FLEXINVERT: An atmospheric Bayesian inversion framework for
 determining surface fluxes of trace species using an optimized grid

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7 Abstract

8 We present a new modular Bayesian inversion framework, called FLEXINVERT, for 9 estimating the surface fluxes of atmospheric trace species. FLEXINVERT can be applied to 10 determine the spatio-temporal flux distribution of any species for which the atmospheric loss (if any) can be described as a linear process and can be used on continental to regional and 11 12 even local scales with little or no modification. The relationship between changes in 13 atmospheric mixing ratios and fluxes (the so-called source-receptor relationship) is described 14 by a Lagrangian Particle Dispersion Model (LPDM) run in a backwards in time mode. In this 15 study, we use FLEXPART but any LPDM could be used. The framework determines the 16 fluxes on a nested grid of variable resolution, which is optimized based on the source-receptor 17 relationships for the given observation network. Background mixing ratios are determined by coupling FLEXPART to the output of a global Eulerian model (or alternatively, from the 18 19 observations themselves) and are also optionally optimized in the inversion. Spatial and 20 temporal error correlations in the fluxes are taken into account using a simple model of 21 exponential decay with space and time and, additionally, the aggregation error from the 22 variable grid is accounted for. To demonstrate the use of FLEXINVERT, we present one case 23 study in which methane fluxes are estimated in Europe in 2011 and compare the results to 24 those of an independent inversion ensemble.

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#### 26 1. Introduction

27 Observations of atmospheric mixing ratios (or concentrations) of trace species (gases or 28 aerosols) contain information about their fluxes between land/ocean and the atmosphere. 29 Atmospheric inversions use this information formally in a statistical optimization to find 30 spatio-temporal distributions of trace gas (or aerosol mass) fluxes (e.g. Tans et al. 1990). This 31 can be done provided that there is a model of atmospheric transport relating changes in fluxes 32 to changes in mixing ratios (or concentrations), that is, the so-called Source-Receptor 33 Relationships (SRRs). Basically, two types of models are used: Eulerian models, in which 34 atmospheric transport and chemistry are calculated relative to a fixed coordinate, or

Lagrangian Particle Dispersion Models (LPDMs), in which diffusion and chemistry are
 calculated from the perspective of air parcels transported by ambient winds.

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4 Eulerian models have been used extensively in atmospheric inversions but have a 5 disadvantage in that SRRs cannot be calculated directly from the model. Instead, the SRRs 6 can be found from multiple runs of the transport model to determine the sensitivity of all 7 receptors to changes in the fluxes in a discrete number of regions (e.g. Fung et al. 1991; 8 Enting, 2002; Rayner et al. 1999). This approach is simple to implement but the 9 computational cost of running the model for each flux region limits the number of regions and thus the resolution that can be used. More recently, adjoints of Eulerian transport models have 10 been developed and can be used as an alternative to calculating SRRs. Adjoints calculate the 11 12 partial derivative of the change in flux in a given grid cell to the change in mixing ratio at a 13 given point and time, which can be used to find the optimal fluxes based on a set of observations (e.g. Kaminski et al. 1999; Chevallier et al. 2005). This approach has the 14 15 advantage that the fluxes can be solved at higher resolution (i.e. the resolution of the transport 16 model). However, deriving adjoint models represents a significant technical challenge, thus 17 adjoints are not available for all transport models and there is a significant lag between forward model developments and these being implemented in the adjoint. A further 18 19 disadvantage is that these systems are computationally demanding, as they require forward 20 and adjoint model runs for every iteration until convergence.

21

22 LPDMs are self-adjoint, i.e., they can track the dispersion of virtual particles representing e.g. 23 an atmospheric gas forward in time from its sources/sinks to receptors (i.e. measurement 24 sites) or backwards in time from receptors to its sources/sinks using the identical model 25 formulation (Stohl et al. 2003; Seibert and Frank 2004; Flesch et al. 1995). The forward and 26 backward calculations are equivalent but one direction can be much more computationally 27 efficient than the other. For instance, if there are few receptors but many sources/sinks, the 28 backwards mode is more efficient. This is the case, for instance, when particles are tracked 29 backwards from a relatively small number of available atmospheric observation sites (i.e. 30 receptors), as in our demonstration case. This feature makes LPDMs very efficient for the 31 purpose of atmospheric inversion and they have been previously used in numerous studies 32 (e.g. Gerbig et al. 2003; Lauvaux et al. 2009; Stohl et al. 2010; Thompson et al. 2011; Keller 33 et al. 2012; Brunner et al. 2012). Lagrangian models may be used on a global scale (e.g. Stohl 34 et al. 2010), sub-continental scale (e.g. Gerbig et al. 2003) or on a regional scale in the order

1 of a few hundred square kilometers (e.g. Lauvaux et al. 2009). Owing to their favorable 2 treatment of atmospheric turbulence in the boundary layer, LPDMs can even be used down to 3 scales of a few hundreds of meters (Flesch et al. 1995) and have been used for inferring 4 source strengths for local sources (e.g. farmsteads and oil spills). A further advantage of 5 LPDMs is that they can be run backward exactly from a measurement site, unlike Eulerian 6 models, where site measurements are represented by the averaged value of the corresponding 7 grid cell. By focusing on local or regional scales, fine resolution may be used without running 8 into problems of too large a number of unknown variables (in this case the fluxes). Fine 9 resolution is desirable as it reduces the model representation error, also known as aggregation error (Kaminski et al. 2001; Trampert and Snieder 1996) but it must be traded-off with the 10 11 total number of flux variables to be determined, which is subject to computational constraints.

12

13 Using LPDMs to solve the inverse problem, however, also has disadvantages. In LPDMs, 14 virtual particles are typically followed backward in time only for the order of days to a few 15 weeks, thus the influence of the atmospheric chemistry and transport and surface fluxes 16 further back in time (the so-called background mixing ratio) must be taken into account 17 separately. Although forward 3D simulations in LPDMs are possible, in order to reproduce 18 background signals, such as seasonal variability, simulations of months to years would be 19 necessary and, therefore, computationally too expensive (Stohl et al. 2009). Alternatively, the 20 background mixing ratio can be accounted for using either observation- or model-based 21 approaches. Observation-based approaches use some filtering method (either statistical or 22 based on meteorological criteria) to identify observations representative of the background i.e. 23 air not (or only minimally) influenced by fluxes during the time of the backwards calculations 24 (e.g. Stohl et al. 2009; Manning et al. 2011). However, the background is strongly influenced 25 by meteorology e.g., air transported from higher latitudes or altitudes may have significantly 26 different mixing ratios compared to air transported from lower latitudes or altitudes even if in 27 both cases no emissions occur during the backward calculation. This makes the determination 28 of an observation-based background difficult. Model-based approaches involve coupling the 29 back-trajectories at their point of termination to the mixing ratios determined from a global 30 model.

31

32 One approach is to run the LPDM on a regional domain and couple this to a global model at 33 the domain boundary. This approach was adopted by Rödenbeck et al. (2009), who use a 2-34 step method to first solve for the fluxes on a coarse grid using a Eulerian model and to

1 calculate the background mixing ratios at the receptors, and second to perform the inversion 2 at regional scale on a finer grid using an LPDM. A similar approach was developed by Rigby 3 et al. (2011) but using a 1-step method. A drawback of both these approaches, however, is 4 that only the coarse resolution Eulerian model is used to calculate the background mixing 5 ratios at the receptors and, thus, is more susceptible to transport errors. We use a different 6 approach and couple the LPDM, run on a global scale, to a Eulerian model at the time 7 boundary, such as done by Koyama et al. (2011). This approach utilizes the more accurate 8 transport of the LPDM to calculate the background at the receptors.

9

In this paper, we present a new framework, called FLEXINVERT, for optimizing fluxes by 10 employing an LPDM that can be coupled to mixing ratio fields from a global (Eulerian) 11 12 model. This method may be used from large continental scales down to local scales and can 13 be used for sparse as well as dense observation networks. In this method, the LPDM is used to 14 transport air masses and, thus, the influence of fluxes, to each receptor. The fluxes inside the 15 domain are optimized on a grid of variable resolution, where finer resolution is used in areas 16 with a strong observation constraint, i.e. close to receptors, and coarser resolution is used 17 elsewhere. FLEXINVERT, as it is presented here, requires that the LPDM is run on a global 18 domain, or at least that the domain is large enough so that trajectories do not exit the domain. 19 In summary, the features of this method are:

20

- atmospheric transport (SRR) is calculated using a single model, i.e. the LPDM

- the LPDM needs only to be run once for each species and receptor to find the SRRs,
  as the output can be applied to optimize the fluxes for any domain and resolution (as
  long as the resolution is no finer than that of the LPDM run)
- the variable resolution grid means that fine resolution close to receptors minimizes
   model representation errors
- background mixing ratios can be provided either by coupling to mixing ratio output
  from a global model or alternatively by using an observation-based method
- the background mixing ratios are optionally included in the optimization
- the influence of fluxes from outside the domain on the mixing ratios at the receptors is
   accounted for without having to solve for them explicitly, thereby reducing the
   dimensionality of the problem
- 33

Variable grid resolution has been used in atmospheric inversions previously such as in the studies of Manning et al. (2003), Stohl et al. (2010) and Wu et al. (2011). Our method for defining the variable grid is based on that of Stohl et al. (2010). However, we have also implemented a re-optimization of the fluxes at variable resolution back to the finest model resolution based on the method of Wu et al. (2011).

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This manuscript is structured as follows: first we describe the inversion framework and the
variable grid formulation and, second, we present an example using real observations of
methane (CH<sub>4</sub>) dry-air mole fractions to optimize CH<sub>4</sub> emissions in Europe.

