Toward a long-term atmospheric CO₂ inversion for elucidating natural carbon fluxes: technical notes of NISMON-CO₂ v2021.1

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**Toward a long-term atmospheric CO₂ inversion for
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NISMON-CO₂ v2021.1**

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Abstract

Global estimates of carbon dioxide (CO₂) fluxes at the earth's surface are imperative to understanding carbon cycle mechanisms and making reliable predictions about the global warming. Furthermore, they can provide valuable science-based information for use in reducing human-induced CO₂ emissions. Inverse analysis is a prominent method for quantitatively estimating spatiotemporal variations in CO₂ fluxes; however, this technique involves a certain degree of uncertainty and requires technical refinement, especially to improve the horizontal resolution so that local fluxes can be compared with other estimates made at the regional or national level. In this study, a newly developed set of inversion schemes was incorporated into a state-of-the-art inverse analysis system named NISMON-CO₂. The introduced schemes involve grid conversion, observational weighting, and anisotropic prior error covariance, the details of which are described in this paper. In addition, pseudo-observation experiments were performed to investigate the effect of the new schemes, as well as to assess the reliability of

NISMON-CO₂ for long-term analysis with practical inhomogeneous observations. The experiment results demonstrate the utility of the grid conversion scheme for high-resolution flux estimates ($1^\circ \times 1^\circ$), and significant improvements were achieved through the observational weighting and anisotropic prior error covariance. Furthermore, the estimated seasonal and interannual variations in regional CO₂ fluxes were confirmed to be reliable, although there is some possible bias in terms of global land–ocean partitioning. These results are useful for interpreting flux variations that result from inverse analysis by NISMON-CO₂ ver. 2021.1 using real observations.

Keywords

Inverse analysis, Carbon dioxide, Carbon cycle, Atmospheric transport model, Data assimilation

1 Introduction

Assessing carbon dioxide (CO₂) fluxes at the earth's surface, both between the atmosphere and land and between the atmosphere and the oceans, is an issue of primary importance in studying the global carbon cycle. Within the atmosphere, the mole fraction of CO₂ has distinct seasonal variations derived from natural CO₂ fluxes that are superimposed on the human-induced trend of overall increasing CO₂. These natural fluxes are induced by dynamic photosynthesis and

respiration activity in the terrestrial biosphere and by sea–air CO₂ exchange driven by temperature, oceanic mixed layers, and biological activity. In addition to seasonal variations, these natural fluxes exhibit significant interannual variations in response to noticeable climate events such as El Niño (e.g., Keeling et al., 1995). On decadal scales, further trends of increase or decrease can be caused by changes in physical or biological conditions on land and in ocean areas. However, the mechanisms underlying natural CO₂ flux variations are not fully understood, resulting in significant uncertainties in global warming predictions made by earth system models equipped with climate–carbon cycle feedbacks (Friedlingstein et al., 2006).

An inverse analysis of atmospheric CO₂, in which an atmospheric transport model is used to connect the surface fluxes to the observed mole fraction of atmospheric CO₂, is a prominent method that quantitatively provides spatiotemporal variations of surface fluxes based on observations. In fact, a number of ground-based stations and other observational platforms have been performing atmospheric CO₂ measurements for several decades; through inverse analyses, they have revealed notable interannual variations of region-by-region CO₂ fluxes (Bousquet et al., 2000; Patra et al., 2005a,b; Baker et al., 2006; Chevallier et al., 2010; Peylin et al., 2013). Over the past decade, the four-dimensional variational method (4D-Var) and the ensemble Kalman smoother (or filter), which are well-known state-of-the-art data assimilation methods in the fields of meteorological and oceanic research, have become popular inverse methods and have made use of a wide variety of observations (Peters et al., 2007; Chevallier et

al., 2010; van der Laan-Luijkx et al., 2017; Niwa et al., 2017b; Liu et al., 2021). However, Friedlingstein et al. (2020) showed that inverse analyses have significantly large ranges of interannual flux variations, even for zonal means of certain latitudinal ranges, demonstrating the limited reliability of such inverse analyses.

However, in recent years, societal momentum to reduce CO₂ emissions has strengthened globally in response to the 1.5°C /2.0°C target of the Paris agreement, and CO₂ flux data from inverse analyses are becoming an important source of information used to independently evaluate national inventories. The latest studies by Chevallier (2021) and Deng et al. (2021) compared inversion fluxes with national inventories based on agriculture, forestry, and other land use changes (LUCs). However, they used data from global-scale inversions with horizontal resolutions generally around hundreds of kilometers. Therefore, further development is required to increase the horizontal resolution so that fluxes can be more consistently compared with national inventories. This is not a trivial task, because a several-decades-long analysis that would be required to investigate long-term trends is computationally demanding.

Niwa et al. (2017a,b) developed a sophisticated and computationally efficient inversion system based on the Nonhydrostatic Icosahedral Atmospheric Model (NICAM: Tomita and Satoh, 2004; Satoh et al., 2008, 2014). The CO₂ inverse analysis using this model is termed the NICAM-based Inverse Simulation for Monitoring CO₂ (NISMON-CO₂) (Niwa et al., 2021), and the CO₂ flux estimates made using this system were included in a multi-inversion

analysis conducted by the Global Carbon Project (GCP) (Friedlingstein et al., 2020, 2021). The general features of NISMON-CO₂ were described by Niwa et al. (2017b), who used pseudo-observations rather than real observations, generated using the same transport model as that used for the inverse analysis. However, that experiment was performed for only a 1-year-long period under ideal conditions; therefore, the practical performance of NISMON for long-term analysis has not yet been demonstrated.

In this paper, we describe the latest inversion settings of NISMON especially for those added to improve the inversion performance and address the aforementioned resolution issue. Furthermore, we performed a long-term inverse analysis using pseudo-observations that emulate actual observation networks. Here, we focused on the use of NISMON in investigating interannual variations of regional CO₂ fluxes and on the effectiveness of the newly introduced techniques. Apart from this study, an inverse analysis using NISMON was performed with the same inversion settings, but using real observations for 1990–2020, which is publicly available as version 2021.1 of NISMON-CO₂ (Niwa 2020). This paper additionally aims to present a detailed description of NISMON-CO₂ ver. 2021.1.

