Bayesian regression and its application to atmospheric emissions estimation

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Presentation Overview

1. introduction
   - atmospheric emissions estimation: what is our goal and what data we have?
   - atmospheric linear inverse problem formulation
2. theory: modeling and estimation methods for atmospheric inversion
   - standard and Bayesian approaches to linear inversion
   - example on ETEX-I test release
3. applications to atmospheric emissions estimation
   - multi-species emissions
   - plume bias correction: towards non-linear regression
   - spatial-temporal emissions estimation:
     - Cs-137 emissions from Chernobyl wildfires
     - atmospheric microplastics
     - towards satellite data inversion: ammonia case

Introduction: linear inverse problem formulation

Our goal:

- to estimate the time-profile of atmospheric emissions, known also as the source term

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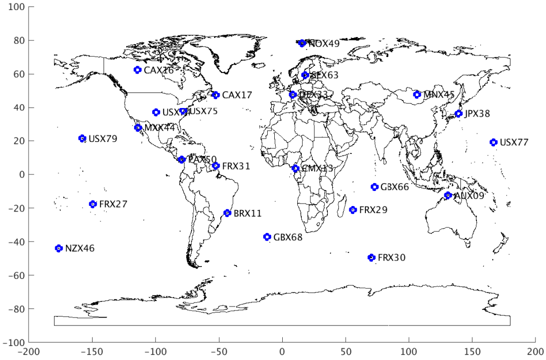
- to estimate the time-profile of atmospheric emissions, known also as the source term,

What data we have:

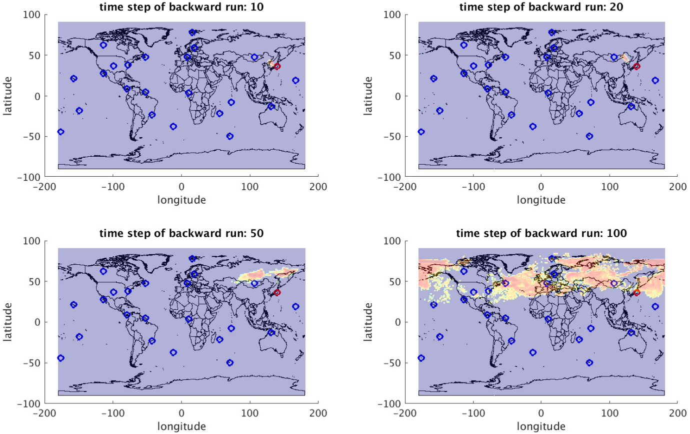
- concentration/deposition measurements
- e.g. Xe133 concentrations from CTBTO stations (map for the year 2014):



Introduction: linear inverse problem formulation

What data we have:

- atmospheric transport model driven by meteorological reanalysis
- e.g. FLEXPART backward runs for each Xe133 observation from the CTBTO network:



Introduction: linear inverse problem formulation

- Suppose that the emissions (source term) are stored in the vector $\boldsymbol{x}$ representing each time-step of temporal discretization

$$\boldsymbol{x} = [x_1, x_2, x_3, \ldots, x_n] \tag{1}$$

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- Assumption: each observation can be explained as a linear combination of simulated coefficients $m_{i,j}$ (called source-receptor-sensitivity (SRS) coefficients) and $\boldsymbol{x}$ as

$$y_i = m_{i,1}x_1 + m_{i,2}x_2 + \cdots + m_{i,n}x_n, \tag{2}$$

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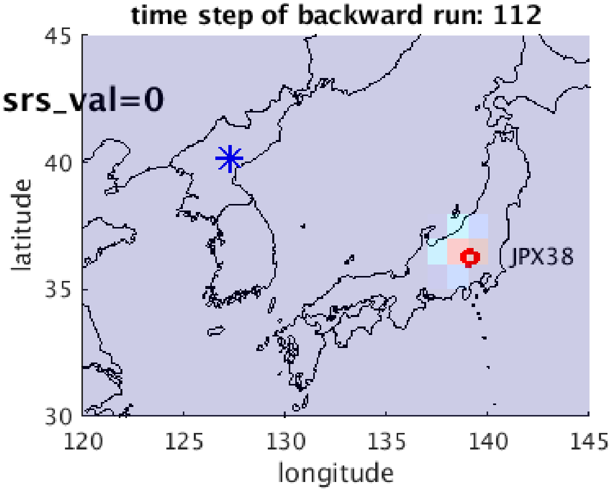
- using atmospheric transport model, we can calculate SRSs coefficients saying:
  - when unit release happen in given location and time-period (sensor, observation y_i), what would be observed on location of interest (emission location) if we go backward in time (SRSs $m_{i,j}$)

Introduction: linear inverse problem formulation

$$y_i = \cdots + m_{i,103}\, x_{103} + m_{i,104}\, x_{104} + m_{i,105}\, x_{105} + \cdots + m_{i,111}\, x_{111} + \underbrace{m_{i,112}}_{0} x_{112},$$



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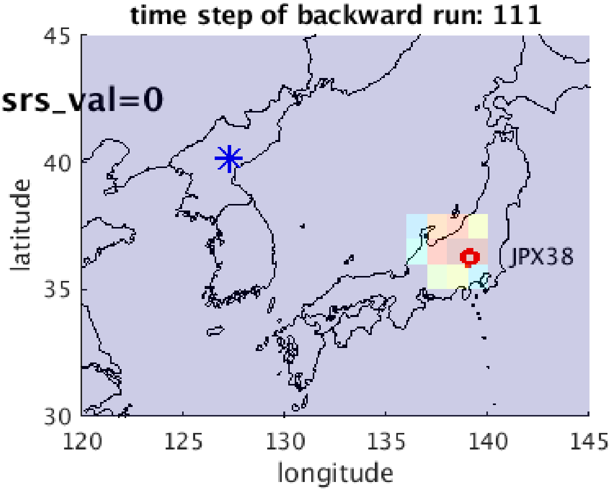

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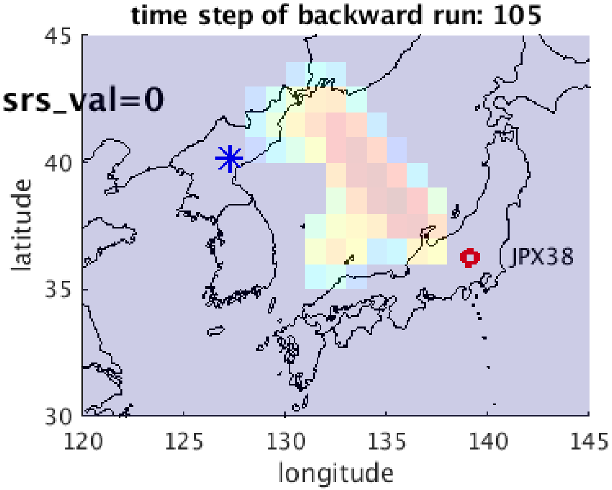

