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

Model uncertainty specified for (depending on application):
- emission rates
- sinks
- boundary conditions
- reaction rates
- ...

$t = t_0$

Forecast state

Forecast

Analysis

Analysed state

Model integration

Observations

Assimilation procedure

Forecast state ($x_f$)

Analysed state ($x_a$)
Assimilation of SO2 and SO4 – a case study

Annual mean for 2003 (Barbu et al, 2009)
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 NO₂
Annual mean modelled and retrieved NO2 distribution

OMI DOMINO v2.0 product

Systematic bias of $1 \times 10^{15}$ molec/cm$^2$
Spatial correlation $R^2 = 0.91$
Temporal correlation statistics
from observation - simulation samples
Spatial correlation lengths from observation - model samples
North Spain

2007-2008 implementation of NOx reduction measures in power plants

amount of observations

assimilated

screened
change in NOx emissions (%) from 2005 to 2009 according to assimilation results
Which emissions are captured in OMI NO2 signal?

MACC inventory
NOx emissions 2005

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

Concentration resulting from Dutch Road Transport at time 0
Which emissions are captured in OMI NO2 signal?

- Power plants
- Industrial combustion
- Road transport
- Other transport
- Agriculture
- Other

Emitted at:
- 2008-10
- 2010-11
- 2011-12
- 2012-13
- 2013-14
- Others
Modelled sector contributions to NO2 columns at OMI overpass in 2005 using a labelling technique

Power generation

Road transport

E-PRTR = European Pollutant Release and Transfer Register
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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