2 Methods

2.1 NICAM-TM and NISMON-CO₂

First, we describe the fundamental features of the NISMON-CO₂ inversion system. One prominent feature of NICAM, on which NISMON is based, is that it adopts an icosahedral grid system and uses the finite volume method to calculate the continuity equation (Tomita and Satoh, 2004). Thanks to the finite volume method, the NICAM-based transport model (NICAM-TM) can completely conserve tracer mass without any numerical mass fixer, which motivated the use of NICAM for simulating atmospheric transport of long-lived species such as CO₂ (Niwa et al., 2011). Simulations with NICAM-TM are performed iteratively in an inverse analysis using NISMON. Further details about NICAM-TM can be found in Niwa et al. (2017a).

The inverse analysis system NISMON uses 4D-Var to optimize surface CO₂ fluxes that are consistent with atmospheric observations (Niwa et al., 2017b). In 4D-Var, the cost function J , whose minimum state is sought, is defined as

$$J(\delta\mathbf{x}) = \frac{1}{2}\delta\mathbf{x}^T\mathbf{B}^{-1}\delta\mathbf{x} + \frac{1}{2}(M(\delta\mathbf{x}) - \mathbf{d})^T\mathbf{R}^{-1}(M(\delta\mathbf{x}) - \mathbf{d}), \quad (1)$$

where $\delta\mathbf{x}$ represents the model parameter to be optimized, which comprises deviations from its prior estimate $\mathbf{x}^{\text{pri}}$ and the initial global mole fraction offset. Meanwhile, $M(\cdot)$ is an operator that represents the model calculation, and $\mathbf{d}$ is an observational vector defined by the observational vector $\mathbf{y}$ as $\mathbf{d} = \mathbf{y} - M(\mathbf{x}^{\text{pri}})$. The matrices $\mathbf{B}$ and $\mathbf{R}$ are the error covariance matrices of prior

131 estimates and the model–observation mismatches, respectively. The iterative calculation of 4D-
 132 Var uses the gradient of the cost function
 133
$$\nabla_{\delta \mathbf{x}} J = \mathbf{B}^{-1} \delta \mathbf{x} + \mathbf{M}^T \mathbf{R}^{-1} (M(\delta \mathbf{x}) - \mathbf{d}), \quad (2)$$

 134 to find the optimal model parameters (i.e., posterior estimates) that minimize the cost function.
 135 $\mathbf{M}^T$ in Eq. (2) represents the transpose of $\mathbf{M}$, which is the tangent linear operator of $M(\cdot)$. In fact,
 136 the relation $\mathbf{M} \Delta \mathbf{x} \approx M(\Delta \mathbf{x})$ is satisfied because the model calculates atmospheric transport
 137 linearly and does not include any chemical reactions. In calculating $M(\cdot)$ and $\mathbf{M}^T$ in the forward
 138 and adjoint simulations, NICAM-TM only simulates CO_2 transport in the so-called offline
 139 mode with prescribed meteorological data. The meteorological data are prepared in the so-
 140 called online mode, in which meteorological fields are simulated by nudging horizontal winds
 141 toward those of reanalysis data. Here, we used the same settings for the model and the same
 142 prescribed meteorological data as those of Niwa et al. (2017a). The forward and adjoint
 143 transport simulations were performed with the horizontal grid level “glevel-5” of NICAM,
 144 whose mean grid interval is 223 km. The model contains 40 layers in vertical up to a height of
 145 approximately 45 km above the sea level.

146 The model operator $M(\cdot)$ includes linear processes other than atmospheric transport;
 147 these include spatiotemporal interpolation from the model grids and temporal steps to
 148 observational locations and times and the conversion of flux data from latitude–longitude grids
 149 to NICAM’s icosahedral grids. Therefore, the matrix $\mathbf{M}$ and its transpose can be written as

$$\mathbf{M} = \mathbf{H}\mathbf{M}'\mathbf{G}, \quad (3)$$

$$\mathbf{M}^T = \mathbf{G}^T\mathbf{M}'^T\mathbf{H}^T, \quad (4)$$

where $\mathbf{H}$ and $\mathbf{G}$ represent the spatiotemporal interpolation and the flux data conversion, respectively. The matrix $\mathbf{M}'$ is the integral calculation of atmospheric transport. This study newly introduces $\mathbf{G}$, which allows the flux data contained in $\delta\mathbf{x}$ to be arrayed in latitude–longitude grids, so the optimization can be performed in that space, but not in the model grid space (i.e., the icosahedral grids). The detailed conversion scheme is described in the next section. In NISMON, the optimization is performed using a quasi-Newton method-based scheme termed the Preconditioned Optimizing Utility for Large-dimensional analyses (POpULar: Fujii and Kamachi, 2003; Fujii, 2005).

160

161 **2.2 Grid conversion scheme**

The icosahedral NICAM grid shapes hexagons or pentagons, which requires an elaborate calculation to convert from latitude–longitude grids, in which the flux input datasets are originally prepared. In a series of the previous studies (Niwa et al., 2011a, 2012, 2017a,b, and 2021), we adopted simple area averaging, in which data from within a certain distance in the surrounding latitude–longitude grids are averaged. Afterward, flux values were slightly modified by scaling to ensure mass conservation. In this study, we updated to a more

sophisticated method of grid conversion, which no longer requires mass-conserving scaling and hence retains linearity.

The new method first divides the latitude–longitude grid data into a finer-scale latitude–longitude grid, as indicated in Fig. 1 (the bold dark-gray mesh becomes the thin light-gray mesh), i.e., rasterizing. Then, each finer grid is allocated to a hexagonal grid whose borders are trigonometrically defined in the latitude–longitude coordinates. In Fig. 1, the colored grid areas are allocated to the centered icosahedral grid, which derives contributions from the surrounding latitude–longitude grids. Consequently, the flux value converted into the i th icosahedral grid f'_i can be written as

$$f'_i = \sum_{j=1}^{N_i} a_{i,j} f(G_j) / A_i, \quad (5)$$

where G_j denotes the original latitude–longitude grid that overlaps the i th icosahedral grid and N_i is the number of those grids ($N_i = 9$ in the case of Fig. 1). The partial area of each original latitude–longitude grid overlapped by the i th icosahedral grid is derived from the finer latitude–longitude grids and represented as $a_{i,j}$ (individual colored areas in Fig. 1). The area of the i th icosahedral grid is denoted as A_i . Because every finer latitude–longitude grid is allocated to one of the surrounding icosahedral grids, the total amount of global data is conserved. Because of the linearity of Eq. (5) and the absence of any mass-conserving scaling, this grid conversion scheme is entirely linear and thus represented by the matrix $\mathbf{G}$ in Eq. (3). This property enables us to easily develop the adjoint code that represents $\mathbf{G}^T$ in Eq. (4). By introducing $\mathbf{G}$ and $\mathbf{G}^T$

into the model operator and its adjoint, respectively, the control variables can be arrayed in latitude–longitude grids.