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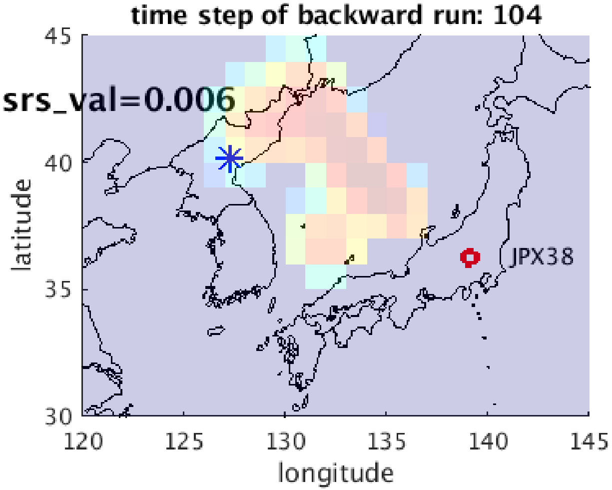

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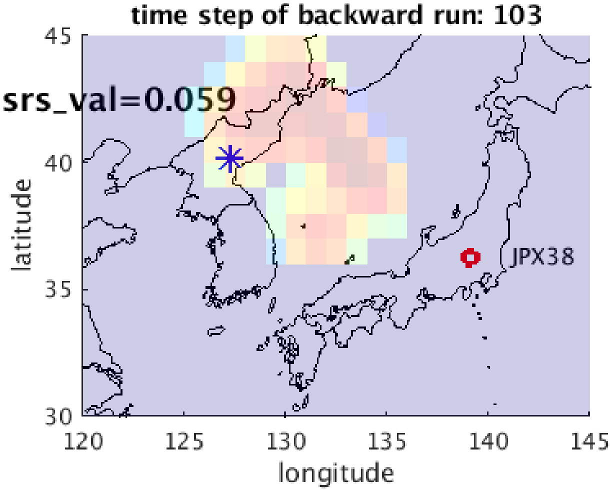

Introduction: linear inverse problem formulation

- we can compare
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  - **predicted** measurements: modeled sensitivities multiplied by source term (common for all) values

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- the comparison in compressed form:

$$
\boldsymbol{y} = \begin{bmatrix} y_1 \\ y_2 \\ \vdots \\ y_p \end{bmatrix} = \begin{bmatrix} m_{11} & m_{12} & \cdots & m_{1n} \\ m_{21} & m_{22} & \cdots & m_{2n} \\ \vdots & \vdots & \ddots & \vdots \\ m_{p1} & m_{p2} & \cdots & m_{pn} \end{bmatrix} \begin{bmatrix} x_1 \\ \vdots \\ x_n \end{bmatrix} = \boldsymbol{Mx},
$$

where the source term vector $\boldsymbol{x}$ is **unknown** and need to be estimated.

Introduction: linear inverse problem formulation

- So far, any questions or comments?

Theory: modeling and estimation

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- least-squares: $\boldsymbol{x}_{\mathrm{LS}} = \left(\boldsymbol{M}^T\boldsymbol{M}\right)^{-1}\boldsymbol{M}^T\boldsymbol{y}$
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  - need for selection of tuning parameters, e.g. α, $\boldsymbol{\theta}$, $\boldsymbol{R}$
  - need for choice of $g(\boldsymbol{x}, \boldsymbol{\theta}) \longrightarrow$ e.g. ridge regression, LASSO, etc.

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- Bayesian modeling
  - more demanding due to model development and parameters estimation
  - we can estimate the shape of $g(\boldsymbol{x}, \boldsymbol{\theta})$ and tuning parameters

Theory: modeling and estimation - Bayesian approach

- Bayes rule (with $\boldsymbol{\theta}$ being the set of all model parameters and $\boldsymbol{y}$ being available data)

$$p(\boldsymbol{x}, \boldsymbol{\theta} | \boldsymbol{y}) \propto p(\boldsymbol{y} | \boldsymbol{x}, \boldsymbol{\theta}) p(\boldsymbol{x}, \boldsymbol{\theta}), \quad (4)$$

  where
  - $p(\boldsymbol{x}, \boldsymbol{\theta} | \boldsymbol{y})$ is the posterior distribution
  - $p(\boldsymbol{y} | \boldsymbol{x}, \boldsymbol{\theta})$ is the data likelihood function (model)
  - $p(\boldsymbol{x}, \boldsymbol{\theta})$ is the prior distribution of model parameters

Theory: modeling and estimation - Bayesian approach

Optimization formulation (with Tikhonov term, $g(\boldsymbol{x}) = \boldsymbol{x}^T \boldsymbol{B}^{-1} \boldsymbol{x}$):

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- its Bayesian counterpart is the prior model:

$$p(\boldsymbol{y}|\boldsymbol{x}, \boldsymbol{R}) = N_{\boldsymbol{y}}(\boldsymbol{M}\boldsymbol{x}, \boldsymbol{R}) \propto \left|\boldsymbol{R}^{-1}\right| \exp\left(-\frac{1}{2}(\boldsymbol{y} - \boldsymbol{M}\boldsymbol{x})^T \boldsymbol{R}^{-1} (\boldsymbol{y} - \boldsymbol{M}\boldsymbol{x})\right)$$

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- we can further select prior models for $\boldsymbol{R}$ and $\boldsymbol{B}$ and estimate them
- analytical solution typically not available –> sampling methods, expectation-maximization-like iterative methods such as Variational Bayes method

Theory: modeling and estimation - Bayesian approach

- Variational Bayes method gives us the form of posterior as

$$\tilde{p}(\theta_i|\boldsymbol{y}) \propto \exp\left(\mathrm{E}_{\tilde{p}(\boldsymbol{\theta}_{/i}|\boldsymbol{y})}\left(\ln p(\boldsymbol{\theta},\boldsymbol{y})\right)\right), \quad i = 1,\ldots,q, \qquad (6)$$