10

#### **2.** Bayesian framework for linear inverse problems

### 12 **2.1 Forward model**

For cases where the atmospheric transport and chemistry are linear, the change in mixing ratio of a given atmospheric species can be related to its fluxes by a matrix operator. Furthermore, the absolute mixing ratio can be related to its fluxes plus the background mixing ratio, which together form the so-called state vector. This is shown in Eq. 1 where  $\mathbf{y}^{mod}_{(M\times 1)}$  is a vector of the modeled mixing ratio at M points in time and space,  $\mathbf{x}_{(N\times 1)}$  is a vector of the N state variables discretized in time and space, and  $\mathbf{H}_{(M\times N)}$  is the transport operator.

$$19 ymod = Hx (1)$$

20 For simplicity, we describe the case where the state variables are optimized for only one time 21 step, although the framework is able to optimize many time steps simultaneously (for an 22 overview of the variables and their dimensions see Tables 1 and 2). We construct the matrix 23 **H** from three components of the atmospheric transport to each receptor: 1) transport of fluxes within a nested domain (i.e. within the global domain),  $\mathbf{H}^{nest}$ , 2) transport of fluxes outside the 24 nested domain, H<sup>out</sup>, and 3) contribution of mixing ratio at the time and location when the 25 back-trajectories terminate, i.e. the initial mixing ratios,  $\mathbf{H}^{bg}$  (see Fig. 1). Similarly, x is 26 constructed from the fluxes inside the domain,  $\mathbf{f}^{nest}$ , fluxes outside the domain,  $\mathbf{f}^{out}$  (there are 27 no common variables between  $f^{nest}$  and  $f^{out}$  so there is no double counting of fluxes), and 28 initial mixing ratios taken from the output of a global model,  $y^{bg}$ . (Note that from hereon we 29 30 refer to the contribution to the observed mixing ratio from where the trajectories terminate as 31 the *initial mixing ratio* and the contribution from the initial mixing ratio plus from the fluxes 32 outside the domain as the *background mixing ratio*, this is explained in section 2.1.2). Equation 1 can thus be expanded to: 33

- $\mathbf{y}^{mod} = \mathbf{H}^{nest} \mathbf{f}^{nest} + \mathbf{H}^{out} \mathbf{f}^{out} + \mathbf{H}^{bg} \mathbf{y}^{bg}$
- 2

### 3 2.1.1 The Source Receptor Relationships (SRRs)

The matrices **H**<sup>nest</sup> and **H**<sup>out</sup> are Jacobians in which each element is a partial derivative of the change in mixing ratio at a given receptor with respect to the change in mass flux in a given grid cell and are built from the SRRs. In this study, we use the LPDM, FLEXPART (Stohl et al. 2005; Stohl et al. 1998) to derive the SRRs, although any other LPDM capable of running in backwards mode could also be used to construct these matrices.

9

In an LPDM, ensembles of particles are released from each receptor point and their displacement is calculated backwards in time based on wind fields from meteorological analysis data. Backwards and forwards calculations are practically equivalent because the transport is time reversible (Seibert and Frank 2004). For a tracer, which undergoes negligible loss in the atmosphere on the time scale of the LPDM calculations, the SRR can be expressed for a receptor and a flux in a given spatio-temporal grid cell (*i*,*n*), as proportional to the average residence time of *J* back trajectories in the grid cell under consideration:

17 
$$\frac{\partial y}{\partial x_{i,n}} = \frac{1}{J} \sum_{j=1}^{J} \frac{\Delta t'_{i,j,n}}{\rho_{j,n}}$$
(3)

where  $\rho_{i,n}$  is the air density in the grid cell and  $\Delta t'_{i,j,n}$  is the residence time of trajectory j in the 18 19 spatio-temporal grid cell (i,n) (see Seibert and Frank 2004). In Eq. 3, the SRR is in units of 20 residence time × volume per unit mass, which is integrated over the height of the surface layer 21 in FLEXPART to be comparable with the surface flux, which is given per unit area. 22 Atmospheric loss processes, such as reaction with the hydroxyl radical, dry and wet 23 deposition, or radioactive decay, can be considered by including a transmission function in 24 the right hand side of Eq. 3, which quantifies the loss. The SRRs for all receptors and fluxes inside the nested domain constitute the elements of H<sup>nest</sup> while the SRRs for all receptors and 25 fluxes outside the domain constitute the elements of H<sup>out</sup>. 26

27

### 28 2.1.2 Initial mixing ratios

The initial mixing ratio is the contribution from the mixing ratios where the LPDM backtrajectories terminate. We include two alternatives for the calculation of the initial mixing ratio. The first alternative uses the sensitivity to the mixing ratio at the points in space and time where the LPDM back-trajectories terminate (calculated in FLEXPART) and the mixing

(2)

1 ratio at those points taken from the output of a global model. The sensitivity to mixing ratio in 2 a grid cell at a given time (*i*) is calculated as the number of particle trajectories that terminate 3 in the grid cell ( $n_i$ ) divided by the total number of particle trajectories released (*J*):

$$4 \qquad \frac{\partial y}{\partial y_i} = \frac{n_i}{J} \tag{4}$$

Again, loss processes can be considered by including a transmission function in the right hand side of Eq. 4. The sensitivity to the mixing ratio in all *P* grid cells (over the global domain) and for all *M* observations is represented by the matrix  $\mathbf{H}^{\mathbf{bg}}_{(M \times P)}$  and the mixing ratios from the global model by the vector  $\mathbf{y}^{\mathbf{bg}}_{(P \times 1)}$ , which has been interpolated to the resolution of the LPDM output. Thus, the initial mixing ratio at all receptors is  $\mathbf{H}^{\mathbf{bg}}\mathbf{y}^{\mathbf{bg}}$ .

10

11 The second alternative approximates the background mixing ratios from the observations 12 themselves. In this case, the background mixing ratio is calculated in one step, i.e. there is no 13 separate calculation of the initial mixing ratio and contribution from outside the nested 14 domain. We have implemented a simple method involving selecting the lower quartile of the observations in a moving time window (e.g. 30 days) over the whole time series. This method 15 16 was chosen as it is robust to the number of observations (i.e. it can be used for in situ as well as discrete measurements) although other more sophisticated background selection algorithms 17 18 exist and could be used instead (e.g. Ruckstuhl et al. 2012; Giostra et al. 2011). The selected 19 observations are the approximation for the contribution to the mixing ratio without any influence from fluxes in the nested domain, thus, the corresponding elements of the prior 20 modelled mixing ratio, H<sup>nest</sup>f<sup>nest</sup>, should be zero. However, this is not always the case, 21 22 therefore, we also subtract the prior simulated mixing ratio from the selected observations so that there is zero contribution from fluxes inside the domain in the background mixing ratio. 23 24 To avoid overestimating the contribution from inside the domain and, hence, underestimating 25 the background mixing ratio, we also select the lower quartile of the prior simulated mixing ratios in a moving time window. Lastly, we calculate a running average of the background 26 27 mixing ratios using a time window of 90 days, which is then linearly interpolated to the timestamp of the observations. This is done for each receptor. Similar methods for the 28 29 background calculation have been used previously for cases where no reliable global model 30 estimate of the mixing ratio was available (e.g. Stohl et al. 2010).

31

#### 32 **2.2** Variable resolution grid

1 To reduce the number of variables in the inversion problem we aggregate grid cells where 2 there is little constraint from the atmospheric observations. In this way, we define a new vector of the fluxes to be optimized,  $\mathbf{f}_{vg}^{nest}$  and transport matrix,  $\mathbf{H}_{vg}^{nest}$  which are on a grid of 3 4 variable resolution (vg = variable grid). The aggregation algorithm is based on time-averaged 5 SRRs optionally convolved with the prior flux estimate. The variable grid is set-up starting 6 with a coarse grid, which is refined in a specified number of steps following the method of 7 Stohl et al. (2009). For example, starting with a coarse resolution of  $4^{\circ} \times 4^{\circ}$  the grid may be refined in two steps to resolutions of  $2^{\circ} \times 2^{\circ}$  and  $1^{\circ} \times 1^{\circ}$ . The refinement is made so that the flux 8 9 sensitivity (optionally multiplied by the prior flux) in each grid cell at its final resolution (e.g.  $1^{\circ} \times 1^{\circ}$ ,  $2^{\circ} \times 2^{\circ}$  and  $4^{\circ} \times 4^{\circ}$ ) is above a given threshold. It is also optional whether or not to make 10 the grid refinement over water bodies and ice so that grid cells with a water/ice area of 99% or 11 12 more are not refined further reflecting cases where the water/ice surface fluxes are either 13 smaller, more homogeneous and/or more certain than the land surface fluxes. For determining which grid cells are land/water/ice we use the International Geosphere Biosphere Programme 14 15 land-cover dataset (IGBP-DIS) (Belward et al. 1999).

16

To convert from the fine to the variable grid, we define a projection operator  $\Gamma_{(W \times K)}$  where *K* is the number of grid cells in the nested domain at the original resolution and, *W*, the number at variable resolution. Each row of  $\Gamma$  corresponds to a cell in the variable grid, and is a summation vector on the fine grid. The row vectors,  $\lambda_i$ , of  $\Gamma$  are orthogonal thus  $\lambda_i \lambda_j^T = 0$  for i  $\neq j$  (since each fine grid cell can only belong to one variable grid cell). The flux vector,  $\mathbf{f}^{\text{nest}}$ and the matrix,  $\mathbf{H}^{\text{nest}}$  on the variable grid can be found according to:

23 
$$\mathbf{f}_{vg}^{\text{nest}} = \Gamma \mathbf{f}^{\text{nest}}$$
 and  $\mathbf{H}_{vg}^{\text{nest}} = \mathbf{H}^{\text{nest}} \Gamma^{\text{T}}$  (5)

where "T" indicates the matrix transpose. The fluxes in  $\mathbf{f}^{nest}$  are weighted by the ratio of the area of fine grid to the variable grid into which it is aggregated. The forward model on the variable resolution grid can thus be written as:

27 
$$\mathbf{y}^{\text{mod}} = \mathbf{H}_{vg}^{\text{nest}} \mathbf{f}_{vg}^{\text{nest}} + \mathbf{H}^{\text{out}} \mathbf{f}^{\text{out}} + \mathbf{H}^{\text{bg}} \mathbf{y}^{\text{bg}} + \boldsymbol{\varepsilon}^{\text{agg}}$$
 (6)

where  $\varepsilon^{agg}_{(M \times 1)}$  is the model representation error from having reduced the resolution of the model (for a schematic for the forward model see Fig. 1). It is also known as the aggregation error and has been described by Trampert and Snieder (1996), Kaminski et al. (2001), and Thompson et al. (2011). We describe the calculation of the aggregation error in section 2.5.