Optimizing fluxes in the latitude–longitude grid has two technical benefits. (a) There is no need to convert flux data from icosahedral grids to latitude–longitude grids after inversion for comparison with other datasets. Such conversion often leads to deterioration of mass conservation and requires additional modification. (b) The flux data are provided at higher resolution than the icosahedral grid data. In this study, we optimized the flux data at a $1^\circ \times 1^\circ$ resolution, which is finer than that at the icosahedral grid used for the transport simulations (~ 223 km). This allows the optimized fluxes to retain high-resolution information originally contained in the prior flux data.

2.3 Flux model

The surface CO₂ fluxes input to NICAM-TM can be described as

$$\begin{aligned}
 f_{\text{CO}_2}(x, t) = & f_{\text{fos}}(x, t) - \beta_{\text{GPP}}(f_{\text{GPP}}(x, t) + \Delta f_{\text{GPP}}(x, t)) + \beta_{\text{RE}}(f_{\text{RE}}(x, t) + \Delta f_{\text{RE}}(x, t)) \\
 & + (1 + \Delta\alpha_{\text{LUC}}(x, t))f_{\text{LUC}}(x, t) + (1 + \Delta\alpha_{\text{fire}}(x, t))f_{\text{fire}}(x, t) \\
 & + f_{\text{ocn}}(x, t) + \Delta f_{\text{ocn}}(x, t),
 \end{aligned} \tag{6}$$

where x and t represent the flux location and time, respectively. Fluxes from fossil fuel use and cement production, gross primary production (GPP) and respiration (RE) of the terrestrial biosphere, LUC, biomass burning, and oceans are denoted as f_{fos} , f_{GPP} , f_{RE} , f_{LUC} , f_{fire} , and f_{ocn} ,

respectively; they are prescribed by flux datasets prepared in advance and here have a monthly temporal resolution.

In Eq. (6), we separate the terrestrial biosphere fluxes into GPP and RE components following the procedure of Lokupitiya et al. (2008), because GPP and RE vary differently. The coefficients β_{GPP} and β_{RE} are scaling factors that reproduce diurnal variations. β_{GPP} and β_{RE} distribute fluxes at a 3-hourly resolution from the monthly f_{GPP} and f_{RE} , respectively, whose values are derived from downward shortwave radiation at the earth's surface and 2 m height air temperatures data from the Japanese 55-year Reanalysis (JRA-55: Kobayashi et al., 2015; Harada et al., 2016). This is similar to the method of Olsen and Randerson (2004). The nature of the terrestrial biosphere causes β_{GPP} and β_{RE} have different diurnal variations (e.g., β_{GPP} is zero at night because photosynthesis is inactive, which it is not the case for β_{RE}).

In the 4D-Var calculation for this study, fossil fuel emissions were not optimized; i.e., they were fixed as f_{fos} , whereas the flux deviations of Δf_{GPP} , Δf_{RE} , and Δf_{ocn} were optimized. In addition, the LUC and biomass burning fluxes vary with scaling factors, whose deviations from 1, $\Delta \alpha_{\text{LUC}}$ and $\Delta \alpha_{\text{fire}}$, were optimized. In the optimization, therefore, the LUC and biomass burning fluxes were not allowed to increase where their basic fluxes are set to zero.

A monthly temporal resolution was chosen for the optimized flux parameters Δf and $\Delta \alpha$, except for the ocean flux Δf_{ocn} , whose temporal resolution is set annual. This is because ocean flux variations are generally small compared with those of terrestrial fluxes and some

erroneous values in the optimized ocean fluxes could arise because of “leakage” of terrestrial signals to oceans. An annual temporal resolution may help mitigate such flux leakages; however, this does not allow the inversion to optimize seasonal variations in ocean fluxes.

In version 2021.1 of NISMON-CO₂, we use fossil fuel emission data from the GCP-Gridded Fossil Emission Dataset (GCP-GridFED: Jones et al., 2021) version 2021.2 for f_{fos} , and the GPP, RE, and LUC data are taken from the Vegetation Integrative Simulator for Trace gases (VISIT: Ito and Inatomi, 2012; Ito, 2019, 2021) for f_{GPP} , f_{RE} , and f_{LUC} , respectively. The biomass burning emission data are taken from the Global Fire Emissions Database (GFED) version 4.1s (van der Werf et al., 2017) for f_{fire} , and the air–sea CO₂ exchange data are taken from the Japan Meteorological Agency (JMA) (Iida et al., 2015, 2021) for f_{ocn} . Except for f_{ocn} , although these data were originally provided at a higher resolution than $1^\circ \times 1^\circ$, here we uniformly aggregated the data at $1^\circ \times 1^\circ$ for ease of analysis.

Note that although the terrestrial flux is separated into GPP, RE, LUC, and biomass burning in this inverse analysis, they are not intended to be fully optimized independently. Inversion can identify drastic flux signals, such as a large-scale biomass burning event, as demonstrated by Niwa et al. (2021); however, estimating fluxes independently for each component is difficult in general because the inversion only uses observations of atmospheric CO₂ mole fractions. The separation of those fluxes is intended to represent the different diurnal

variations of GPP and RE and to consider future possible constraints such as those from carbonyl sulfide (Campbell et al., 2008).

2.4 Prior error covariance

A 4D-Var calculation requires not only a prior estimate but also its prior error covariance matrix, as denoted by $\mathbf{B}$ in Eqs. (1) and (2). In the inverse experiment of Niwa et al. (2017b), the prior error covariances (i.e., the off-diagonal elements of $\mathbf{B}$) were simply designed with a Gaussian function that is commonly used for the globe, in which a correlation scale length was arbitrarily determined. However, that is too simple an assumption, because the error covariances should dynamically vary in space and time according to the flux mechanisms.

