Using FlexPart For Modelling GHGs

Rona Thompson, NILU

Applications of modelling GHGs

Observations of GHGs (CO_2, CH_4, N_2O, etc.) are used to:

1) determine radiative forcing and thus warming effect
2) guide for realistic forcing of climate models
3) verification of GHG emissions

Model simulations of GHGs can:

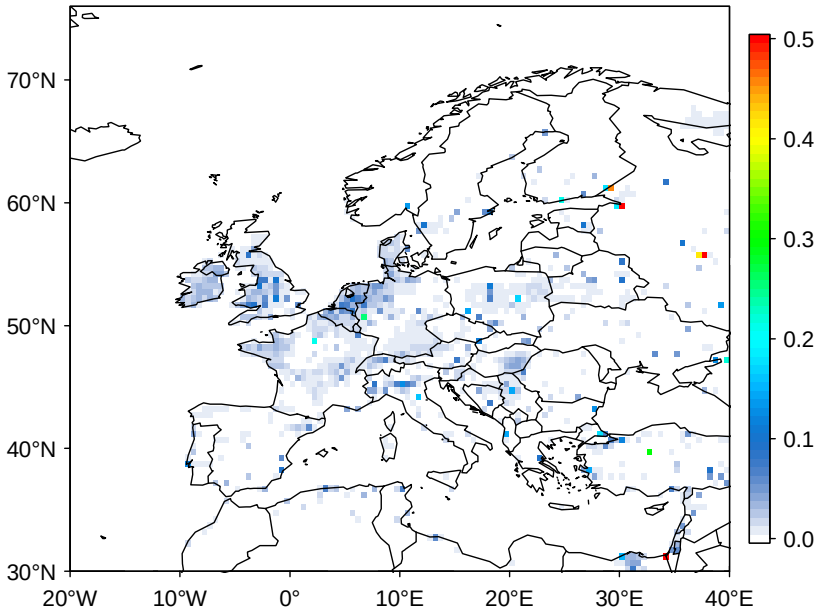
- interpolate sparse observations to help with (1) and (2)
- relate *prior* emission estimates to changes in atmospheric mixing ratios for comparison with observations to help with (3)
- provide *prior* estimates for satellite retrievals of GHGs
- help better understand observed variability in observations

Considerations for modelling GHGs

- Most GHGs are long-lived species (CH_4 lifetime ~10 yrs, N_2O lifetime ~120 yrs)
  - ✓ need to account for the *history* of the atmosphere up to the point where FlexPart simulation begins
  - ✓ this can be done through adding an *initial condition* to the simulated change in mixing ratio
- GHGs have many point sources as well as diffuse sources
  - ✓ emission estimates are discretized to grid-cells and in time
  - ✓ consider efficiency of simulation: compute each grid-cell as separate source OR use backward time simulation from observation?

Example emissions for CH_4

Total microbial emissions (g m^{-2} day^{-1})