33 2.3 Aggregation of the background mixing ratios

1 The contribution of fluxes outside the domain to the change in mixing ratios at the receptor points (i.e.  $\mathbf{H}^{out}\mathbf{f}^{out}$ ) can be added to the initial mixing ratio ( $\mathbf{H}^{bg}\mathbf{y}^{bg}$ ). The contribution to the 2 modelled mixing ratio (i.e. y<sup>mod</sup>), which is not accounted for by the SRRs and fluxes inside 3 the domain (i.e. the background mixing ratio), is then defined by a new matrix,  $\mathbf{M}_{cg(M \times R)}$ , on a 4 5 coarse grid (cg), which has rows corresponding to M observations and columns corresponding to R grid cells or latitudinal bands. When the initial mixing ratio is calculated using the 6 sensitivity matrix, H<sup>bg</sup> and mixing ratio fields, y<sup>bg</sup> from a global Eulerian transport model, 7 then  $M_{cg}$  is defined as: 8

9 
$$\mathbf{M}_{cg} = \left(\mathbf{H}^{out} \circ \mathbf{F}^{out} + \mathbf{H}^{bg} \circ \mathbf{Y}^{bg}\right) \Gamma_{cg}^{\mathrm{T}}$$
 (7)

10 where  $\circ$  indicates the Hadamard matrix product,  $\mathbf{F}^{out}_{(M \times P)}$  has *M* rows of  $(\mathbf{f}^{out})^{\mathrm{T}}$ , and  $\mathbf{Y}^{bg}_{(M \times P)}$ 11 has *M* rows of  $(\mathbf{y}^{bg})^{\mathrm{T}}$ . The matrix,  $\Gamma_{cg(R \times P)}$ , is a projection operator from the Eulerian model 12 resolution to a coarse resolution of *R* grid cells (note  $\Gamma_{cg} \neq \Gamma$ ). Noteworthy, is that the matrix 13 multiplication  $\mathbf{H}^{out} \circ \mathbf{F}^{out}$  is made using the original resolution of the LPDM and fluxes and 14 that the conversion to the coarse grid is performed only on the mixing ratios, thus avoiding an 15 aggregation error in this component. When the background is calculated using the 16 observations themselves, then  $\mathbf{M}_{cg}$  is defined as:

17 
$$\mathbf{M}_{cg} = \operatorname{diag}(\mathbf{y}^{bg})\Gamma_{bg}^{\mathrm{T}}$$
 (8)

18 where  $\Gamma_{bg(R \times M)}$  is an operator to map the background mixing ratios to a matrix where the 19 background for each measurement is allocated to one of *R* latitudinal bands. Note that the 20 contribution from grid cells outside the domain is not explicitly included as it is assumed that 21 this contribution is incorporated into the definition of  $\mathbf{y}^{bg}$  when it is calculated from the 22 observations (see section 2.1.2).

23

For both methods, the columns of  $M_{cg}$  correspond to the mixing ratios in each of the *R* coarse grid cells (or latitudinal bands when using the observation-based method) such that the sum of each row gives the total background mixing ratio for each measurement (note that for the observation-based method there is only one non-zero entry in each row). The spatial distribution of the contribution to the background mixing ratio (dimension *R*) is maintained as it is these contributions that are optimized in the inversion.

30

31 We then define a new transport operator,  $\mathbf{H}_{(M \times N)}$  by concatenating the matrices  $\mathbf{H}^{\text{nest}}_{\mathbf{vg}}$  and  $\mathbf{M}_{cg}$ . 32 Similarly, we define the state vector,  $\mathbf{x}_{(N \times 1)}$  by concatenating  $\mathbf{f}^{\text{nest}}_{\mathbf{vg}(W \times 1)}$  (the flux variables 33 inside the nested domain) and  $\mathbf{a}_{cg(Rx1)}$  (scalars of the mixing ratios in the columns of  $\mathbf{M}_{cg}$ ):

1 
$$\mathbf{H} = \begin{bmatrix} \mathbf{H}_{vg}^{\text{nest}} \mathbf{M}_{cg} \end{bmatrix}$$
 and  $\mathbf{x} = \begin{bmatrix} \mathbf{f}_{vg}^{\text{nest}} \mathbf{a}_{cg} \end{bmatrix}$  (9)

2 The prior value of  $\mathbf{a}_{cg}(i)$  (i = 1 to R) is 1. After inversion, the optimized values of  $\mathbf{a}_{cg}$ 3 determine the posterior background mixing ratios.

4

# 5 2.4 Optimization of the fluxes and background mixing ratios

6 The uncertainty in the initial mixing ratios and in the contribution to the mixing ratio from 7 fluxes outside the domain can be considerable. Therefore, we include this component in the 8 optimization problem. The prior state vector for optimization,  $\mathbf{x}_{b}$  thus contains variables for 9 the surface fluxes (on the variable resolution grid) and variables for the optimization of the 10 mixing ratios (on the coarse resolution grid defined by  $\Gamma_{cg}$ ).

11

Based on Bayes' theorem, the most probable solution for  $\mathbf{x}$  is the one that minimises the difference between the observed and modelled mixing ratios while also depending on the prior state variables,  $\mathbf{x}_{\mathbf{b}}$  and their uncertainties (for details on Bayes' theorem see e.g. Tarantola (2005)). Assuming that the uncertainties have a Gaussian probability density function (*pdf*) this can be described by the cost function:

17 
$$J(\mathbf{x}) = \frac{1}{2} (\mathbf{x} - \mathbf{x}_{\mathbf{b}})^{\mathrm{T}} \mathbf{B}^{-1} (\mathbf{x} - \mathbf{x}_{\mathbf{b}}) + \frac{1}{2} (\mathbf{H}\mathbf{x} - \mathbf{y}^{\mathrm{obs}})^{\mathrm{T}} \mathbf{R}^{-1} (\mathbf{H}\mathbf{x} - \mathbf{y}^{\mathrm{obs}})$$
(10)

where  $\mathbf{B}_{(N \times N)}$  is the prior flux error covariance matrix (see section 2.5),  $\mathbf{R}_{(M \times M)}$  is the 18 observation error covariance matrix (see section 2.6), and y<sup>obs</sup> is a vector of the observed 19 mixing ratios. There exist a number of methods to find the x for which Eq. 10 is at a 20 21 minimum; we use the approach of finding the first derivative of Eq. 10 and solving this for x. 22 By rearrangement, x can be found according to Eq. 11. Equation 11 has a number of 23 alternative formulations and the one we use is the most efficient when the number of 24 observations is smaller than the number of unknowns, since the size of the matrix to invert (**HBH**<sup>T</sup>+**R**) has dimensions of  $M \times M$ : 25

26 
$$\mathbf{x} = \mathbf{x}_{\mathbf{b}} + \mathbf{B}\mathbf{H}^{\mathrm{T}} (\mathbf{H}\mathbf{B}\mathbf{H}^{\mathrm{T}} + \mathbf{R})^{-1} (\mathbf{y}^{\mathrm{obs}} - \mathbf{H}\mathbf{x}_{\mathbf{b}})$$
 (11)

The inverse of  $(\mathbf{HBH}^{T}+\mathbf{R})$  is found by Cholesky factorization (using the DPOTRF and DPOTRI routines from the LAPACK library). The corresponding posterior error covariance matrix,  $\mathbf{A}_{(N \times N)}$  is the inverse of the second derivative of the cost function, J'':

30 
$$\mathbf{A} = (J'')^{-1} = \mathbf{B} - \mathbf{B}\mathbf{H}^{\mathrm{T}} (\mathbf{H}\mathbf{B}\mathbf{H}^{\mathrm{T}} + \mathbf{R})^{-1} \mathbf{H}\mathbf{B}$$
(12)

#### **1 2.5 Prior error covariance matrix**

Errors in the prior flux estimates are correlated in space and time owing to correlations in the biogeochemistry model, upscaling model, or anthropogenic emission inventory that was used to produce these estimates. Most often, there is little known about the true temporal and spatial error correlation patterns. Here we define the spatial error correlation for the fluxes as an exponential decay over distance, such that each element in the spatial correlation matrix,  $C_s$  is:

8 
$$c_{\mathrm{S}(i,j)} = \exp\left(-\frac{d_{ij}}{k_{\mathrm{S}}}\right)$$
 (13)

9 where  $d_{ii}$  is the distance between grid cells i and j in a given timestep and  $k_{\rm S}$  is the spatial 10 correlation scale length on land or ocean (we assume that fluxes on land and ocean are not 11 correlated with one another). The temporal error correlation matrix,  $C_T$  is described similarly using the time difference between grid cells in different timesteps. The full temporal and 12 13 spatial correlation matrix, C is given by the Kronecker product:  $C_T \otimes C_S$ . The error covariance matrix for the fluxes,  $\mathbf{B}^{\mathbf{flux}}_{\mathbf{vg}(W \times W)}$  is the matrix product of correlation pattern, C and the error 14 covariance of the prior fluxes,  $\sigma\sigma^{T}$  where  $\sigma$  is a vector of the flux errors. We calculate the 15 16 error on the flux in each grid cell (on the fine grid) as a fraction of the maximum value out of that grid cell and the 8 surrounding ones. Finally, the  $\mathbf{B}^{\mathbf{flux}}_{\mathbf{yg}}$  matrix is scaled so that the square 17 root of its sum is consistent with a total error value assigned for the whole domain. This error 18 19 estimate may be from e.g. comparisons of independent biogeochemistry modelled fluxes or 20 flux inventories. The correlation matrix could be calculated for the fine grid and converted to the variable grid using the prolongation operator as  $\Gamma B^{flux} \Gamma^T$ . However, we calculate  $B^{flux}_{vg}$ 21 directly for the variable grid (dimensions  $W \times W$ ) as the multiplication step  $\mathbf{B}^{\mathbf{flux}} \mathbf{\Gamma}^{\mathrm{T}}$  is very slow 22 if K is large and/or if there are many timesteps. In addition,  $\mathbf{B}^{\mathbf{flux}}_{(K \times K)}$  is calculated for the fine 23 grid for a single time step only, as it is needed in the calculation of the aggregation error (see 24 25 section 2.6) and for the optimization of the posterior fluxes back to the fine grid (section 2.8). 26 We assume that the errors for the scalars of the background mixing ratios (i.e.  $a_{cg}$ ) are uncorrelated and have a fixed prior value (e.g. 1%). The error variance for these scalars is 27 appended to  $\mathbf{B}^{\mathbf{flux}}_{\mathbf{vg}}$  to give  $\mathbf{B}_{(N \times N)}$ . 28

29

#### 30 2.6 Aggregation error

1 The aggregation incurred by reducing the spatial resolution of the model can be calculated by 2 projecting the loss of information in the state space into the observation space (Kaminski et al. 3 2001). The full aggregation error covariance matrix  $\mathbf{E}^{agg}_{(M \le M)}$  is given by:

4 
$$\mathbf{E}^{\text{agg}} = \mathbf{H} \Gamma_{\mathbf{B}} \mathbf{B}^{\text{flux}} \Gamma_{\mathbf{a}}^{\mathrm{T}} \mathbf{H}^{\mathrm{T}}$$
 (14)

5 where  $\Gamma_{-}$  is the projection of the loss of information in the variable grid compared to the fine 6 grid. The matrix  $\Gamma_{-}$  can be calculated simply from the rows vectors  $\lambda_{i}$  of the projection 7 operator  $\Gamma_{-}$  which are weighted by the square root of the row sum so as to have unit length:

8 
$$\Gamma_{-} = \mathbf{I} - \sum_{i=1}^{W} \lambda_i \lambda_i^T$$
 (15)

9 where **I** is the identity matrix. As  $\lambda_i \lambda_i^T$  is a matrix of size  $P \times P$ , where *P* can be on the order 10 of 10,000 to 100,000, it is not calculated directly but rather **H** $\Gamma$ . as follows:

11 
$$\mathbf{H}\Gamma_{-} = \mathbf{H} - \sum_{i=1}^{W} \mathbf{H}\lambda_{i}\lambda_{i}^{\mathrm{T}}$$
(16)

12

#### 13 **2.7 Observation error covariance matrix**

14 The errors in the observation space incorporate measurement as well as model transport and 15 representation errors. For the measurement errors, we use values of the measurement 16 repeatability as given by the data providers. The measurement errors can be given as a single 17 value or for each observation, in which case it is read from the observation files. Transport 18 errors are extremely difficult to quantify and depend not only on the model but also on the 19 input data, resolution and location. Therefore, we do not quantify the full transport error, but 20 only the part of it that can be estimated from the model FLEXPART, i.e. the stochastic 21 uncertainty, which arises by the representation of transport with a limited number of particles 22 (see Stohl et al. (2005)). The stochastic error, however, is likely to be much smaller than the 23 full transport error. It is possible, however, to include an additional estimate of the transport 24 error into Eq. 17, if this information were available. The error in the modelled mixing ratio is 25 calculated using the stochastic uncertainty in the same way that the mixing ratios themselves 26 are calculated. We consider two types of representation error: observation representation error 27 and model aggregation error (discussed above). The observation representation error is 28 calculated from the standard deviation of all measurements available in a user-specified 29 measurement averaging time interval, based on the idea that if the measurements are 30 fluctuating strongly within that interval then their mean value is associated with higher 31 uncertainty than if the measurements are steady (e.g. Bergamaschi et al. 2010). If only one

measurement is available during this interval, then a user-defined minimum error is used instead. The measurement and transport errors are assumed to be uncorrelated. Although this is a common assumption, correlations likely exist between e.g. hourly observations owing to errors in the modelled boundary layer height and wind fields, which could lead to temporal correlations. However, in the current version of FLEXINVERT, we do not account for these correlations, hence, we define a diagonal matrix with elements equal to the quadratic sum of the measurement, transport model and measurement representation errors:

8 
$$\sigma^2 = \sigma_{meas}^2 + \sigma_{repr}^2 + \sigma_{repr}^2$$
(17)

9 Another assumption that is made is that the observed – modelled mixing ratio residuals have a 10 Gaussian distribution (Eq. 10 is based on this assumption). Therefore, in cases where the 11 distribution is highly skewed, observations corresponding to the tail of the distribution will 12 have a strong influence on the result of the inversion. FLEXINVERT does not include any 13 component to deal with skewed distributions; however, the influence of observations in the 14 tail of the distribution may be reduced by increasing their uncertainty. For more details about 15 dealing with skewed distributions we refer the reader to Stohl et al. (2009).

16

17 The observation error covariance matrix,  $\mathbf{R}_{(M \times M)}$  is the sum of this diagonal matrix plus the 18 aggregation error covariance matrix,  $\mathbf{E}^{agg}$ .

19

### 20 **2.8 Optimization of the fluxes to fine resolution**

The optimal solution of the fluxes,  $\mathbf{f}^{nest^*}_{vg}$  is found for the variable grid according to Eq. 11 21 22 and the corresponding posterior error covariance matrix, A is found according to Eq. 12. 23 However, it is not possible to directly apply the inverse of the projection operator to retrieve the optimal emissions at fine resolution since the operation from the variable to the fine 24 25 resolution is ambiguous; there is insufficient information to redefine the fluxes at fine resolution. To find the optimal emissions at fine resolution,  $\mathbf{f}^{\mathbf{nest}^*}_{(K \times 1)}$ , we use an adaptation of 26 27 the method of Wu et al. (2011). This method involves a second Bayesian optimization step, 28 which uses the prior information about the distribution of the fluxes within each grid cell on 29 the variable resolution grid:

30 
$$\mathbf{f}^{\text{nest}*} = \mathbf{f}_{b}^{\text{nest}} + \mathbf{B}_{naw}^{\text{flux}} \Gamma^{\text{T}}_{\text{unit}} \left( \Gamma \mathbf{B}^{\text{flux}} \Gamma^{\text{T}} + \mathbf{A}^{\text{flux}} \right)^{-1} \left( \mathbf{f}_{\text{vg}}^{\text{nest}*} - \Gamma \mathbf{f}_{b}^{\text{nest}} \right)$$
 (18)

31 (see Appendix A for the derivation of Eq. 18 and 19). Since we only optimize the fluxes, i.e. 32  $f^{nest^*}$ , the matrices  $B^{flux}$  and  $A^{flux}$  represent only the parts of the error covariance matrices 33 corresponding to flux errors. We have introduced a new error covariance matrix,  $B^{flux}_{naw}$ ,

which is the non area-weighted (naw) version of  $\mathbf{B}^{flux}$ , i.e. calculated using the flux errors not 1 2 weighted by the ratio of the grid cell areas on the fine and coarse grid. Also, we have 3 introduced  $\Gamma_{unit}$ , which is equivalent to  $\Gamma$  but with each row vector normalized by the row 4 sum so that they have unit length. Our method departs from Wu et al. (2011) in that for the 5 error in posterior state vector on the variable grid we use the error covariance of the posterior solution on the variable grid A, rather than a Dirac distribution. The inverse of 6  $(\Gamma B^{flux}\Gamma^T + A^{flux})$  is found by Singular Value Decomposition (SVD) using the DGESDD 7 routine from the LAPACK library. We find the posterior error covariance matrix  $A^{flux^*}_{(K \times K)}$ 8 9 also for the fine resolution fluxes according to:

10 
$$\mathbf{A}^{\mathbf{flux}^*} = \left(\mathbf{B}^{\mathbf{flux}-1}_{\mathbf{naw}} + \Gamma^{\mathrm{T}}_{\mathbf{unit}} \mathbf{A}^{\mathbf{flux}-1} \Gamma_{\mathbf{unit}}\right)^{-1}$$
(19)

11 The inverse of the matrices  $\mathbf{B}^{\mathbf{flux}}_{\mathbf{naw}}$ ,  $\mathbf{A}^{\mathbf{flux}}$ , and  $(\mathbf{B}^{\mathbf{flux}}_{\mathbf{naw}})^{-1} + \Gamma_{\mathbf{unit}}^{\mathbf{T}} \mathbf{A}^{\mathbf{flux}} - \Gamma_{\mathbf{unit}}$ ) are also found by 12 SVD, which can also be used for matrices that are non-positive definite. This optimization to 13 the fine grid should be carefully evaluated if used. Alternatively, we also include a simple 14 mapping back to the fine grid by distributing the flux in a coarse grid to the corresponding 15 fine grid cells based on the prior relative flux distribution at fine resolution.

16

#### 17 **2.9 Inequality constraints**

18 For some atmospheric species, there are physical restrictions on the values of the fluxes. For 19 example, for anthropogenic species, such as halocarbons and SF<sub>6</sub>, the fluxes can only be positive over land, while there are no appreciable fluxes (positive or negative) over ocean. 20 21 Using an inequality constraint in the cost function Eq. 10 would mean that the first derivative 22 would be undefined. Therefore, we adopt a "truncated Gaussian" approach following Thacker 23 (2007), in which inequality constraints are applied by treating these as error-free observations. 24 The inequality constraints are applied to the posterior fluxes derived previously (i.e. with no 25 inequality constraint). This is described by the following equation, which is analogous to Eq. 11: 26

27 
$$\mathbf{f}^{\text{nest}^{**}} = \mathbf{f}^{\text{nest}^{*}} + \mathbf{A}^{\text{flux}} \mathbf{P}^{\text{T}} \left( \mathbf{P} \mathbf{A}^{\text{flux}} \mathbf{P}^{\text{T}} \right)^{-1} \left( \mathbf{c} - \mathbf{P} \mathbf{f}^{\text{nest}^{*}} \right)$$
(20)

where  $\mathbf{P}_{(Q \times K)}$  is a matrix operator to select the *Q* variables that violate the inequality constraint and **c** is a vector of the inequality constraints of length *Q*. The inequality constraint does not only affect the grid cells with negative values but there is also some adjustment to other cells according to the correlations described by the posterior error covariance matrix,  $\mathbf{A}^{\mathbf{flux}}$ . The posterior error covariance matrix, however, is unchanged as the observation error covariance matrix in this case is zero. To apply the inequality constraint requires running a second code,
 which uses the output of FLEXINVERT.

3

4 A brief description of the software, its inputs, and outputs, is provided in Appendix B.

5

# 6 **3.** Case study: estimation of CH<sub>4</sub> fluxes in Europe

7 We provide a case study using FLEXINVERT for the estimation of methane (CH<sub>4</sub>) fluxes in 8 Europe. Methane was chosen, as it is an important greenhouse gas with an atmospheric 9 lifetime of approximately 10 years (Denman et al. 2007) and since its loss in the troposphere is principally by reaction with the OH radical, which can be approximated as a linear process. 10 The fluxes of CH<sub>4</sub> are mostly positive (i.e. from the surface to the atmosphere) although small 11 12 negative fluxes of CH<sub>4</sub> by oxidation in soils are also possible (Ridgwell et al. 1999). Europe 13 was chosen as it is reasonably well covered by observations, both discrete air sampling and 14 in-situ measurements. The important sources of CH<sub>4</sub> in Europe are mostly anthropogenic, 15 namely agriculture, landfills, and oil and gas exploitation (including fugitive emissions as 16 well as those from incomplete combustion). Natural sources of CH<sub>4</sub> are less important in 17 Europe and principally wetlands and mostly in the higher latitudes. In this case study, we estimate the total fluxes of CH<sub>4</sub> in the nested domain from 30°N to 70°N and 15°W to 45°E at 18 19 monthly resolution for the year 2011.