In this study, we define the error covariance matrix using an ensemble set of prior fluxes as follows. First, we prepare a long-term flux dataset and consider each year of data as a member of the ensemble. Therefore, the number of years equals the number of ensemble members, and each member contains 12 temporal components for monthly data. For a prior flux component n , we approximate the covariance matrix using m members as

$$\tilde{\mathbf{B}}_{n,\text{cyc}} = \frac{1}{m-1} \sum_{k=1}^m \delta \mathbf{x}_n^k (\delta \mathbf{x}_n^k)^T, \quad (7)$$

where the subscript “cyc” represents cyclostationary, whose matrix is applied repeatedly for each year. The superscript k is an ensemble index, and $\delta \mathbf{x}_n^k$ describes the deviation of the flux of the k th member from the ensemble mean (i.e., the long-term mean). However, due to the

limited number of ensemble members (the number of years), erroneous correlations arise in remote areas, i.e., so-called sampling errors. Therefore, to localize covariances, we apply the Schur product of $\tilde{\mathbf{B}}_{n,cyc}$ with a correlation matrix $\mathbf{C}$ as

$$\mathbf{B}_{n,cyc} = \gamma_n \tilde{\mathbf{B}}_{n,cyc} \circ \mathbf{C}. \quad (8)$$

Here, we use the Gaussian function for each element of $\mathbf{C}$ as

$$C_{ij} = \exp \left(-\frac{l_{ij}^2}{L^2} \right), \quad (9)$$

where the indices i and j represent different flux locations, whose distance is denoted by l_{ij} . L is a constant and is set to 2000 km here. In practice, we let $C_{ij} = 0$ where $l_{ij} > 4000$ km to minimize the computational burden. In Eq. (8), the scaling factor γ_n is introduced as a tuning parameter to adjust the degree to which fluxes are subjected to change by observational constraints. Furthermore, we replaced the resulting negative covariances with zeros, as we consider negative correlations to be primarily generated by sampling errors. The technique described in Eq. (8) has been used as “the covariance localization” in an ensemble Kalman filter assimilation (Lorenc, 2003), but we have applied it here for the 4D-Var inverse analysis.

For the covariances of the prior terrestrial biosphere GPP and RE, we used a long-term VISIT simulation for 1901–2020, generating 120 members for the covariance calculation (Eq. 7). After applying the correlation matrix, the resulted covariance was inflated by $\gamma_n = 4$ so that the globally integrated variance is approximately $(2 \text{ Pg C yr}^{-1})^2$. The defined uncertainty of 2 Pg C yr^{-1} is derived from the difference between the global total net flux of the prior data

and that estimated from the growth rate of the atmospheric observations under the assumption that terrestrial flux is the dominant driver of the global flux variations. To determine the error covariance of the prior ocean flux, we used the JMA ocean flux data for 1990–2019. Here, because we optimize the annual means of ocean fluxes, an annual mean is considered one ensemble member (with 30 members in total). The covariance for the ocean fluxes was derived similarly as that of the terrestrial biosphere using Eqs. (8) and (9) with $\gamma_n = 2$; the integrated variance is approximately $(0.17 \text{ Pg C yr}^{-1})^2$; this number is rather arbitrary compared with that for the terrestrial flux, but it was determined by several trial experiments performed beforehand.

For the other flux components, LUC and biomass burning, we do not consider error correlations and define each prior flux error covariance matrix $\mathbf{B}_{n,\text{cyc}}$ as a diagonal matrix, whose diagonal elements (i.e., variance) are all set to 4.0; this yields errors of 200%. We combined all $\mathbf{B}_{n,\text{cyc}}$ and applied it to every year, whose values are arrayed in the same grid and resolution as those of the prior flux, i.e., $1^\circ \times 1^\circ$. Then, by including the variance of the global initial mole fraction offset of $(0.5 \text{ ppm})^2$, we finally obtained the full matrix of $\mathbf{B}$.

Examples of the prior flux error covariance are given in Fig. 2, which shows error correlations ($= B_{ij}/(B_{ii}^{1/2} B_{jj}^{1/2})$) of a given prior GPP for April and July. In both months, the original error correlations were significant, even at a great distance. That feature is more pronounced in July, whereas larger correlations are present at a short distance in April (gray crosses in Fig. 2a and b). After performing the calculation in Eq. (8), those correlations become

localized, whereas the aforementioned feature is maintained (red crosses in Fig. 2a and b). The error correlations generated in this way have anisotropic spatial structures, as shown in Fig. 2c-f. Furthermore, they differ by month and location. In fact, these anisotropic correlation features arise from year-to-year variations in the long-term GPP data of VISIT, which are driven by changes of meteorological conditions (shortwave radiation, temperature, moisture etc.). We expect them to reflect actual biogeochemical mechanisms better than the simple isotropic correlations previously used.

2.5 Observations

The observational data input to NISMON was derived from the dataset named Observation Package (ObsPack)-GLOBALVIEWplus and the related near-real-time version ObsPack-NRT, provided through the National Oceanic and Atmospheric Administration (Masarie et al., 2014). In addition, we used other independently provided data; all data are listed in Niwa (2020). In version 2021.1 of NISMON-CO₂, versions 6.1_2021_03-01 (Schuldt et al., 2021a) and 6.1.1_2021-05-17 (Schuldt et al., 2021b) of ObsPack-GLOBALVIEWplus and ObsPack-NRT, were used, respectively. To demonstrate the potential performance of NISMON, we performed pseudo-observation inversions for this study, for which we emulated observational data at actual sites and times. The locations of the observations are shown in Fig. 3. For this dataset, we used all available observations provided from selected institutes (Niwa, 2020), of which

some sites have a constant data record over the entire period and some sites do not. Therefore, many temporal observational gaps are present; we used the pseudo-observation experiment to investigate the effect of these data gaps on flux estimations.

