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GLOBEMISSION

Regional-scale particulate matter emission estimates using satellite observations and variational inversion

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Outline

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    - Emission adjustments for SO2 and wildfire PM
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Introduction

- **The problem: estimate anthropogenic PM emissions a with inverse modelling**
  - emission inventory adjusted to provide optimal agreement between satellite observations and the SILAM chemistry transport model
- **Gridded emission estimates, monthly averaging**
  - Europe, 0.5 degree resolution
- **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 inversion method

- **Emission inversion: combine statistically a priori emissions (inventory or model) with observations (direct or indirect)**
- **Observations y defined as operators acting on the state variable x: y = Hx**
- **The variational method:**
  - split the computed period into assimilation windows
  - find the emission minimizing a quadratic cost function measuring the deviation from a priori and observed values over the assimilation window
- **The adjoint observation operator propagates observed discrepancy y-Hx to the model state variable (concentration)**
- **The adjoint model propagates the state variable to emission forcing**
  - allows adjusting primary species using the observations of secondary species

Satellite data and observation operators

- **AOD product from MODIS (Aqua + Terra)**
- **Mass extinction coefficients required by observation operator**
  - evaluated on the fly to match the model aerosol configuration
- **L2 data, no further aggregation/averaging**

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!**

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The emission inversion, v1.0: setup

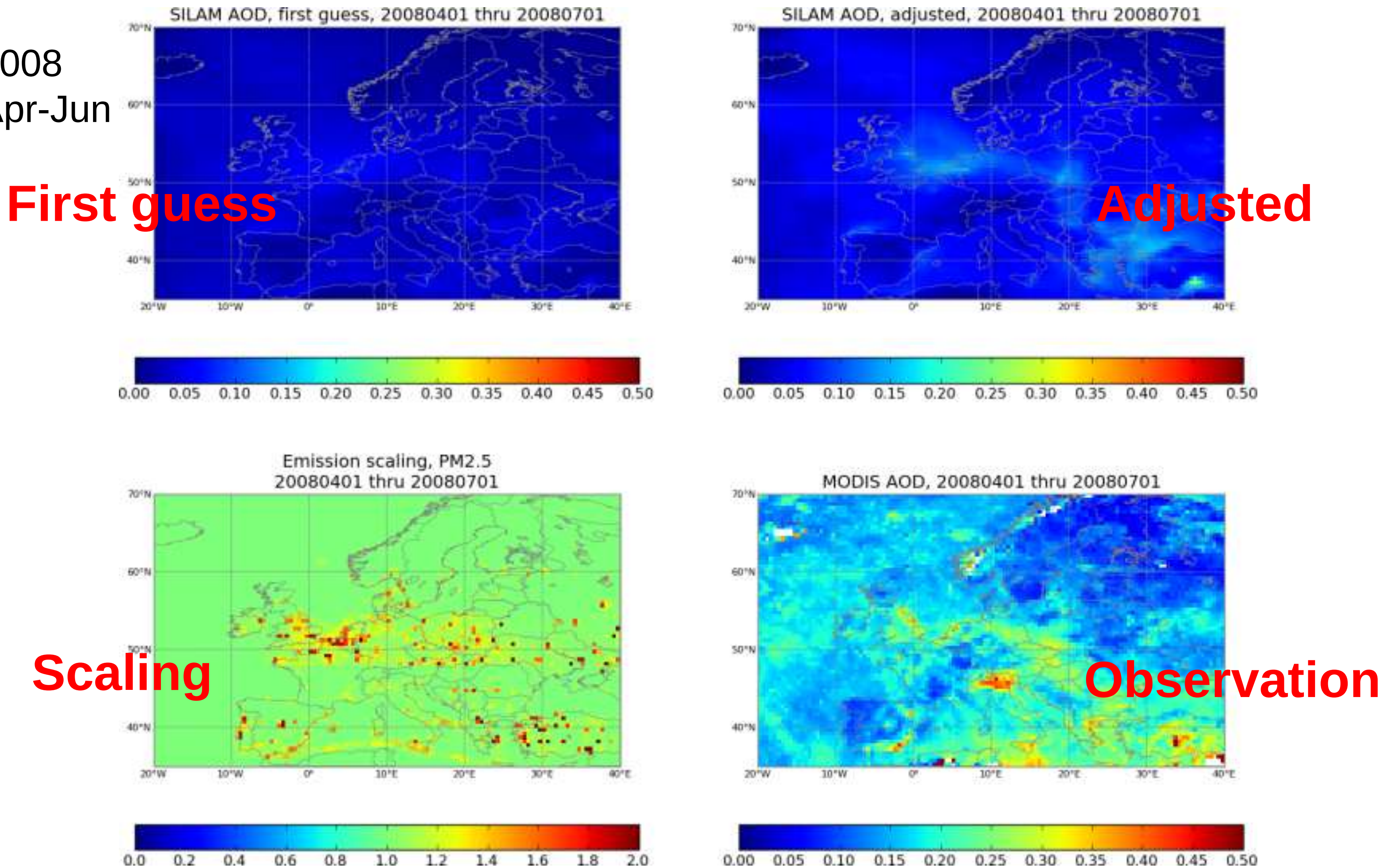
- **The inverse emission estimates generated for year 2008 in European domain**
  - 0.5° horizontal resolution
  - DMAT chemical scheme
    - only sulphur chemistry in the inversion, but other SIA estimated separately and subtracted from the observed values
- **A priori PM emission sources:**
  - Anthropogenic – TNO/MACC 2007
  - Wildfires – Sofiev et al. (2009)
  - Sea salt – Sofiev et al. (2012)
  - dust – currently not included
- **72 h assimilation window – dictated by PM atmospheric lifetime**
- **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