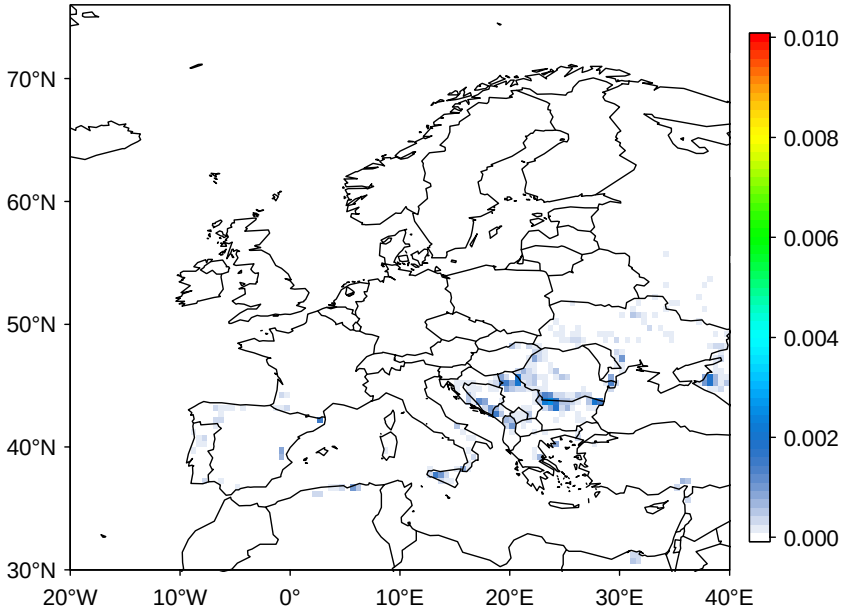


Combination of diffuse and point sources discretized on a grid

Includes emissions from:

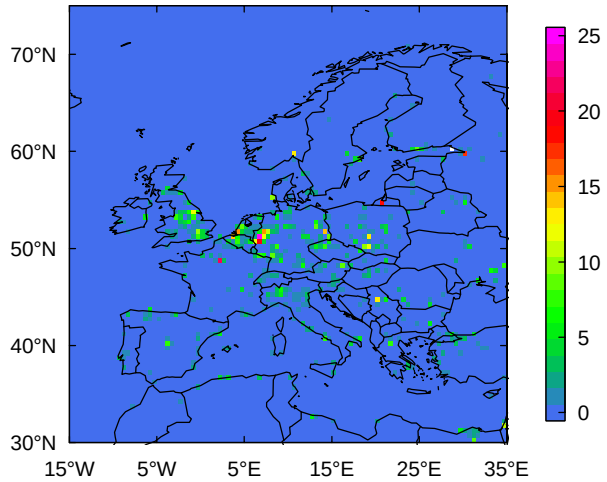
- wetlands
- agricultural
  - enteric fermentation
  - manure management
- landfills

Biomass burning emissions (g m^{-2} day^{-1})