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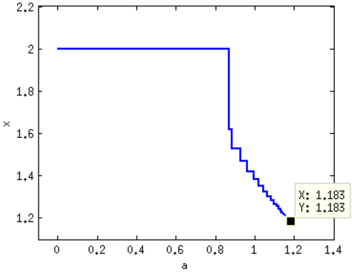
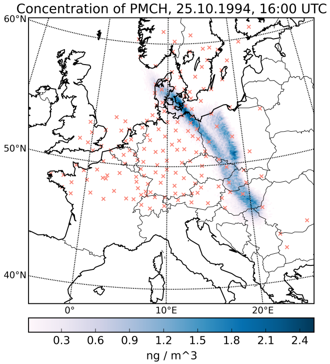
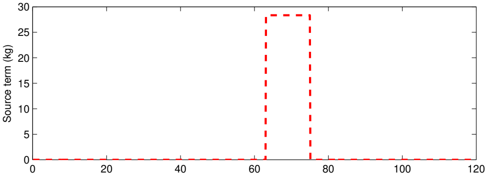
- see simple scalar example for model $d = ax + e$



Theory: modeling and estimation - ETEX example

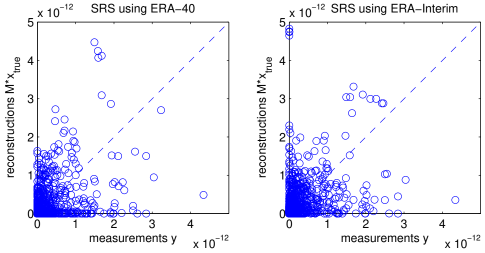
- ETEX-I (European tracer experiment) in 1994.
- 340 kg of inert perfluoromethylcyclohexane (PMCH) was released during 12 hours in Brittany, France.
- 168 stations over the Europe.





Theory: modeling and estimation - ETEX example

- example reconstruction with the **true** release and two different meteorological reanalysis



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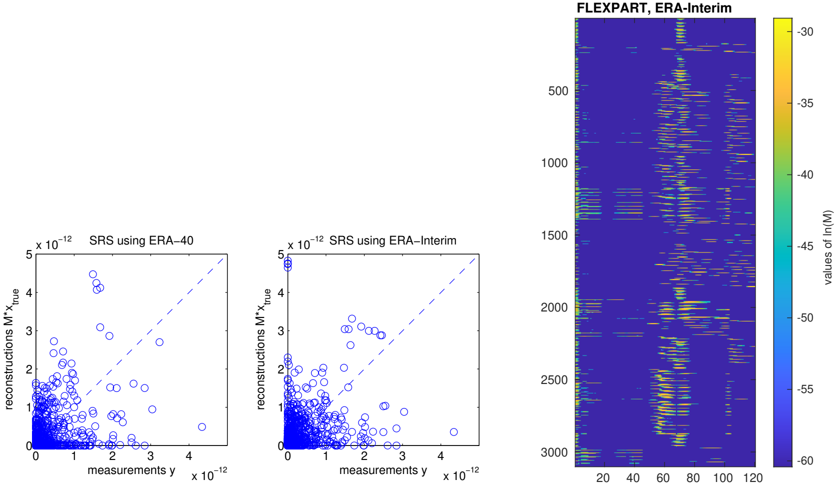
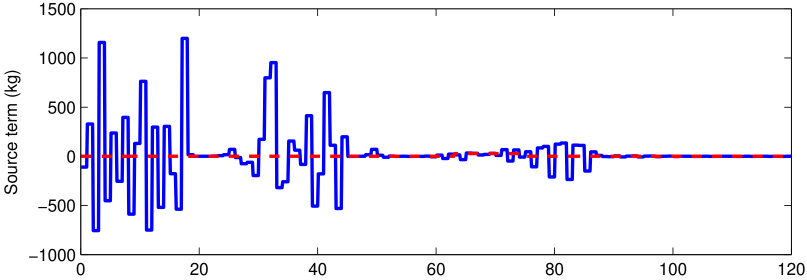


Theory: modeling and estimation - ETEX example

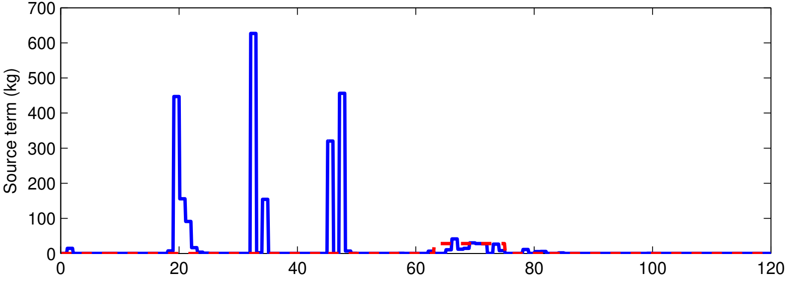
- least-squares solution,
  $\boldsymbol{x}_{\mathrm{LS}} = \arg\min_x \left((\boldsymbol{y} - \boldsymbol{Mx})^T \boldsymbol{R}^{-1} (\boldsymbol{y} - \boldsymbol{Mx}) \right)$, for $\boldsymbol{R} = \boldsymbol{I}$:

$$\boldsymbol{x}_{\mathrm{LS}} = \left(\boldsymbol{M}^T \boldsymbol{M} \right)^{-1} \boldsymbol{M}^T \boldsymbol{y} \tag{7}$$



Theory: modeling and estimation - ETEX example

- non-negative least-squares solution (s.t. $\boldsymbol{x} \geq 0$)



Theory: modeling and estimation - ETEX example

- LASSO: use of L1 norm in regularization $\longrightarrow$ assumption of sparsity,
  $\boldsymbol{x}_{\text{LASSO}} = \arg\min_x \left((\boldsymbol{y} - \boldsymbol{M}\boldsymbol{x})^T \boldsymbol{R}^{-1} (\boldsymbol{y} - \boldsymbol{M}\boldsymbol{x}) + \alpha ||\boldsymbol{x}||_1 \right)$,
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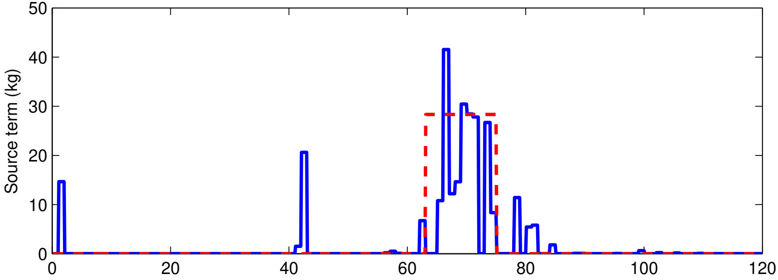

Theory: modeling and estimation - ETEX example - LS-APC algorithm

Least Square with Adaptive Prior Covariance (LS-APC) model:

- source term vector model
  - we assume correlation between neighboring source term elements

$$p\left(x_{j+1}\right) = N\left(-l_j x_j, v_j\right) \tag{8}$$

with prior model $p(l_j)$ favoring sparse ($l_j \rightarrow 0$) or smooth ($l_j \rightarrow -1$) solution

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  - the covariance structure shape, matrix $\boldsymbol{B}$, of the prior model $p\left(\boldsymbol{x}\right) = N\left(0, \boldsymbol{B}\right)$

$$\boldsymbol{B} = \boldsymbol{L}\boldsymbol{V}\boldsymbol{L}^T, \text{ where } \boldsymbol{L} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ l_j & 1 & 0 & 0 \\ 0 & \ddots & \ddots & 0 \\ 0 & 0 & l_{n-1} & 1 \end{pmatrix} \tag{9}$$

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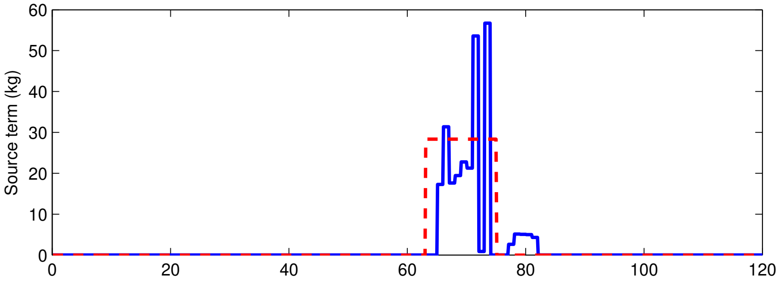
  - non-negativity of emissions is achieved by the choice of the prior model $p(\boldsymbol{x})$ as the truncated Gaussian distribution with non-negative support
- variational Bayes solution leading to an iterative algorithm

Theory: modeling and estimation - ETEX example

- LS-APC solution



[Tichý, O., Šmídl, V., Hofman, R., and Stohl, A.: LS-APC v1.0: a tuning-free method for the linear inverse problem and its application to source-term determination, Geosci. Model Dev., 2016.]

Theory: modeling and estimation

- So far, any questions or comments?

Applications

- multi-species emissions
- plume bias correction: towards non-linear regression
- spatial-temporal emissions estimation:
  - Cs-137 emissions from Chernobyl wildfires
  - atmospheric microplastics
  - towards satellite data inversion: ammonia case

Applications: multi-species emissions

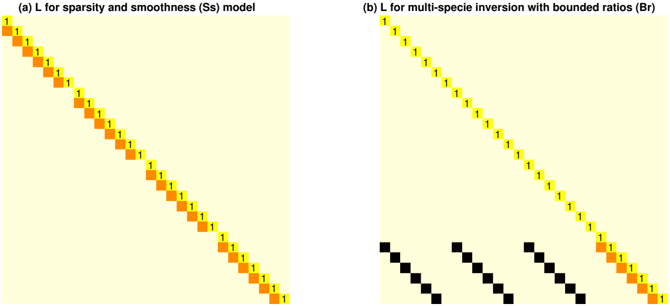
- often, we do not have one emitted specie, but a mixture of different species:
  - complex multi-nuclide emissions in the case of nuclear accidents
  - different size of fractions, e.g. from fires
  - different emissions altitudes
- it makes sense that these species are (e.g. for a given time-step) correlated

Applications: multi-species emissions

- recal the the covariance structure of the LS-APC model,

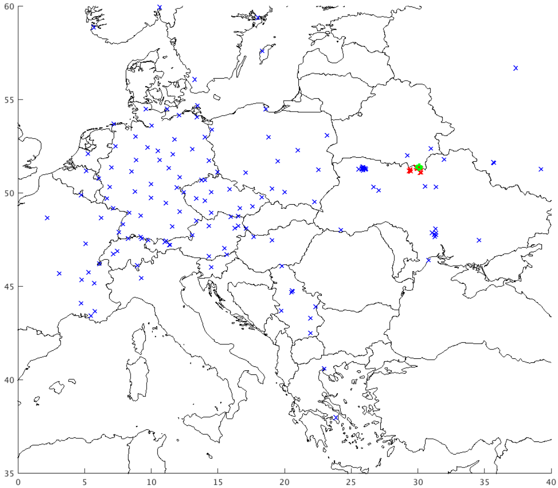
$$\boldsymbol{B} = \boldsymbol{L}\boldsymbol{V}\boldsymbol{L}^T, \text{ where } \boldsymbol{L} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ l_1 & 1 & 0 & 0 \\ 0 & \ddots & \ddots & 0 \\ 0 & 0 & l_{n-1} & 1 \end{pmatrix}$$

- we can model general structure of the matrix $\boldsymbol{L}$, such as



Applications: multi-species emissions - wildfires in Chernobyl

- substantial amount of Cs-137 has been emitted during April 2020 wildfires around Chernobyl and measured across Europe:



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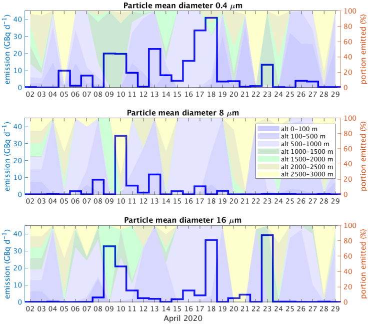

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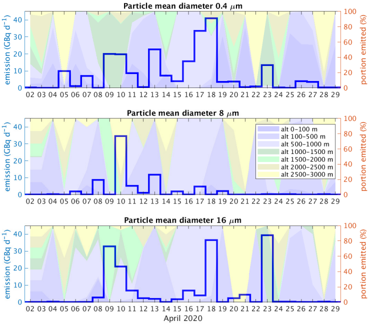
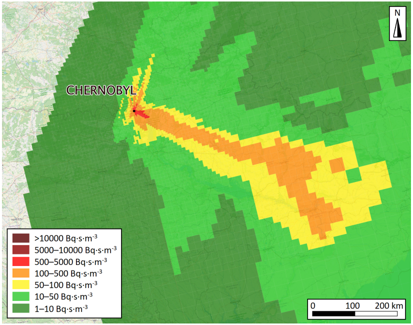




[Tichý, O., Evangeliou, N, Selivanova, A., and Šmídl, V.: Inverse modeling of 137Cs during Chernobyl 2020 wildfires without the first guess, submited to Atmospheric Pollution Research, 2024.]