20

### 21 **3.1 Inversion set-up**

### 22 3.1.1 FLEXPART runs

23 FLEXPART (version 8.1) (Stohl et al. 1998; Stohl et al. 2005) was used to generate the SRRs 24 by running 10-day backwards mode simulations from each of the receptors (i.e. the 25 observation sites). FLEXPART was run at  $1.0^{\circ} \times 1.0^{\circ}$  resolution with meteorological analyses 26 from the European Centre for Medium-Range Weather Forecasts (ECMWF). Backwards 27 ("retro-plume") simulations were made by releasing 20,000 virtual particles in 3-hourly 28 intervals and the SRRs (or equivalently emission sensitivities) were saved as 24-hour 29 averages. Particles were released from the sampling inlet height at each observation site (see 30 Table 3). The loss of CH<sub>4</sub> by reaction with the OH radical was also included in the backwards 31 simulations even though the loss is very small over a 10-day period. Figure 2a shows the 32 combined total emission sensitivity for all observation sites in 2011. The total emission 33 sensitivity was used to determine the variable resolution grid (Fig. 2b) with grid cells ranging 34 in size from  $1.0^{\circ} \times 1.0^{\circ}$  to  $4.0^{\circ} \times 4.0^{\circ}$ .

### 2 **3.1.2.** Observations

3 We used measurements of CH<sub>4</sub> from approximately weekly samples in the National Oceanic 4 and Atmospheric Administration Global Monitoring Division (NOAA GMD) Carbon Cycle 5 and Greenhouse Gases (CCGG) network. These measurements are made using Gas 6 Chromatographs fitted with Flame Ionization Detectors (GC-FID). In addition, we used data 7 from a number of in-situ measurement sites. These included in-situ GC-FID instruments operated by the Umweltbundesamt (UBA), the Institute for Atmospheric Sciences and 8 9 Climate (ISAC) and the Advanced Global Gases Experiment (AGAGE) as well as in-situ Cavity Ring Down Spectrometers (CRDS) operated by EMPA and the Finnish 10 Meteorological Institute (FMI). All measurements were reported as dry-air mole fractions in 11 12 parts-per-billion (abbreviated as ppb) on the NOAA2004 calibration scale, except AGAGE 13 data, which were reported on the Tohoku University scale but were converted to the 14 NOAA2004 scale using a conversion factor of 1.0003 (see Table 3).

15

In-situ measurements were assimilated as averages of the afternoon observations (12:00 to 18:00) at low altitude sites and as averages of night-time observations at mountain sites (00:00 to 06:00) and the corresponding FLEXPART SRRs were selected and averaged in the same way. Discrete measurements were assimilated as available and matched with the closest available 3-hourly SRR to the sampling time. The measurement error was defined as 5 ppb based on the repeatability of the measurements and, in the case of the in-situ data, the representation error was defined as the standard deviation of the afternoon observations.

23

### 24 **3.1.3.** Prior fluxes and initial mixing ratios

25 The prior flux was composed from estimates of anthropogenic and natural emissions from a number of different models and inventories (see Table 4 for details) and the total global 26 source amounted to 610 TgCH<sub>4</sub> y<sup>-1</sup>. Methane fluxes were resolved monthly in the wetland, 27 ocean, termite, wild animal, soil, and biomass burning estimates, while the anthropogenic and 28 29 geological flux estimates were only resolved annually. For the anthropogenic and biomass 30 burning sources, the 2010 estimates were used, as no estimates were available for 2011. For 31 the remaining natural sources, climatological estimates were used. All fluxes were used at a 32 spatial resolution of  $1.0^{\circ} \times 1.0^{\circ}$ .

Prior flux error covariance matrix,  $\mathbf{B}^{\text{flux}}$ , was calculated as described in section 2.5 using a spatial correlation length of 500 km,  $k_s = 500$ , and a temporal correlation length of 90 days,  $k_T$ = 90. For the calculation of the flux errors we used a fraction of 0.5 of the maximum flux out of the cell of interest and the 8 surrounding ones.

5

6 The background mixing ratios may be estimated either from the observations themselves or 7 by coupling FLEXPART to a global model (see sections 2.1.2 and 2.3). For the latter method, 8 FLEXPART calculates the sensitivity to the mixing ratio at the termination point of the 9 virtual particles. These sensitivities were coupled to daily 3D fields of CH<sub>4</sub> mixing ratios from 10 the atmospheric chemistry transport model, TM5, in order to calculate the initial mixing ratios. The TM5 model was run at  $6.0^{\circ} \times 4.0^{\circ}$  horizontal resolution with 25 eta pressure levels using 11 12 pre-optimized fields of CH<sub>4</sub> fluxes (Bergamaschi et al. 2010). Atmospheric loss of CH<sub>4</sub> by 13 reaction with OH radicals is calculated in TM5 using monthly fields of OH concentration 14 (Bergamaschi et al. 2005) resulting in mean atmospheric lifetime of CH<sub>4</sub> of 10.1 years, which 15 is close to the IPCC recommended value of 9.7 (±20%) years (Denman et al. 2007). The 16 initial mixing ratios were added to the change in mixing ratios from fluxes outside the domain, together forming the background mixing ratio matrix,  $\mathbf{M}_{cr(M \times R)}$ . The background was 17 optimized at a resolution of  $30^{\circ} \times 15^{\circ}$  (longitude by latitude) over the global domain (i.e. R =18 19 144). The uncertainty in the scalars of the background mixing ratio was set to 0.2% equivalent 20 to approximately 4 ppb.

21

## 22 **3.2** Sensitivity tests

We performed six inversions to test the sensitivity of the posterior fluxes and error reduction to the spatial correlation scale length (S1 to S3), to the optimization of the background (S4), to the filtering and averaging of the observations (S5), as well as to the background estimation method (S6). The tests are summarized in Table 5.

27

### 28 **3.3 Results**

The inversions were run on a Linux Ubuntu machine with 62 GB memory. The maximum and
mean memory usage was 18 and 6.4 GB, respectively, and each inversion took approximately
1.8 days to complete.

32

Figure 3 shows the observed CH<sub>4</sub> mixing ratios at in-situ measurement sites compared with
 those simulated with the TM5 model and FLEXPART using the prior and posterior fluxes

1 from test S1. At high altitude sites, namely, CMN, JFJ, and SSL, the global model tends to underestimate the synoptic variability largely due to the coarse resolution. This can be 2 3 quantified by the Normalized Standard Deviation (NSD) (i.e. the SD of the model normalized 4 by the SD of the observations), which for TM5 was 0.46, 0.81 and 0.71, compared with 0.81, 5 0.75, and 1.07 for FLEXPART, for the three sites respectively. On the other hand, TM5 6 overestimated the variability at MHD, a coastal site, with a NSD of 2.53 compared with 0.97 7 in FLEXPART. Again, this is likely to be due to the coarse resolution in TM5, which cannot 8 accurately resolve the location of MHD and overestimates the influences of land fluxes at this 9 site.

10

To examine the differences between the two methods of estimating the background mixing 11 12 ratios, we compare the background determined in test S1 (model-based method) and test S6 13 (observation-based method). The results are shown in Fig. 4 at the in-situ measurement sites. 14 The two methods compare reasonably well with one another with the mean difference 15 between the two backgrounds being between -7 and 4 ppb for the different sites. At MHD, 16 however, observation-based background is considerably lower than the model-based one (difference of -11 ppb). This departure is caused by an overestimation of the prior 17 18 contribution to the mixing ratio from fluxes inside the nested domain, and since this is 19 subtracted from the observations that have been identified as being representative of the 20 background, this leads to an overall too low background estimate at this site (see section 2.1.2 21 for details).

22

23 The model performance at the measurement sites for test S1, a priori and a posteriori, is summarized in Table 6, which compares the correlation coefficient (R), Root Mean Square 24 25 Error (RMSE) and NSD of the simulated mixing ratios versus the observations. As expected, 26 the mixing ratios a posteriori agree better with observations. To assess the assumptions made about the uncertainties and error correlation scales used in **B** and **R**, we look at  $\chi^2$ , which has 27 28 the value of the cost function at the optimum (equivalently the weighted sum of squares divided by the number of observations). Ideally,  $\chi^2$  would be equal to 1 indicating that the 29 posterior solution is within the limits of the prescribed uncertainties. In actual fact, the  $\chi^2$ 30 values are larger than 1.  $\chi^2$  increased with increasing spatial correlation scale length with 31 values of 2.24, 2.56, and 2.97 with  $k_s$  of 200, 300 and 500 km, respectively, which is as 32 33 expected since a longer correlation scale length corresponds to fewer degrees of freedom. Using all observations, as in test S5, resulted in a  $\chi^2$  of 2.05, the lowest value, as this also 34

resulted in larger SDs over the averaging interval (1 day) and, hence, larger uncertainties in
 the observation space.

3

4 The posterior fluxes and the flux increments (posterior minus prior fluxes) for the six 5 sensitivity tests are shown in Fig. 5. The posterior fluxes and flux increments from tests S1 to 6 S5 are quite similar, however, on close inspection there are a few notable differences. 7 Decreasing the spatial correlation scale length from 500 to 200 km (tests S1 to S3) resulted in 8 a more heterogeneous pattern of flux increments as the greater degrees of freedom allowed 9 smaller spatial scales to be modified in the inversion. Although, overall the patterns of flux increments from all tests were consistent with lower emissions, relative to the prior, over 10 11 France, Italy and the UK, and higher emissions over Austria, Hungary, and eastern Europe. 12 When the background mixing ratios are also optimized (test S4) there is only a small change 13 with respect to test S1; namely, in S4 the emissions are slightly lower over the United 14 Kingdom, France and the Iberian Peninsular. Lower emissions are found as the background 15 mixing ratios have been increased (by approximately 0.2%) to minimise the observation – 16 model error and, hence, smaller increments were needed in the emissions. Furthermore, using 17 all observations (test S5), compared to only afternoon ones at low altitude sites and only 18 night-time ones at high altitude sites, made almost no difference to the posterior fluxes. Test 19 S6, which used the observation-based approach for the background estimation, differed the 20 most from the other tests. Notably, higher emissions, compared to the other inversions, were 21 found in France, Germany, the Czech Republic and the UK and, correspondingly, the flux 22 increments were more positive in these regions as well. This difference is a direct result of the 23 lower background mixing ratios estimated at a number of sites with the observation-based 24 method and highlights the challenge of obtaining robust background estimates.

25

Figure 6 shows the error reduction for the six sensitivity tests. The largest error reductions are found using  $k_s = 500$  km, i.e. in tests S1, S4, S5 and S6, for which the error reduction is almost identical. The error reductions in tests S2 and S3 are smaller and more limited to central Europe as compared to S1. Again, this is because increasing the correlation scale length results in fewer degrees of freedom for the inversion and effectively spreads the atmospheric information over a larger area.

