Technical note: A high-resolution inverse modelling technique for estimating surface CO₂ fluxes based on the NIES-TM - FLEXPART coupled transport model and its adjoint.

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Abstract

We developed a high-resolution surface flux inversion system based on the global Lagrangian-Eulerian coupled tracer transport model composed of National Institute for Environmental Studies Transport Model (NIES-TM) and FLEXible PARTicle dispersion model (FLEXPART). The inversion system is named NTFVAR (NIES-TM-FLEXPART-variational) as it applies variational optimisation to estimate surface fluxes. We tested the system by estimating optimized corrections to natural surface CO₂ fluxes to achieve best fit to atmospheric CO₂ data collected by the global in-situ network, as a necessary step towards capability of estimating anthropogenic CO₂ emissions. We employ the Lagrangian particle dispersion model (LPDM) FLEXPART to calculate the surface flux footprints of CO₂ observations at a 0.1° × 0.1° spatial

resolution. The LPDM is coupled to a global atmospheric tracer transport model (NIES-TM). Our inversion technique uses an adjoint of the coupled transport model in an iterative optimization procedure. The flux error covariance operator is being implemented via implicit diffusion. Biweekly flux corrections to prior flux fields were estimated for the years 2010-2012 from in-situ CO_2 data included in the Observation Package (ObsPack) dataset. High-resolution prior flux fields were

5   prepared using Open-Data Inventory for Anthropogenic Carbon dioxide (ODIAC) for fossil fuel combustion, Global Fire Assimilation System (GFAS) for biomass burning, the Vegetation Integrative SImulator for Trace gases (VISIT) model for terrestrial biosphere exchange and Ocean Tracer Transport Model (OTTM) for oceanic exchange. The terrestrial biospheric flux field was constructed using a vegetation mosaic map and separate simulation of CO_2 fluxes at daily time step by the VISIT model for each vegetation type. The prior flux uncertainty for terrestrial biosphere was scaled

10   proportionally to the monthly mean Gross Primary Production (GPP) by the Moderate Resolution Imaging Spectroradiometer (MODIS) MOD17 product. The inverse system calculates flux corrections to the prior fluxes in the form of a relatively smooth field multiplied by high-resolution patterns of the prior flux uncertainties for land and ocean, following the coastlines and vegetation productivity gradients. The resulting flux estimates improve fit to the observations at continuous observations sites, reproducing both the seasonal variation and short-term concentration variability, including

15   high CO_2 concentration events associated with anthropogenic emissions. The use of high-resolution atmospheric transport in global CO_2 flux inversion has the advantage of better resolving the transport from the mix of the anthropogenic and biospheric sources in densely populated continental regions and shows potential for better separation between fluxes from terrestrial ecosystems and strong localised sources such as anthropogenic emissions and forest fires. Further improvements in the modelling system are needed as the posterior fit is better than that by the National Oceanic and Atmospheric

20   Administration (NOAA) CarbonTracker only for a fraction of the monitoring sites, mostly at coastal and island locations experiencing mix of background and local flux signals.

1   Introduction

Inverse modelling of the surface fluxes is implemented by using chemical transport model simulations to match

25   atmospheric observations of greenhouse gases. CO_2 flux inversions studies started from addressing large scale flux distributions (Enting and Mansbridge, 1989; Tans et al., 1990; Gurney et al., 2002; Peylin et al., 2013 and others) using background monitoring data and global transport models at low and medium resolutions targeting extraction of the information on large and highly variable fluxes of carbon dioxide from terrestrial ecosystems and oceans. Merits of improving the resolutions of global transport simulations to 9-25 km have been also discussed by previous studies, such as

30   Agusti-Panareda et al. (2019) and Maksyutov et al. (2008). However, global inverse modelling studies have never been conducted at these spatial resolutions. On the other hand, regional scale fluxes, such as national emissions of non-CO_2

greenhouse gases (GHGs), have been estimated using inverse modelling tools relying on regional (mostly Lagrangian) transport algorithms capable of resolving surface flux contributions to atmospheric concentrations at resolutions from 1 to 100 km (Vermeulen et al., 1999; Manning et al., 2011; Stohl et al., 2009; Rodenbeck et al., 2009; Henne et al., 2016; He et al., 2018; Schuh et al., 2013; Lauvaux et al., 2016 and others). Extension of the regional Lagrangian inverse modelling

5   to the global scale based on combination of three-dimensional (3-D) global Eulerian model and Lagrangian model have been implemented in several studies (Rugby et al., 2011; Zhuravlev et al., 2013; Shirai et al., 2017), which demonstrated an enhanced capability of resolving the regional and local concentration variability driven by fine scale surface emission patterns, while using inverse modeling schemes relying on regional and global basis functions that yield concentration responses of regional fluxes at observational sites. A disadvantage of using regional basis functions in inverse modeling is

10 the flux aggregation errors as noted by Kaminski et al. (2001), is addressed by developing grid-based inversion schemes based on variational assimilation algorithms that yield flux corrections that are not tied to aggregated flux regions (Rodenbeck et al., 2003; Chevallier et al., 2005; Baker et al., 2006, and others). In order to implement a grid-based inversion scheme suitable for optimizing surface fluxes using a high-resolution atmospheric transport capability of the Lagrangian model, an adjoint of a coupled Eulerian-Lagrangian model is needed, such as one reported by Belikov et al. (2016).

15 In thus study, we applied an adjoint of the coupled Eulerian-Lagrangian transport model, which is a revised version of Belikov et al. (2016), to the problem of surface flux inversion based on transport model with spatial resolution of 0.1° longitude-latitude. While global higher resolution transport runs, such as ~1 km, can be done with coupled Eulerian-Lagrangian models (Ganshin et al., 2012), the choice of the model resolution in our inversion system is often dictated mainly by the availability of the prior surface CO_2 fluxes and the size of available computer memory. A practical need for

20 running high-resolution atmospheric transport simulations at global scale is currently driven by expanding GHG observing capabilities towards quantifying anthropogenic emissions by observing at the vicinity of emission sources (Nassar et al., 2017), including observations in both background and urban sites, with tall towers, commercial airplanes, and satellites. At the same time, the focus of inverse modeling is evolving towards studies of the anthropogenic emissions, with a target of making better estimates of regional and national emissions in support of national and regional GHG emission reporting

25 and control measures (Manning et al., 2011; Henne et al., 2016; Lauvaux et al., 2016). In that context, global-scale high-resolution inverse modeling approaches have advantage in closing global budgets, while regional and national scale inverse modeling approaches with limited area models require boundary conditions normally provided by global model simulations with optimized fluxes. Often there is an additional degree of freedom introduced by allowing corrections to the boundary concentration distribution to improve a fit at the observation sites (Manning et al., 2011). As a result, the global total of

30 regional emission estimates does not necessarily match the balance constrained by global mean concentration trends. A global coupled Eulerian-Lagrangian model (e.g. Ganshin et al., 2012), has potential for addressing both the objectives, that

is closing the global balance and operating at range of scales from a single city (Lauvaux et al., 2016) to large country or continental scale. Here we report developing a high-resolution inverse modeling technique suitable for application at a broad range of spatial scales and apply it to the problem of estimating the distribution of CO_2 fluxes over the globe that provides best fit to the observations. In separate studies, the same inversion systems were applied to inverse modeling of

5   methane emissions (Wang et al., 2019; Janardanan et al., 2020).

In this paper, we report its application to the inverse problem of the natural CO_2 flux estimation as a first step towards estimating the fossil CO_2 emissions where the advantage of high-resolution approach is more evident. The paper is composed as follows: this Section 1 provides Introduction; Section 2, the transport model and its adjoint; Section 3 introduces the Prior fluxes, observational dataset, and gridded flux uncertainties; Section 4 gives the formulation of the

10   inverse modeling problem and numerical solution; Section 5 presents simulation results and discussion, which is followed by the Summary and Conclusions.