Applications: Plume bias correction

- motivation: wind angle, the case of low-level inversion over the Central Europe in November 2011

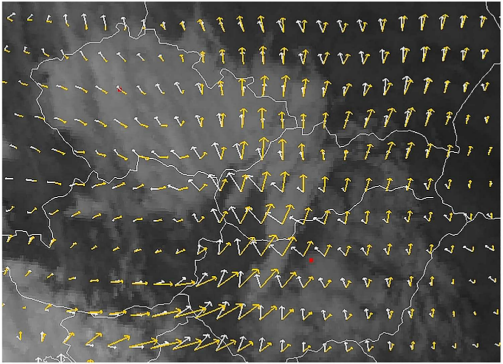
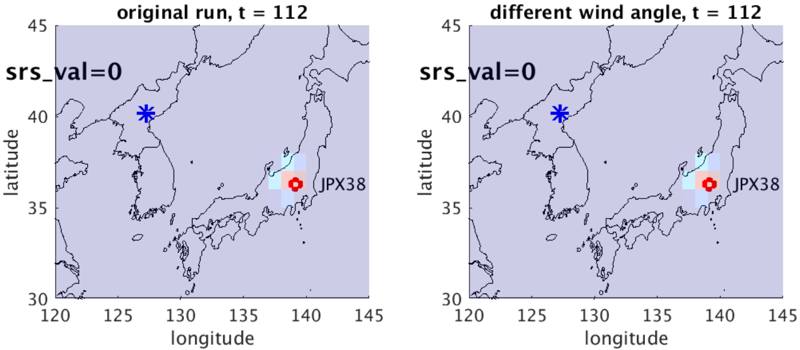


Fig 6. Visible satellite image at 12 UTC, 4 November 2011. Fog and stratus clouds covering the Czech Republic indicates the presence of a strong low-level inversion. Wind below the inversion layer (950 hPa, white arrows) differs significantly from the wind above the inversion layer (850 hPa, orange arrows). Red dots show the locations of Prague and Budapest. Data obtained from EUMETSAT and GFS.

- few degrees in wind orientation can make a big difference in SRS coefficients, i.e. the matrix $\boldsymbol{M}$

[Leelosy et al., Numerical simulations of atmospheric dispersion of iodine-131 by different models, PloS One, 2017]

Applications: Plume bias correction



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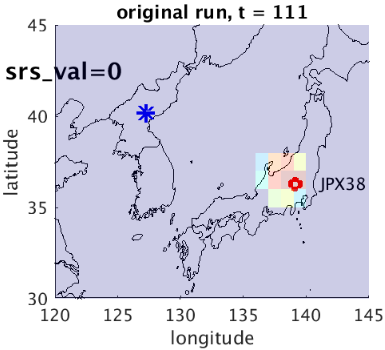

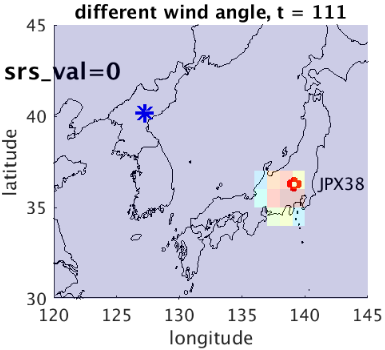

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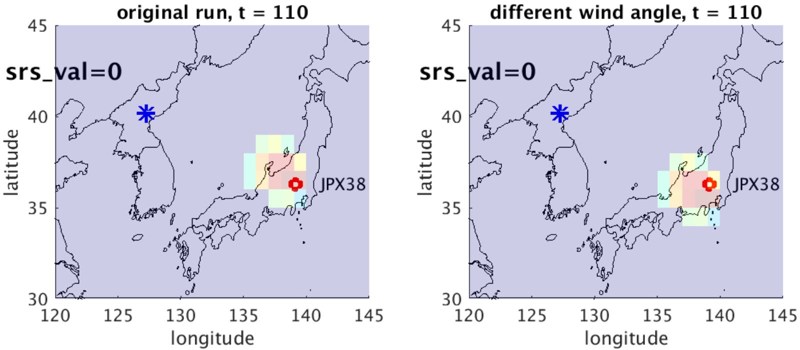

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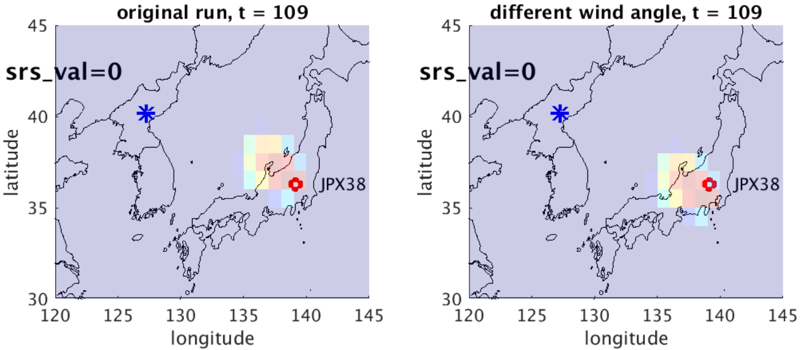

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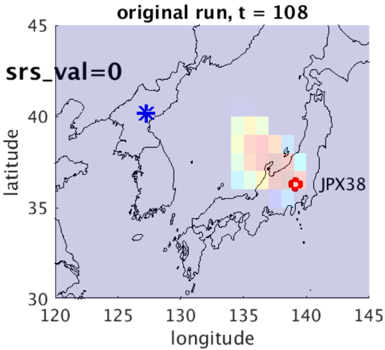

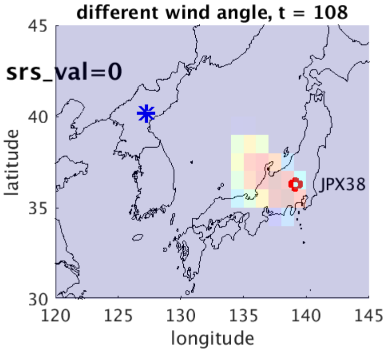

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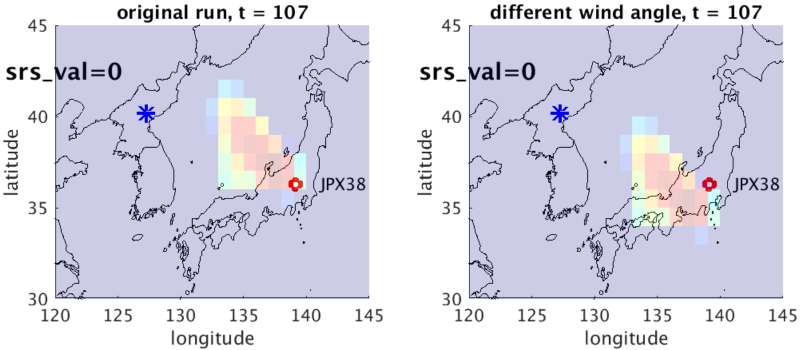