32

Lastly, we compare the simulated mixing ratios using the a priori and a posteriori fluxes(from test S1) with observations at an independent site, i.e. one that was not included in the

1 inversion, Puy de Dôme, France (PUY). Figure 7 shows the observed, prior, posterior and 2 background mixing ratios at the time-stamp of the observations at PUY. Both the prior and 3 posterior mixing ratios overestimate the observed variability with NSD of 2.24 and 2.04, 4 respectively. This is most probably owing to both the topography (the station is located on a 5 volcanic cone, which represents a very abrupt change in topography) as well as the fact that 6 there are significant emissions in the prior around the station. A likely explanation is that 7 FLEXPART overestimates the BL height at PUY and thus overestimates the influence of 8 local emissions on this site. Despite the model transport errors at this site, using the posterior 9 fluxes improves the RMSE (23.1) and correlation coefficient (0.18) compared to the prior 10 (26.4 and 0.16, respectively).

11

### 12 **3.4. Discussion**

The results for the sensitivity tests S1 and S6, using the model and observation based background mixing ratios, respectively, highlight the challenge of robustly identifying the background and the influence that this has on the optimized fluxes (see Fig. 5). There are different problems associated with each method and warrant further discussion here.

17

First, using an optimized global model (in our case study, the chemistry-transport model, 18 19 TM5) to derive the prior background may lead to problems of circularity, i.e. if the 20 background is included in the optimization and the same observations used to constrain the 21 global model are also used in the Lagrangian inversion. If different sets of observations are 22 used then this is not a problem, however, if there is overlap then the prior information (about 23 the background) and the observations are no longer completely independent. The degree of 24 overlap should, however, be small since the background calculated for the Lagrangian model 25 is an extremely smoothed version of the global modelled mixing ratio fields, as the sensitivity 26 to the background even for a single measurement is distributed over large parts of at least one 27 hemisphere. Furthermore, in the global model, the background is constrained mainly by 28 measurements from outside the region of interest. The degree of circularity is minimized even 29 further if new observations are included in the Lagrangian inversion, which may also 30 encompass assimilating observations from the same sites but at higher temporal resolution in 31 the Lagrangian model if observations from no additional sites are available. In any case, the 32 model-based background should be from a pre-optimized model or optimized in the 33 Lagrangian inversion, as biases in the background will be propagated into biases in the 34 optimized fluxes.

2 Second, using a filtering of the observations to derive the background will also lead 3 circularity, i.e. if the same observations are also used to optimize the background in the 4 inversion, and this case should be avoided. When the observations are used to derive the 5 background, biases only arise in the detection of the background signal. The background 6 mixing ratio may fluctuate depending on the altitude and latitude of the air masses' origin. In 7 addition, if the site is in an area of strong local fluxes, a background signal may not detectable. 8 Analysing the modelled back trajectories in such cases may help determine if candidate 9 observations for the background calculation are likely to have been influenced by fluxes in the 10 domain or not. Furthermore, the observation-based method for determining the background is not appropriate for species such as CO<sub>2</sub>, which have a strong diurnal cycle and thus no 11 12 definable background.

13

14 We compare our posterior CH<sub>4</sub> flux estimates with those derived from independent inverse 15 models, specifically, with the results of a recent inverse model ensemble for CH<sub>4</sub> fluxes over 16 Europe from the NitroEurope project (Bergamaschi et al. 2014). This ensemble consisted of 17 four independent models, including two Eulerian and two Lagrangian ones. Although the time period covered by the ensemble (2006 to 2007) differs from our study (2011), the fluxes of 18 19 CH<sub>4</sub> in Europe are thought to have been fairly stable between both periods and, hence, the 20 differences are likely to represent differences in the model set-ups rather than only changes in 21 the fluxes. Table 6 compares the prior and posterior emission totals from this study with those 22 of Bergamaschi et al. (2014). Overall, the posterior fluxes from this study are within the range 23 from the ensemble, despite differences in the time period and the atmospheric observations 24 used. There is only one exception, i.e. BENELUX, where our estimate is 24% lower than the 25 lowest limit of the ensemble range. This may be due, at least in part, to real changes in 26 emissions. However, it may also be due to differing distributions of the posterior emissions 27 close to the boundaries of BENELUX with France and Germany, which considering the small 28 area of BENELUX, may become important in the calculation of the total emission. Another 29 contributing factor may also be that in the inversions in the Bergmaschi et al (2014) study, the 30 station, Cabauw (52.0°N, 4.9°E), in the Netherlands, was included (whereas it was not 31 included in our inversion), which likely also has a strong influence on the posterior fluxes in 32 BENELUX.

33

#### 34 5. Summary and conclusions

We have presented a new Bayesian inversion framework, FLEXINVERT, for the estimation 1 2 of surface to atmosphere fluxes of atmospheric species. The framework is based on Source -3 Receptor Relationships (SRR), which describe the relationship between changes in mixing 4 ratio at a receptor "point" and changes in fluxes, calculated by the Lagrangian Particle 5 Dispersion Model, FLEXPART. Fluxes may be optimized at any given temporal resolution 6 and on a nested grid of variable spatial resolution. The variable grid is determined using the 7 information of the integrated SRRs and has finer resolution where there is a strong 8 observational constraint and coarser resolution where there is a weak constraint. In this 9 framework, the background mixing ratio, i.e. the contribution to the mixing ratio at the receptors not accounted for by transport and fluxes inside the nested domain, is calculated by 10 11 coupling FLEXPART to the output of a global Eulerian model (or alternatively, in the case 12 that no such model output is available, it is calculated from the observations themselves). The 13 background mixing ratios are also included in the optimization problem.

14

15 We demonstrated the performance of FLEXINVERT in a case study estimating CH<sub>4</sub> fluxes 16 over Europe in 2011. The posterior fluxes were found to compare well to the results from an 17 independent inversion ensemble consisting of four different transport models and inversion frameworks. Although, we have only presented an example for CH<sub>4</sub>, FLEXINVERT can be 18 19 applied to any species for which atmospheric loss (if any) can be described as a linear process 20 such as radioactive decay, dry and wet deposition, and oxidative chemistry. Furthermore, the 21 framework can be used on continental, regional and even local scales with little or no 22 modification.

23

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10

### 1 Appendix

2

# 3 A. Optimization of the posterior fluxes to the fine grid resolution

4 Optimization of the posterior fluxes,  $\mathbf{f}^{\text{nest}^*}_{\text{vg}}$  on the variable resolution grid to the fine 5 resolution grid by applying Bayes' theorem. (Note that to simplify the notation we have used 6  $\mathbf{f} = \mathbf{f}^{\text{nest}^*}_{\text{vg}}$ ,  $\mathbf{f}_{b} = \mathbf{f}^{\text{nest}}$ , and  $\mathbf{f}^* = \mathbf{f}^{\text{nest}^*}$  i.e. the optimized fluxes on the fine grid).

7 
$$\rho(\mathbf{f}^*|\mathbf{f}) = \frac{\rho(\mathbf{f}|\mathbf{f}^*)\rho(\mathbf{f}^*)}{\rho(\mathbf{f})}$$
 (A1)

8 where  $\rho(\mathbf{f}^*|\mathbf{f})$  is the posterior *pdf* that  $\mathbf{f}^*$  lies in the interval  $(\mathbf{f}^*, \mathbf{f}^* + d\mathbf{f}^*)$  when  $\mathbf{f}$  (the 9 "observation") has a given value. Assuming a Gaussian *pdf*, and taking the natural logarithm 10 we can express  $\rho(\mathbf{f}|\mathbf{f}^*)$  as:

11 
$$-2\ln\rho(\mathbf{f}|\mathbf{f}^*) = (\mathbf{f} - \Gamma \mathbf{f}^*)^{\mathrm{T}} \mathbf{A}^{\mathbf{flux}-1} (\mathbf{f} - \Gamma \mathbf{f}^*)$$
(A2)

12 (where  $\mathbf{A}^{\mathbf{flux}}$  is the posterior error covariance matrix and  $\Gamma$  is the projection operator) and we 13 can express  $\rho(\mathbf{f}^*)$  as:

14 
$$-2\ln\rho(\mathbf{f}^*) = (\mathbf{f}^* - \mathbf{f}_b)^T \mathbf{B}^{\mathbf{flux}-1}(\mathbf{f}^* - \mathbf{f}_b)$$
(A3)

15 (where  $\mathbf{B}^{\mathbf{flux}}$  is the prior error covariance matrix on the fine grid) and by substituting Eq. 2 and 16 3 into Eq. 1 we derive the expression for  $\rho(\mathbf{f}^*|\mathbf{f})$ :

17 
$$-2\ln\rho(\mathbf{f}^*|\mathbf{f}) = (\mathbf{f}^* - \mathbf{f}_b)^{\mathrm{T}} \mathbf{B}^{\mathbf{flux}-1} (\mathbf{f}^* - \mathbf{f}_b) + (\mathbf{f} - \Gamma \mathbf{f}^*)^{\mathrm{T}} \mathbf{A}^{\mathbf{flux}-1} (\mathbf{f} - \Gamma \mathbf{f}^*)$$
(A4)

18 the cost function can be thus be defined as:

19 
$$J(\mathbf{f}^*) = \frac{1}{2} (\mathbf{f}^* - \mathbf{f}_b)^{\mathrm{T}} \mathbf{B}^{\mathrm{flux}-1} (\mathbf{f}^* - \mathbf{f}_b) + \frac{1}{2} (\mathbf{f} - \Gamma \mathbf{f}^*)^{\mathrm{T}} \mathbf{A}^{\mathrm{flux}-1} (\mathbf{f} - \Gamma \mathbf{f}^*)$$
(A5)

20 and the first derivative:

21 
$$J'(\mathbf{f}^*) = \mathbf{B}^{\mathbf{flux}-1}(\mathbf{f}^* - \mathbf{f}_{\mathbf{b}}) - \Gamma^{\mathrm{T}}\mathbf{A}^{\mathbf{flux}-1}(\mathbf{f} - \Gamma\mathbf{f}^*)$$
 (A6)

22 thus we can derive the expression for  $\mathbf{x}^*$  at the minimum:

23 
$$\mathbf{f}^* = \mathbf{f}_b + \mathbf{B}_{naw}^{flux} \Gamma_{unit}^T \left( \Gamma \mathbf{B}^{flux} \Gamma^T + \mathbf{A}^{flux} \right)^{-1} \left( \mathbf{f} - \Gamma \mathbf{f}_b \right)$$
 (A7)

24

#### **B. Description of the software**

### 26 **B.1 General description**

The code corresponding to the inversion framework described in this paper is called
FLEXINVERT and is available from the website: http://flexinvert.nilu.no under a GNU
General Public License. FLEXINVERT is coded in Fortran90 and has been tested with the

gfortran compiler and the Linux Ubuntu operating system and a makefile for gfortran is
 included. To run FLEXINVERT, the LAPACK and NetCDF libraries for Fortran must be
 installed. The current version of FLEXINVERT can be run directly with output from
 FLEXPART 9.2.

