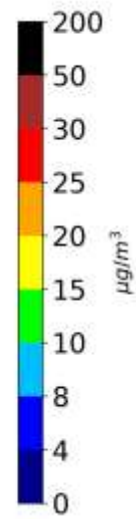
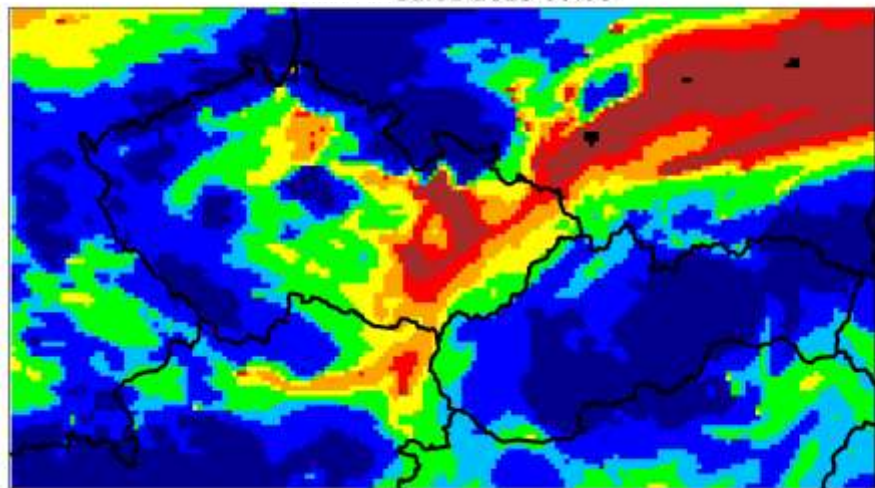
Full model simulations 01.01.2015 00:00



Model included only Slovak emissions 01.01.2015 00:00



Method1 (without Slovak emissions) 01.01.2015 00:00



## 1.1. Difference between the two methods

The real concentration  $C_R$  of given pollutant at given point can be in principle divided to three parts as


$$C_R = C_{ext} + C_{source} + I_{ext,source}$$

from external emissions  
originated outside the source

from the source emissions

from the interaction between  
the source and the external  
emissions

Model is not full reality, therefore we introduce the superscript  $M$

$$C_M = C_{ext}^M + C_{source}^M + I_{ext,source}^M$$

## 1.1. Difference between the two methods

The transboundary concentrations calculated by the two methods can be write as

$$T_M(\text{Method1}) = C_{ext}^M$$

$$T_M(\text{Method2}) = C_M - C_{SK}^M$$

$$\text{with } C_M = C_{ext}^M + C_{SK}^M + I_{ext,SK}^M,$$

1. The transboundary concentrations are estimated using  $T_M(\text{Method1})$  which is unambiguously attributed to foreign sources
2. The pollution which cannot by unambiguously attributed to the foreign sources is identified as  $I_{ext,SK}^M$  . It is calculated by diference between between  $T_M(\text{Method2})$  and  $T_M(\text{Method1})$ .