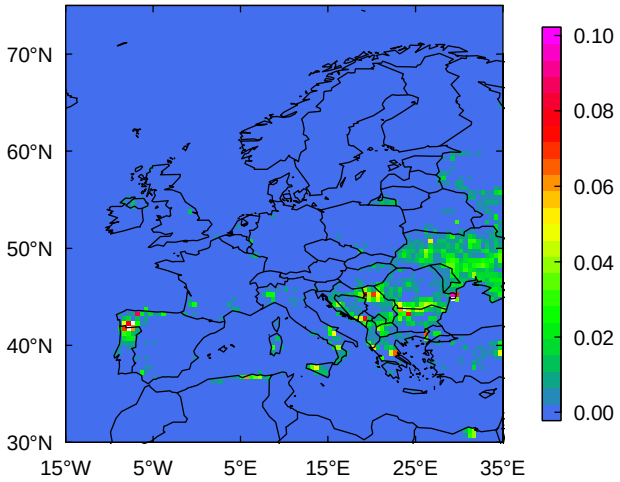


Example fluxes for CO_2

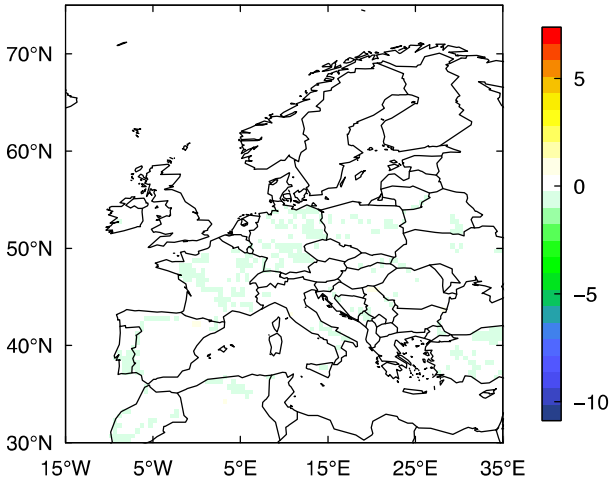
Fossil fuel (g/m^2/day): EDGAR-v4.3.2



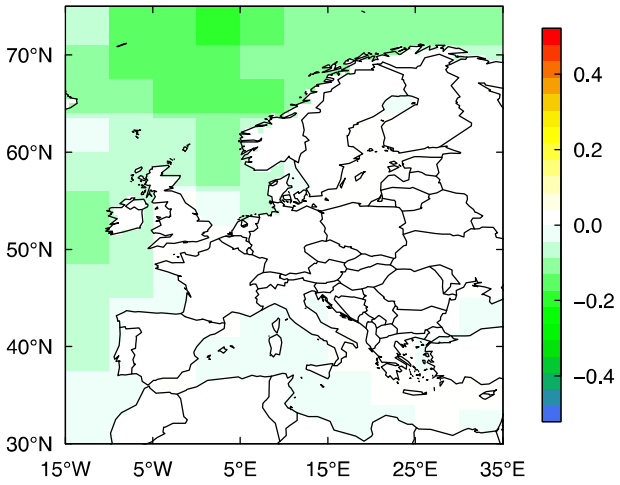
Biomass burning (g/m^2/day): GFED-v41s



NEE (g/m^2/day): ORCHIDEE



Ocean (g/m^2/day): SOCAM



How to set-up FlexPart for GHGs?

Using the backwards time mode:

- in COMMAND: ldirect = -1
- particles released from observation times and locations (these are the RELEASES)
- particles will follow wind fields backwards in time (backwards in time dispersion)
- calculation of residence times of particles in the first vertical layer to establish the SOURCE-RECEPTOR-RELATIONSHIP (SRR) (see: *Seibert and Frank, Atmos. Chem. Phys. 2004*)
- SRRs written to grid_time files that can be used with gridded emission estimates to calculate the change in mixing ratio at the observation times and locations

Files to prepare

In the “options” folder:

- COMMAND: generic settings for run
- RELEASES: times and locations of observations (i.e. releases)
- OUTGRID: specification of output grid
- AGECLASSES: lifetime of virtual particles (determines length of backward trajectories)
- SPECIES_XXX: defines species characteristics
- pathnames: in/out paths for flexpart

Understanding output options

- COMMAND file settings for output units for backward runs

Direction	IND_SOURCE	IND_RECEPTOR	INPUT	OUTPUT
backward	1	1	1	s
backward	1	2	1	s m^3 kg^{-1}
backward	2	1	1	s m^3 kg^{-1}
backward	2	2	1	s

- Use units of “s” if modelling concentrations (kg m^{-3}):

  SRR × flux × 1/z = (s) × (kg m^{-2} s^{-1}) × m^{-1} = kg m^{-3}

- Use units of “s m^3 kg^{-1}” if modelling mixing ratios (ppt):

  SRR × flux × 1/z = (s m^3 kg^{-1}) × (kg m^{-2} s^{-1}) × m^{-1} = ppt (by mass)

Understanding output options

COMMAND settings for output files

- LOUTSTEP: time step for the output (grid_time files) in sec
- LOUTAVER: averaging time for the output in sec
- NESTED_OUTPUT: output also for a nested grid (0 or 1)
- LINIT_COND: output also initial conditions files (1)
- LNETCDFOUT: output (grid_time files etc.) as NetCDF (0 or 1)
- SURF_ONLY: for grid_time files write only surface layer (1)

Output files

- grid_time_YYYYMMDDHHMMSS
  - ✓ one file per SRR (or “footprint”) (sparse matrix format)
  - ✓ dimensions: lon × lat × height × release
  - ✓ also contains optional dimensions for ageclass and species
- grid_initial_001
  - ✓ one file at the termination footprint
  - ✓ dimensions: lon × lat × height × release
  - ✓ also contains optional dimensions for ageclass and species