5

# 6 **B.2. Input data**

FLEXINVERT uses two definition files, the first specifies the paths, filenames, and other filerelated information (files.def), and the second specifies the settings for each inversion run,
such as the domain, dates, and uncertainties (control.def).

10

11 - FLEXPART files

12 FLEXINVERT looks for FLEXPART output files for each receptor in directories with the following naming convention: /STN NET/YYYYMM/ where STN NET is the name of the 13 14 receptor and must be the same as that given in the station list file and in the prefix of the 15 observation files. The FLEXPART files required are: header, grid time (and 16 grid initial when computing the background using global model output). It is 17 important to note that if the full 3D SRR fields are saved in the grid time files, the 18 reading of these files becomes considerably slower, therefore, it is recommended to save 19 only the surface layer of the SRR fields in the grid time files. However, if the 20 grid initial files are used, these need all layers. (An option for this grid time and 21 grid initial was added into FLEXPART 9.2). Also, note that FLEXINVERT expects 22 the stochastic errors to be written to the grid time files, if these are not written then a 23 minor modification is required in readgrid.f90.

24 - Station list file

This file specifies the receptors (where there are observations) to include in the inversion The default file has the following format: receptor name, latitude, longitude, altitude, observation type (either CM for continuous or FM for flask measurement) and a character string of up to 55 characters describing the receptor. However, only the receptor name and type are actually used in the inversion.

30

31 ID LAT LON ALT TYP STATIONNAME
32 STN\_NET XX.XX XXX.XX XXX XX Station Name, Country

- 33
- 34 Observations

1 The sub-routine readobs.f90 reads the observations from a separate ASCII file for each 2 receptor. Again, FLEXINVERT looks for the file prefix STN\_NET. The files contain 6 3 columns: year, month, day, hour, minute, mixing ratio, and optionally the measurement 4 error estimate.

5 - Prior fluxes

6 The sub-routine reademissions.f90 reads the prior fluxes (or equivalently prior 7 emissions) from a NetCDF file containing a 3D floating variable for the fluxes with 8 dimensions time, latitude and longitude, and the corresponding dimension variables. The 9 name of the floating point variable is specified in files.def by the variable emisname.

10 - Landcover file

FLEXINVERT uses high-resolution landcover data to specify areas of water when
determining the variable resolution grid. By default, FLEXINVERT uses the IGBP data,
which is included in the tar archive.

14 - Land-sea mask file

15 A land-sea mask file is used in FLEXINVERT to determine which grid-cells are on 16 land/ocean when calculating the covariance matrix. The default land-sea mask is at 17  $0.125^{\circ} \times 0.125^{\circ}$  resolution and is converted to the needed resolution automatically.

18 - 3D concentration fields

For the calculation of the initial mixing ratios from a global model, its 3D concentration fields are needed. FLEXINVERT includes routines for reading the output of the models LMDZ4 and TM5 in NetCDF format, which can be used as templates for reading data from other models.

23

### 24 B.3 Output data

25 At the end of an inversion run, FLEXINVERT writes the output into the following files:

26

• obsread.txt

ASCII file containing the observed, prior, posterior and background mixing ratios at the same timestamp as the observations. Note that if the background is not optimized, then the observed, prior and posterior mixing ratios are the difference from the background and the values in the column BGND\_POST are zero. The obsread.txt file has the following format:

33

34 ID DATE OBS PRIOR POST BGND\_PRIOR BGND\_POST ERROR

STN\_NET YYYYMMDDHH F11.3 F11.3 F11.3 F9.3 F9.3 F9.3

2

3 • modout.nc

NetCDF file containing floating point variables for the prior and posterior mixing ratios
(ypri and ypos, respectively) as well as the prior and posterior background mixing ratios
(bgpri and bgpos, respectively). These mixing ratios are computed using the fluxes at
the finest resolution and at the timestamp of the FLEXPART trajectories. The variables
have dimension of time and receptor.

9 • analysis.nc

NetCDF file containing floating point variables for the prior and posterior fluxes
 (emis\_prior and emis\_post, respectively) as well as the prior and posterior flux errors
 (error\_prior and error\_post, respectively). The variables are in dimensions of
 longitude, latitude and time and have units of kg m<sup>-2</sup> s<sup>-1</sup>.

14 • covb.nc

15 NetCDF file containing a floating point variable of the prior error covariance matrix 16 (covb) and units (kg m<sup>-3</sup> s<sup>-1</sup>)<sup>2</sup>. Note that the errors are scaled by the numerical scaling 17 factor defined in mod var.f90.

18 • cova.nc

19 As for covb.nc but containing the posterior error covariance matrix (cova).

20 • covr.nc

21 NetCDF file containing a floating point variable of the observation error covariance matrix 22 (covr) with units of mixing ratio squared (e.g. ppb<sup>2</sup>).

• nbox\_xy.nc

NetCDF file containing a floating point variable of the mapping of the fine to the variable
resolution grid with dimensions of the number of longitudinal by latitudinal grid cells.

26

For testing purposes, the following files are also written but in most cases will not berequired:

- 29
- 30 gain.nc
- 31 NetCDF file containing the Gain matrix,  $\mathbf{BH}^{\mathrm{T}}(\mathbf{HBH}^{\mathrm{T}} + \mathbf{R})^{-1}(\mathbf{y} \mathbf{H}\mathbf{x}_{\mathbf{b}})$ .
- 32 covbfin.nc

1	N	NetCDF file of the prior error covariance matrix on the fine grid (covbfin) with units
2	(1	$kgm^{-3} s^{-1})^2$ . Note that the errors are scaled by the numerical scaling factor.
3	• c	ovbfinaw.nc
4	А	As for covbfin.nc but containing the area-weighted errors (covbfinaw).
5	• c	ovagg.nc
6	N	NetCDF file of the aggregation errors in units of mixing ratio squared (e.g. ppb <sup>2</sup> ).
7	• g	rid_operator.nc
8	N	NetCDF file of the projection operator, $\Gamma$ , from the fine to the variable grid.
9	• g	rid_coarse.nc
10	N	NetCDF file of the projection operator, $\Gamma_{cg}$ , from the fine to the coarse grid.
11	• e	misflex.nc
12	N	NetCDF file of the prior emissions converted to the FLEXPART (i.e. fine) grid in units of
13	k	$g m^{-3} s^{-1}$ .
14	• k	nest_finobs.nc
15	N	$MetCDF$ file of the transport operator, $H^{nest}$ , for the fine grid and averaged to the
16	0	bservation averaging interval.
17	• k	nest_obs.nc
18	N	$HetCDF$ file of the transport operator, $H^{nest}$ , for the variable grid and averaged to the
19	0	bservation averaging interval.
20	• k	nest_trim.nc
21	N	$HetCDF$ file of the transport operator, $H^{nest}$ , for the variable grid with rows matching
22	0	bservations.
23	• k	out_obs.nc
24	N	JetCDF file of the initial mixing ratio contributions, H <sup>out</sup> y <sup>out</sup> , for the coarse grid and
25	a	veraged to the observation averaging interval.
26	• ir	nmr.nc
27	N	NetCDF file of the 3D initial mixing ratios from the global model (for option bgmethod =
28	2	only) interpolated to the FLEXPART resolution (first timestep only).
29	• at	rea_box.txt
30	A	ASCII file containing a vector of the variable grid cell areas $(m^2)$ .
31	• p	rior.txt
32	А	ASCII file containing the prior state vector, $\mathbf{x}_{\mathbf{b}}$
33	• p	osterior.txt

- 1 ASCII file containing the posterior state vector, **x**
- 2 bgscalars.txt
- 3 ASCII file containing the prior and posterior scalars of the background mixing ratios and
- 4 their errors with the format:
- 5 PRIOR POST PRIOR\_ERROR POST\_ERROR

```
6 F6.4 F6.4 F6.4 F6.4 F6.4
```

# 8 C. Applying inequality constraints

9 After running FLEXINVERT, a separate code may be run to apply inequality constraints. The
10 inequality constraint code is similarly written in Fortran90 and has been tested with the
11 gfortran compiler and the Linux operating system. To run the code, the LAPACK and
12 NetCDF libraries for Fortran must be installed. This code is available from the website:
13 http://flexinvert.nilu.no.

Variable	Dimension	Description
v <sup>mod</sup>	$M \times 1$	modelled atmospheric mixing ratios
x	$N \times 1$	state vector
Н	$M \!\!\times\! N$	complete atmospheric transport operator
f <sup>nest</sup>	$K \times 1$	fluxes inside the nested domain
H <sup>nest</sup>	$M \!\!\times\! K$	atmospheric transport operator for inside the nested domain
f <sup>out</sup>	$P \times 1$	fluxes outside the nested domain
H <sup>out</sup>	$M \!\!\times\! P$	atmospheric transport operator for outside the nested domain
$v^{bg}$	$P \times 1$	mixing ratios from the global model interpolated to the fine grid globally
$\mathbf{H}^{bg}$	$M \!\!\times\! P$	sensitivity to initial mixing ratios from the global model
Γ	$W \!\!\times\! K$	prolongation operator from the fine to the variable grid in the nested
		domain
f <sup>nest</sup> vg	$W \times 1$	fluxes inside the nested domain on the variable grid
H <sup>nest</sup> vg	$M \!\!\times\! W$	atmospheric transport operator for fluxes on the variable grid
M <sub>cg</sub>	$M \!\!\times\! R$	total background mixing ratios
$\Gamma_{cg}$	$R \times P$	prolongation operator from fine to coarse grid
$\Gamma_{bg}$	$R \!  imes \! M$	prolongation operator for observation-based background mixing ratios
acg	$R \times 1$	background mixing ratio scalars
Xb	$N \times 1$	prior state vector of fluxes and background mixing ratio scalars
σ	$W \times 1$	prior flux error vector
В	$N \!\!\times\! N$	prior error covariance matrix on the variable grid
<b>B</b> <sup>flux</sup> vg	$W\!\!\times\!W$	prior error covariance matrix for the fluxes on the variable grid
<b>B</b> <sup>flux</sup>	$K \!\!\times\! K$	prior error covariance matrix for fluxes on the fine grid
R	$M \!\!\times\! M$	observation error covariance matrix
С	$W\!\!\times\!W$	spatial and temporal correlation matrix
Cs	$W \!\!\times\! W$	spatial correlation matrix
CT	$1 \times 1$	temporal correlation matrix (trivial case when only 1 time step is used)
Α	$N \!\!\times\! N$	posterior error covariance matrix
A <sup>flux</sup>	$W \!\!\times\! W$	posterior error covariance matrix for fluxes on the variable grid
f <sup>nest*</sup>	$K \times 1$	posterior fluxes optimized on the fine grid
A <sup>flux*</sup>	$N \!\!\times\! N$	posterior error covariance matrix for fluxes on the fine grid
Р	$Q \times K$	operator to select variables that violate the inequality constraint
c	Q	a vector of inequality constraints

1 Table 1. Overview of the variables used in this manuscript

<sup>3</sup> Table 2. Overview of the dimension notation and their values in the case study (test S1).