## 2. Estimation of the real transboundary concentrations

**NO<sub>2</sub> 2015**

r 0.66  
BIAS -6.44 µg/m<sup>3</sup>  
RMSE 7.73 µg/m<sup>3</sup>  
MQI 0.989

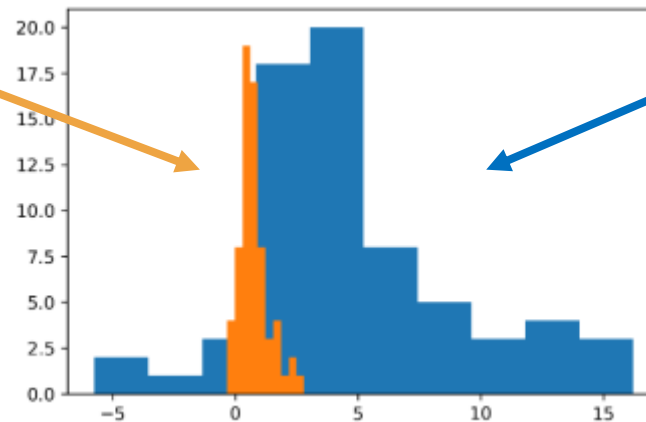
$$C_R = C_M + \Delta$$

rewritten more suitably

$$C_R = C_M + c \times C_M$$

for 101 background stations within domain

variation c

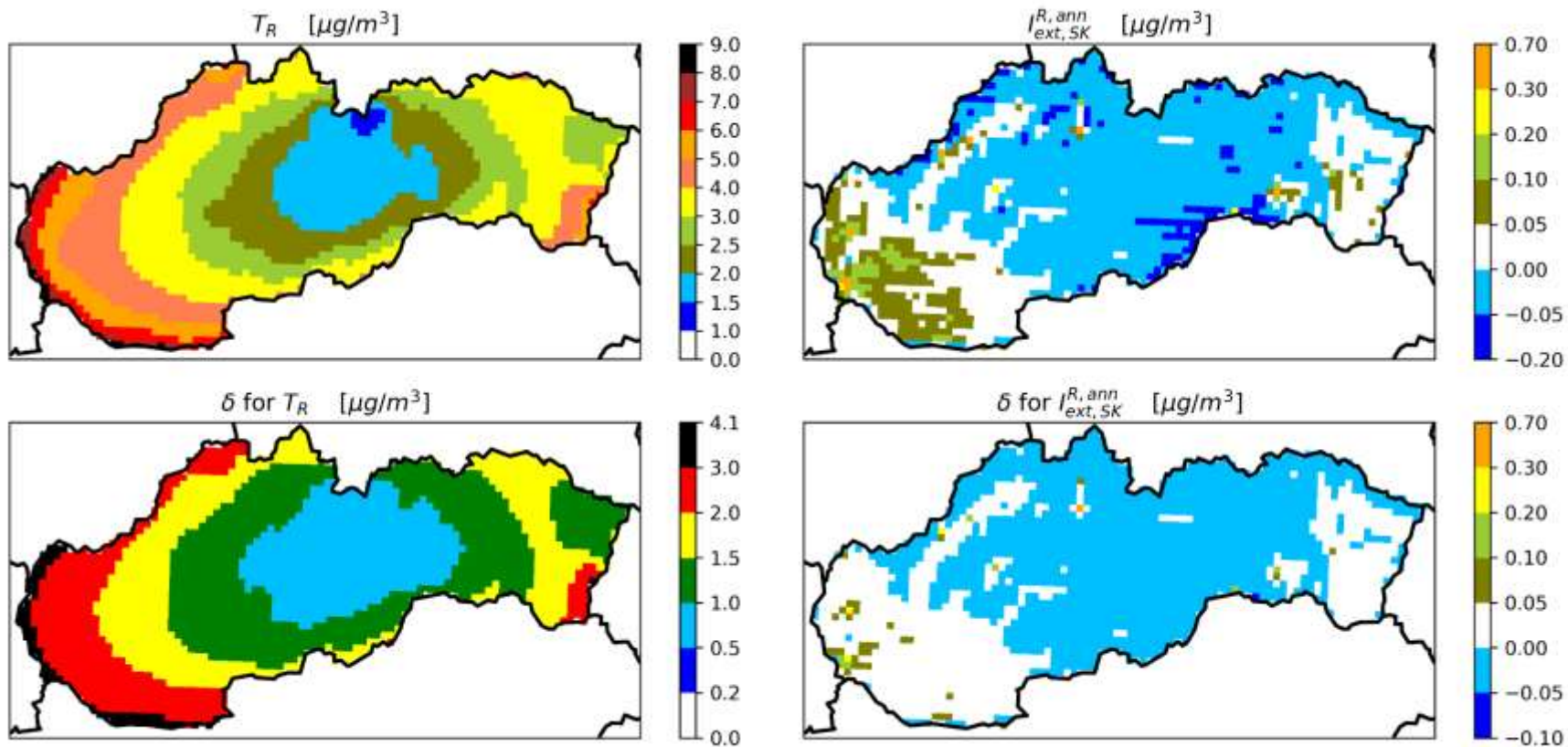


$$c_{\text{rem}} = 1.11 \pm 0.96$$

$$T_R = T_M + t \times T_M$$

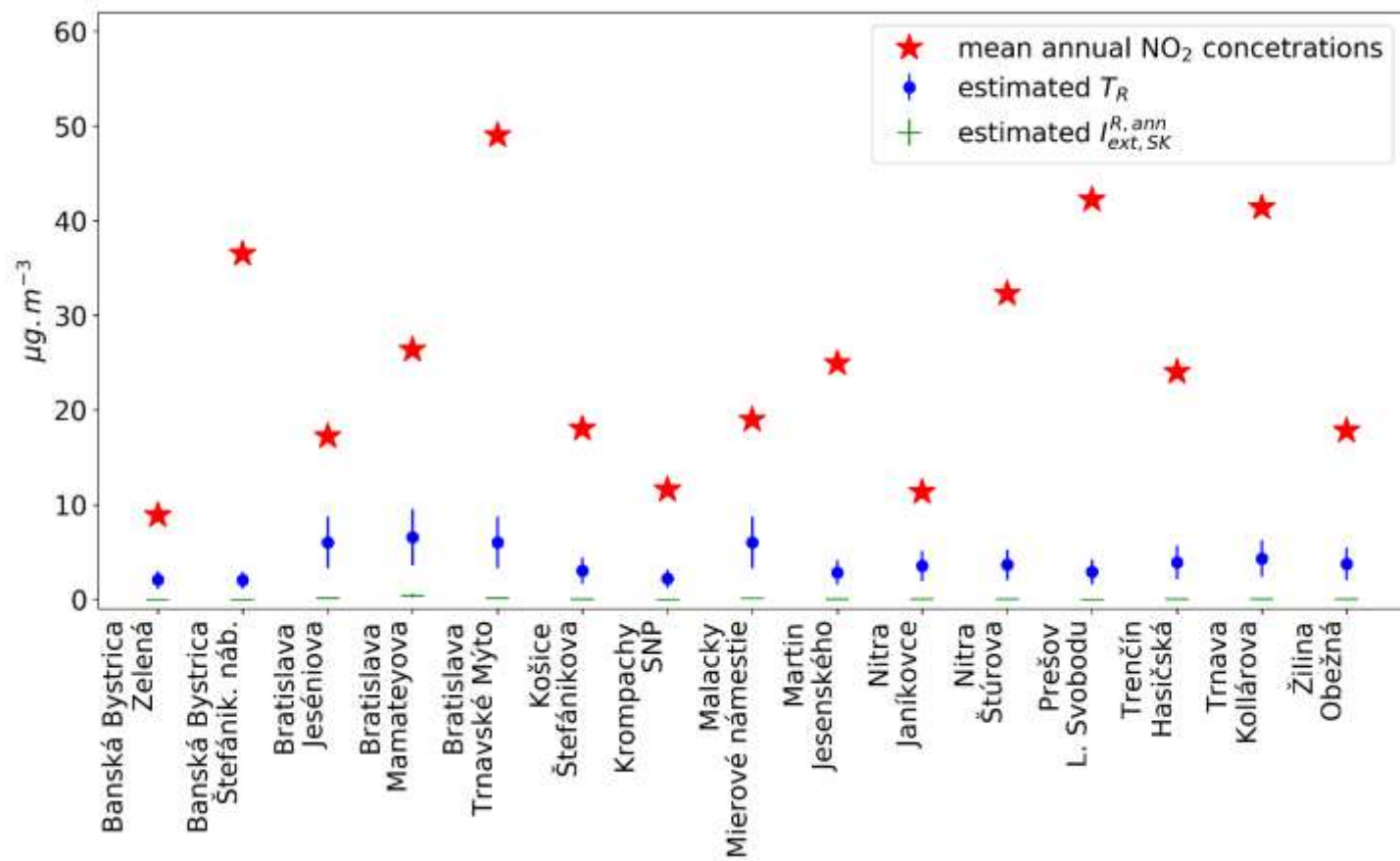
$$t \equiv c_{\text{rem}}$$

### 3. Results for NO<sub>2</sub> 2015

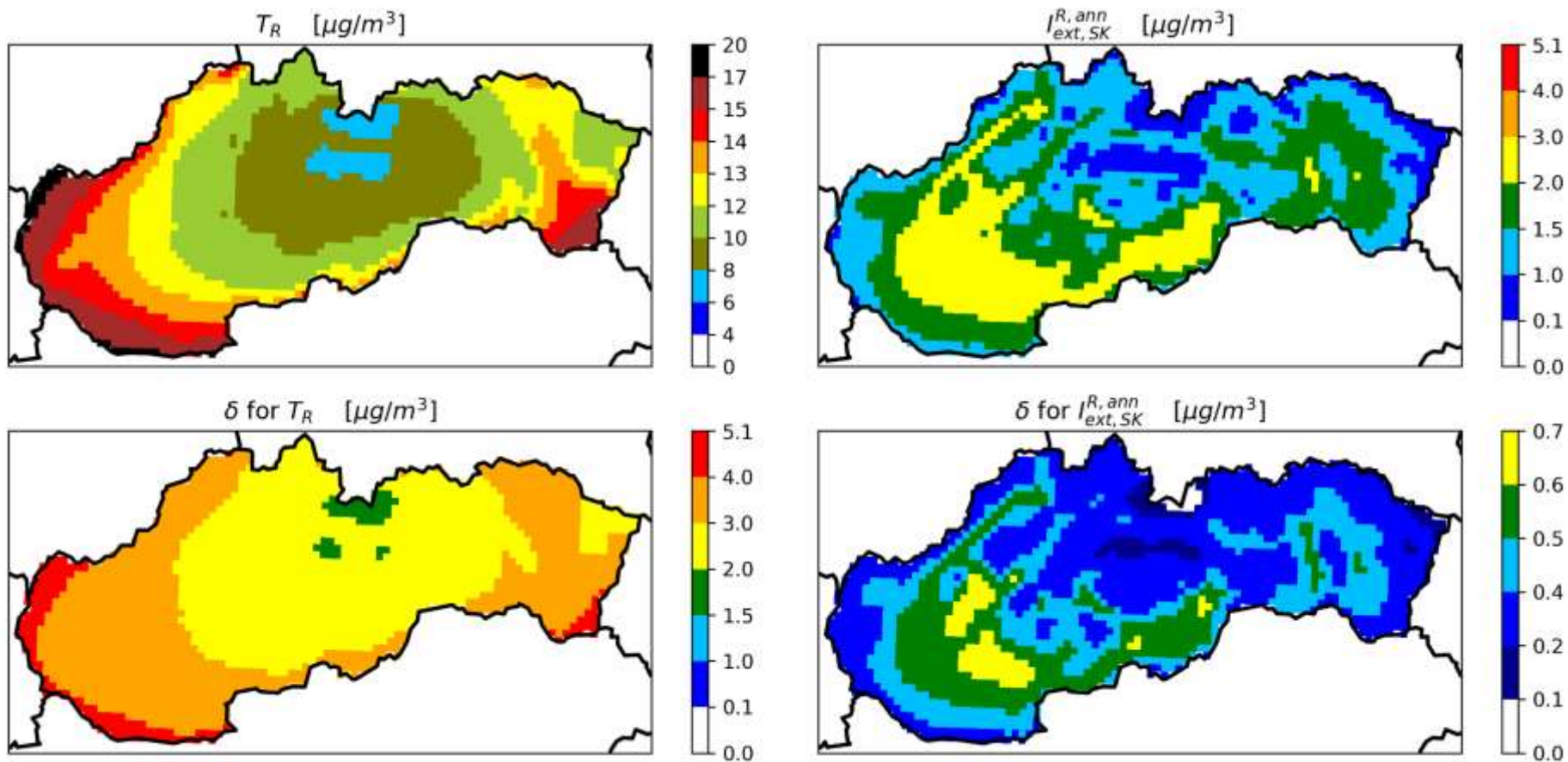


Concentrations which comes from the presence of the non-linear interaction term is small in comparison with the uncertainty which comes from the model itself and its input. However, we shown that the interaction term expressed in hourly concentrations can be very large in some episodes.

