#### Estimation of emission trends using data assimilation

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Assimilation of atmospheric trace gas observations is a useful tool in estimating surface emissions. To estimate absolute emission rates, it is necessary to have a model that accurately describes all physical processes between source and receptor, including transport, sinks, and chemistry. If some of these are only partly known, it is not possible to attribute observed concentrations completely to the known sources. In spite of this, it is however still possible to obtain information on emission trends, assuming that biases in the absolute estimates are invariant on the considered time scale.

This has been illustrated by experiment with the LOTOS-EUROS region air quality model simulating NO2 concentrations over Europe for 2005-2012. Observations of tropospheric NO2 columns from the OMI satellite instrument were assimilated with the simulations using an Ensemble Kalman Filter technique, assuming that the main model uncertainty is in the anthropogenic emissions of NOx. The assimilation system includes a screening mechanism that rejects observations from analysis if the value is too far of from the model forecast taking into account the observation, representation, and model uncertainty. Assessment of the fraction of rejected observations pointed to two specific regions where the model and observations significantly differ from each other: the Italian Po-valley and the costal areal of northern Spain, where persistent biases were found over a large area. Assimilation parameters such as temporal correlation in emission uncertainty and analysis localization length scales were estimated from observation-minus-forecasts statistics over the entire period, leading to a significant increment in the number of observations accepted for assimilation.

The assimilation provides an 8 year time series of correction fields for the NOx emissions, based on the observations and taking into account the uncertainties. The figure shows a map of the emission trend with respect a reference year, and illustrates consistent trends over large regions.



Relative change in emissions in 2009 with respect to 2005 as estimated from assimilation of NO2 tropospheric columns.







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### Data assimilation in LOTOS-EUROS: EnKF







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#### Assimilation of SO2 and SO4 – a case study









#### SO2 annual cycle over all assimilation stations



Need for specification of uncertainty in SO2/SO4 conversion rates !







#### **First conclusions**

- Data assimilation is feasible, but every application is different!
- Emission estimation is hampered by relatively large biases and other model shortcomings.
- To estimate emissions challenges are:
  - To disentangle the uncertainty due to the emission input from other model uncertainties
  - To combine different sources of data multi component
  - Model error specification is key It is not enough to put uncertainties to input parameters or boundary conditions
- Without this estimation of emission trends is most feasible
  - Example for NO<sub>2</sub>





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# Annual mean modelled and retrieved NO2 distribution

OMI DOMINO v2.0 product

Systematic bias of  $1.10^{15}$  molec/cm<sup>2</sup> Spatial correlation R<sup>2</sup> = 0.91







-# days









#### **Temporal correlation statistics from observation - simulation samples**



0.00 0.10 0.20 0.30 0.40 0.50 0.60 0.70 0.80 0.90 1.00 1-day temporal correlation of OMI meas











## Spatial correlation lenghts from observation - model samples



0.00 25.00 50.00 75.00 100.00 125.00 150.00 175.00 200.00 225.00 250.00 Spatial correlation length for OMI for period total







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M. Schaap Fossil Fuel pilot

### Which emissions are captured in OMI NO2 signal ?

MACC inventory NOx emissions 2005



- Power generation
- House holds
- Industrial combustion
- Production processes
- Extraction of fossil fuels
- Solvent use
- Road transport
- Off-road trnsport
- Waste incineration
- Agriculture

Label definition:

- 6 source sectors
- 5 hours of the day between 9 and 14
- Boundary conditions



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Concentration resulting from Dutch Road Transport at time 0











#### Which emissions are captured in OMI NO2 signal ?







## Modelled sector contributions to NO2 columns at OMI overpass in 2005 using a labelling technique

Power generation



Road transport

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Adjusted gridding of point sources by year in TNO-MACC-II inventory pre-2003 using EPER 2001; 2003-2005 with EPER 2004; 2006-2007 with E-PRTR 2007; after 2007 annual E-PRTR.









#### Conclusions

- Feasible to apply data assimilation to estimate long term trends
- Provides insight in dominant sources (given good quality a-priory data)
- Target: monitor energy transition impacts
- Based on these analysis feedback given to MACC inventory





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### Thank you for you attention



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