Inversion experiments: preliminary results

- **Model initially underestimates AOD, emission adjustments typically positive**
- **long assimilation window, ill-posed inverse problem**
  - daily results require averaging down 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



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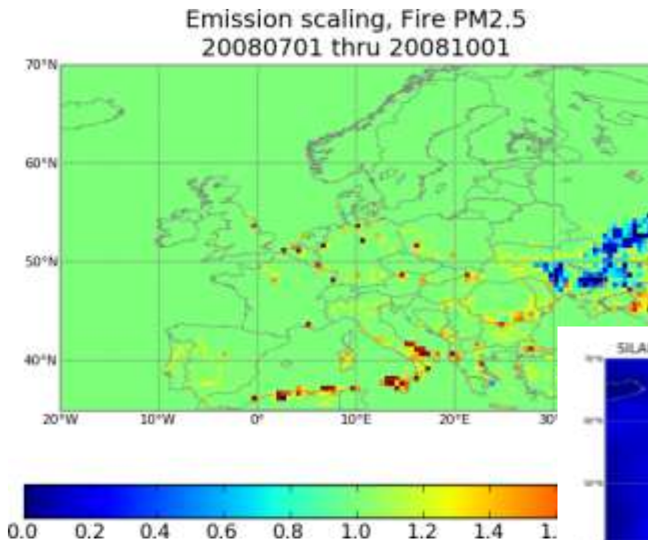
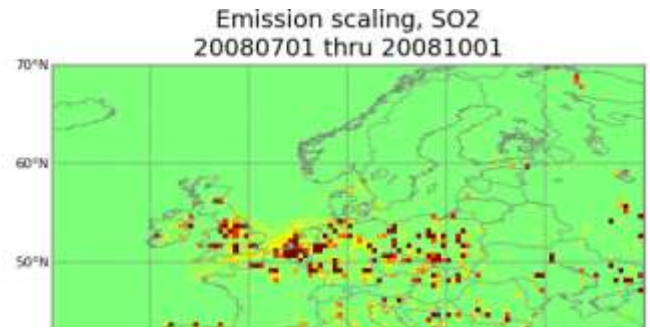
Adjustments to SO2 and wildfire emissions