### 3. Results for NO<sub>2</sub> 2015

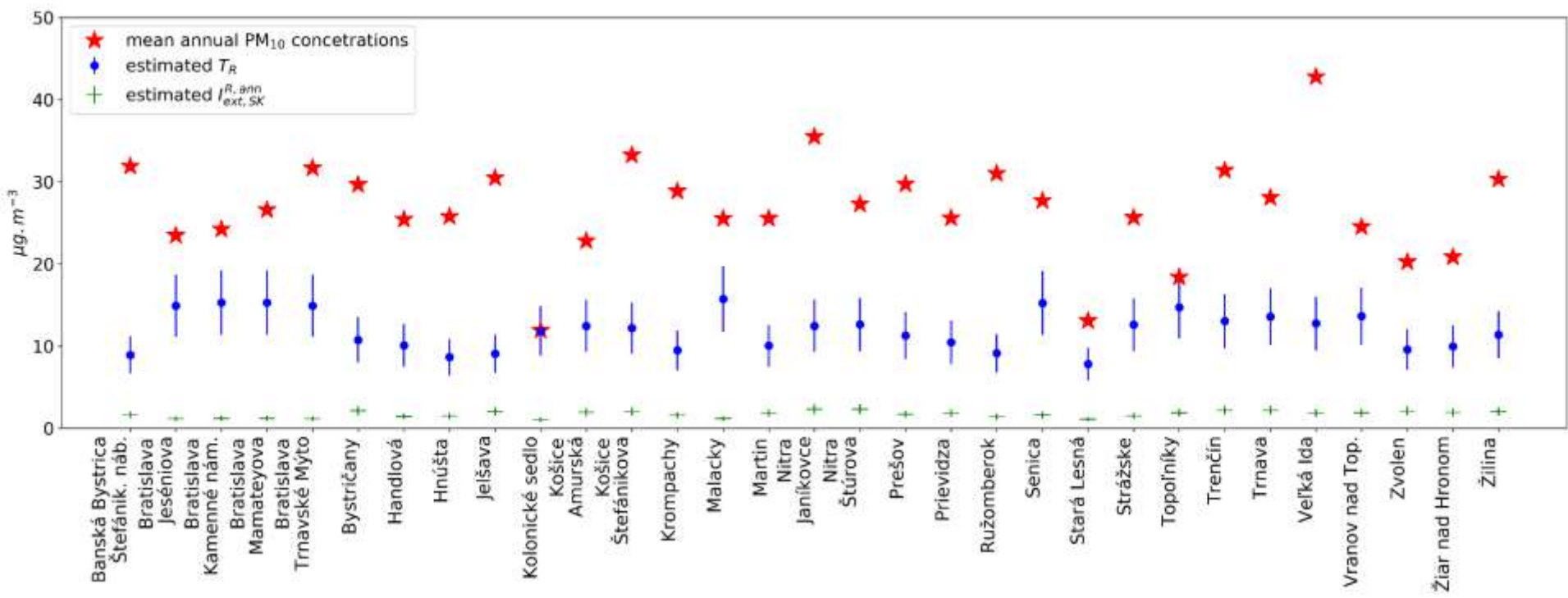


### 3. Results for PM<sub>10</sub> 2015



In the calculation of annual mean transboundary PM<sub>10</sub> 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<sub>10</sub> the non-linearity is important even in case of annual mean calculations of transboundary pollution and can reach  $2.7 \mu\text{g}/\text{m}^3$  and up to 25% of the calculated transboundary pollution

# 3 Results for PM<sub>10</sub> 2015

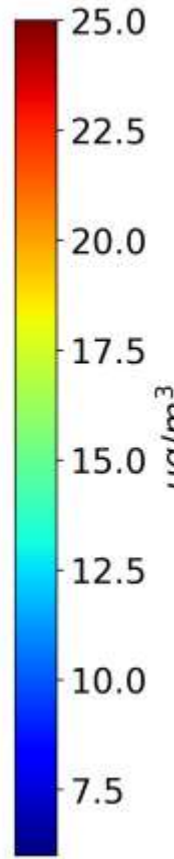
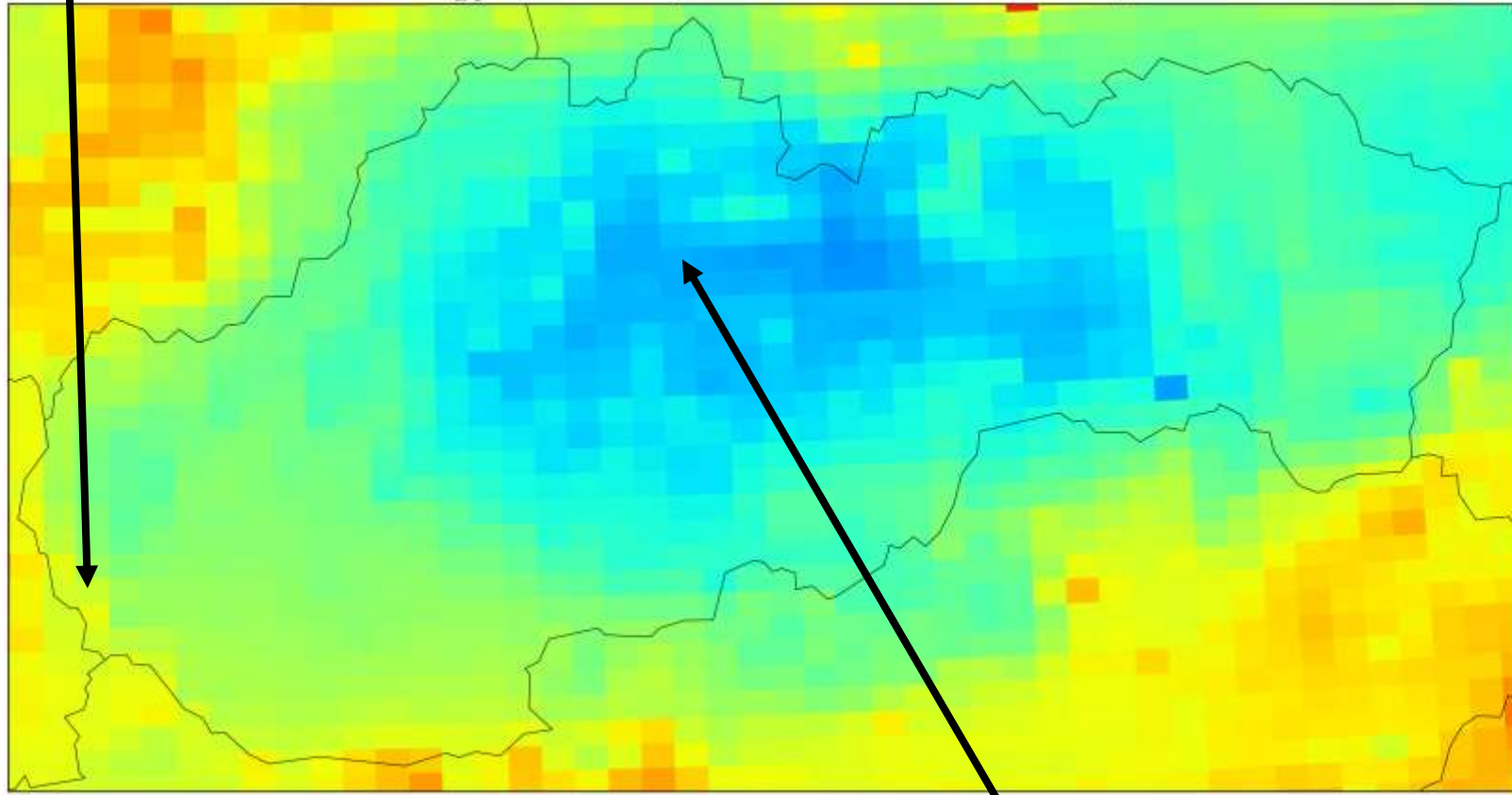


