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GLOBEMISSION

Variational aerosol emission inversion in regional scale using MODIS observations

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Outline

- **Introduction**
  - The SILAM variational inversion and assimilation system
- **Emission inversion for particulate matter using MODIS observations**
  - Setup
  - Initial results
    - Emission adjustments
    - Evaluation: Airbase, Aeronet
    - Further output:
      - Emission adjustments for SO2 and wildfire PM
- **Conclusions**

The SILAM variational assimilation and inversion system

- **The SILAM eulerian regional to global chemistry transport model**
  - CB4 photochemistry, SOx and inorganic aerosols after DMAT (Sofiev, 2000)
- **4D-Var assimilation:**
  - adjoint model with continuous adjoint code for advection, discrete for all other processes
  - flexible control variables: initial condition, multiplicative or additive emission adjustments
  - minimization with the L-BFGS-B method, positivity constraint for emission rates
  - spatial covariance model shared with the 3D-Var code
- **Applied to short-term forecasting of SO2 in Vira & Sofiev (2012)**
- **Adapted for volcanic source term inversion, including vertical profiles (poster: Vira et al., 2013)**
- **Adjoint code for gas-phase chemistry implemented in 2013, using the Kinetic PreProcessor (KPP)**

PM emission inversion for year 2008 using MODIS observations

- **Gridded emission estimates, monthly averaging**
  - Europe, 0.5 degree resolution
- **Assimilation of the MODIS 550 nm AOD product (Aqua + Terra)**
- **Mass extinction coefficients required by observation operator**
  - evaluated on the fly to match the model aerosol configuration
- **In the following, we aim to assess**
  - technical feasibility of variational emission inversion in yearly and regional scales
  - sensitivity to errors and the need for regularization

The emission inversion method

- **Based on 4D-Var, quadratic cost function**
  - control vector: gridded emission estimates as multiplicative factors to the a priori inventory
  - emission adjustments kept constant over assimilation window
  - 72 h assimilation window – needed to cover species lifetime
- **Simplified aerosol chemistry (only 1st order processes)**
  - only sulphur chemistry in the inversion: NH4, NO3 simulated in a separate run and subtracted from the observed values

The emission inversion, v1.0: setup

- **The inverse emission estimates generated for year 2008 in European domain**
  - 0.5° horizontal resolution
  - DMAT chemical scheme
- **A priori PM emission sources:**
  - Anthropogenic – TNO/MACC 2007
  - Wildfires – Sofiev et al. (2009)
  - Sea salt – Sofiev et al. (2012)
  - dust – currently not included
- **Emission sector dependent a priori uncertainty:**
  - up to 100% for PM2.5, PM10 emission
  - up to ~30% for SO2 emission
  - 100% for fires, 50% for sea salt

Attributing the PM observations to emissions

- **The contribution of various PM components to the AOD not unique**
- **The max-likelihood solution determined dynamically based on the sensitivities of**
  - the observation operator:
    - mass extinction coefficient depending on particle size and composition
  - the model:
    - spatial segregation of emitted components
    - the relative contribution of each component at a given location
    - the a priori emission uncertainty
- **Highest sensitivity to concentrated emissions sources of fine particles!**

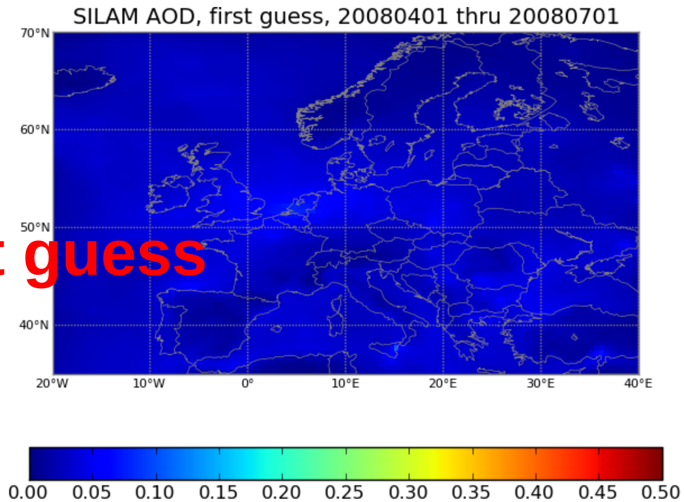
Inversion experiments: preliminary results