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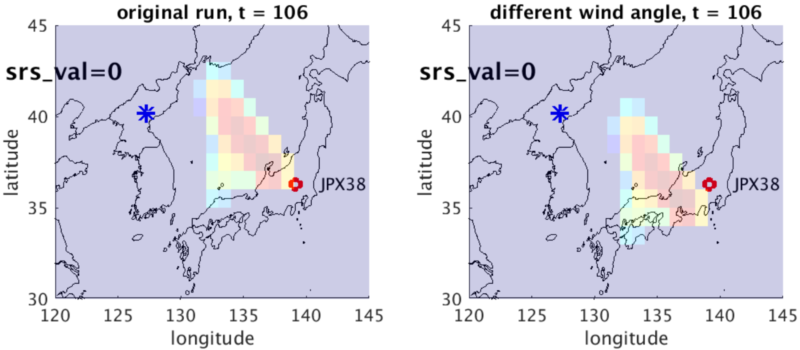

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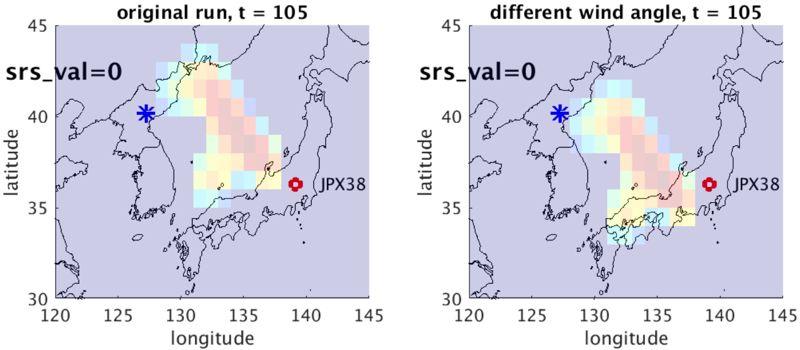

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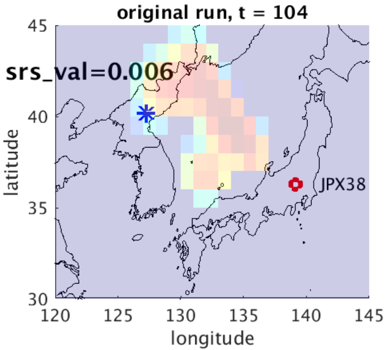

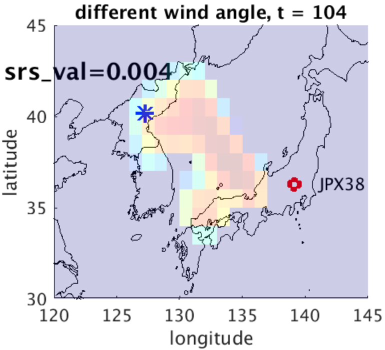

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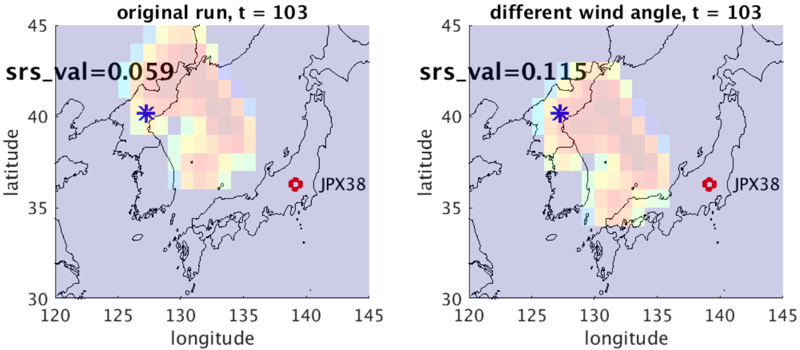

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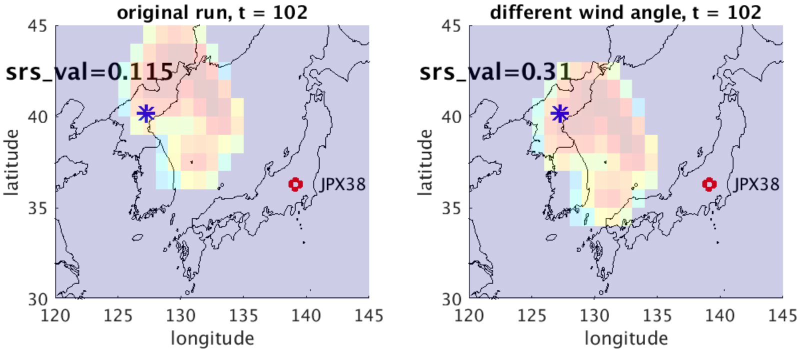

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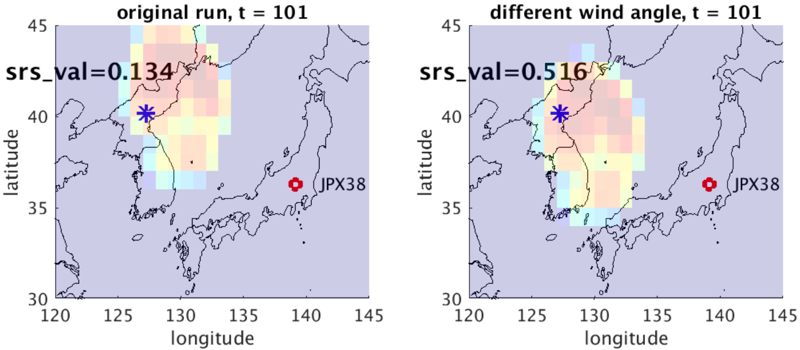

Applications: Plume bias correction

- atmospheric transport model provide more than point concentration predictions
  - we can read predicted concentrations also around each measurement