# 4. Results for PM<sub>10</sub> with SHERPA model

Quite realistic near border

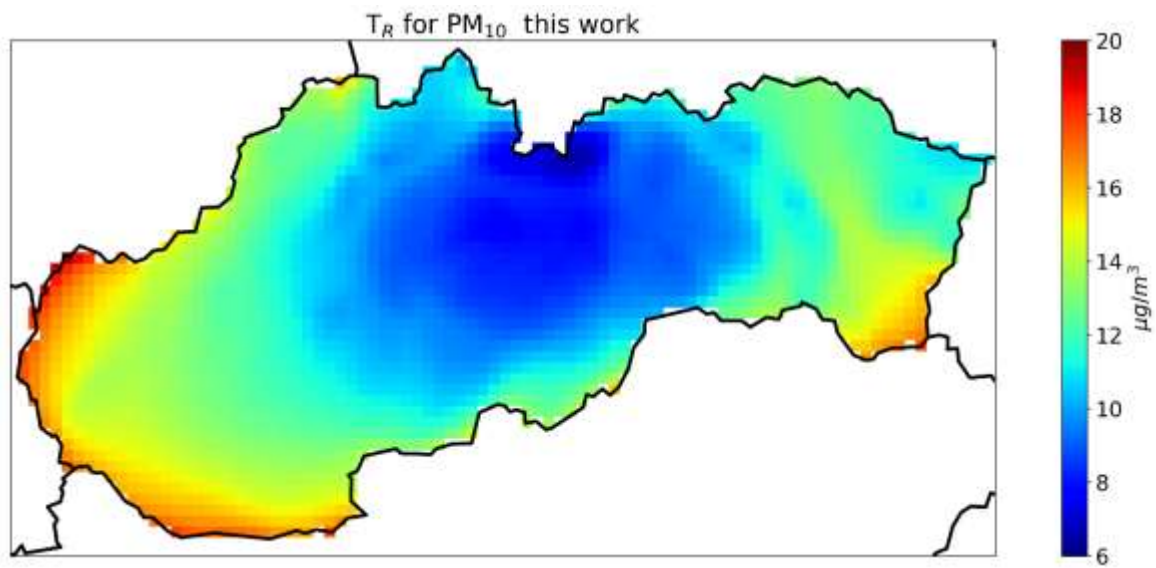
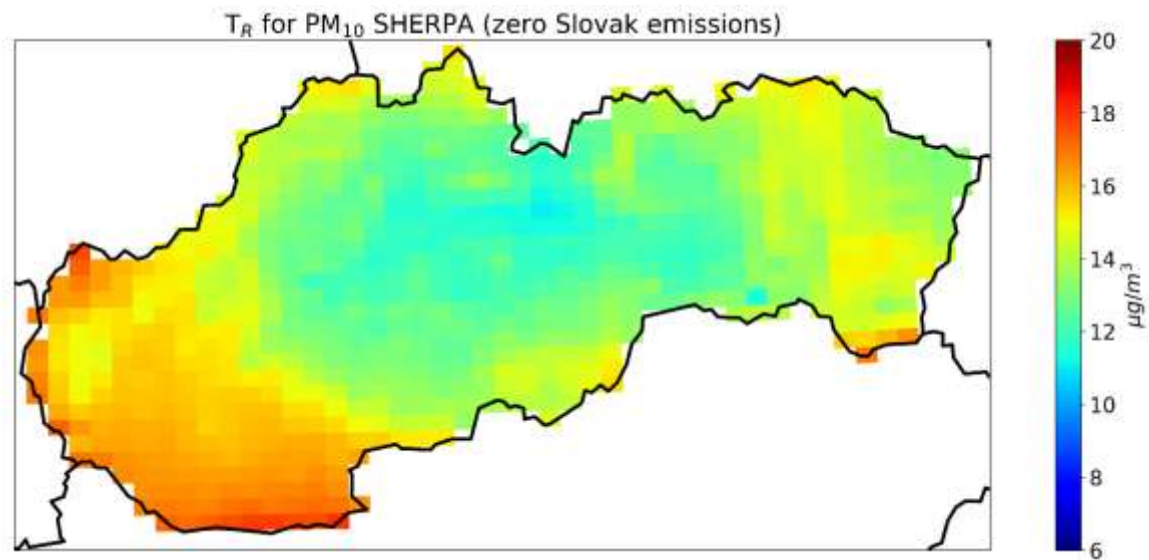
Based on 50 % emissions reduction and then downscaled

PM<sub>10</sub> SHERPA (zero Slovak emissions)

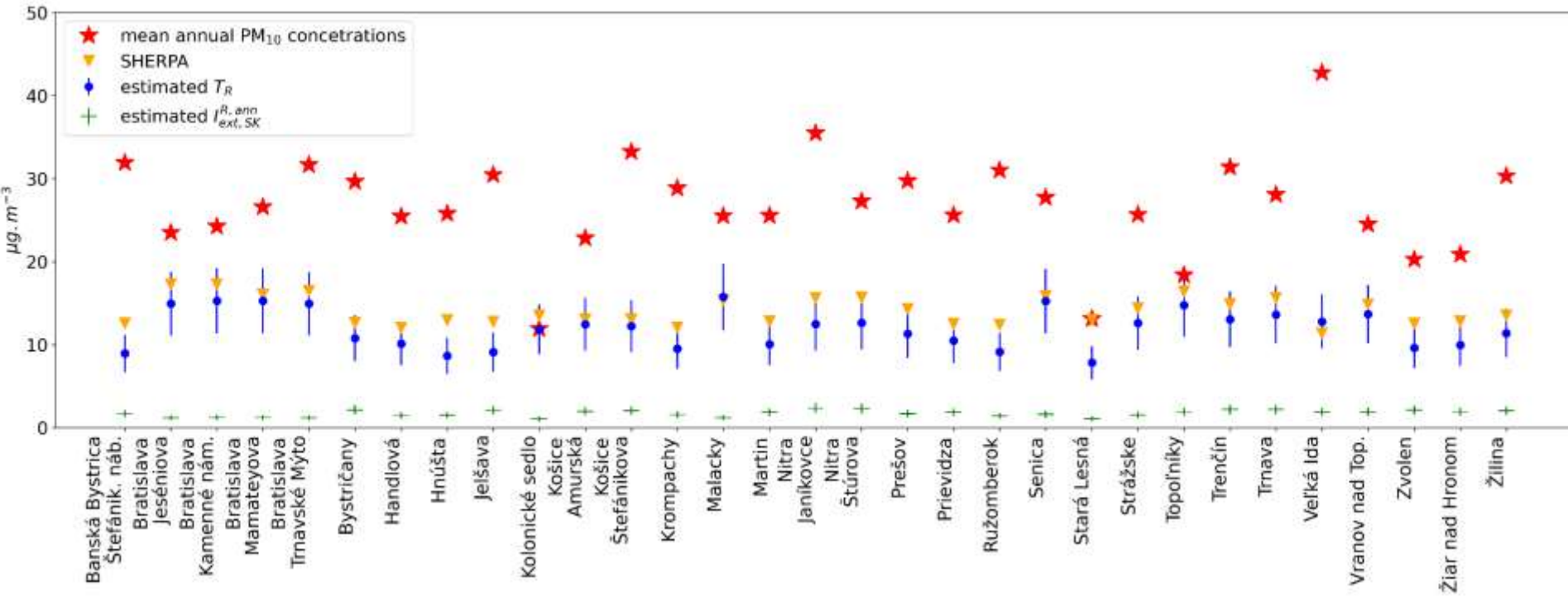


Overestimate the concentrations far from the border and on higher altitudes: i.e Chopok concentrations  $\approx 4-5 \mu\text{g}/\text{m}^3$  but SHERPA without SK emissions  $\approx 12 \mu\text{g}/\text{m}^3$