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

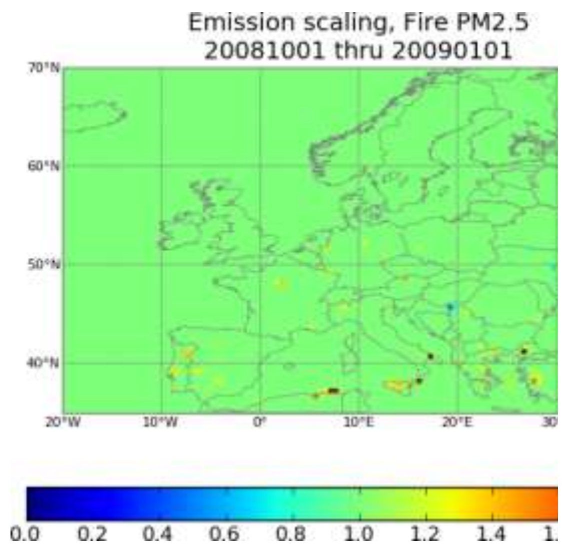
Fire PM and SO2 scaling, July-December 2008

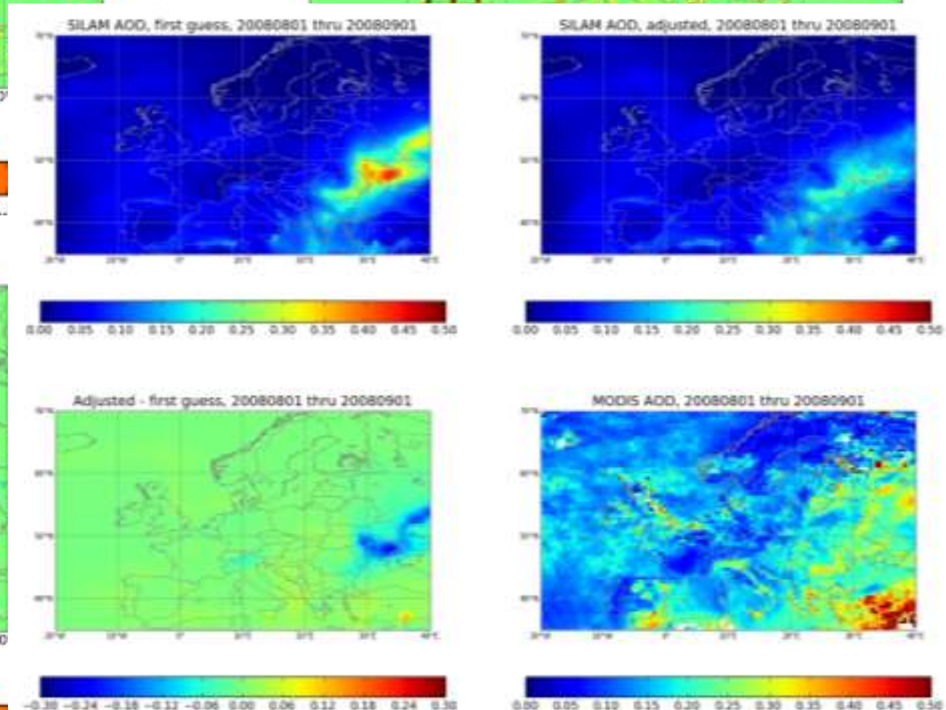
Jul
Aug
Sep



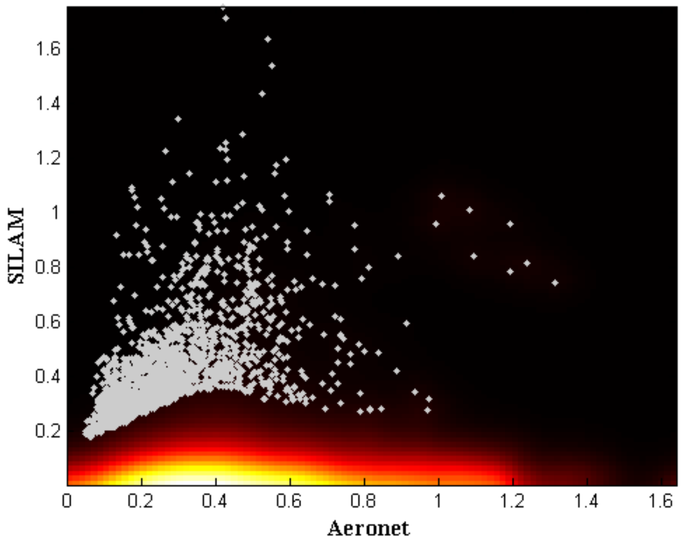


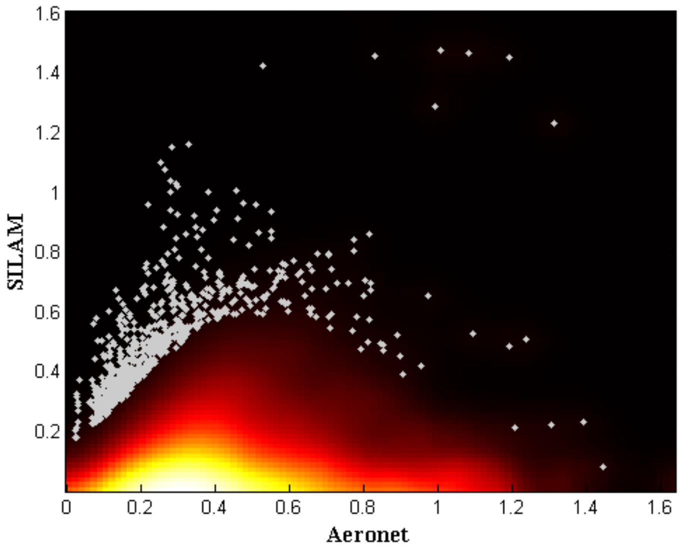
Oct
Nov
Dec





A posteriori validation: comparison with Aeronet

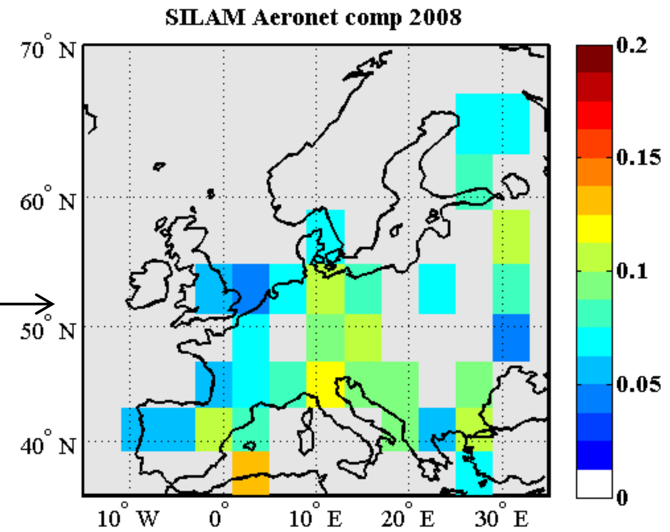