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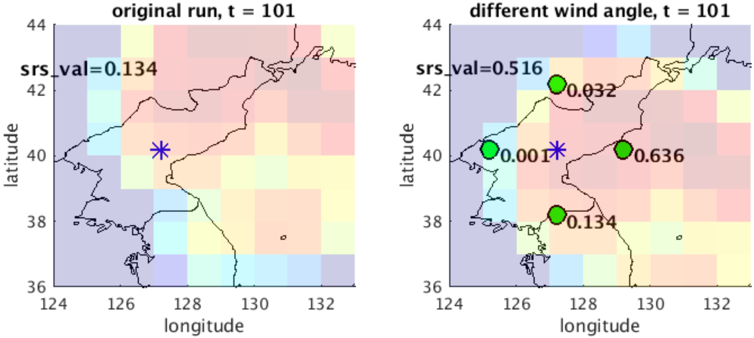
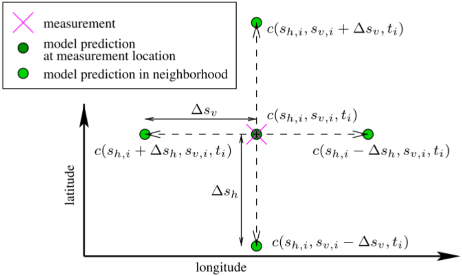


Applications: Plume bias correction

- what information we have



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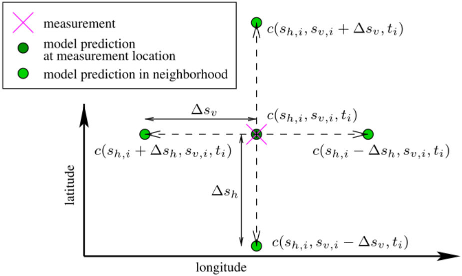

- we can formalize this information to SRS-like matrices as horizontal, vertical, and temporal gradients around each measurement, yielding

$$\boldsymbol{M}_h, \boldsymbol{M}_v, \boldsymbol{M}_t$$

of the same size as the original SRS matrix $\boldsymbol{M}$

Applications: Plume bias correction - towards non-linear regression

- formally:

$$\boldsymbol{y} = \overbrace{\left(\boldsymbol{M} + \boldsymbol{H}_h\boldsymbol{M}_h + \boldsymbol{H}_v\boldsymbol{M}_v + \boldsymbol{H}_t\boldsymbol{M}_t\right)}^{\widetilde{\boldsymbol{M}}}\boldsymbol{x} \qquad (10)$$

- $\boldsymbol{M}_h$, $\boldsymbol{M}_v$, and $\boldsymbol{M}_t$ are **simulated** sensitivities of horizontal, vertical, and temporal gradients around each measurement
- $\boldsymbol{H}_h$, $\boldsymbol{H}_v$, and $\boldsymbol{H}_t$ are matrices with **unknown** weights (*bias corrections*) on diagonals

$\Longrightarrow$ bilinear problem

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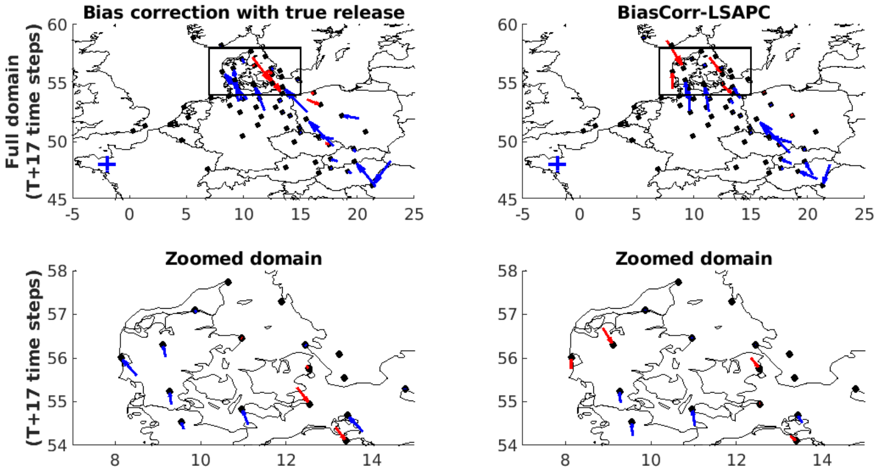
$\Longrightarrow$ bilinear problem

- in Bayesian models for $\boldsymbol{H}$s: assumptions such as
  - only small corrections are allowed in time and space
  - bias corrections of neighboring sensors are correlated

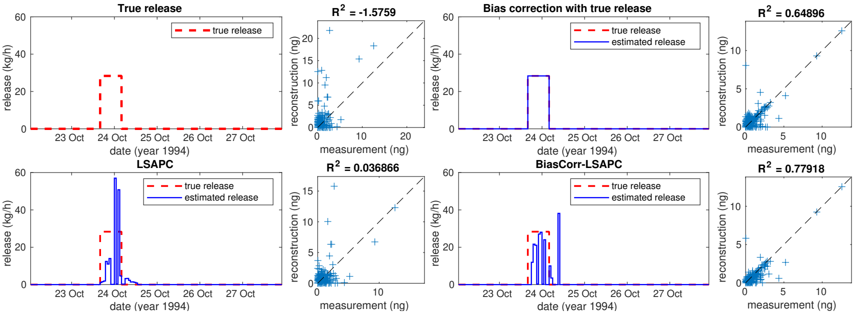
[Tichý, O., Šmídl, V. and Evangeliou, N.: Source term determination with elastic plume bias correction, Journal of Hazardous Materials, vol.425, 2022.]

Applications: Plume bias correction - ETEX experiment

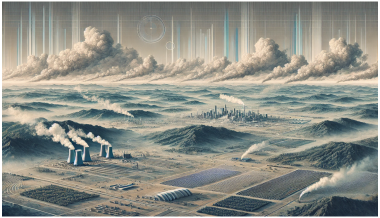
Example corrections (comparison with true release)



Applications: Plume bias correction - ETEX experiment

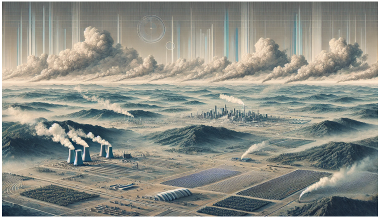


Applications: spatial-temporal emissions



- so far, we assumed point-source emissions
- the emissions, however, can come from (part of) spatial domain, introducing the whole new complexity to the estimation problem
- measurements are then formed by contributions from possible many locations

Applications: spatial-temporal emissions



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- the emissions, however, can come from (part of) spatial domain, introducing the whole new complexity to the estimation problem
- measurements are then formed by contributions from possible many locations
- examples:
  - emissions from wildfires
  - atmospheric microplastics
  - satellite data: ammonia emissions

Applications: spatial-temporal emissions - Chernobyl wildfires

- Chernobyl wildfires have spatial-temporal character, see map based on FIRMS satellite data (Fire Information for Resource Management System, NASA):