- **Model initially underestimates AOD, emission adjustments typically positive**
- **long assimilation window => ill-posed inverse problem**
  - daily results require averaging down to eg. monthly
- **Adjustments > 2 for anthropogenic PM2.5 emission**
- **Only minor adjustments for coarse PM (small contribution to AOD) and sea salt (widespread and varying source area)**
- **Strong adjustments for wildfire PM and SO2…**
- **Significant contribution by dust for some months**

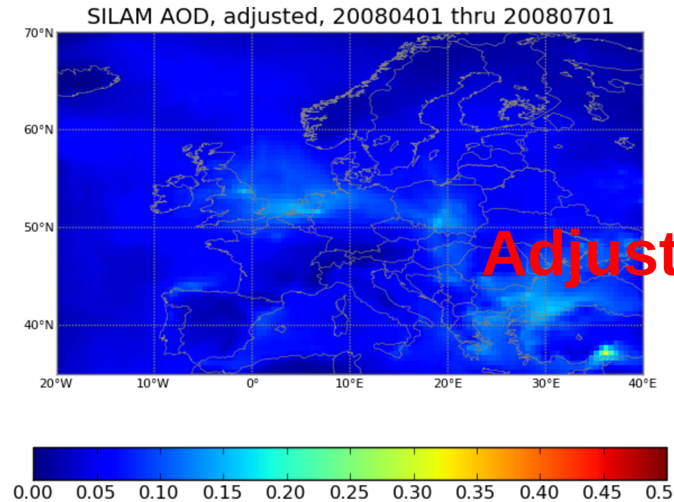
Aerosol optical depth and PM2.5 scaling

2008
Apr-Jun

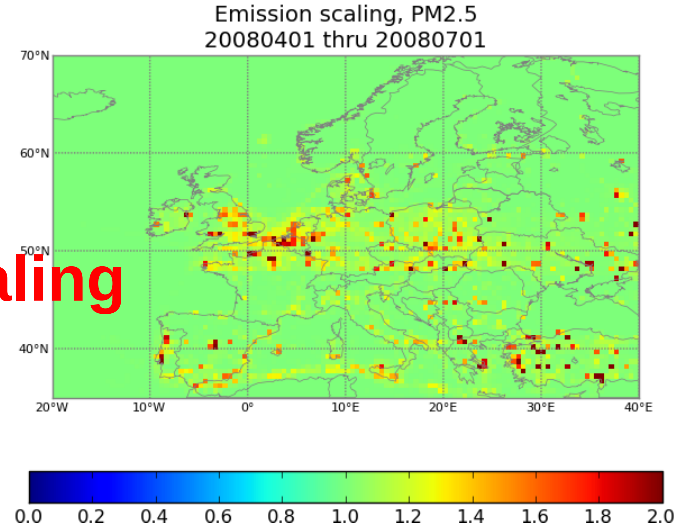
First guess



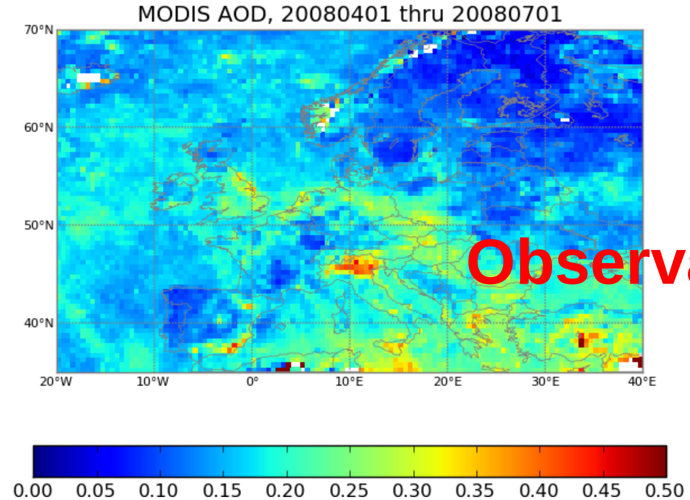