Simulating GHG mixing ratios

1) change in mixing ratio using grid_time files
   - $y_i^{chg} = \boldsymbol{h}_i \boldsymbol{x}$
   - for a single observation, y_i^{chg}, is a dot product of vectors $\boldsymbol{h}_i$ and $\boldsymbol{x}$, the SRR (divided by height z) and fluxes, respectively
   - if choose units for mixing ratio, y_i^{chg} is in ppt (by mass) need to convert to pptv (by volume) using: 28.97/molmass
2) initial conditions using grid_initial files
   - $y_i^{ini} = \boldsymbol{h}_i^{ini} \boldsymbol{y}^{cini}$
   - for a single observation, y_i^{ini}, is dot product of $\boldsymbol{h}_i^{ini}$ and $\boldsymbol{y}^{cini}$, a weighting vector (defined by grid_initial) and a vector specifying the 3D concentration field (from e.g. a Eulerian model), respectively
3) total modelled mixing ratio
   - $y_i^{mod} = y_i^{chg} + y_i^{ini}$

First order chemistry – OH reaction

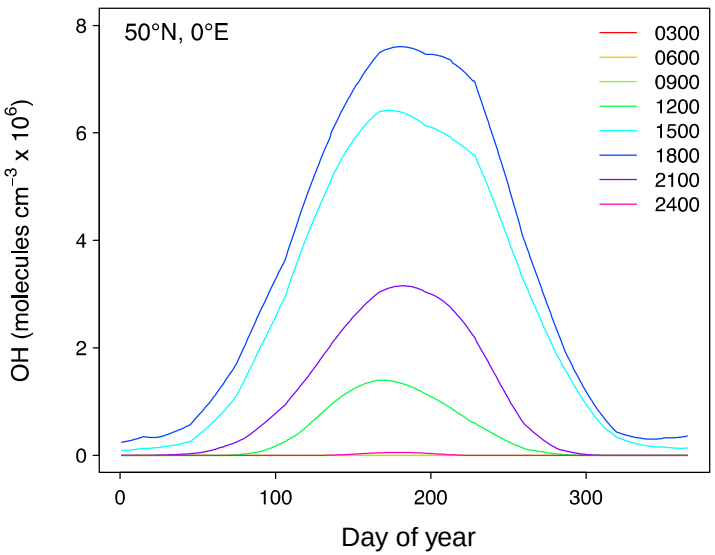
- OH is major tropospheric oxidant oxidizing many species (e.g. CH_4, CO, VOCs)
- OH dominant sink process of CH_4 (~90% of atmospheric loss)
- OH oxidation is a first-order reaction i.e. rate depends only on concentration of one species (i.e. not on OH itself)
- Can assume OH is not depleted significantly by reaction with other species – therefore this chemistry is a linear process
- Rate of OH reaction depends on reagent and temperature
- Amount of OH depends on photochemical production (availability of light) and is fairly well buffered in the atmosphere

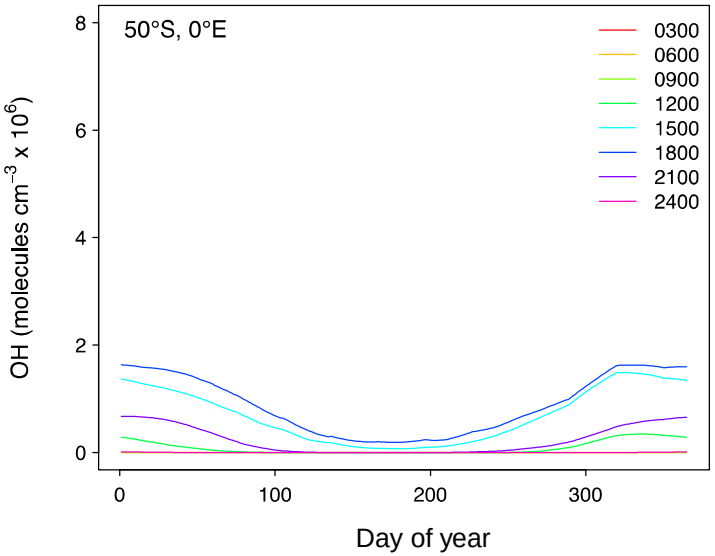
OH reaction in FlexPart

- Uses pre-calculated monthly OH fields (from GEOS-Chem)
- Correct monthly fields for the photolysis rate based on the solar zenith angle (gethourlyOH.f90):

$$OH_{i,j,k}^{hour} = OH_{i,j,k}^{month} \times \frac{j_{i,j,k}}{\bar{j}_{i,j,k}}$$