## 4. Results for PM<sub>10</sub> with SHERPA model



## 4. Results for PM<sub>10</sub> with SHERPA model



SHERPA includes different meteorology, emissions and methodology, but one can see that SHERPA results are more uniform (smaller variation within the domain)

It performs similarly near the borders, but overestimate results far from the borders

It has some artefacts (station Velka Ida has lowest transboundary concentrations, but it is close to the border )



## 5. The interaction term and the non-linearity

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  $\alpha$  reduction of source emissions on the concentrations is defined as

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

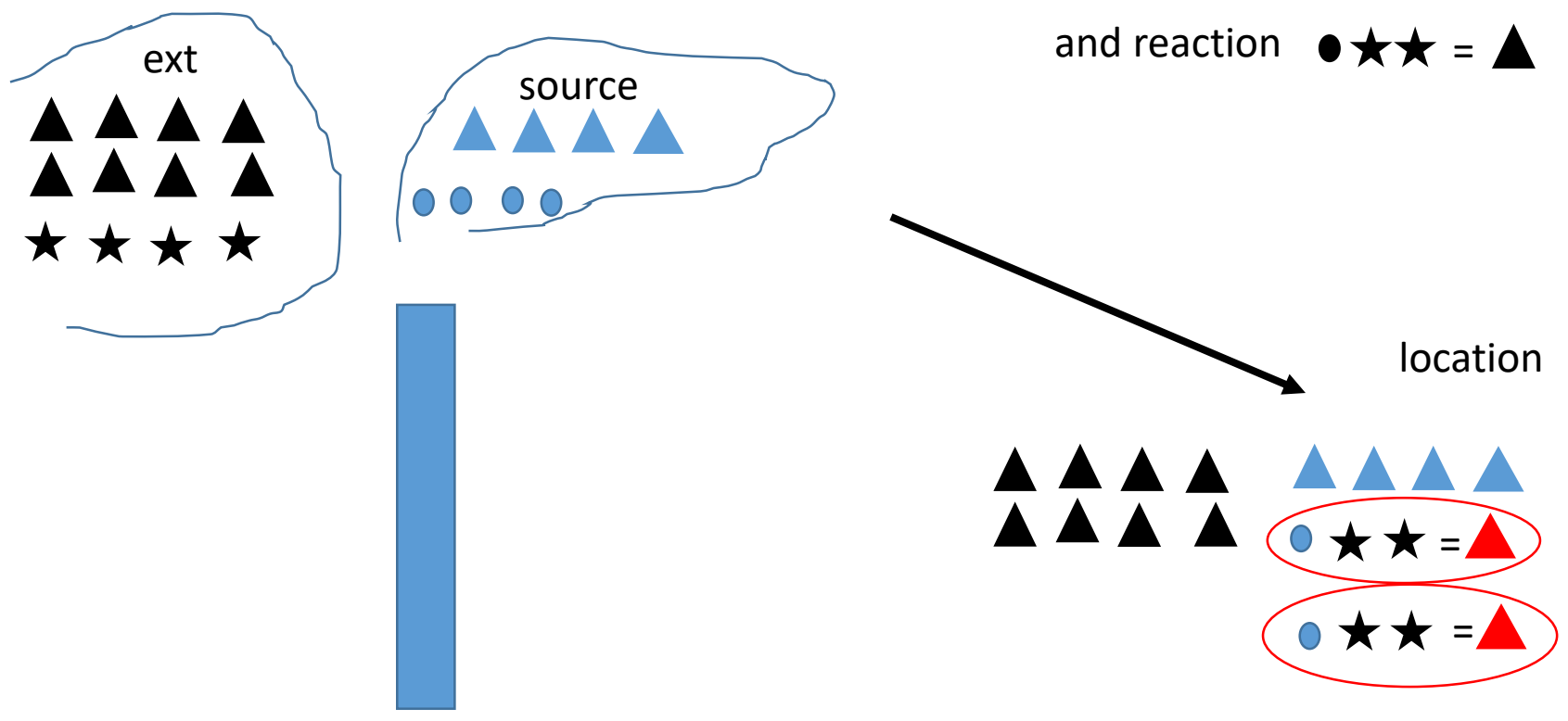
and, similarly, the impact of  $\alpha$  reduction of external emissions on the concentrations can be written as

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

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

# 5. Example

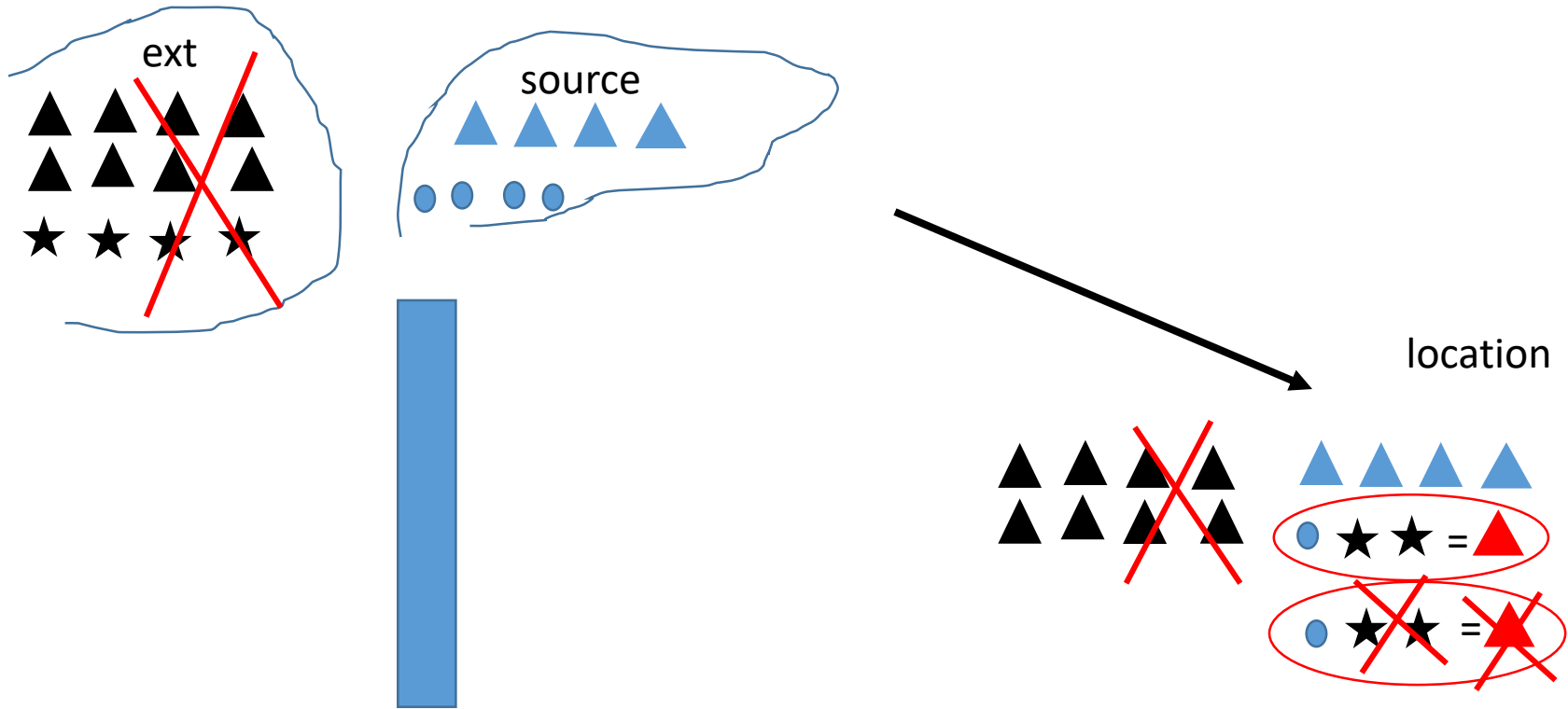
Consider concentrations of pollutant ▲ and reaction ●★ ★ = ▲