2   The coupled tracer transport model, its adjoint and the implementation

15   For simulation of the CO_2 transport in the atmosphere we used a coupled Eulerian-Lagrangian model NIES-TM-FLEXPART, which is a further modification of the model described by Belikov et al. (2016). The coupled model combines NIES-TM v08.1i with horizontal resolution of $2.5° \times 2.5°$ and 32 hybrid-isentropic vertical levels (Belikov et al., 2013) and FLEXPART model v.8.0 (Stohl et al., 2005) run in backward mode with surface flux resolution of $0.1° \times 0.1°$. Both models use the Japan 25-year reanalysis (JRA-25)/JMA Climate Data Assimilation System (JCDAS) meteorology (Onogi

20   et al., 2007), with 40 vertical levels interpolated to a $1.25° \times 1.25°$ grid. The use of low-resolution wind fields for high resolution transport is better justified for cases of nearly geostrophic flow over flat terrain (Ganshin et al., 2012), but was also considered applicable in regions with more complex topography (Ware et al., 2019). The coupled transport model adjoint was derived from the Global Eulerian-Lagrangian Coupled Atmospheric transport model (GELCA) (Ganshin et al., 2012; Zhuravlev et al., 2013; Shirai et al., 2017). To facilitate model application in our iterative inversion algorithm, all

25   the components of the model – Eulerian model and the coupler are integrated in one executable (online coupling) as described in Belikov et al., (2016), while the original GELCA model implements Eulerian and Lagrangian components sequentially, and then applies the coupler (off-line coupling). The changes in the current version with respect to the version presented by Belikov et al. (2016) include an adjoint code derivation for model components using the adjoint code compiler Tapenade (Hascoet and Pascual, 2013), instead of using the TAF compiler (Giering and Kaminski, 2003). Additionally,

30   the indexing and sorting algorithms for the transport matrix were revised to allow an efficient memory use for processing large matrices of LPDM-driven responses to surface fluxes arising in the case of high-resolution surface fluxes and large

number of observations, especially when using satellite data. A manually derived adjoint of the NIES-TM v08.1i is used as in Belikov et al. (2016), due to its computational efficiency. In the version of the model that includes manually coded adjoint, only the second order van Leer algorithm (van Leer, 1977) is implemented, as opposed to third order algorithm typically used in forward model (Belikov et al., 2013).

3   Prior fluxes, flux uncertainties and observations

Prior CO_2 fluxes, were prepared as a combination of monthly-varying fossil fuel emissions by the Open-Data Inventory for Anthropogenic Carbon dioxide (ODIAC), available at a global 30 arc second resolution (Oda et al., 2018), ocean-atmosphere exchange by the Ocean Tracer Transport Model (OTTM) 4D-var assimilation system, available at a 1° resolution (Valsala and Maksyutov, 2010), daily CO_2 emissions by biomass burning by Global Fire Assimilation System (GFAS) dataset provided by Copernicus services at a 0.1° resolution (Kaiser et al., 2012), and daily varying climatology of terrestrial biospheric CO_2 exchange simulated by optimized Vegetation Integrative SImulator for Trace gases (VISIT) model (Ito, 2010; Saito et al., 2014). Figure 1 presents samples of the four prior flux components (fossil, vegetation, biomass burning and ocean) used in the forward simulation.

3.1   Emissions from fossil fuel

For fossil fuel CO_2 emissions (emissions due to of fossil fuel combustion and cement manufacturing) we used ODIAC data product (Oda and Maksyutov, 2011, 2015; Oda et al., 2018) at 0.1° × 0.1° resolution on monthly basis. The version 2016 of the ODIAC-2016 data product (ODIAC2016, Oda et al., 2018) is based on global and national emission estimates and monthly estimates made at monthly resolution by the Carbon Dioxide Information Analysis Center (CDIAC) (Boden et al., 2016; Andres et al., 2011). For spatial disaggregation it uses the emission data for powerplant emissions by the CARbon Monitoring and Action (CARMA) database (Wheeler and Ummel, 2008), while the rest of the national total emissions on land were distributed using spatial patterns provided by night-time lights data collected by the Defence Meteorological Satellite Program (DMSP) satellites (Elvidge et al., 1999). The ODIAC fluxes were aggregated to a 0.1° resolution from the high-resolution ODIAC data. The ODIAC emission product is suitable for this type of studies because the global total emission is constrained by updated estimates while providing a high-resolution emission estimate. Thus, it can be applied to carbon budget problems across different scales.

3.2   Terrestrial biosphere fluxes

CO_2 fluxes by the terrestrial biosphere at a resolution of $0.1°$ were constructed using a vegetation mosaic approach, combining the vegetation map data by synergetic land cover product (SYNMAP) dataset (Jung et al., 2006), available at a 30 arc second resolution, with terrestrial biospheric CO_2 exchanges simulated by optimized VISIT model (Saito et al., 2014) for each vegetation type in every $0.5°$ grid at daily time step. The area fraction of each vegetation type is derived from SYNMAP data for each $0.1°$ grid. The CO_2 net ecosystem exchange (NEE) fluxes on $0.1°$ grid were prepared by combining the vegetation type-specific fluxes with vegetation area fraction data on $0.1°$ grid. By averaging the daily flux data for period of 2000-2005 the flux climatology was derived for use in the recent years, when the VISIT model simulation based on JRA-25-JCDAS reanalysis data is not available.

3.3   Emissions from biomass burning

Daily biomass burning CO_2 emissions by Global Fire Assimilation System (GFAS) dataset relies on assimilating Fire Radiative Power (FRP) observations from the MODIS instruments onboard the Terra and Aqua satellites (Kaiser et al., 2012). The fire emissions at $0.1°$ resolution are calculated from FRP with land cover-specific conversion factors compiled from a literature survey. The GFAS system adds corrections for observation gaps in the observations, and filters spurious FRP observations of volcanoes, gas flares and other sources.

3.4   Oceanic exchange flux

The air-sea CO_2 flux component for the flux inversion used an optimized estimate of oceanic CO_2 fluxes by Valsala and Maksyutov (2010). The dataset is constructed with a variational assimilation of the observed partial pressure of surface ocean CO_2 (pCO_2) available in Takahashi et al. (2017) database into the OTTM (Valsala et al., 2008), coupled with a simple one-component ecosystem model. The assimilation consists of a variational optimization method which minimizes the model to observation differences in the surface ocean dissolved inorganic carbon (DIC) (or pCO_2) within two-month time window. The OTTM model fluxes produced on a $1° \times 1°$ grid at monthly time step are interpolated to a $0.1° \times 0.1°$ grid, taking into account, the land fraction map derived from 1 km resolution MODIS landcover product.

3.5   Flux uncertainties for land and ocean.

CO_2 flux uncertainties are needed for both land and ocean. Climatological, monthly-varying flux uncertainties for land were set to 20% of MODIS gross primary productivity (GPP) by MOD17A2 product available on a $0.05°$ grid at monthly

resolution (Running et al., 2004). Oceanic flux uncertainties are assigned based on the standard deviation of the OTTM assimilated flux from climatology by Takahashi et al. (2009), plus the monthly variance of the interannually-varying OTTM fluxes (Valsala and Maksyutov, 2010), with a minimum value of 0.02 $gCm^{-2}day^{-1}$, in the same way as in the lower spatial resolution inverse model by Maksyutov et al. (2013). Oceanic flux uncertainties were first estimated on a $1° \times 1°$ resolution

5   at monthly time step, and then interpolated to a $0.1° \times 0.1°$ grid, with the same procedure as for the oceanic fluxes.

3.6   Atmospheric CO₂ observations.

We used CO₂ observation data distributed as the ObsPack-CO₂ GLOBALVIEWplus v2.1 (Cooperative Global Atmospheric Data Integration Project, 2016). The data from the flask sites were used as average concentration for a pair of flasks. Afternoon (15:00 to 16:00 local time) average concentrations were used for continuous observations over land

10   and for remote background observation sites. For the continuous mountain top observations, we used early morning observations (05:00 to 06:00 local time). Geographical local time is used, as defined by UTC time with longitude dependent offset. The list of the observation locations, with ObsPack site ID, site names, data providers and data references appear in the Table A1 in the Appendix. Aircraft observational data collected by NOAA Aircraft Program at Briggsdale, Colorado (CAR), Cape May, New Jersey (CMA), Dahlen, North Dakota (DND), Homer, Illinois (HIL), Worcester, Massachusetts,

15   (NHA), Poker Flats, Alaska (PFA), Rarotonga, (RTA), Charleston, South Carolina (SCA), Sinton, Texas (TGC) (Sweeney et al., 2015), and by the CONTRAIL project over West Pacific (CON) (Machida et al., 2008) were grouped into averages for each 1 km altitude bin, altitude counted from sea level. Within the 1 km altitude range, average of both concentration and the altitude are taken. Aircraft observations were not assimilated, only intended for use in validation.