2.6 Observation–model mismatch errors

We defined the observation-model mismatch error covariance matrix $\mathbf{R}$ as

$$R_{ii} = \frac{1}{(\beta r_i)^2 N_i}, \quad (10)$$

where r_i denotes the standard deviation of the mole fractions around the i th observation and N_i represents the number of observations within a certain spatiotemporal range of the i th observation. Here, we apply a spatiotemporal range of within 1 week, a 1000-km horizontal diameter circle, and a 1-km vertical depth. Furthermore, we introduce a scaling factor β so that $\chi^2 (:= \frac{2J_{\min}}{m})$, where m is the number of observations and $J_{\min}$ is the minimum of the cost function, should be <1 (Tarantola, 2005). We calculated r_i from simulated daily mole fraction variations of CO_2 at the site of observation i ranging from 1 week before and 1 week after the observational timing; the simulation was performed using NICAM-TM with prior fluxes. As denoted by Eq. (10), $\mathbf{R}$ is a diagonal matrix, which assumes that all observations are independent from each other. However, this is a simplistic assumption, especially where observations were obtained with high density. For such areas, we therefore inflate the variances by introducing N_i , which lessens the weights of such dense observations in space and time. For

instance, this variance inflation prevents continuous *in situ* observations from imposing overly strong constraints compared with discrete flask sampling observations. For NISMON-CO₂ ver.2021.1, we adopted a strategy to use as many available observations as possible. In fact, this led to duplicated observations, because several institutes share the same observational platforms; N_i places a single-site constraint on such collocated observations as well.

2.7 Pseudo-observation inversion experiment

To investigate the general performance of NISMON and the effectiveness of the newly introduced inversion settings, we performed an inverse analysis under ideal conditions; that is, we know the “true” state of fluxes. We prepared two datasets for the true and prior fluxes. First, we performed a forward transport simulation of atmospheric CO₂ with the true flux dataset and extracted the simulated CO₂ mole fractions at the aforementioned observational points and times. Then, we performed inverse experiments incorporating those pseudo-observations and optimized the fluxes from the prior estimates. A similar experiment was conducted by Niwa et al. (2017b); however, the experiment in this study was designed more practically, using a mixture of continuous *in situ* and discrete flask measurements (Fig. 3) and covering a longer analysis period. Furthermore, by virtue of the grid conversion scheme implemented, we optimized fluxes in a latitude–longitude grid of $1^\circ \times 1^\circ$, rather than in the icosahedral grid.

The forward simulation used to generate the pseudo-observations (true run) employed the same NICAM-TM settings used for the inversion, except for the horizontal resolution. The horizontal resolution is “glevel-6”, whose mean grid interval is 112 km. This is one level higher than that used for the inverse analysis (glevel-5, 223 km). Therefore, the true run can simulate much smaller mole fraction variations. This can provide insights into the influence of errors in the model representativeness, although the resolution is not high enough to fully consider this aspect.

The true fluxes used here were taken from the posterior fluxes of NISMON-CO₂ ver. 2021.1 (Niwa, 2020), which were produced from real observations; therefore, the generated pseudo-observations of atmospheric CO₂ mole fractions have realistic seasonal and interannual variations and the true fluxes are assumed to reflect this as well. We made moderate changes to the prior fluxes from those in NISMON-CO₂ v2021.1 (described in Section 2.3), except for the fossil fuel emissions, which were taken from the same GCP-GridFED dataset. For the terrestrial fluxes (f_{GPP} , f_{RE} , f_{LUC} and f_{fire}), we used the same VISIT and GFEDv4.1s datasets as in NISMON-CO₂ v2021.1, but they were climatological ones averaged for 2006–2018. Meanwhile, the ocean fluxes (f_{ocn}) were based on different ocean flux data from Takahashi et al. (2009), which provide climatological fluxes for the reference year 2000. Therefore, other than the fossil fuel emissions, the prior fluxes do not contain interannual variations.

The inversion calculation was performed over the period from January 2007 to March 2018, where the first 12 months and last 3 months were disregarded as the spin-up and spin-down times of the analysis, respectively. Therefore, the inversion fluxes were evaluated over a 10-year period. It should be noted that this analysis period is almost three times shorter than that of NISMON ver. 2021.1 (1990–2020) for the ease of performing the multiple inverse experiments described in the next section.

2.8 Sensitivity tests

In addition to the control experiment that was performed with the aforementioned settings (referred as CTL), we performed four inversions with different settings. The first (referred as NO_ERR) was conducted using the pseudo-observations produced by the model with the same horizontal resolution as the inversion, which ensures that the model representation error is no longer present, i.e., a so-called identical twin experiment, which is the experiment performed by Niwa et al. (2017b). The second inversion (referred as ICO) was performed by optimizing fluxes on the icosahedral grids, as done previously. The third experiment (referred as NO_WTS) did not consider observational weighting applied to the observation–model mismatch error, i.e., $N_i = 1$ in Eq. (10). Here, for the fair comparison with the CTL case, we changed β in Eq. (10) so that the cost function (Eq. 1) is nearly equivalent to that of CTL at the beginning of the iterative calculation. The fourth inversion (referred as ISO_ERR) employed

the isotropic prior error covariance that was simply defined using a Gaussian function with correlation scale lengths of 500 km and 1000 km for land and ocean fluxes, respectively, which was also previously adopted (Niwa et al., 2017b). In this experiment, also for the purpose of fair comparison, each γ_n in Eq. (8) was tuned so that the global integrated value of the resulting prior error covariance was nearly equivalent to that of CTL.

3 Results

3.1 Cost function and number of iterations

In the inverse analysis of Niwa et al. (2017b), the optimization for estimating 12 monthly fluxes required approximately 50 iterations. That experiment assumed weekly observation sampling at 65 sites. The optimization would be expected to require more than 50 iterations when using an expanded observation network with over a hundred sites consisting of continuous measurements and flask air samplings (Fig. 3). In fact, it requires a few of hundreds of iterations. Figure 4 shows the cost function changes from the beginning to the 300th iteration for the CTL experiment and the other four sensitivity cases. As Fig. 4 shows, all cost functions decrease smoothly but do not fully converge at the 300th iteration. However, we confirmed that seasonal and interannual variations in the optimized fluxes, which will be discussed later on, had mostly but not perfectly converged at the 300th iteration. Hereafter, we show results from fluxes optimized by 300 iterations for all experiments.