Adjusted



Scaling



Observation



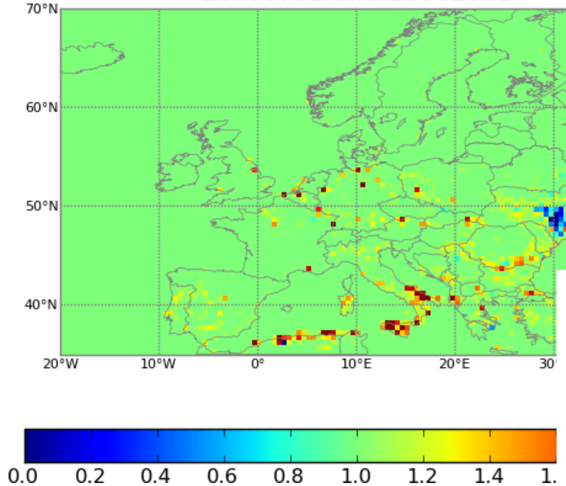
Adjustments to SO2 and wildfire emissions

- **Main focus has been on primary anthropogenic PM emissions**
- **The emission inversion also includes**
  - SO2 emissions via sulphate formation
  - Wildfire PM emissions
- **AOD observations have high sensitivity to SO2 emissions**
  - adjustments as strong or stronger than PM2.5

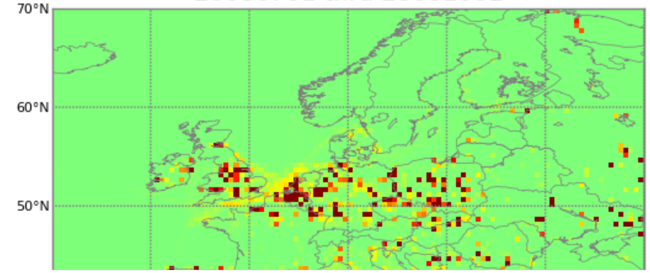
Fire PM and SO2 scaling, July-December 2008

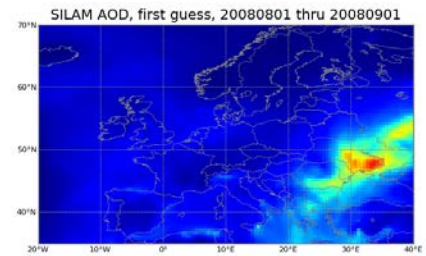
Jul
Aug
Sep

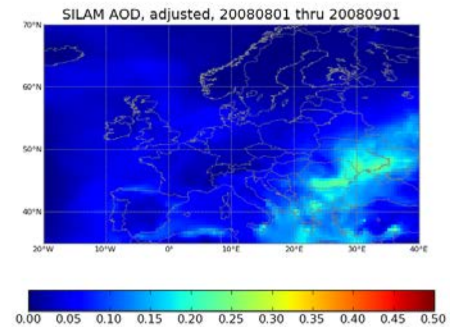
Emission scaling, Fire PM2.5
20080701 thru 20081001