20   **4   Inverse modelling algorithm**

4.1   Flux optimization problem

Inverse problem of atmospheric transport is formulated by Enting (2002) as finding the surface fluxes that minimize misfit between transport model simulation $H \cdot (f + x)$ and the vector of observations y. The equation $y = H \cdot (f + x)$ has to be solved for unknown flux correction x, where f is known flux, called pre-subtracted by Gurney et al. (2002), and x is

25   solved for at the transport model grid scale (Kaminski et al., 2001). By introducing residual misfit vector $r = y - H \cdot f$, the problem can be formulated as minimizing a norm of difference $(r - H \cdot x)$. As the observation data alone are not sufficient to uniquely define the solution x, additional regularization is required. By introducing additional constraints on

the amplitude and smoothness of the solution, the inverse modelling problem is formulated (Tarantola, 2005) as solving for optimal value of vector x at the minimum of a cost function $J(x)$:

$$J(x) = \frac{1}{2}(H \cdot x - r)^T \cdot R^{-1} \cdot (H \cdot x - r) + \frac{1}{2}x^T \cdot B^{-1} \cdot x \qquad (1)$$

where x is optimised flux, R is a covariance matrix for observations and B is a covariance matrix for surface fluxes. By introducing a decomposition of B as $B = L \cdot L^T$ (construction of matrix L explained in detail in Section 4.2) and a variable substitution $x = L \cdot z$ the second term in Eq. (1) is simplified. At the same time, by assuming that R can be decomposed into $R = \sigma^T \cdot \sigma$, where σ is a vector of data uncertainties, and introducing expressions $b = \sigma^{-1} \cdot (r - H \cdot x)$, and

10 $A = \sigma^{-1} \cdot H \cdot L$, the new form of Eq. (1) is introduced:

$$J(z) = \frac{1}{2}((A \cdot z - b)^T(A \cdot z - b) + z^T \cdot z) \qquad (2)$$

The solution minimizing $J(z)$ can be obtained by forcing the derivative $\partial J(z)/\partial z = A^T(A \cdot z - b) + z$ to zero, which results in

$$(A^T A + I) \cdot z = A^T b \qquad (3)$$

15 An optimal solution z at the minimum of the cost function $J(z)$ is found iteratively with the Broyden–Fletcher–Goldfarb–Shanno (BGFS) algorithm (Broyden, 1969; Nocedal, 1980), as implemented by Gilbert and Lemarechal (1989). The method requires ability to accurately estimate the cost function $J(z)$ and its gradient $A^T(A \cdot z - b) + z$, and has modest memory storage demands. Given the solution z, flux correction vector x is then found by reversing variable substitution as $x = L \cdot z$.

20 The convergence of the solution may be affected by accuracy of the adjoint. The result of duality test defined as norm of difference between NIES-TM-FLEXPART forward and adjoint modes estimated as $(< y, H \cdot x > - < H^T \cdot y, x >)/(< y, H \cdot x >)$ was found to be in the order of 10^{-9}, while for Lagrangian component based on receptor sensitivity matrices prepared with FLEXPART, it is about 10^{-15} when double precision is used in calculations, same as in (Belikov et al., 2016). The formulation of the minimization problem as presented by Eq. (2) is convenient for the derivation of the flux

25 uncertainties, as it is possible to solve Eq. (3) via the truncated singular value decomposition (SVD) and estimate regional flux uncertainties based on derived singular vectors (Meirink et al., 2008). Alternatively, as mentioned by Fisher and Courtier (1995) it is also possible to use the flux increments derived at each iteration of the BFGS algorithm in place of the singular vectors. Although we did not use SVD for constructing the posterior covariances in this study, we tested solving the optimisation problem with SVD. We derived SVD of $A^T A$ using a code by Wu and Simon (2000), which implements

30 an algorithm by Lanczos (1950), and confirmed that we get practically the same solution as one obtained with BFGS

algorithm. Lanczos (1950) algorithm is a commonly used SVD technique applied in case of large sparse matrix or a linear operator, when it is impractical to directly make SVD of A. A truncated SVD of A is given by expression $A \approx U\Sigma V^T$, where Σ is diagonal matrix of n singular values, while U and V are matrices of left and right singular vectors. Variable substitutions

$$z = V^T s, d = U^T b, \tag{4}$$

transforms z into a space of singular vectors s and reduces Eq. (3) to $(\Sigma^T\Sigma + I) \cdot s = \Sigma^T d$, resulting in a solution

$$s = \Sigma^T d/(\Sigma^T\Sigma + I), \tag{5}$$

which is evaluated directly, as Σ is diagonal. In case of having only n largest singular values, the elements of solution s are given by $s_i = \lambda_i d_i/(\lambda_i^2 + 1)$, for all $i \leq n$. Once the solution (5) is found, it is taken back to the space of dimensional fluxes z by applying variable substitutions (4). For fluxes, we have $x = L \cdot z, z = V^T s, d = U^T b$, thus solution is provided by

$$x = LV \cdot \frac{\Sigma^T}{(\Sigma^T\Sigma + I)} \cdot U^T b. \tag{6}$$

Another variant of SVD approach may be more memory efficient in the case of very large dimension of a flux vector, then applying SVD to AA^T instead of $A^T A$ can save some memory as in a representer method (Bennett, 1992). It gives the same solution as SVD of $A^T A$ using less intermediate memory storage when the dimension of the observation vector y is lower compared to that of the flux vector x.

The forward and adjoint mode simulations with transport model needed to implement iterative optimization are composed of several steps:

1. Running the Lagrangian model FLEXPART to produce source-receptor sensitivity matrices. For each observation event, a backward transport simulation with FLEXPART model is implemented, to produce daily average surface flux footprints at a $0.1° \times 0.1°$ resolution and the 3-D concentration field footprint, taken at the end of the 3-day backward simulation run (stopping at 0 GMT). The surface flux sensitivity data are recorded in the unit of ppm(gCm^{-2}day^{-1})$^{-1}$.

2. Running the coupled transport model forward, which includes:

   a. Running the 3-D Eulerian model NIES-TM from the 3-D initial concentration field, with the prescribed surface fluxes. Sampling the 3-D field at model coupling times for each observation according to 3-D concentration field footprints, calculated at the first step by FLEXPART. NIES-TM reads the same $0.1°$ fluxes as Lagrangian transport model, and remaps them onto its $2.5° \times 2.5°$ grid, before including in the simulation.

b. Use two-dimensional (2-D) surface flux footprints prepared with Lagrangian model to calculate the surface flux contribution to the simulated concentrations for the last 3 days.

c. Combining the concentration contributions produced by Eulerian (a) and Lagrangian (b) component to give total simulated concentration.

3. In the inverse modelling, the transport model is run in three modes:

a. The forward model is first run with prescribed prior fluxes, starting from the 3-D initial CO_2 concentration field, to calculate differences between the observation and the model simulation (residual misfit).

b. at the inverse modelling/optimization step, only flux corrections are propagated in forward model runs, which are optimized to fit the observation-model misfit. The prescribed prior fluxes are not used (switched off) at this step. The model starts from a zero 3-D initial concentration field and runs forward with flux corrections updated by the optimization algorithm at every iteration, to produce simulated concentrations. Corrections to the 3-D initial concentration field are not estimated, and not included into the control vector. Instead, the model is given three months of spin-up period before the target flux estimation period to adjust simulated concentration to observations.

c. in the adjoint mode, the adjoint mode atmospheric transport is simulated backward in time starting from the vector of residuals to produce a gradient of the cost function (defined as eq. (1)) with respect to the surface fluxes. Given the gradient, the optimization algorithm provides the new flux corrections field. For convenience, the transport model and its adjoint are implemented as callable procedures suitable for direct communication mode.