$$C_R = C_{ext} + C_{source} + I_{ext,source}$$

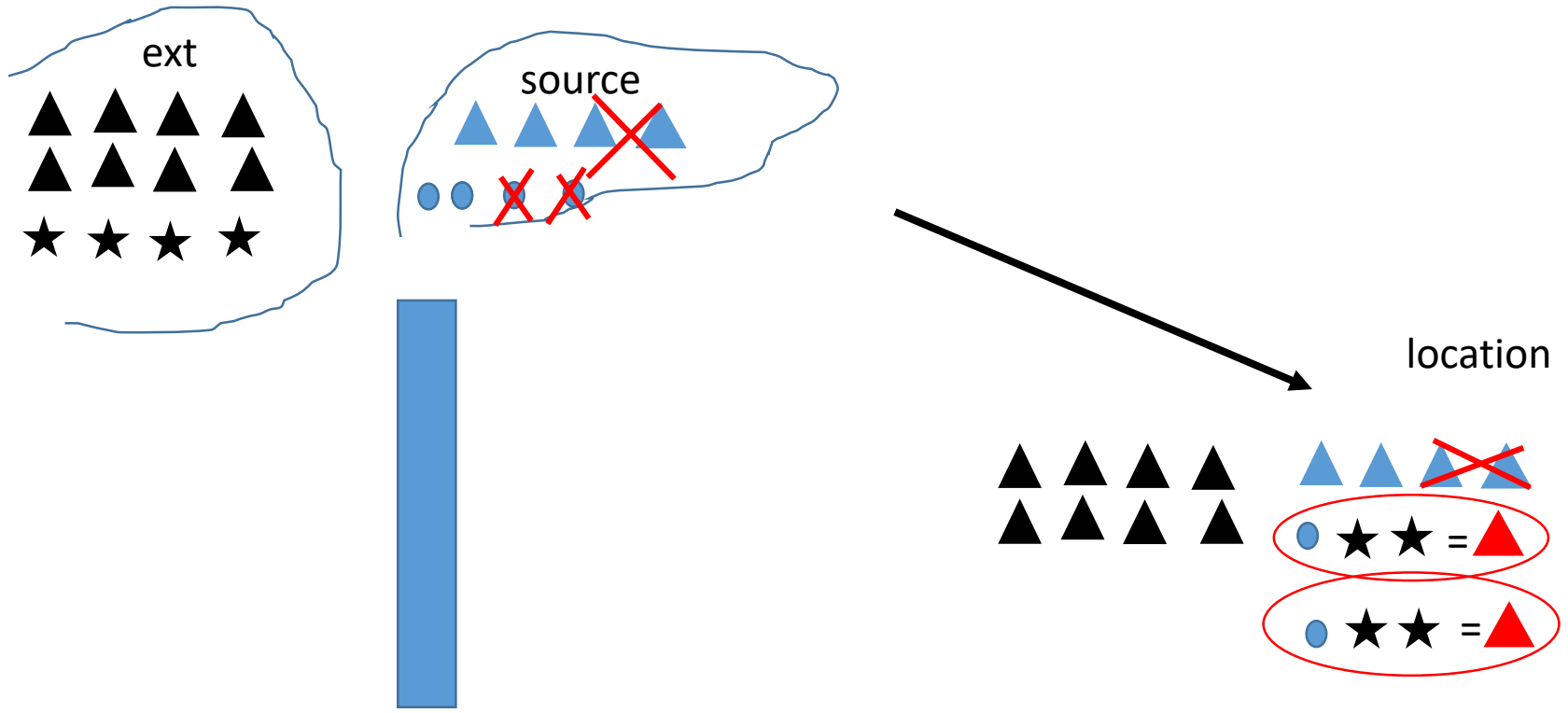
$$14 = 8 + 4 + 2$$

# 5. Example : $\alpha = 50\%$ reduction



$$[extIM_{0.5}]_{\%} = \frac{\Delta C_{ext(0.5)}}{0.5C_R} \times 100\% = \frac{14-9}{0.5 \cdot 14} \times 100\% = \frac{5}{7} \times 100\% = 71\%$$

# 5. Example : $\alpha = 50\%$ reduction

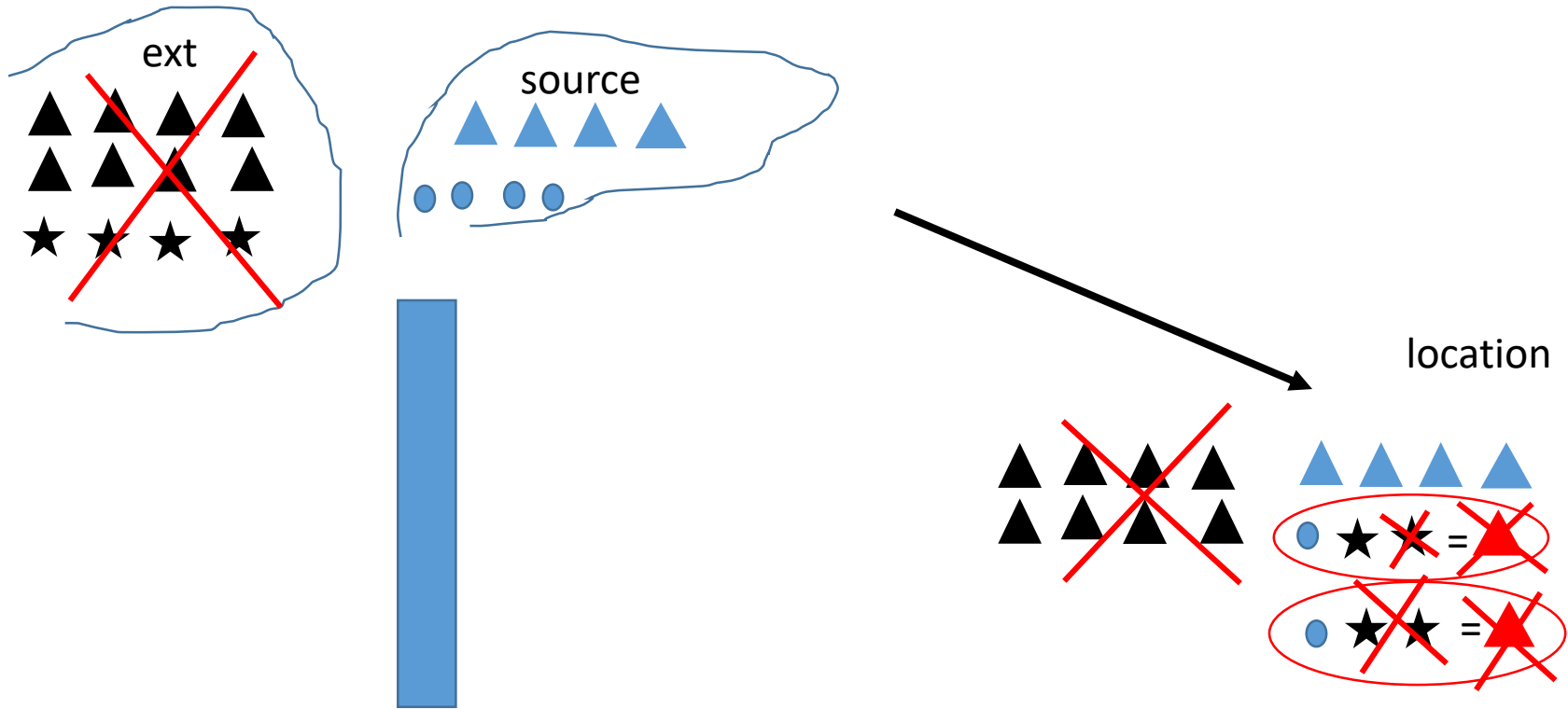


$$[extIM_{0.5}]_{\%} = \frac{\Delta C_{ext(0.5)}}{0.5C_R} \times 100\% = \frac{14-9}{0.5 \cdot 14} \times 100\% = \frac{5}{7} \times 100\% = 71\%$$