- **Independent datasets needed for evaluation of results**
- **Top right: model vs observed AOD, a priori**
- **Bottom right: model vs observed AOD, a posteriori**
- **Improved agreement due to assimilation, but highest AODs remain strongly underestimated**

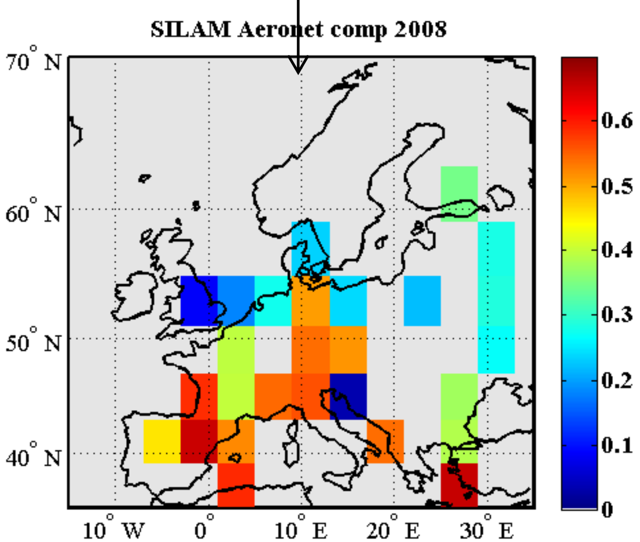
Aeronet: spatial pattern of model bias (obs – model)