Emission scaling, SO2
20080701 thru 20081001

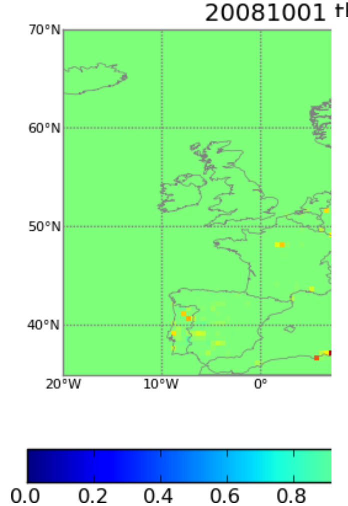


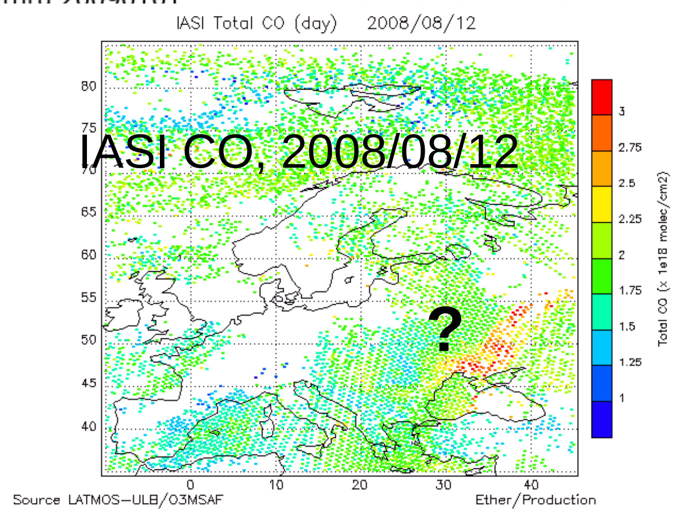


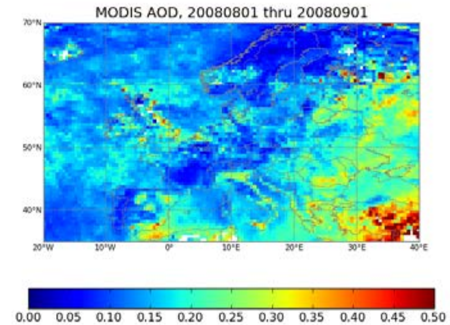


Oct
Nov
Dec

Emission scaling, Fire PM2.5
20081001 thru 20090101

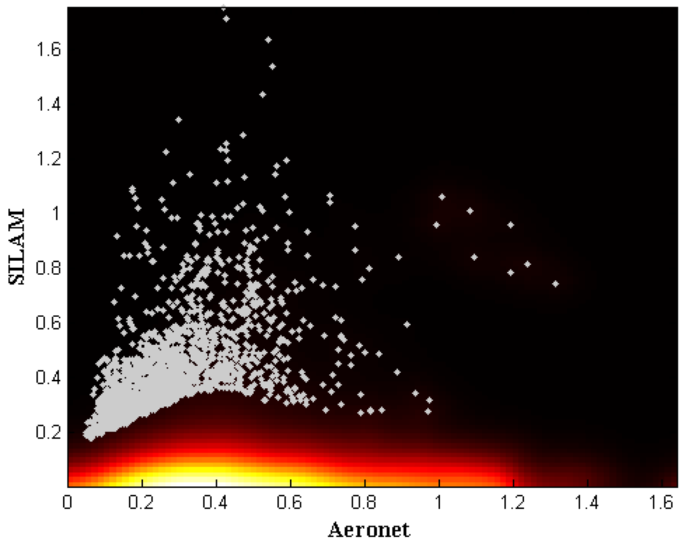


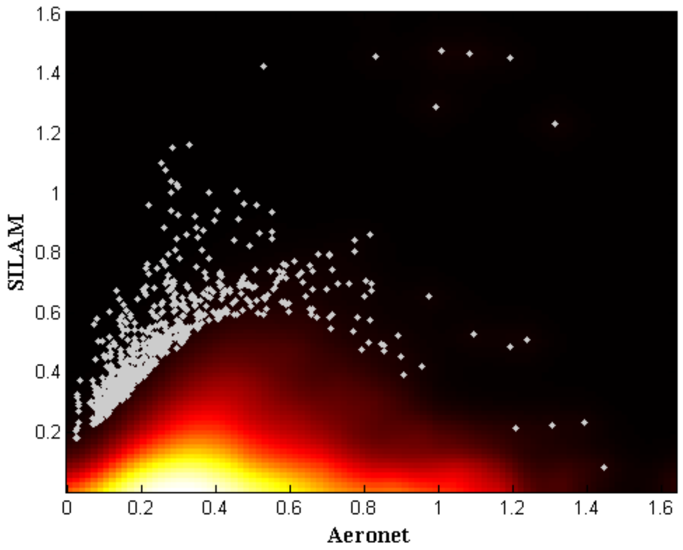




A posteriori validation: comparison with Aeronet

- **Independent datasets needed for evaluation of results**