412

413 **3.2 CO₂ flux maps and comparison with the icosahedral grid optimization**

414 An example of the optimized fluxes is presented in Fig. 5, showing the distributions of CO₂
415 fluxes averaged for July 2011. (Fossil fuel emissions are excluded.) Here, we compare the
416 optimized flux of CTL with that of ICO as well as the true (i.e., NISMON-CO₂ v2021.1) and
417 prior fluxes. It can be clearly seen from the continental sinks in Eurasia and the east side of
418 North America, significant sources in central Africa and South Asia, and the southwest side of
419 North America that both the CTL and ICO cases successfully retrieved the true flux pattern
420 from the prior fluxes, which exhibit very different patterns. Furthermore, the pattern of ocean
421 fluxes was also retrieved to some extent, although the ocean fluxes were optimized annually.
422 The tongue shape of the sources in the eastern equatorial Pacific, which is persistent throughout
423 an entire year, was suitably retrieved and its source strength was increased toward the true
424 fluxes, even though the ocean fluxes are much smaller than the terrestrial fluxes. Furthermore,
425 it should be noted that CTL, in which fluxes were optimized in the latitude–longitude grid,
426 showed a similar global distribution to the ICO case while resolving smaller flux distributions.
427 This fact demonstrates that the latitude–longitude grid optimization newly introduced in this
428 study works reasonably and could provide more highly resolved fluxes than the resolution of
429 the atmospheric transport model with the help of prior flux datasets.

430

3.3 Seasonal and interannual variations of regional CO₂ fluxes

Here we present seasonal and interannual variations in CO₂ fluxes by different numbers of iterations to demonstrate how those temporal variations converge as the iterative calculation proceeds. Figure 6 shows the seasonal variations of non-fossil fuel CO₂ fluxes in the 11 TransCom terrestrial regions (Gurney et al., 2002; Baker et al., 2006) averaged for 2008–2017. In most areas, the optimized seasonal variation is highly consistent with that of the true flux. Furthermore, the optimization converges well within 50 iterations for northern high-latitude areas, which contain dense observations (Europe, Boreal Asia, and Boreal N. America). Even in other regions, the flux variations converged within 100 iterations.

However, for interannual variations, convergence requires more iterations. In Fig. 7, interannual variations of the regional CO₂ fluxes had not converged at 100 iterations, even in northern high-latitude areas. Toward the 300th iteration, the interannual variations converge much more slowly than the seasonal variations do. Furthermore, significant differences still exist between 200 and 300 iterations (e.g., Boreal N. America), suggesting that 300 iterations may not be fully sufficient for convergence. That is why the cost function continues to decrease at the 300th iteration (Fig. 4).

Nevertheless, after 300 iterations, the posterior flux shows interannual variations consistent with those of the true flux, especially in northern high-latitude areas. In other areas, notable peaks and troughs of the interannual variations were more or less well retrieved,

although some noticeable errors are present (e.g., consistently smaller than the true flux in South America).

The slower convergence speed of the interannual variations than that of the averaged seasonal variations is attributed to their small degree of variation. For example, the fluxes in the northern high-latitude areas have a seasonal amplitude of approximately 10 Pg C yr^{-1} , whereas their annual averages vary within approximately 1 Pg C yr^{-1} . These results indicate that such small flux variations require a large number of iterations to be optimized. This is also true for the ocean fluxes. Figure 8 shows yearly variations in ocean fluxes for the 11 TransCom ocean regions. Although the posterior ocean fluxes changed even more gradually, they approached the true fluxes from the prior data. In fact, the ocean flux variations are much smaller than those of terrestrial fluxes; the range of interannual variations is at most 0.4 Pg yr^{-1} . However, the inversion mostly reproduced those interannual variations and 10-year trends (generally decreasing trends), although some areas contain persistent biases (approximately $0.1\text{--}0.2 \text{ Pg C yr}^{-1}$). However, the posterior fluxes do not fully represent the recent uptake increases in the North Pacific and North Atlantic (Fig. 8b and d) despite the fact that those regions have relatively dense observation networks of marine stations and ships (Fig. 3). This is probably because these areas are located on the downwind sides of the continents. Terrestrial flux signals from the continents are transported over the oceans with larger magnitudes than

those of ocean fluxes, which might make it difficult to clearly distinguish ocean fluxes in the inversion.

Figures 6–8 also show seasonal and interannual variations in the posterior fluxes that were derived under the assumption of no model error (NO_ERR, blue dotted line). As shown in Fig. 6, the posterior flux of NO_ERR shows nearly the same patterns in terrestrial seasonal variations as CTL. Although the NO_ERR case is better than the CTL case for the interannual variations of the terrestrial fluxes, it does not perfectly follow the true flux (Fig. 7). Conversely, NO_ERR shows little improvement in the interannual variations over the oceans (Fig. 8). Those NO_ERR features can be confirmed by Figs. 9 and 10, in which the root-mean-square errors of the yearly posterior flux variations for NO_ERR are better over terrestrial areas and comparable with those of CTL over the oceans (see blue and cyan bars).

3.4 Global land–ocean partitioning

The global total, land, and ocean budgets are summarized in Table 1, and their interannual variations are depicted in Fig. 11. For 2008–2017, the global mean true sinks over the land and the ocean are 2.62 and 2.04 Pg C yr⁻¹, respectively (estimated using NISMON-CO₂ v2021.1 with real observations), and the prior estimates are 4.61 and 1.41 Pg C yr⁻¹, respectively. Meanwhile, the inversion estimates by CTL are 3.39 and 1.29 Pg C yr⁻¹, respectively; that is, a bias of approximately 0.8 Pg C yr⁻¹ exists in the estimated global land–ocean partitioning,

whereas the global total sink is nearly identical to that of the true flux; i.e., mass conservation is ensured. In fact, this bias is coherently present over the entire analysis period (Fig. 11). Nevertheless, Fig. 11 also shows that the inversion successfully reproduced the interannual variation patterns from the constant prior estimates, even for the ocean, which changes moderately. Furthermore, Fig. 11 indicates that 100–150 iterations are required for convergence of the land–ocean partitioning, which are less than those required for estimates of regional interannual variations (Figs. 7 and 8). Compared with the CTL case, the NO_ERR case does not necessarily provide better estimates of land–ocean partitioning and displays the same coherent bias and similar interannual variations (Table 1 and Fig. 11).

3.5 Sensitivity to inversion settings

As shown in Table 1, the root-mean-square error for the overall fluxes during 2008–2017, which were calculated against the true fluxes, was smallest in NO_ERR ($2.74 \times 10^{-1} \text{ gC m}^{-2} \text{ day}^{-1}$), and the second smallest value was obtained in CTL ($3.49 \times 10^{-1} \text{ gC m}^{-2} \text{ day}^{-1}$). The smaller root-mean-square error of NO_ERR compared with that of CTL is attributed to the fluxes over land, indicating that the horizontal resolution of the atmospheric transport is more influential in estimates of terrestrial fluxes.