$$[sourceIM_{0.5}]_{\%} = \frac{\Delta C_{source(0.5)}}{0.5C_R} \times 100\% = \frac{14-12}{0.5 \cdot 14} \times 100\% = \frac{2}{7} \times 100\% = 29\%$$

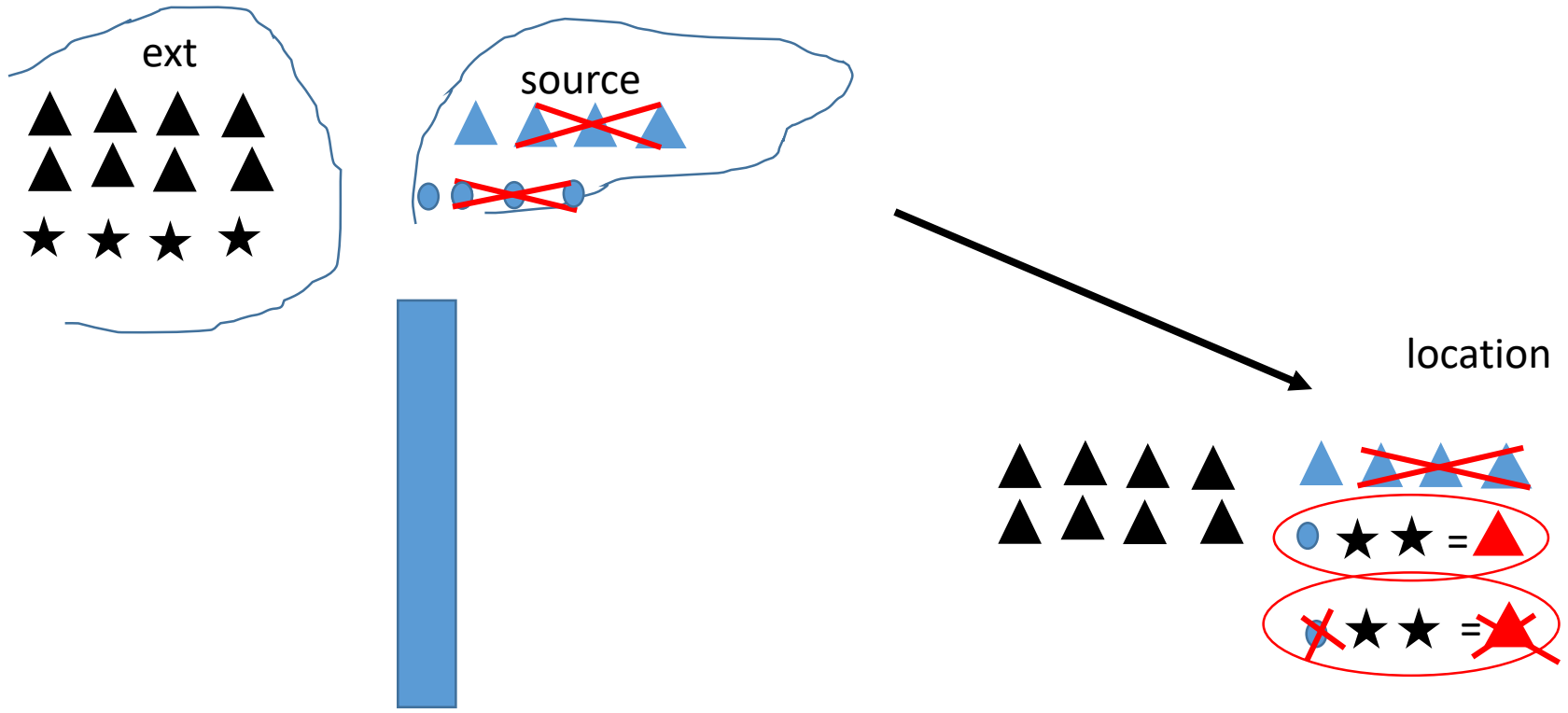
$$[extIM_{0.5}]_{\%} + [sourceIM_{0.5}]_{\%} = 100\% \rightarrow \hat{I}_{ext,source(0.5)} = 0$$

# 5. Example : $\alpha = 75\%$ reduction



$$[extIM_{0.75}]_{\%} = \frac{\Delta C_{ext(0.75)}}{0.75C_R} \times 100\% = \frac{14-6}{0.75 \times 14} \times 100\% = \frac{8}{10.5} \times 100\% = 76\%$$