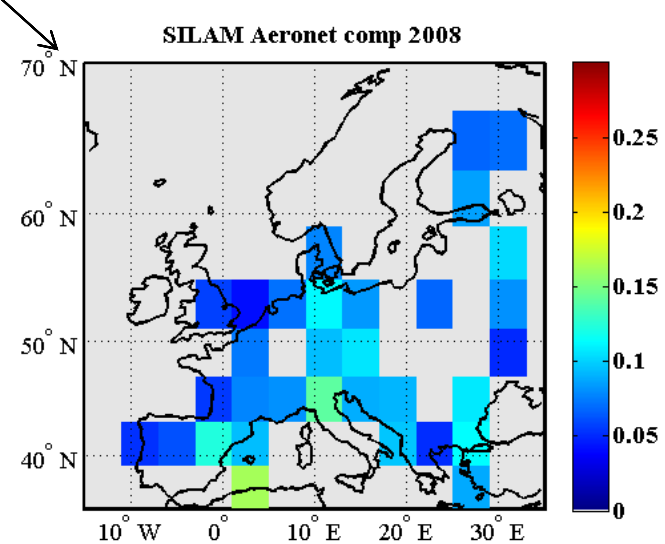
All observations



Observed AOD < 0.5

Observed AOD > 0.5





Comparison with in-situ measurements

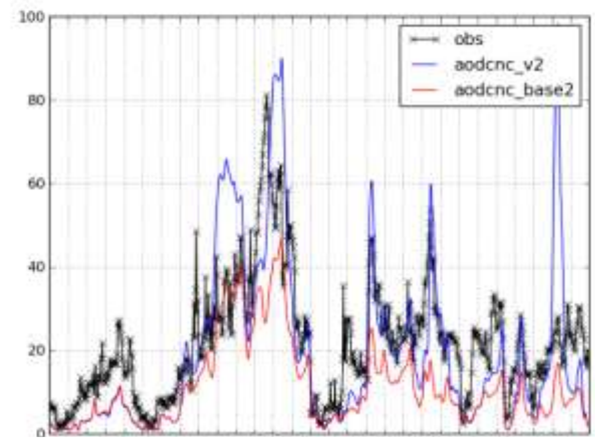
- **Statistics for PM2.5, rural background stations only**

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

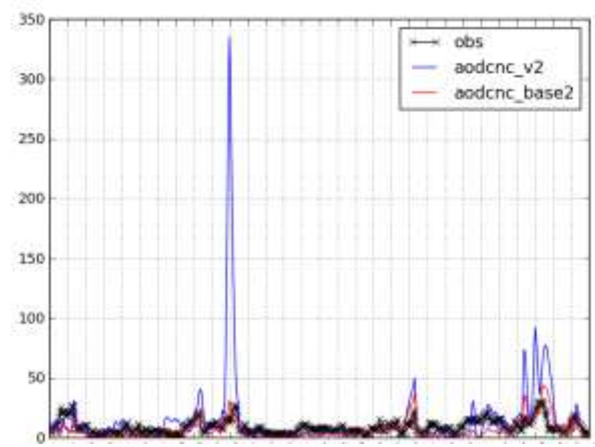
- **The run with adjusted emission suffers from short but strong spurious peaks**
  - Inversion is error-amplifying – need stronger/smarter regularization to suppress model/observation errors?
  - Need better observation error characterization or quality control?

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

DEBB007
Feb 2008



DEBB0063
Sept 2008



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Conclusions and outlook

- **Variational emission inversion is feasible on timescales up to years**
- **Satellite AOD observations sensitive to also SO_2 emissions**
- **Challenges in inversion:**
  - Ill-posed inverse problem due to lifetime of fine PM
  - A priori covariance matrix poorly specified
  - Large first-guess model bias
- **Emission inversion needs model-independent evaluation**
  - consider temporal/spatial trends in inverse estimates against the AIS shipping emissions
- **Outlook for future work**
  - Revise the inversion algorithm and model setup:
    - a priori covariance modeling for emission fluxes
  - Include dust emissions in the model