- where $j_{i,j,k}$ is the hourly photolysis rate for a given location (i,j,k) and $j^{av}_{i,j,k}$ is the monthly mean photolysis rate (precalculated)





OH reaction in FlexPart

- The OH reaction rate is calculated using the temperature dependent formulation (ohreaction.f90):

$$k_{OH} = CT^N \exp\left(\frac{-D}{T}\right)[OH]$$

- where *C* and *D* are species specific (assigned in SPECIES) in units of [cm^3 molecule^{-1} s^{-1}] and [K], respectively, *T* is temperature and [*OH*] the OH concentration [molecule cm^{-3}]. The constant *N* has a default value of 2 but may vary for some species.
- The mass of a given species, *x* after reaction with OH is given as:

$$x_{t+t'} = x_t \exp(-k_{OH} t')$$

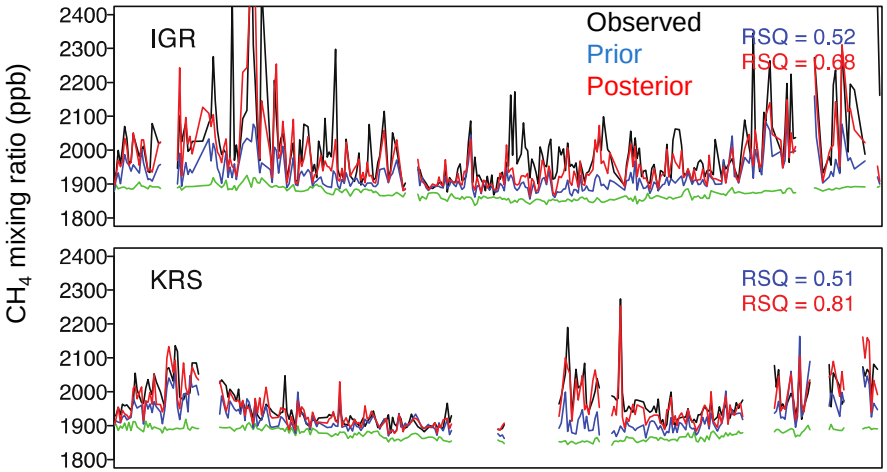
- where *t'* is the time interval of reaction (ltsample)

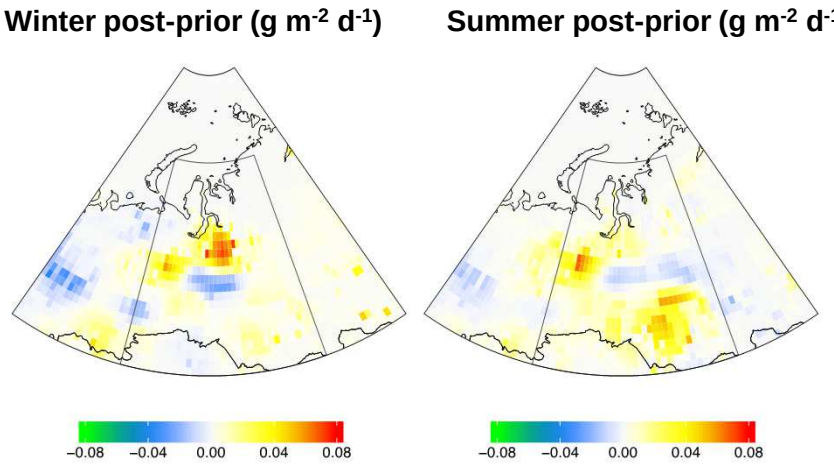
Ref: Atkinson, R. (1997) *J. Phys. Chem. Ref. Data, 26*(2), 215-290