- **Top right: model vs observed AOD, a priori**
- **Bottom right: model vs observed AOD, a posteriori**
- **Improved agreement due to assimilation, but highest AODs remain strongly underestimated**





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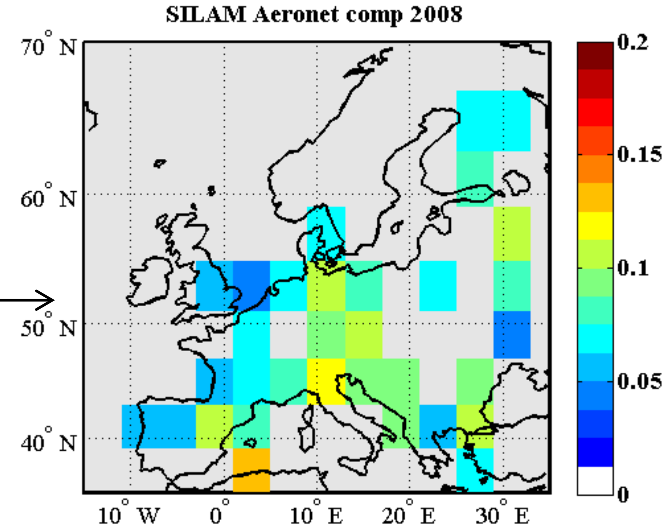
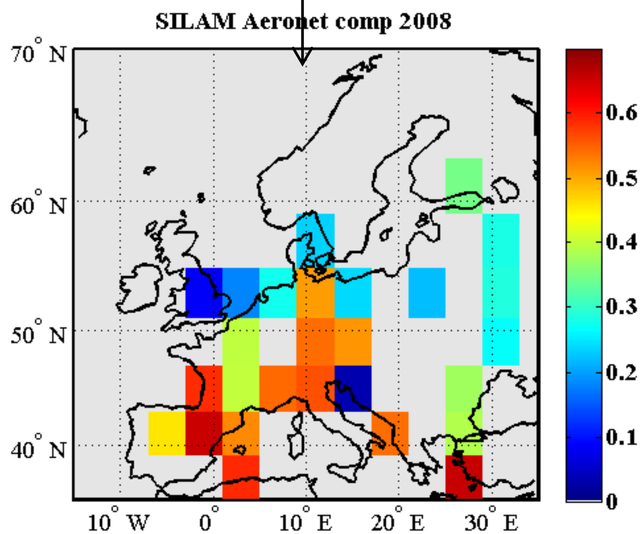
Aeronet: spatial pattern of model bias (obs – model)