Dimension	Description	Value
M	total number of observations	1602
N	total number of state variables (12 time steps)	13,896
Κ	number of fine resolution grid cells in 1 time step	2400
W	number of variable resolution grid cells in 1 time step	1158
Р	number of grid cells in global domain in 1 time step	2700
R	number of coarse grid cells for the background mixing ratio	144

Site ID	Organization	Latitude	Longitude	Altitude (m)	Туре	Description
PAL	FMI	68.0°N	24.1°E	572	СМ	Pallas, Finland
ICE	NOAA	63.3°N	20.3°W	127	FM	Heimay, Vestmannaeyjar, Iceland
BAL	NOAA	55.4°N	17.2°E	28	FM	Baltic Sea, Poland
MHD	AGAGE	53.3°N	9.9°W	40	СМ	Mace Head, Ireland
OXK	NOAA	50.0°N	11.8°E	1185	FM	Ochsenkopf, Germany
SSL	UBA	47.9°N	7.9°E	1213	СМ	Schauinsland, Germany
HPB	NOAA	47.8°N	11.0°E	990	FM	Hohenpeissenberg, Germany
HUN	NOAA	47.0°N	16.7°E	344	FM	Hegyhatsal, Hungary
JFJ	EMPA	46.6°N	8.0°E	3590	СМ	Jungfraujoch, Switzerland
PUY <sup>a</sup>	NOAA	45.8°N	3.0°E	1475	FM	Puy de Dôme, France
BSC	NOAA	44.2°N	28.7°E	3	FM	Black Sea, Constant, Romania
CMN	ISAC	44.2°N	10.7°E	2165	СМ	Monte Cimone, Italy
CIB	NOAA	41.8°N	4.9°W	845	FM	CIBA, Spain
LMP	NOAA	35.5°N	12.6°E	50	FM	Lampedusa, Italy

1 Table 3. Atmospheric observation sites for  $CH_4$  mole fraction used in the case study. The 2 altitude is given in meters above sea-level.

- 3 <sup>a</sup>Only used for independent validation
- 4

5 Table 4. Methane flux estimates used in the prior in the case study.

Source	Dataset	Total (TgCH <sub>4</sub> y <sup>-1</sup> )
anthropogenic:		
- agriculture <sup>b</sup>	EDGAR-4.2 FT2010 <sup>a</sup>	152.8
- industrial processes <sup>c</sup>	EDGAR-4.2 FT2010	0.3
- residential and transport <sup>d</sup>	EDGAR-4.2 FT2010	13.4
- waste <sup>e</sup>	EDGAR-4.2 FT2010	61.5
- oil, coal and gas <sup>f</sup>	EDGAR-4.2 FT2010	129.7
natural:		
- wetlands	LPJ DGVM <sup>g</sup>	175.0
- biomass burning	GFED-3.1 <sup>h</sup>	19.3
- geological seeps	(Etiope et al. 2008)	55.3
- soils	(Ridgwell et al. 1999)	-37.9
- wild animals	(Houweling et al. 1999)	5.0
- termites	(Sanderson et al. 1996)	19.3
- ocean	(Lambert and Schmidt, 1993)	17.0
total		610.0

<sup>a</sup>Emission Database for Global Atmospheric Research (http://edgar.jrc.ec.europa.eu)

<sup>b</sup>IPCC categories: 4A, 4B, 4C

8 <sup>c</sup>IPCC categories: 2

9 <sup>d</sup>IPCC categories: 1A3, 1A4

10 <sup>e</sup>IPCC categories: 6A, 6B, 6C

11 <sup>f</sup>IPCC categories: 1A1, 1A2, 1B1, 1B2, 7A

12 <sup>g</sup>Lund-Potsdam-Jena Dynamic Global Vegetation Model (LPJ DGVM)

13 <sup>h</sup>Global Fire Emissions Database (http://www.falw.vu/~gwerf/GFED.html)

Test ID	Spatial correlation	Observations	Background <sup>a</sup>
S1	500 km	afternoon/night only <sup>b</sup>	model-based, not optimized
S2	300 km	afternoon/night only	model-based, not optimized
S3	200 km	afternoon/night only	model-based, not optimized
S4	500 km	afternoon/night only	model-based, optimized
S5	500 km	all <sup>c</sup>	model-based, not optimized
S6	500 km	afternoon/night only	observation-based, not optimized

1 Table 5. Overview of the sensitivity tests.

2 <sup>a</sup>The method of calculation and whether or not the background mixing ratios were optimized in the

3 inversion 4 <sup>b</sup>Low altit

<sup>b</sup>Low altitude sites averaged afternoon data, high altitude sites averaged night data

5 <sup>c</sup>Averaged all data over 24 h.

6

7	Table 6 Statistics	of the cimulated	variant abcorriad CL	I mixing ration	from tost S1
/	Table 0. Statistics (	of the simulated	versus observed Cr	14 IIIIXIIIg Tatios	nom test S1.

Site ID	Prior			Posterior		
	NSD	R	RMSE	NSD	R	RMSE
PAL	0.99	0.69	16.2	1.04	0.82	11.9
ICE	1.01	0.26	9.4	0.90	0.24	9.2
BAL	1.14	0.66	16.6	0.96	0.72	13.7
MHD	1.18	0.57	9.2	0.97	0.63	7.8
<b>OXK</b> <sup>a</sup>	-	-	42.3	-	-	7.34
SSL	1.21	0.52	28.2	1.07	0.71	19.3
HPB	0.61	0.49	44.3	0.67	0.73	33.7
HUN	0.54	0.69	47.5	0.96	0.88	27.1
JFJ	1.04	0.30	21.4	0.75	0.33	20.3
BSC	1.10	0.24	51.1	0.91	0.39	35.2
CMN	1.00	0.56	26.4	0.81	0.68	21.6
CIB	0.89	0.50	20.9	0.91	0.68	15.6
LMP	2.02	0.34	35.0	1.68	0.45	24.0

8 <sup>a</sup>insufficient observations for calculating R and NSD

<sup>10</sup> Table 7. Comparison of CH<sub>4</sub> emissions (TgCH<sub>4</sub> y<sup>-1</sup>) from this study with the range of values 11 from an inversion ensemble for 2006 and 2007 (Bergamaschi et al. 2014). The prior and 12 posterior emissions are shown from test S1 and include the 1- $\sigma$  SD prior and posterior 13 uncertainties. NW Europe includes UK, Ireland, BENELUX, France, and Germany, and E 14 Europe includes Hungary, Poland, Czech Republic, and Slovakia, according to the definition 15 in Bergamaschi et al. (2014).

	Prior	Posterior	Bergamaschi et al. 2014
UK + Ireland	$2.66 \pm 0.84$	$2.41 \pm 0.33$	2.32 - 4.57
<b>BENELUX</b> <sup>a</sup>	$1.18 \pm 0.80$	$1.09 \pm 0.26$	1.44 - 2.29
France	$4.33 \pm 1.37$	$3.14 \pm 0.42$	2.02 - 4.94
Germany	$2.22 \pm 1.16$	$2.48 \pm 0.33$	2.35 - 3.51
NW Europe	$10.39 \pm 4.17$	$9.12 \pm 1.34$	8.13 - 14.44
Hungary	$0.37 \pm 0.62$	$0.50 \pm 0.17$	0.34 - 0.73
Poland	$2.81 \pm 1.05$	$2.62 \pm 0.38$	1.84 - 2.87
Czech Republic + Slovakia	$1.18 \pm 0.94$	$1.27 \pm 0.27$	1.12 - 1.63
E Europe	$4.36 \pm 2.61$	$4.39 \pm 0.82$	3.59 - 4.90
NW + E Europe	$14.75 \pm 4.17$	$13.51 \pm 2.16$	11.71 - 19.34

<sup>a</sup>BENELUX = Belgium, The Netherlands and Luxembourg

<sup>9</sup> 

1 Figure 1. Schematic showing how the forward model is defined. The black dots represent 2 receptors, the solid boxes represent gridded fluxes and the dotted box represents gridded 3 mixing ratios from global model output. The arrows indicate transport to a receptor (which 4 may be either inside or outside the nested domain): solid arrows show transport from fluxes 5 within the nested domain, dashed arrows indicate transport from fluxes outside the nested 6 domain, and the dotted arrows indicate transport of the mixing ratio at the point of back-7 trajectory termination. Each arrow can be thought of as an element (i.e. a partial derivate) in the transport matrix,  $\mathbf{H}^{nest}$ ,  $\mathbf{H}^{out}$ , and  $\mathbf{H}^{bg}$ , respectively. 8





Figure 2. Total emission sensitivity for 2011 in units of log(s m<sup>-3</sup> kg<sup>-1</sup>) calculated using FLEXPART and used to determine the variable grid (note that for this no weighting is applied for the number of observations available from each site) (a) and the variable resolution grid used in the inversion (b). The points indicate the positions of the observation sites.



Figure 3. CH<sub>4</sub> mole fractions (ppb) as observed (black) and simulated from the prior (blue)
and posterior models (red). Also shown is the background CH<sub>4</sub> mole fraction (green) and CH<sub>4</sub>
calculated by the TM5 Eulerian model (light blue).



- 1 Figure 4. Comparison of the background CH<sub>4</sub> mole fraction, calculated using the observation-
- 2 based (purple) and the model-based methods (green), with the observed mixing ratio (black).
- 3



- 1 Figure 5. A posteriori fluxes of CH<sub>4</sub> (left) and the flux increments (i.e. a posteriori a priori
- 2 fluxes) (right) for each of the sensitivity tests (in units of  $gCH_4 m^{-2} day^{-1}$ ).
- 3



1 Figure 6. Error reduction for the CH<sub>4</sub> fluxes for each of the sensitivity tests.



- Figure 7. Comparison of the prior (blue), posterior (red) and background (green) simulated
   CH<sub>4</sub> mixing ratios (ppb) with observations (black) at the independent site, PUY. Results are
- 3 shown for test S1.