The other sensitivity tests, ICO, NO_WTS, and ISO_ERR, provided similar values for global land and ocean uptakes, i.e., similar biases; however, their root-mean-square errors for

the overall fluxes are larger than those of CTL (Table 1). In the ICO case, the larger root-mean-square error comes from the terrestrial fluxes, a feature that can be also found in interannual flux variations (see Fig. 9). This result indicates that the latitude-longitude optimization introduced in this study not only provided the higher-resolution fluxes but also improved the inversion estimates. The NO_WTS case shows larger root-mean-square errors not only over the land but also over the ocean, which is true for the overall fluxes (Table 1) and regional interannual variations (Figs. 9 and 10). This impact is the largest among all the sensitivity tests, suggesting that the spatiotemporal weighting of the observation–model mismatch errors plays an important role in the inversion. The ISO_ERR case also has a larger root-mean-square error than the CTL case for the overall terrestrial fluxes (Table 1). However, this is not necessarily the case for the regional interannual variations; ISO_ERR has smaller errors in some regions than CTL (Fig. 9). These results suggest that the anisotropic error covariance improved small-scale variations of the estimated flux.

4 Discussion

In this study, temporal variations in CO₂ fluxes were retrieved well, demonstrating the reliability of the flux estimates by NISMON. However, areas in the southern hemisphere and tropical areas still have significant errors in flux estimates, which are probably due to the sparse observational network there. However, it is promising that the weakly decreasing trend was

retrieved in some ocean regions, because detecting such small flux changes is important for monitoring carbon sink capacities of natural reservoirs and to provide insights into the future global carbon cycle under warming condition. The persistent bias in the global carbon budgets suggests difficulty in separating the land and ocean budgets. This land–ocean partitioning error might be mitigated by using additional O₂/N₂ data, as Rödenbeck et al. (2008) did, but this is left for future studies.

It should be noted here that we did not select observational sites based on their temporal coverage. Therefore, well-reproduced interannual variations indicate that such temporal gaps in the observational data would not significantly affect the estimation of interannual variations. In other words, some erroneous interannual variations might be affected by the observational gaps. As shown in Figs. 6 and 7, this study demonstrated that the interannual variations in those large-scale regions can be well reproduced in general, suggesting that small the observational gaps have small influences. In contrast, some inversion studies select only observational sites that have sufficient temporal coverage (e.g., Chevallier et al., 2010). However, observational constraints may differ not only in terms of temporal data coverage but also by meteorological conditions. When wind direction at a site changes from year to year, its observational constraints may also change. As such, our inversion strategy is to use as many observations as possible without considering any site-specific selection in terms of temporal data coverage. This study demonstrates that such a strategy is reasonable.

In contrast to the previous study of Niwa et al. (2017b), we used a higher-resolution model to prepare pseudo-observations (~112 km). This can provide some insight into the representative error of the model, although the actual degree of the model representation error is likely much larger. The inversion performance in retrieving seasonal and interannual variations in surface fluxes shown here is probably overestimated because of the small model errors. Nevertheless, the difference between CTL and NO_ERR offers valuable insight into the effect of model resolution, from which we can predict a degree of improvement in a future inversion with a higher-resolution model.

One major update to the inversion settings of Niwa et al. (2017b) is the implementation of a grid conversion in the inverse calculation, which enables a high-resolution latitude–longitude grid optimization. The computational time needed for the grid conversion is minute, and the convergence speed is almost the same as that of ICO (Fig. 4); therefore, this implementation does not induce any significant increase in computational costs. Because it uses the same transport model resolution as before, the amount of information that can be extracted from the observations does not necessarily increase. Nevertheless, it is valuable because the original information in the prior flux datasets does not deteriorate. Furthermore, such highly resolved fluxes would be more useful for regional- or national-scale budget analyses, demand for which has recently increased for use in monitoring and reducing national carbon emissions. This scheme has a broad utility, because it is also applicable to inversion systems that use a

conventional latitude–longitude grid model. The grid conversion scheme can be applied to convert from higher- to lower-resolution latitude–longitude grids.

Other updates to the inversion settings include the observational weighting and the anisotropic prior error covariance. Although both of these updates improved the inversion results, the impact of observational weighting is much larger. Weighting is performed using Eq. (10) with three parameters (horizontal and vertical lengths and temporal range for counting nearby observations), which is much simpler than changing the model resolution or the prior flux error covariance design. Therefore, modifying this equation or parameter sweeping would be an efficient way to improve the inversion. The impact of the anisotropic prior error covariance is comparable with that of the observational weighting on the root-mean-square-error of the overall fluxes (Table 1), whereas the root-mean-square errors of the regional temporal variations are marginal (Figs. 9 and 10), suggesting improvements in small-scale spatial variations. In fact, the better result from CTL than ISO_ERR is partially attributed to the fact that the true flux was derived from the inversion flux of NISMON v2021.1, which used the same inversion settings, including the prior error covariance, as those of CTL. This might have given CTL some advantages. Nevertheless, the difference between CTL and ISO_ERR suggests potential improvements made by the anisotropic error covariance; inversion estimates could be improved by shaping the error covariance according to real flux variations. Because the prior error covariance is derived from the long-term prior flux datasets, prior flux datasets have

become more important for inverse analyses by NISMON. Furthermore, the anisotropic error covariance is also valuable for providing high-resolution fluxes with the grid conversion scheme. If isotropic error covariance is used, the patterns of flux increments from prior fluxes would be blurred even when the optimization is performed in the latitude–longitude grid, because the isotropic error covariance works on spatial smoothing. However, as shown in Fig. 2, the anisotropic error covariance maintains the fine scale patterns derived from flux dynamics.