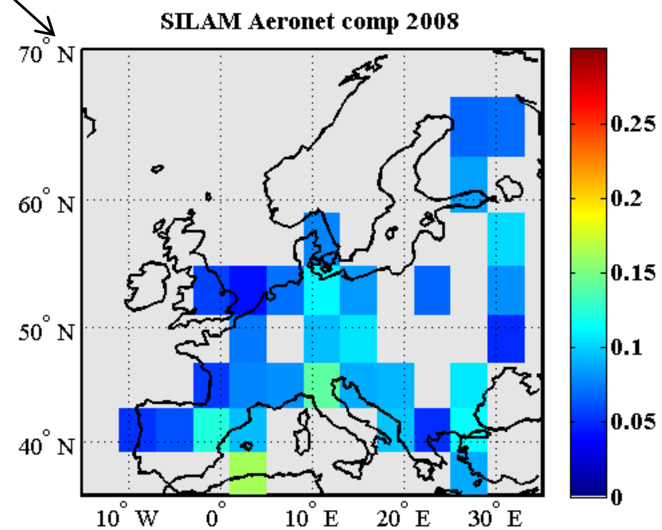
All observations



Observed AOD < 0.5

Observed AOD > 0.5





Comparison with in-situ measurements

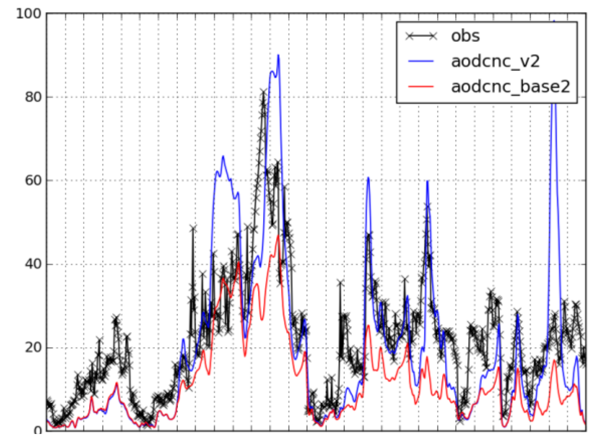
- **Statistics for PM2.5, rural background stations only**

	RMSE	Bias	Corr
Adjusted	12.14	-1.82	0.41
Original	11.01	-3.40	0.40

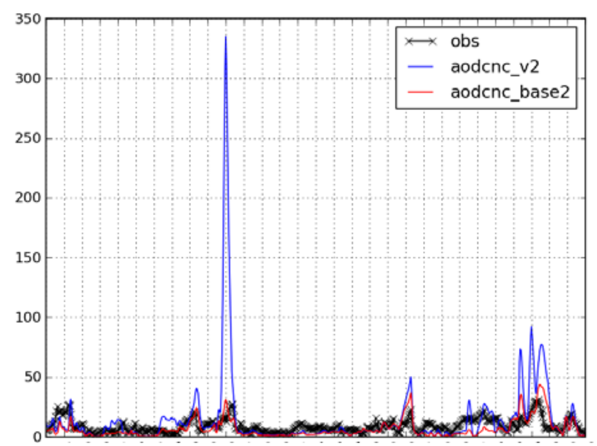
- **The run with adjusted emission suffers from short but strong spurious peaks**
  - Inversion is error-amplifying – need stronger/smarter regularization to suppress model/observation errors?
  - Need better observation error characterization or quality control?

PM2.5 concentration, µg/m3
observed a priori adjusted

DEBB007
Feb 2008



DEBB0063
Sept 2008



Conclusions and outlook

- **Variational emission inversion is feasible on timescales up to years**
- **Challenges inversion of PM emissions:**
  - Large first-guess model bias
- **Problematic assimilation window:**
  - PM lifetime favors longer windows (> days)
  - but inversion becomes ill-posed due to increasing footprint of each observation
  - temporal aggregation of the emission adjustment could help
    - but strong, intermittent emission sources require sufficient temporal resolution
- **AOD observations sensitive to SO_2 emissions**
  - large a posteriori emission increments unless constrained directly
- **Future work: more focused inverse studies**
  - dust
  - fire episodes

References

- **Sofiev, M., A model for the evaluation of long-term airborne pollution transport at regional and continental scales, Atmos. Environ., 34(15), 2481–2493, 2000**
- **Vira, J., Sofiev, M., 2012. On variational data assimilation for estimating the model initial conditions and emission fluxes for short-term forecasting of SOx concentrations. Atmos. Environ. 46, 318–328.**
- **Vira, J., Hakkarainen, J., Sofiev, M., 2013. Variational inversion of SO2 emission flux in the 2011 Grimsvötn eruption. Poster presentation, ESA/EUMETSAT volcanic ash and aviation User Workshop, Dublin, 4-6 March 2013. http://vast.nilu.no/media/cms_page_media/15/3A-15_vira_1.pdf**