Steps 1 is carried out the same way as in other versions of the coupled transport model (Zhuravlev et al., 2013; Shirai et al., 2017). At steps 2 and 3 the procedure of running the forward and adjoint model is organised differently. At the beginning of the transport model runs, all the data prepared by the Lagrangian model are stored in computer memory, in order to save the time for reading and re-sorting the data at each iteration.

To create the initial concentration field, we used a 3-D snapshot of CO_2 concentration for the same day from a simulation of previous year, which are already optimised (usually Oct 1st, or Jan 1st)), and correct it globally for the difference between years using the NOAA monthly mean data for South Pole as representative for the global mean concentration.

4.2 Implementation of covariance matrices L and B.

We optimized surface flux fields separately for two sets of fluxes in every grid globally, for land and ocean, following the approaches done by Meirink et al. (2008) and Basu et al. (2013), who suggested optimizing for global surface flux fields separately for each optimized flux category. Separating the total flux into independent flux categories, each with its own

5   flux uncertainty pattern, results in using homogenous spatial covariance matrices, significantly simplifying the coding of the matrix B. The matrix B can be given as a product of a diagonal matrix of flux uncertainties and a matrix with 1.0 as diagonal elements, while non-diagonal elements are exponentially declining with squared distance between grid points (Meirink et al., 2008). In practice, an extra scaling of the uncertainty is needed for balancing the constraint on fluxes with the data uncertainty, which also impacts the regional flux uncertainties. Several empirical methods are in use, where tuning

10   parameters are horizontal scale (Meirink et al., 2008) and uncertainty multiplier (Chevallier et al., 2005; Rodenbeck, 2005). In our B matrix design, we follow Meirink et al. (2008) in representing B matrix as multiple of non-dimensional covariance matrix C and diagonal matrix of flux uncertainty D as $B = D^T \cdot C \cdot D$. C matrix is commonly implemented as band matrix with non-diagonal elements declining as $\sim \exp\left(-x^2/l^2\right)$ with distance x between the grid cells, as in 2-D spline algorithms (Wahba and Wendelberger, 1980). Applying multiplication by matrix C becomes computationally costly at a

15   high spatial resolution in the cases when the correlation distance l is much larger than the size of the model grid. The correlation distance used here is 500 km for land and ocean, and two weeks in time. In that case, the use of implicit diffusion with directional splitting to approximate the Gaussian shape appears to be computationally efficient, as the number of floating-point operations per grid point do not grow with the ratio of correlation distance l to the grid size. The covariance matrix based on diffusion operator is popular in many ocean data assimilation systems, as a convenient way to deal with

20   coastlines (e.g. Derber and Rosati, 1989; Weaver and Courtier, 2001). Applying diffusion operator for the covariance matrix helps achieving the spatial homogeneity between polar and equatorial regions, as diffusion produces theoretically uniform effect on flux field regardless of the polar singularity. The diffusion operator works as a low-pass filter, selectively suppressing all the wavelengths shorter than the covariance length scale. As we need to construct the covariance matrix B in the form $B = L \cdot L^T$, we choose to construct L first and then derive its transpose L^T. The factorization of L is given

25   by $L = u_F \cdot (L_{xy} \otimes L_t) \cdot m$, where L_t is the one-dimensional covariance matrix for time dimension, $\otimes$ is a Kronecker product. We approximate the two-dimensional diffusion by splitting the diffusion into two dimensions, latitude and longitude, as in Chua and Bennett, (2001), and apply several iterations of this process. The horizontal covariance L_{xy} is implemented in $N = 3$ iterations of one-dimensional diffusion so that $L_{xy} = (L_x \otimes L_y)^N$. L_x and L_y are diffusion operators for longitude and latitude directions respectively, while u_F is a diagonal matrix of flux uncertainty for each grid

30   cell and each flux category (land and ocean), and m is a diagonal matrix of map factor which is introduced to scale

contributions to the cost function by model grid area, with diagonal elements given by $\cos^{-1/2}\theta$ (where θ is latitude). The adjoint operators L_x^T, L_y^T are derived by applying the adjoint code compiler Tapenade (Hascoet and Pascual, 2013) to the Fortran code of modules approximating operators L_x and L_y by implicit diffusion. L_t and its transpose L_t^T are of lower dimension and are designed as in Meirink et al. (2008) by deriving a square root of the Gaussian-shaped time covariance

5   matrix with direct SVD (Press et al., 1992).

The important merit of the algorithm is that it makes minimal use of computer memory, avoiding allocation of the memory space larger than several times the dimension of observation and flux vectors, making it suitable for ingesting large amount of surface and space-based observations. It should be mentioned that the computer memory demand for accommodating surface flux sensitivity matrices for massive space-based observations can be limited as discussed by Miller et al. (2019).

10 **4.3   Inversion setup**

Combination of coupled transport model NIES-TM-FLEXPART (as described in Section 2) with variational optimization algorithm (Sections 4.1-4.2) constitutes the inverse modeling system NIES-TM-FLEXPART-VAR (NTFVAR). We test the inversion algorithm presented in pervious sections on a problem of finding a best fit to CO_2 observations provide by

15 ObsPack dataset by optimizing corrections to land and ocean fluxes. By the design of our inverse modeling system, we produce smoothed fields of scaling factors that are multiplied by fine resolution flux uncertainty fields to give flux corrections. We derive the surface CO_2 flux corrections at $0.1°$ resolution and half-month time step. Our purpose is to demonstrate that we can optimize fluxes to improve fit to the observations using iterative optimization procedure, based on high-resolution coupled transport model and its adjoint. Our report is limited to technical development towards

20 achieving capability of estimating anthropogenic CO_2 emissions based on atmospheric observations, and we do not elaborate on the impact of improvement in simulating the tracer transport at high resolution on the quality of the optimized natural fluxes, which requires additional study. The flux optimization is applied in a short time-window of 18 months, for each optimized year, and simulation starts on October 1ˢᵗ, three months ahead of the target year. Three-month spin-up period is given to let inversion adjust the modeled concentration to the observations, so that the balance is achieved between

25 fluxes, concentrations and concentration trends. The simulation is continued until reaching the limit of 45 cost function gradient calls, by that time M1QN3 procedure by Gilbert and Lamarechal (1989) is able to complete 30 iterations. We optimize fluxes for three years from 2010 to 2012, and analyze simulated concentration fit at the observation sites. The average root mean squared misfits (RMSE) between optimized concentration and observations are compared between forward simulation with prior fluxes and optimized simulation, while for evaluation, we use statistics of optimized

simulations by the operational CarbonTracker inverse modeling system (ObsPack_co2_1_CARBONTRACKER_CT2017_2018-05-02; Peters et al., 2007).

5   Results and discussion.

5.1 Analysis of the posterior model fit to the observations

5   We compared the results of forward simulation with prior and optimized fluxes with the processed observations for ground observation sites, as shown in Table A1, and for airborne vertical profiles, used for independent validation (Table A2). Figure 2 shows the observations with forward (prior) and optimized simulations at Barrow (BRW), Jungfraujoch (JFJ), Wisconsin (LEF), Pallas (PAL), Yonagunijima (YON), and Syowa (SYO). Optimization leads to improved seasonal variation of the simulated concentration, including phase and amplitude at most sites. At SYO we find synoptic scale

10   variations with amplitude in the order of few tenths of a ppm that were to a large extent captured by the model. Plots for BRW and JFJ show the ability of the inversion to correct the seasonal cycle, while the difference between model and observations in the southern hemisphere (SYO) is contributed by interannual variations of the carbon cycle. The model-observation mismatch for surface sites included in the ObsPack is presented in Figure 3. The model was able to reduce the model to observation mismatch for most background sites, where the seasonal cycle is affected mostly by natural terrestrial

15   and oceanic fluxes, while average reduction of the mismatch from forward to optimized simulation is 14%. The reason for relatively small reduction is the addition of climatological flux corrections to the prior, estimated by inverse modeling of two years of data, 2009 and 2010. As a result, the inversion starts from the initial flux distributions already adjusted to fit the seasonal cycle of observed concentration. The correction for difference in global concentration trend between years is not made, thus there are visible differences between prior and optimized simulations in southern hemispheric background

20   sites. At most of the Antarctic sites, the mean posterior (after optimization) mismatch (reported as RMSE) is at the level of 0.2 ppm. Over the land, closer to anthropogenic sources, there is less relative reduction of mismatch on average at annual mean scale. One of the reasons for seeing little improvement is keeping fossil CO_2 emissions fixed and optimizing only the natural fluxes (while the strong signal from fossil emission is not affected by flux corrections). Another possible contributor to the large mismatch over land is neglecting the diurnal cycle under assumption of using only observations at

25   well mixed condition, and also the limited ability of the low-resolution reanalysis dataset to capture frontal processes in extratropical continental atmosphere, as discussed by Parazzoo et al. (2011).