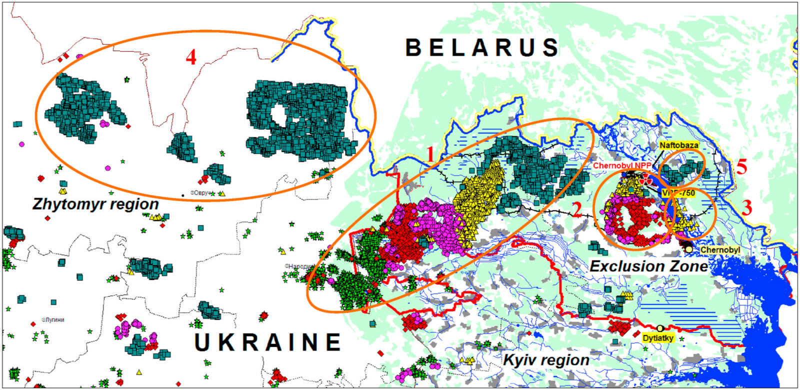

Fig. 1. Location of active fires from April 3–20, 2020, according to satellite data (★ - April 3–7, ◆ - April 8–9, ● - April 10–11, △ - April 12–13, ■ - April 16–20). The main areas of fires are shown by ovals: 1- Polissia district; 2- near the villages of Chystohalivka and Kopachi; 3 - the cooling pond of ChNPP; 4- Ovruch district; 5 - left bank of the Pripyat River.

[Talerko, M., et al.: Simulation study of radionuclide atmospheric transport after wildland fires in the Chernobyl Exclusion Zone in April 2020, Atmospheric Pollution Research, 2021]

Applications: spatial-temporal emissions - Chernobyl wildfires

- the spatial-temporal inverse problem can be formulated as

$$\boldsymbol{y} = \sum_{\text{lon, lat}} \tilde{\boldsymbol{M}}_{\text{lon, lat}} \boldsymbol{x}_{\text{lon, lat}} + \boldsymbol{e}, \tag{11}$$

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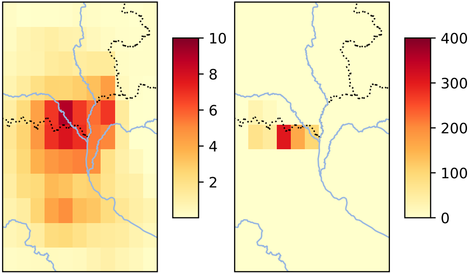
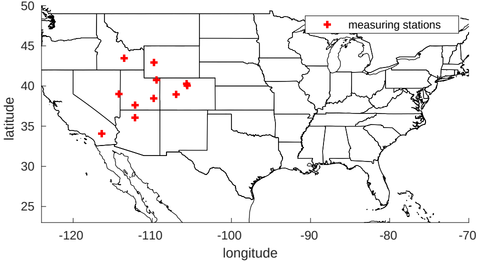
- work in progress



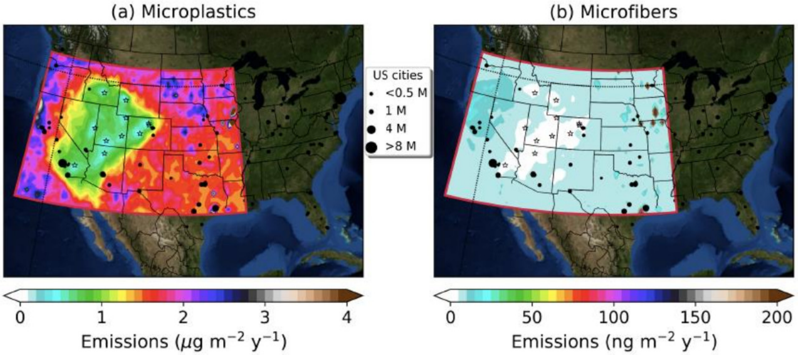
Applications: spatial-temporal emissions - microplastics

- atmospheric microplastics: relatively difficult to measure and analyze
- see example dataset of microplastics and microfibers data:



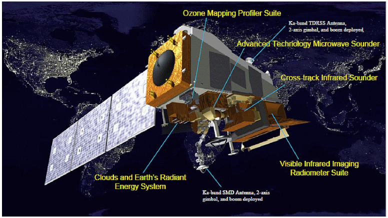
[Brahney, J., et al.: Plastic rain in protected areas of the United States, Science, 2020]

Applications: spatial-temporal emissions - microplastics



[Evangeliou N., Tichý O., Eckhardt S., Groot Zwaaftink C., Brahney J. , Sources and fate of atmospheric microplastics revealed from inverse and dispersion modelling; from global emissions to deposition, Journal of Hazardous Materials, 2022.]

Applications: spatial-temporal emissions - ammonia from satellite data



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- measurement available (almost) for each spatial grid-point and given vertical level

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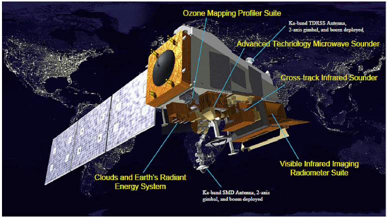

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- non-linear observation model for each grid-point

$$\ln \boldsymbol{v}^{\text{sat}} = \ln \boldsymbol{v}^{a} + \boldsymbol{A}\left(\ln \boldsymbol{v}^{\text{model}} - \ln \boldsymbol{v}^{a}\right) \qquad (12)$$

- $\boldsymbol{v}^{\text{model}}$ is modeled concentration from with contributions from the whole domain

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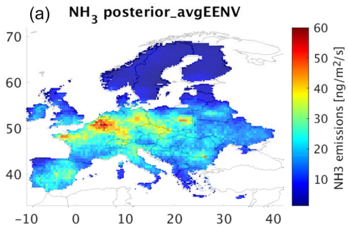

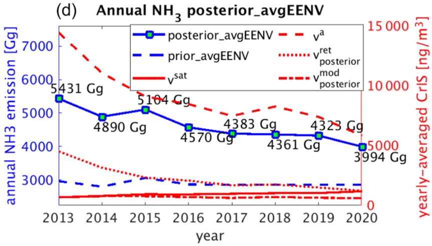

[Tichý O., Eckhardt S., Balkanski Y., Hauglustaine D., and Evangeliou N., Decreasing trends of ammonia emissions over Europe seen from remote sensing and inverse modelling, Atmospheric Chemistry and Physics, 2023.]

Conclusions remarks

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  - no need for extensive tuning of parameters
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Thank you for your attention.
Feel free to ask.