# 5. Example : $\alpha = 75\%$ reduction

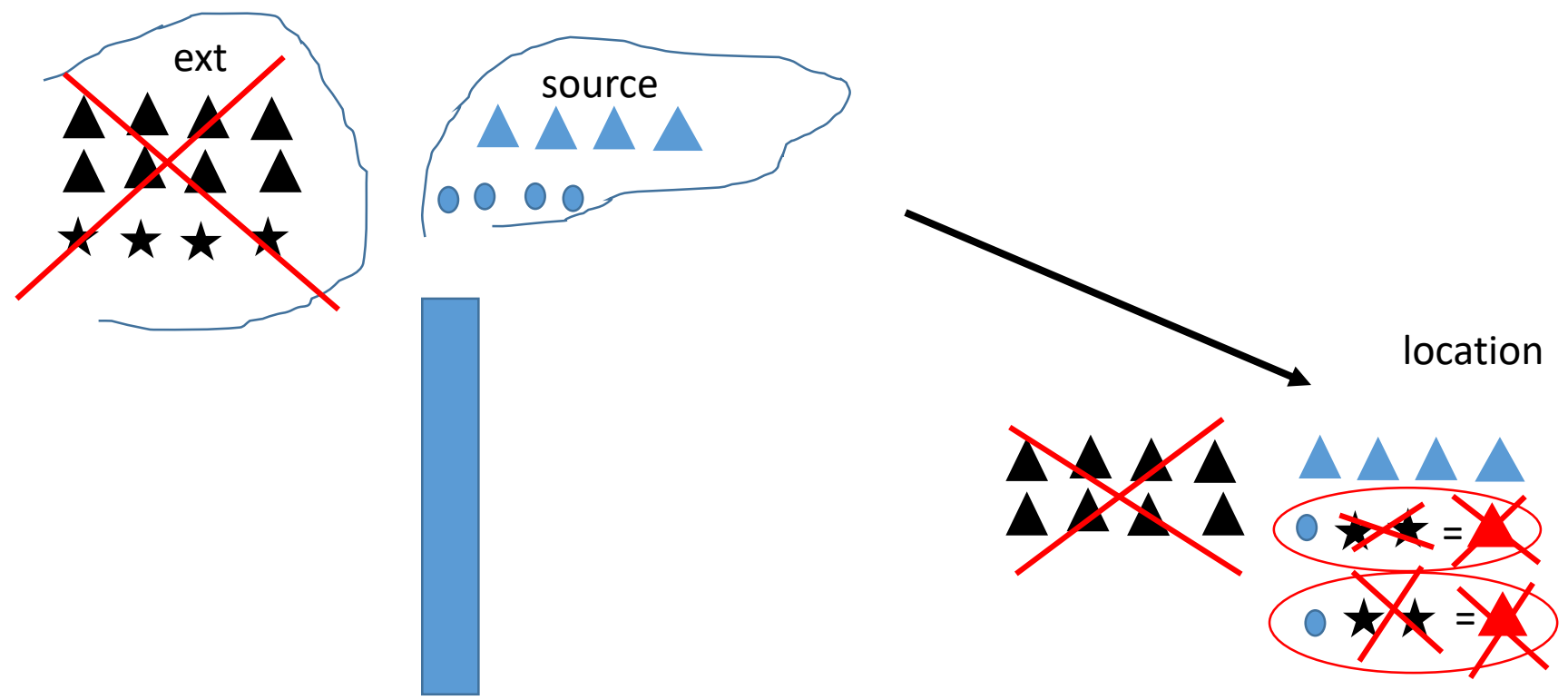


$$[extIM_{0.75}]_{\%} = \frac{\Delta C_{ext(0.75)}}{0.75C_R} \times 100\% = \frac{14-6}{0.75 \cdot 14} \times 100\% = \frac{8}{10.5} \times 100\% = 76\%$$

$$[sourceIM_{0.75}]_{\%} = \frac{\Delta C_{source(0.75)}}{0.75C_R} \times 100\% = \frac{14-10}{0.75 \cdot 14} \times 100\% = \frac{4}{10.5} \times 100\% = 38\%$$

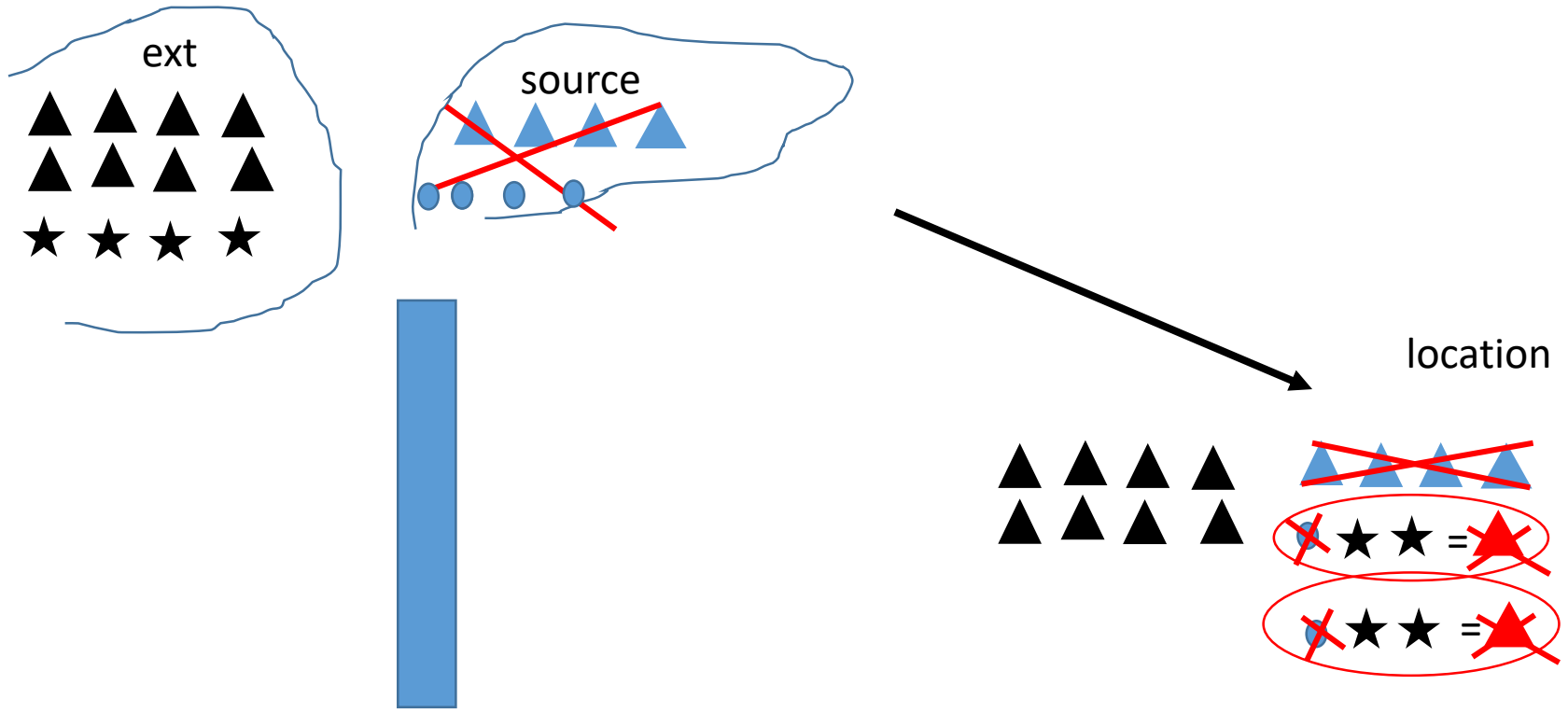
$$[extIM_{0.75}]_{\%} + [sourceIM_{0.75}]_{\%} = 114\% \rightarrow \hat{I}_{ext,source(0,75)} = 1.5$$

# 5. Example : $\alpha = 100\%$ reduction



$$[extIM_1]_{\%} = \frac{\Delta C_{ext(1)}}{C_R} \times 100\% = \frac{14-4}{14} \times 100\% = \frac{10}{14} \times 100\% = 71\%$$

# 5. Example : $\alpha = 100\%$ reduction



$$[extIM_1]_{\%} = \frac{\Delta C_{ext(1)}}{C_R} \times 100\% = \frac{14-4}{14} \times 100\% = \frac{10}{14} \times 100\% = 71\%$$

$$[sourceIM_1]_{\%} = \frac{\Delta C_{source(1)}}{C_R} \times 100\% = \frac{14-8}{14} \times 100\% = \frac{6}{14} \times 100\% = 43\%$$

$$[extIM_1]_{\%} + [sourceIM_1]_{\%} = 114\% \rightarrow \hat{I}_{ext,source(1)} = -2$$



## 5. Example : results

From the above example we have :

$$\begin{aligned}\hat{I}_{ext,source}(0.5) &= 0 \\ \hat{I}_{ext,source}(0.75) &= -1.5 \\ \hat{I}_{ext,source}(1) &= -2 = -I_{ext,source}\end{aligned}$$

Generally, one can obtained :

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

So that the highest absolute value of the non-linear interaction term from (Thunis et al. 2019) is equal to the interaction term defined in our work.

## 6. Conclusions

The comparison of two methods in calculating transboundary concentrations of  $\text{NO}_2$  and  $\text{PM}_{10}$  using CMAQ chemical transport model was presented.

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.

While for  $\text{NO}_2$  the annual mean interaction term was less than 3% of the total concentrations, for  $\text{PM}_{10}$  it can be as high as 16% of the total concentrations and around 25% of the calculated transboundary contribution.

It was shown that this interaction term expressed in hourly concentrations can be very large in some episodes.

It was proposed that the real transboundary pollution  $T_R$  could be estimated by de-biasing transboundary concentration computed by the model  $T_M$  as  $T_R = T_M + c_{rem} T_M$ , where the coefficient  $c_{rem} = (C_R - C_M) / C_M$  is calculated comparing the full model run  $C_M$  concentrations against  $C_R$  concentrations measured at the rural and suburban background stations.

The connection between interaction term introduced in our work and the non-linear interaction term discussed in Thunis et al. (2019) is presented.