Figure 3 also shows, for comparison, the statistics of the average misfit for optimized simulation by CarbonTracker, for the same period and same monitoring stations. The comparison is useful for understanding the strength and weaknesses of the inversion system presented here. Over the background monitoring sites, the high-resolution model does not show

advantage over CarbonTracker in terms of the fit between optimized model simulation and observations, which may indicate a better performance by the Eulerian model TM5 used in CarbonTracker. On the other hand, several sites where the high-resolution model shows better fits to observations over CarbonTracker are located inland or near the coast, closer to anthropogenic and biogenic sources. Lower misfit is achieved by high-resolution model at Key Biscaine (KEY), Baring

5   Head (BHD), Marianna island (GMI) and Cape Kumukahi (KUM), among others, which can be attributed to coastal/island locations, while there is little or no advantage at mountain sites like Mauna Loa (MLO) or Jungfraujoch (JFJ). This result may be influenced by differences in the model physics between NIES-TM-FLEXPART and TM5 in the lower troposphere, near the top of the boundary layer and in shallow cumuli. The mismatch between our optimized model and observations for sites used in the inversion is only 4% lower on average than that by CarbonTracker. For average misfit comparison, all

10   data, both assimilated and not assimilated, are included for sites shown in Figure 3. The results for CGO were not counted, due to the use of different datasets, as our system used only NOAA flask data, which underwent background selection (by wind direction) at the time of sampling.

For independent validation, a comparison of the unoptimized and optimized simulation to the vertical profile data is shown in Figure 4. For each vertical profile site, the observations were grouped according to altitude range, at 1 km steps. Altitude

15   code (e.g. 050, 015, 025, 035, …) to be added to site the identifier is constructed as altitude of midlevel multiplied by 10. The observations at PFA (Poker Fat Alaska) between surface and 1 km are grouped as PFA005 (mid altitude 0.5 km), while those in 5 to 6 km range are designated as PFA055 (mid altitude 5.5 km). As for optimized surface data in Figure 3, we show RMSE for forward simulation with prior fluxes, optimized simulation and CarbonTracker. CarbonTracker shows better fit at most altitudes except for the lowest 1 km where the results shown by the two systems are similar. Comparison

20   to CarbonTracker (CT2017) suggests that free tropospheric performance can be improved by implementing more detailed vertical mixing processes in the Lagrangian and Eulerian component models.

6 Summary and conclusions

A grid-based flux inversion system was developed, which is suitable for inverse estimation of the surface fluxes at biweekly time step and 0.1° spatial resolution. To implement the high-resolution capability, several developments were completed.

25   High-resolution prior fluxes were prepared for three surface flux categories: fossil emissions by ODIAC dataset are based on point source database and nightlights, biomass burning emissions (GFAS) are based on MODIS observations of fire radiative power and biosphere exchange is based on mosaic representation of landcover and process-based VISIT model simulation. High-resolution transport for global set of observations is achieved by combining short-term simulations with high-resolution Lagrangian model FLEXPART with global three-dimensional simulation with medium-resolution Eulerian

model NIES-TM. Use of variational optimization with a gradient-based method in the inversion helps avoiding the need to invert large matrices with dimensions dictated by the number of optimized grid fluxes or the number of the observations. Accordingly, the adjoint of the coupled transport model was developed to apply the variational optimization. Computationally efficient implementation of flux error covariance operator is achieved by using implicit diffusion

5   algorithm. Overall, the presented algorithm demonstrates feasibility of high-resolution inverse modeling at global scale, extending the capabilities achieved by regional high-resolution modeling approaches used for estimating the national greenhouse gas emissions for comparison to the national greenhouse gas inventories. Comparison of the optimized simulation to the observations shows some improvement over lower resolution CarbonTracker model for some continental and coastal observation sites, located closer to anthropogenic emissions and strong biospheric fluxes, but also demonstrates

10   the need for further improvement of the inverse modeling system components. Transport model errors can be reduced by improving transport modeling algorithms in Eulerian and Lagrangian model and using combination of recent higher resolution reanalysis data with high-resolution wind data simulations by regional models in the regions of interest. Inverse modeling algorithm can be improved by tuning the uncertainty scaling, and spatial and temporal covariance distances. Prior fluxes can be improved by developing high-resolution diurnally varying biospheric fluxes, developing a more detailed

15   fossil emission inventory, and developing updates to biomass burning and oceanic fluxes.

Code and data availability.

The inverse model and forward transport model code can be made available to research collaborators. Observed CO_2 concentrations ObsPack dataset is available from NOAA/ESRL (https://www.esrl.noaa.gov/gmd/ccgg/obspack/), ODIAC fossil fuel emissions - from CGER/NIES database (http://db.cger.nies.go.jp/dataset/ODIAC/).

20   **Author contributions.**

SM developed the inverse and transport model algorithms and model codes, ran the model, analysed the results and wrote the manuscript. TO developed the anthropogenic emission inventory, MS developed biospheric flux dataset, JK provided the biomass burning emission fluxes, VV prepared the oceanic CO_2 fluxes. RJ contributed to model testing and data preparation, DB contributed to the development of the NIES transport model, coupled model and coupled adjoint, RZ and

25   AG developed the Lagrangian response simulation system based on FLEXPART model. AA, LC, ED, LH, TM, TN, MR, RL, CS, DR contributed the observational data. All the authors contributed to the development of the manuscript.

Competing interests.

The authors declare no competing interests.

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Appendix

Table A1. List of the observation site included in the ObsPack dataset

Site ID	Lat	Lon.	Site name	Lab name	Sampling	Reference
ALT	82.45	-62.51	Alert	EC	insitu	Worthy et al. 2003
ALT	82.45	-62.51	Alert	NOAA	flask	Conway et al. 1994
AMS	-37.8	77.54	Amsterdam Island	LSCE	insitu	Gaudry et al. 1991
AMT	45.03	-68.68	Argyle	NOAA	insitu	Andrews et al. 2014
ARA	-23.86	148.47	Arcturus	CSIRO	flask	Francey et al. 2003
ASC	-7.97	-14.40	Ascension Island	NOAA	flask	Conway et al. 1994
ASK	23.26	5.63	Assekrem	NOAA	flask	Conway et al. 1994