5 Conclusions

In this paper, we described updated inversion settings for NISMON-CO₂. Furthermore, to demonstrate the more practical application of an inverse analysis over a long-term period, we conducted 10-year inversion experiments with pseudo-observations that emulate an inhomogeneous observation network consisting of continuous measurements and flask air samplings with actual data gaps. Although the land–ocean partitioning contained a persistent global bias, the seasonal and interannual variations were sufficiently retrieved in the experiment, demonstrating the reliability of the NISMON-CO₂ v2021.1 product. Furthermore, sensitivity experiments elucidated the flux estimation improvements attributed to grid conversion, observational weighting, and anisotropic prior error covariance, which have been newly introduced to NISMON here. These schemes will be used as the default settings of NISMON-

600 CO₂ and can also be applied to inversion analyses of other atmospheric constituents (e.g.,
601 methane).

602 Since the adoption of the Paris Agreement in 2015, the world, for the first time, is
603 looking in the same direction to reduce emissions of greenhouse gases with the common goal
604 of limiting global warming to well below 2°C, preferably below 1.5°C, compared with pre-
605 industrial level. To verify achievements in CO₂ emission reductions, consecutive evaluations
606 of surface CO₂ fluxes is imperative. To this end, we will operationally conduct inversion
607 analysis using NISMON-CO₂ and provide science-based estimates of CO₂ fluxes, as well as
608 generating further developments in the system.

609

610 **Abbreviations**

611 NICAM: Nonhydrostatic Icosahedral Atmospheric Model

612 NISMON-CO₂: NICAM-based Inverse Simulation for Monitoring CO₂

613 NICAM-TM: NICAM-based Transport Model

614 4D-Var: four-dimensional variational method

615 GCP: Global Carbon Project

616 GPP: gross primary production

617 RE: respiration

618 LUC: land use change

619 GridFED: Gridded Fossil Emission Dataset

620 VISIT: Vegetation Integrative Simulator for Trace gases

621 GFED: Global Fire Emissions Database

622 JMA: Japan Meteorological Agency

623 ObsPack: Observation Package

624

625 **Declarations**

626 **Availability of data and material**

627 The following flux datasets used in this study are available at:

628 GridFED v2021.2: <https://doi.org/10.5281/zenodo.5565199>

629 VISIT: <https://doi.org/10.17595/20210521.001>

630 JMA ocean flux:

631 https://www.data.jma.go.jp/gmd/kaiyou/english/co2_flux/co2_flux_data_en.html

632 GFED v4.1s: <https://www.globalfiredata.org/>

633 NISMON-CO₂ v2021.1: <https://doi.org/10.17595/20201127.001>

634

635 **Competing interests**

636 The authors declare that they have no competing interest.

637

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Authors' contributions

YN conceived and designed the study and carried out the inversion experiments. KI developed the grid-conversion scheme. AI and YI made the important prior flux data of VISIT and the JMA ocean flux, respectively, and helped to interpret land and ocean flux variations in the analysis. All authors read and approved the final manuscript.

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669

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860 **Figure legends**

Figure 1. Illustration of flux conversion from latitude-longitude grids to icosahedral grids. The black line indicates boundaries of the icosahedral grids, and dark gray and light gray lines represent original and finer latitude-longitude grids, respectively. In the conversion, fluxes in colored areas are allocated to the centered icosahedral grid.

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Figure 2. Error correlations of a prior GPP flux at near 60.5°N, 100.5°E along with distance for April (a) and July (b). Gray and red crosses denote correlations before and after the localization (Eq. 8), respectively. Spatial structures of the localized error correlations of (a) and (b) are

shown in (c) and (d), respectively. (e) and (f) are the same as (c) and (d) but for a different prior GPP flux at near 30.5°N, 110.5°E.

Figure 3. Locations of observations used in the pseudo-observation inversion experiment. The observations are the same as those of NISMON-CO₂ v2021.1 (Niwa, 2020), but for those available during 2007-2018. Flask air sampling and in-situ observations at ground-based stations are depicted by closed magenta and open purple triangles, respectively. Shipboard observational locations are denoted by light blue circles. Tower observation sites are denoted by cyan crosses.

Figure 4. Cost function changes with iterations for the CTL case (blue) and the other four sensitivity tests of NO_ERR (light blue), ICO (light green), NO_WTS (orange), and ISO_ERR (pink).

Figure 5. Distributions of CO₂ fluxes averaged for July 2011 from the true (a) and prior (b) flux datasets, and from the posterior fluxes of the CTL (c) and ICO (d) cases. White color areas depict fluxes close to zero. Here, the fossil fuel emissions, which were not optimized, were excluded.

Figure 6. Monthly variations of non-fossil fuel CO₂ fluxes averaged for 2008–2017 for the 11 terrestrial regions of TransCom (Gurney et al., 2002; Baker et al., 2006). Light gray and dark gray solid lines are those of the prior and true fluxes, respectively. The other colors indicate posterior fluxes from different iterations (from 10 to 300) of CTL. Blue dot line denotes the 300th iteration result in the case where model errors do not exist (NO_ERR).

Figure 7. The same as Fig. 6, but for yearly variations from 2008 to 2017

Figure 8. The same as Fig. 7, but for the 11 ocean regions of TransCom

Figure 9. Root-mean-square errors (RMSE) of the yearly fluxes against the true fluxes for the TransCom terrestrial regions. The results of the CTL and the other four sensitivity tests are shown.

Figure 10. Same as Fig. 9, but for the TransCom ocean regions

Figure 11. Yearly variations of the global land (a), ocean (b) and total (c) CO₂ fluxes (fossil fuel emission are not included). Light gray and dark gray solid lines are those of the prior and true fluxes, respectively. The other colors indicate posterior fluxes from different iterations

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Tables

Table 1. Global scale net sinks and errors for 2008-2017. The sink values do not include fossil fuel emissions. The errors are derived from root-mean-square errors against the true fluxes over the land, the ocean, and the global total.

Flux data	Sinks			Root-mean-square errors		
	[Pg C yr ⁻¹]			[× 10 ⁻¹ g C m ⁻² day ⁻¹]		
	Land	Ocean	Total	Land	Ocean	Total
True	2.62	2.04	4.67	n/a	n/a	n/a
Prior	4.61	1.41	6.02	5.92	0.33	5.93
CTL	3.39	1.29	4.68	3.48	0.29	3.49
NO_ERR	3.38	1.29	4.67	2.73	0.29	2.74
ICO	3.38	1.31	4.69	4.19	0.26	4.19
NO_WTS	3.39	1.26	4.65	4.61	0.32	4.62
ISO_ERR	3.38	1.31	4.69	4.52	0.29	4.53

Figures

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Illustration of flux conversion from latitude-longitude grids to icosahedral grids. The black line indicates boundaries of the icosahedral grids, and dark gray and light gray lines represent original and finer latitude-longitude grids, respectively. In the conversion, fluxes in colored areas are allocated to the centered icosahedral grid.

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