Real-life examples

CH_4 emissions in Western Siberia

- Prior emissions underestimate mixing ratios at e.g. IGR and KRS stations
- Indicates likely underestimate of emissions
- Atmospheric inversion (FLEXINVERT+) adjusts prior emission to best match observations



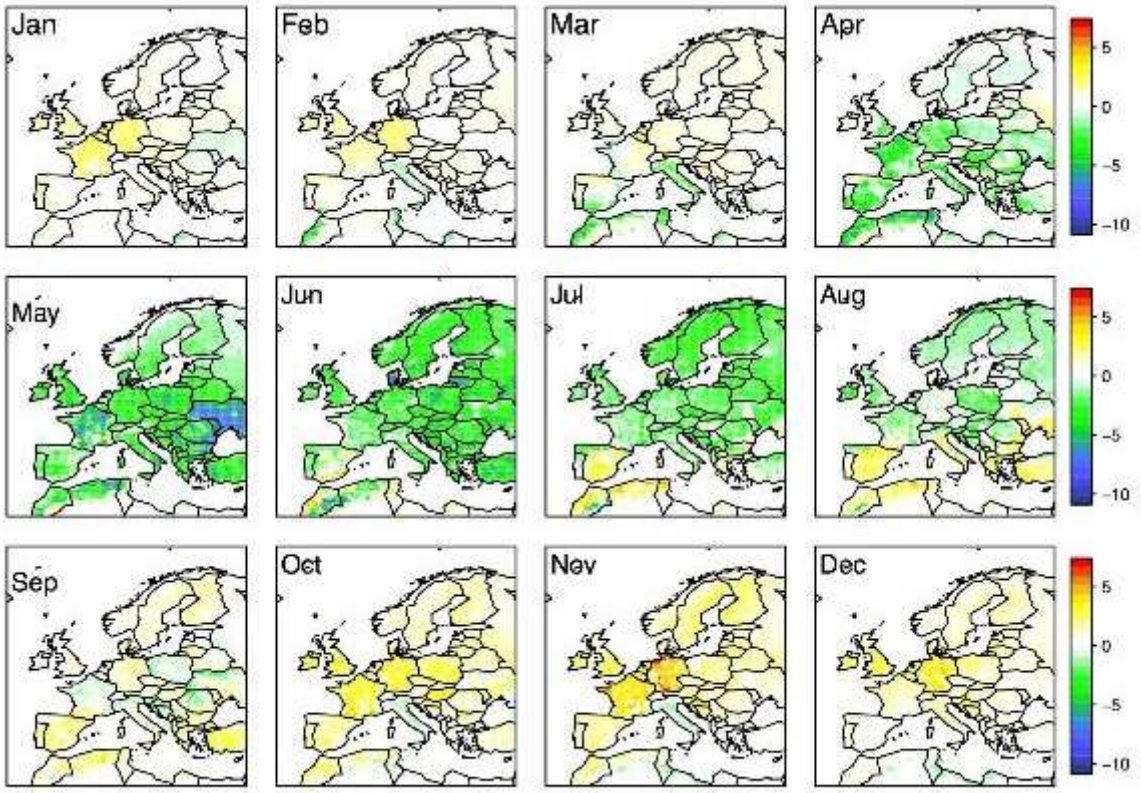


Fully study described in: Thompson et al., *Atmos. Chem. Phys*. 2017

Real life examples

CO_2 land-biosphere fluxes in Europe

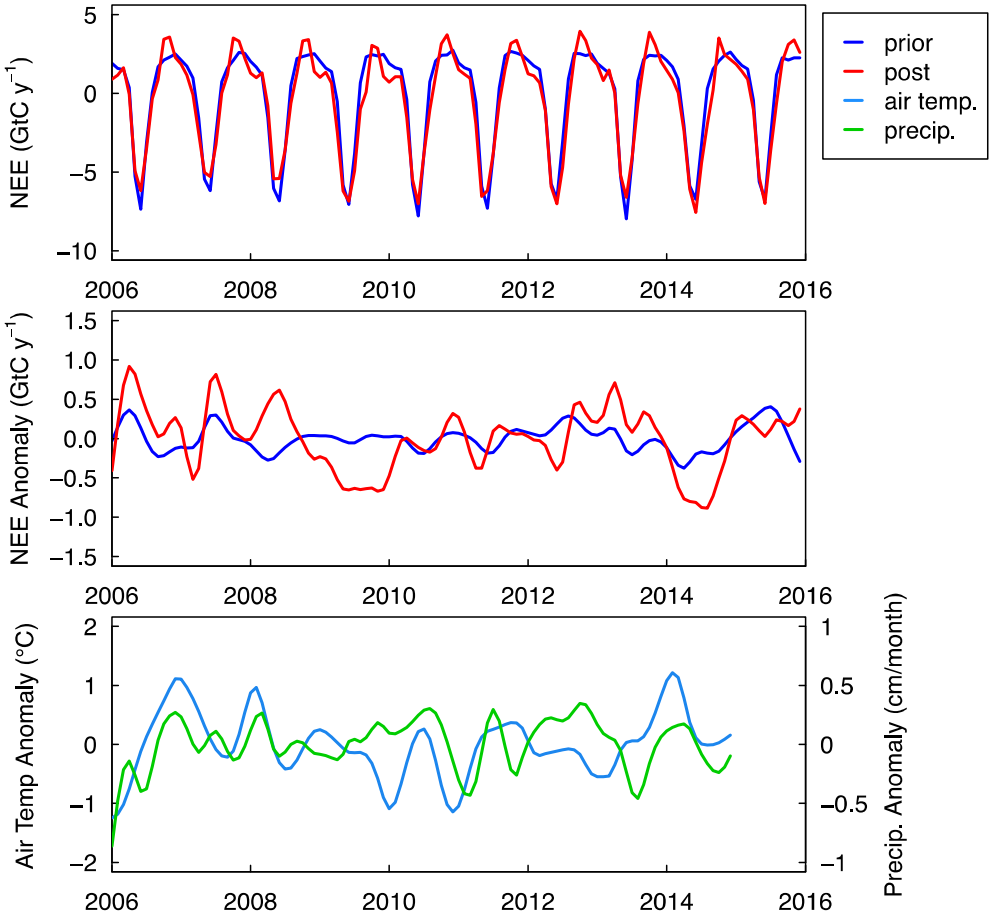
- Atmospheric inversions of CO_2 to determine the net ecosystem exchange (NEE) flux



Real life examples

CO_2 land-biosphere fluxes in Europe

- Preliminary results find the European biosphere is a net sink of CO_2 of 0.41 ± 0.3 GtC/y
- Previous studies find a sink of between 0.3 and 1.0 GtC/y
- Annual NEE is very uncertain since it is a small residual signal compared to the gross fluxes



Comment on setting-up FlexPart

- Real-world problems will involve 1000s to 10000s of observations
- Need to automate generation of RELEASES (and other *options* files)
- For computational efficiency split runs by e.g. observation site and month (run these separate jobs in parallel)

Comment on forward time mode

- GHGs can of course also be simulated in forward time mode in FlexPart
- Generate grid_conc (for concentration) and grid_pptv (for volume mixing ratios)
- Need to create RELEASES file containing *ALL* fluxes for all grid cells (or modify to read directly from ncdf file – this modification has been made by S. Henne at EMPA)
- For atmospheric inversions, the grid_conc/grid_pptv files are not sufficient (but could be used for ensemble inversion approaches)

Inverse modelling

FlexInvert+

- A Bayesian inversion framework developed at NILU
- Uses FLEXPART for atmospheric transport
- Suitable for long-lived species
- See: http://flexinvert.nilu.no