AZR	38.77	-27.38	Terceira Island	NOAA	flask	Conway et al. 1994
BAO	40.05	-105.00	Boulder Atmospheric Observatory	NOAA	insitu	Andrews et al. 2014
BCK	-116.1	62.80	Bechoko	EC	insitu	Worthy et al. 2003
BHD	-41.41	174.87	Baring Head Station	NOAA	flask	Conway et al. 1994
BHD	-41.41	174.87	Baring Head Station	NIWA	insitu	Brailsford et al. 2012
BMW	32.27	-64.88	Tudor Hill	NOAA	flask	Conway et al. 1994
BRA	51.2	-104.7	Bratt's Lake Saskatchewan	EC	insitu	Worthy et al. 2003
BRW	71.32	-156.61	Barrow	NOAA	flask	Conway et al. 1994
BRW	71.32	-156.61	Barrow	NOAA	insitu	Peterson et al. 1986
CBA	55.21	-162.72	Cold Bay	NOAA	flask	Conway et al. 1994
CES	51.97	4.93	Cesar	ECN	insitu	Vermeulen et al. 2011
CGO	-40.68	144.69	Cape Grim	NOAA	flask	Conway et al. 1994
CHL	58.75	-94.07	Churchill	EC	insitu	Worthy et al. 2003
CHR	1.70	-157.15	Christmas Island	NOAA	flask	Conway et al. 1994
CIB	41.81	-4.93	Centro de Investigacion de la Baja Atmosfera	NOAA	flask	Conway et al. 1994
CPT	-34.35	18.49	Cape Point	NOAA	flask	Conway et al. 1994
CPT	-34.35	18.49	Cape Point	SAWS	insitu	Brunke et al. 2004
CRI	15.08	73.83	Cape Rama	CSIRO	flask	Francey et al. 2003
CRZ	-46.43	51.85	Crozet Island	NOAA	flask	Conway et al. 1994
CYA	-66.28	110.52	Casey	CSIRO	flask	Francey et al. 2003
DRP	-59.12	-63.63	Drake Passage	NOAA	ship flask	Conway et al. 1994
EGB	44.23	-79.78	Egbert	EC	insitu	Worthy et al. 2003
EIC	-27.15	-109.45	Easter Island	NOAA	flask	Conway et al. 1994
ESP	49.38	-126.54	Estevan Point	EC	insitu	Worthy et al. 2003
EST	51.66	-110.21	Esther	EC	insitu	Worthy et al. 2003
ETL	54.35	-104.98	East Trout Lake	EC	insitu	Worthy et al. 2003
FSD	49.88	-81.57	Fraserdale	EC	insitu	Worthy et al. 2003
GMI	13.39	144.66	Mariana Islands	NOAA	flask	Conway et al. 1994
GPA	-12.25	131.04	Gunn Point	CSIRO	flask	Francey et al. 2003

HBA	-75.61	-26.21	Halley Station	NOAA	flask	Conway et al. 1994
HDP	40.56	-111.65	Hidden Peak (Snowbird)	NCAR	insitu	Stephens et al. 2011
HPB	47.80	11.02	Hohenpeissenberg	NOAA	flask	Conway et al. 1994
HUN	46.95	16.65	Hegyhatsal	HMS	insitu	Haszpra et al. 2001
HUN	46.95	16.65	Hegyhatsal	NOAA	flask	Conway et al. 1994
INX	-86.02	39.79	INFLUX (Indianapolis Flux Experiment)	NOAA	flask	Conway et al. 1994
IZO	28.31	-16.50	Izana	NOAA	flask	Conway et al. 1994
IZO	28.31	-16.50	Izana	AEMET	insitu	Gomez-Pelaez et al. 2011
JFJ	46.55	7.99	Jungfraujoch	KUP	insitu	Uglietti et al. 2011
KAS	49.23	19.98	Kasprowy Wierch	AGH	insitu	Necki et al. 2003
KEY	25.66	-80.16	Key Biscayne	NOAA	flask	Conway et al. 1994
KUM	19.52	-154.82	Cape Kumukahi	NOAA	flask	Conway et al. 1994
LEF	45.95	-90.27	Park Falls	NOAA	insitu	Andrews et al. 2014
LJO	32.87	-117.26	La Jolla	SIO	flask	Keeling et al. 2005
LLB	54.95	-112.45	Lac La Biche	EC	insitu	Worthy et al. 2003
LLB	54.95	-112.45	Lac La Biche	NOAA	flask	Conway et al. 1994
LMP	35.52	12.62	Lampedusa	NOAA	flask	Conway et al. 1994
LUT	53.4	6.35	Lutjewad	RUG	insitu	van der Laan et al. 2009
MAA	-67.62	62.87	Mawson Station	CSIRO	flask	Francey et al. 2003
MEX	18.98	-97.31	High Altitude Global Climate Observation Center	NOAA	flask	Conway et al. 1994
MHD	53.33	-9.9	Mace Head	NOAA	flask	Conway et al. 1994
MHD	53.33	-9.9	Mace Head	LSCE	insitu	Ramonet et al. 2010
MID	28.21	-177.38	Sand Island	NOAA	flask	Conway et al. 1994
MLO	19.54	-155.58	Mauna Loa	NOAA	flask	Conway et al. 1994
MLO	19.54	-155.58	Mauna Loa	NOAA	insitu	Thoning et al. 1989
MNM	24.28	153.98	Minamitorishima	JMA	insitu	Tsutsumi et al. 2005

MQA	-54.48	158.97	Macquarie Island	CSIRO	flask	Francey et al. 2003
NAT	-5.51	-35.26	Farol De Mae Luiza Lighthouse	NOAA	flask	Conway et al. 1994
NMB	-23.58	15.03	Gobabeb	NOAA	flask	Conway et al. 1994
NWR	40.05	-105.59	Niwot Ridge	NOAA	flask	Conway et al. 1994
NWR	40.05	-105.59	Niwot Ridge	NCAR	insitu	Stephens et al. 2011
OTA	-38.52	142.82	Otway	CSIRO	flask	Francey et al. 2003
OXK	50.03	11.81	Ochsenkopf	NOAA	flask	Conway et al. 1994
PAL	67.97	24.12	Pallas-Sammaltunturi	NOAA	flask	Conway et al. 1994
PAL	67.97	24.12	Pallas-Sammaltunturi	FMI	insitu	Hatakka et al. 2003
POC			Pacific Ocean Cruise	NOAA	flask	Conway et al. 1994
PSA	-64.92	-64	Palmer Station	NOAA	flask	Conway et al. 1994
RPB	13.16	-59.43	Ragged Point	NOAA	flask	Conway et al. 1994
RYO	39.03	141.82	Ryori	JMA	insitu	Tsutsumi et al. 2005
SCT	33.41	-81.83	Beech Island	NOAA	insitu	Andrews et al. 2014
SEY	-4.68	55.53	Mahe Island	NOAA	flask	Conway et al. 1994
SGP	36.8	-97.5	Southern Great Plains	NOAA	flask	Conway et al. 1994
SHM	52.72	174.1	Shemya Island	NOAA	flask	Conway et al. 1994
SMO	-14.25	-170.56	Tutuila	NOAA	flask	Conway et al. 1994
SMO	-14.25	-170.56	Tutuila	NOAA	insitu	Halter et al. 1988
SNP	38.62	-78.35	Shenandoah National Park	NOAA	insitu	Andrews et al. 2014
SPL	40.45	-106.73	Storm Peak Laboratory (Desert Research Institute)	NCAR	insitu	Stephens et al. 2011
SPO	-89.98	-24.8	South Pole	NOAA	flask	Conway et al. 1994
SPO	-89.98	-24.8	South Pole	NOAA	insitu	Gillette et al. 1987
STR	37.76	-122.45	Sutro Tower	NOAA	flask	Andrews et al. 2014
SUM	72.6	-38.42	Summit	NOAA	flask	Conway et al. 1994
SYO	-69.01	39.59	Syowa Station	NOAA	insitu	Morimoto et al. 2003
TAP	36.73	126.13	Tae-ahn Peninsula	NOAA	flask	Conway et al. 1994
THD	41.05	-124.15	Trinidad Head	NOAA	flask	Conway et al. 1994
USH	-54.85	-68.31	Ushuaia	NOAA	flask	Conway et al. 1994

UTA	39.9	-113.72	Wendover	NOAA	flask	Conway et al. 1994
UUM	44.45	111.1	Ulaan Uul	NOAA	flask	Conway et al. 1994
WAO	52.95	1.12	Weybourne	UEA	insitu	Forster and Bandy, 2006
WBI	41.73	-91.35	West Branch	NOAA	insitu	Andrews et al. 2014
WGC	38.27	-121.49	Walnut Grove	NOAA	insitu	Andrews et al. 2014
WIS	30.86	34.78	Weizmann Institute of Science	NOAA	flask	Conway et al. 1994
WKT	31.32	-97.33	Moody	NOAA	insitu	Andrews et al. 2014
WLG	36.29	100.9	Mt. Waliguan	NOAA	flask	Conway et al. 1994
WSA	43.93	-60.02	Sable Island	EC	insitu	Worthy et al. 2003
YON	24.47	123.02	Yonagunijima	JMA	insitu	Tsutsumi et al. 2005
ZEP	78.91	11.89	Ny-Alesund	NOAA	flask	Conway et al. 1994

Table A2. Validation sites. Aircraft data collected by NOAA/ESRL (Sweeney et al., 2015) and NIES (Machida et al., 2008)

Site ID	Lat.	Lon.	Site/project name	Territory	Lab name
ACG	68	-165	Alaska Coast Guard	Alaska	NOAA
CAR	41	-104	Briggsdale	Colorado	NOAA
CMA	39	-74	Offshore Cape May	New Jersey	NOAA
CON			CONTRAIL	West Pacific	NIES
DND	47	-99	Dahlen	North Dakota	NOAA
ESP	49	-127	Estevan Point	British Columbia	NOAA
ETL	54	-105	East Trout Lake	Saskatchewan	NOAA
INX	40	-86	Indianapolis Flux Experiment	Indianapolis	NOAA
LEF	46	-90	Park Falls	Wisconsin	NOAA
HIL	40	-88	Homer	Illinois	NOAA
NHA	43	-71	Offshore Portsmouth	New Hampshire	NOAA
PFA	65	-148	Poker Flat	Alaska	NOAA
RTA	-21	-160	Rarotonga	Rarotonga	NOAA

SCA	33	-79	Offshore Charleston	South Carolina	NOAA
SGP	37	-98	Southern Great Plains	Oklahoma	NOAA
TGC	28	-97	Offshore Corpus Christi	Texas	NOAA

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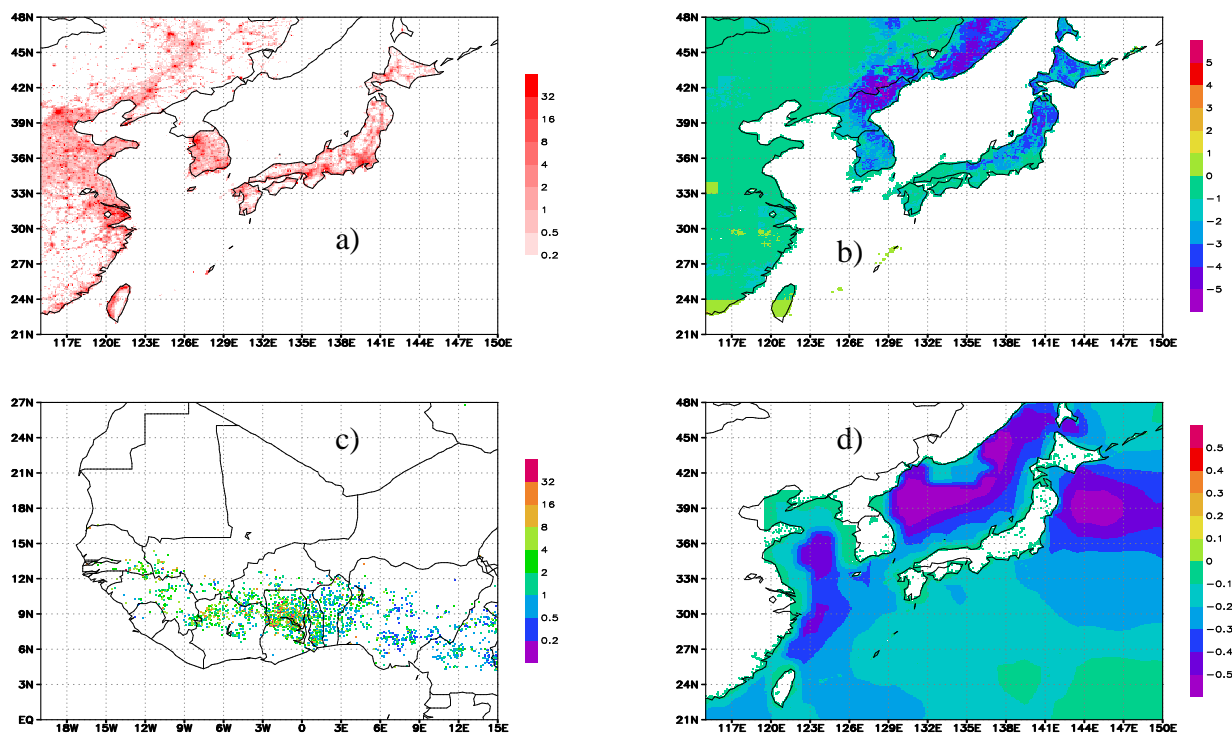
Figure 1: Prior CO_2 fluxes (unit: gCm^{-2}day^{-1}): a) emissions from fossil fuel burning by ODIAC (Jan 2011), b) fluxes from
15 **terrestrial biosphere by optimized VISIT model (day 160, Jun 9), c) emissions from biomass burning by GFAS in Africa (Jan 10, 2011), d) fluxes due to ocean-atmosphiere exchange by the OTTM assimilation model (Jan 2011)**
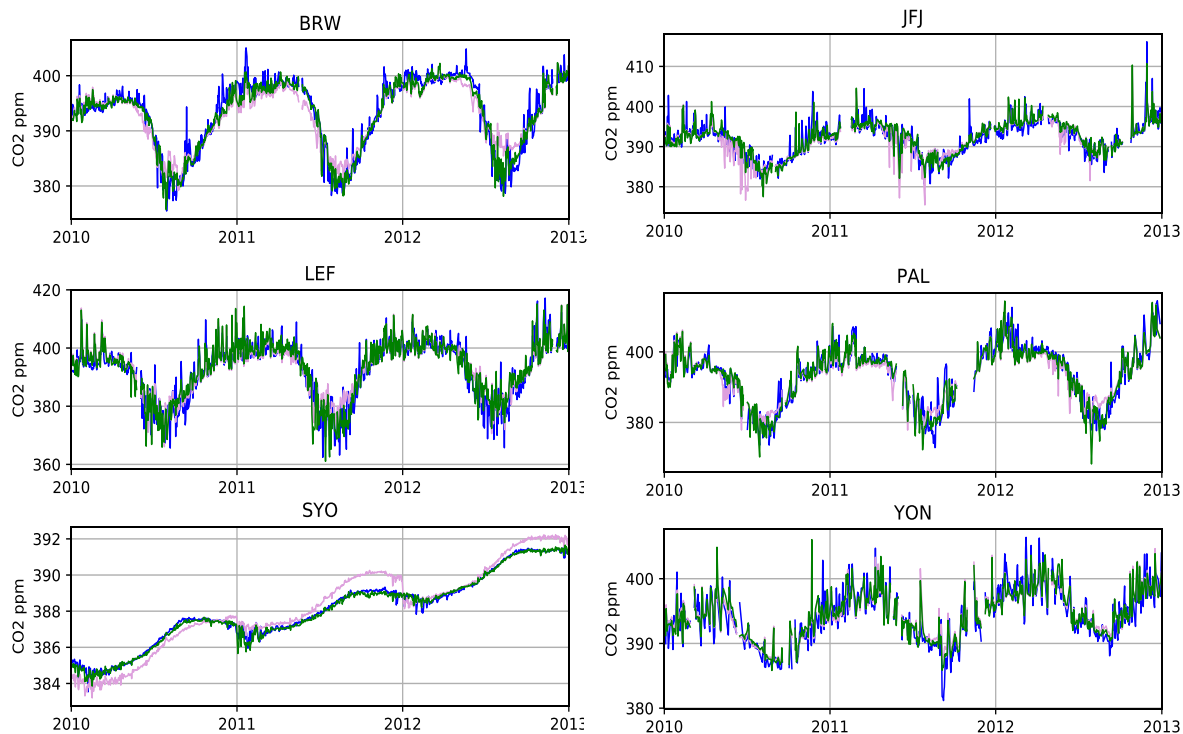
Figure 2: Time series of simulated and observed concentrations (blue - observed, plum -forward (unoptimized), green – optimized) at Barrow (BRW), Jungfraujoch (JFJ), Wisconsin (LEF), Pallas (PAL), Syowa (SYO), and Yonagunijima (YON).
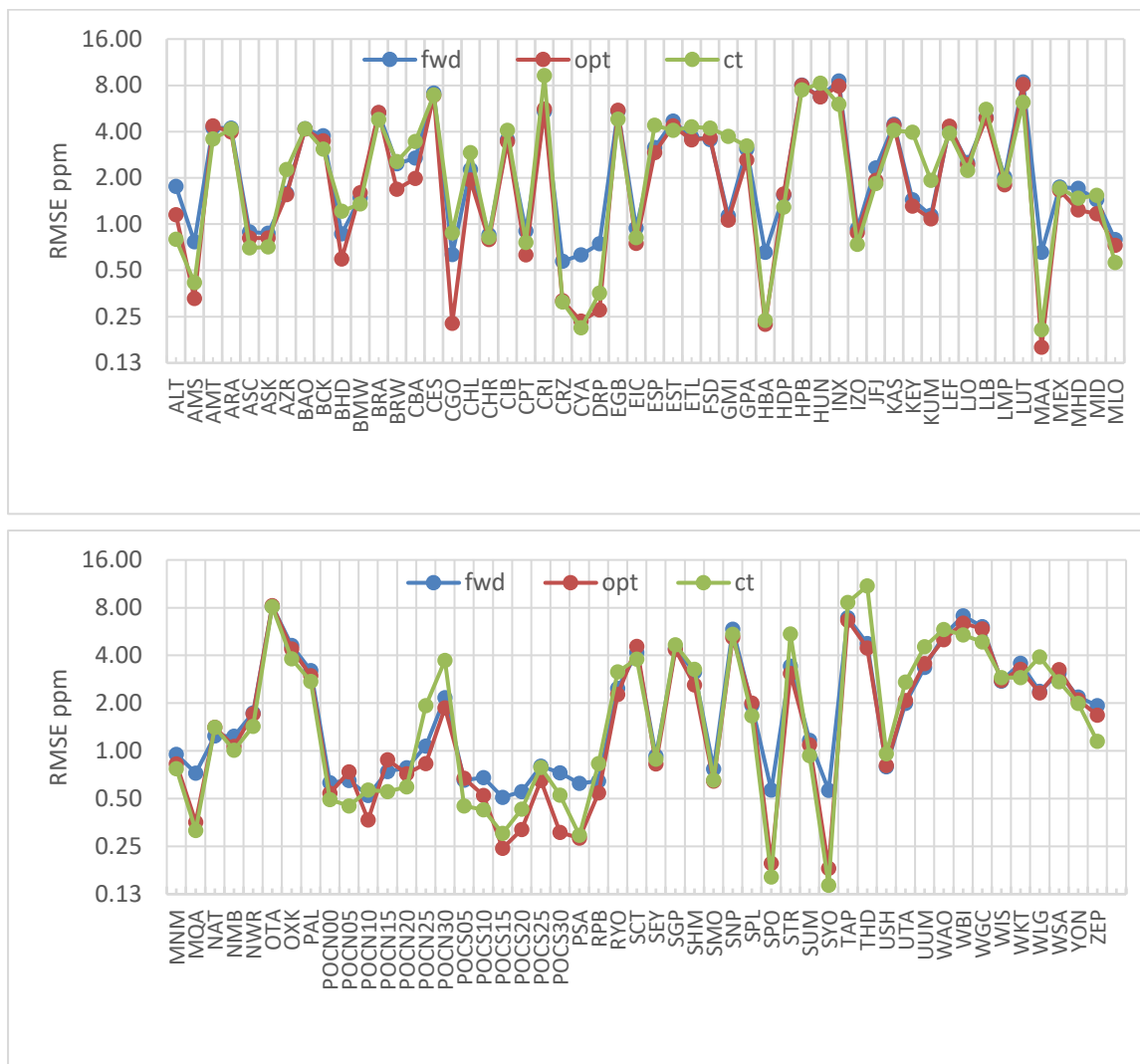
Figure 3: RMS difference between model and observations in 2010-2012 for (surface) sites included in inversion (blue – prior, red – optimized, green – CT2017)
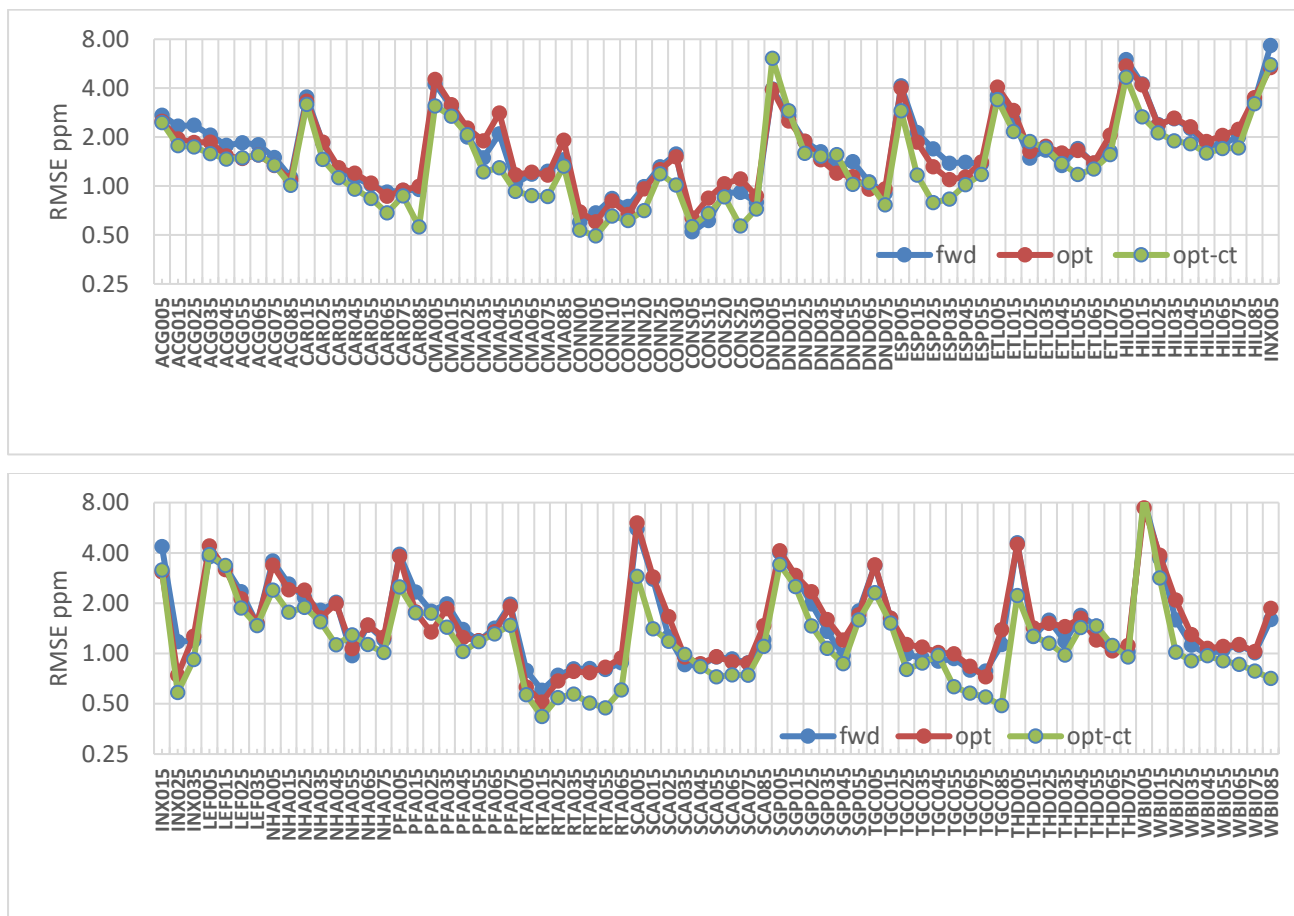
Figure 4: RMS difference between model and observations in 2010-2012 for aircraft sites, not included in inversion (blue – prior, red – optimized, green – CT2017)