Performance of a local ensemble transform Kalman filter for the analysis of atmospheric circulation and distribution of long-lived tracers under idealized conditions

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Received 10 February 2009; revised 7 July 2009; accepted 10 July 2009; published 10 October 2009.

A data assimilation system for the analysis of atmospheric circulation and long-lived tracer distributions in the troposphere and stratosphere has been developed and tested by using a local ensemble transform Kalman filter (LETKF), which has been applied to assimilate both meteorological fields and tracer concentrations into a general circulation model and an atmospheric transport model. Assimilated meteorological fields are used for driving the transport model. The performance of the LETKF data assimilation system is assessed under idealized conditions by assuming that the forecast models provide a perfect representation of atmospheric conditions. The LETKF meteorological analysis facilitates the study of atmospheric transport characteristics and provides high-quality tracer transport simulations, reflecting its flow-dependent and physically well balanced analysis. In particular, eddy mixing features are better analyzed by LETKF than by an analysis that employs a conventional assimilation scheme (i.e., nudging technique). The conventional analysis causes excessive tracer eddy dispersions, which were commonly observed in previous studies using three-dimensional variational analyses. Further improvement in tracer analysis can be achieved by assimilating the tracer concentration within the LETKF. The assimilation of tracer concentration effectively reduces the tracer background error caused by initial distribution and surface flux errors. Tracer analysis can also be improved by considering the covariance with wind fields in a background error matrix, in which wind observation directly impacts the tracer states, reducing 20% of the tracer analysis error in the free troposphere. The sensitivity of the tracer analysis to assimilation parameters and model errors is discussed to obtain an optimal data assimilation framework.


1. Introduction

Numerical simulations using atmospheric transport models or chemical transport models (CTMs) have been widely used for investigating spatial and temporal variations in atmospheric chemical constituents. Although such models have provided insight into variously scaled atmospheric constituent variability, insufficient representation of chemical and transport processes has caused uncertainties in the results. Further improvement of the constituent simulation is possible through the use of an advanced data assimilation technique that allows high-quality meteorological analysis and performs efficient chemical constituent assimilation.

The quality of meteorological analysis significantly affects atmospheric constituent simulation results. Successful constituent simulations can be performed with high-quality meteorological analyses by improving the representation of atmospheric transport [e.g., Bregman et al., 2006; Kinnison et al., 2007; Monge-Sanz et al., 2007]. However, currently available reanalysis products, which are mostly created using three-dimensional variational data assimilation (3D-VAR) [e.g., Lorenc, 1986] systems, are known to cause considerable problems in transport simulations. In the simulations with 3D-VAR meteorological analysis, a rapid upward transport in the tropics and excessive mixing between the tropics and midlatitudes causes the underestimation of the age of air in the extratropics in the stratosphere [van Noije et al., 2004; Scheele et al., 2005; Chipperfield, 2006]. The 3D-VAR system produces an additional force without maintaining physical balance in the momentum and thermodynamic equations because of its isotropic and instantaneous analysis increment. Similar to the 3D-VAR analysis, the analysis carried out using the Physical Space Statistical Analysis (PSSA) scheme in the Goddard Earth Observing System (GEOS) data assimilation system [Cohn et al., 1998] leads to excessive subtropical transport and the underestimation of the age of air in the extratropical stratosphere [Douglass et al., 2003; Schoeberl et al., 2003; Tan et al., 2004]. The physically unbalanced analysis increment is considered to lead to...
unrealistic circulations in reanalyses, such as excessive mixing and unrealistically strong residual circulations [e.g., Douglass et al., 2003]. The excessive subtropical mixing is probably associated with the proliferation of eddy features [Tan et al., 2004].

[4] Several recent studies have demonstrated that meteorological analyses conducted using advanced data assimilation systems (e.g., 4D-VAR system) improve atmospheric transport simulations [van Noije et al., 2004; Scheele et al., 2005; Monge-Sanz et al., 2007]. The 4D-VAR system is conceptually similar to the 3D-VAR system, except that the cost function includes an additional term that is a function of time (i.e., background error covariance evolves implicitly in 4D-VAR), producing a physically balanced analysis field at each model time step [e.g., Courtier and Talagrand, 1990]. Monge-Sanz et al. [2007] demonstrated that a highly realistic Brewer-Dobson circulation and subtropical mixing can be achieved when ECMWF operational data (i.e., 4D-VAR product) are used instead of ERA-40 analyses, a 3D-VAR product, and thus a 4D-VAR system corrects the underestimation of the age of air.

[5] An ensemble Kalman filter (EnKF) is one of the recently developed advanced data assimilation techniques [Evensen, 1994; Burgers et al., 1998], in which the forecast error covariance is advanced by using the model itself (i.e., flow-dependent forecast error covariance), rather than by estimating the forecast (or background) error as a constant covariance matrix. An EnKF allows the production of physically balanced analysis fields, similar to the 4D-VAR system. The algorithm of the EnKF does not require an adjoint model and is easier to implement than a 4D-VAR system (detailed descriptions and comparisons between EnKF and 4D-VAR are provided by Lorenc [2003] and Kalnay et al. [2007]). The EnKF system has been widely tested and successfully employed for meteorological data assimilation [e.g., Houtekamer et al., 2005; Szunyogh et al., 2005, 2008]. The flow-dependent analysis of EnKF may have advantages over conventional approaches when used to analyze atmospheric circulation and transport properties. Considering the possibility that future atmospheric reanalyses will be carried out using EnKF data assimilation systems, it is important to investigate the ability of EnKF meteorological analysis as a driver of atmospheric transport simulations. It must be noted that a local ensemble transform ensemble Kalman filter (LETKF) [Ott et al., 2004; Hunt et al., 2007] has already been tested for application to meteorological reanalysis in the AGCM for Earth Simulator (AFES)-LETKF experimental ensemble reanalysis (ALERA) project [Miyoshi et al., 2007]. The LETKF has considerable conceptual and computational advantages over the original EnKF. By employing a localization technique the LETKF removes sampling errors caused by the limited ensemble size (note that Houtekamer et al. [2005] applied a similar localization technique for the covariance inflation in the EnKF).

[6] Meanwhile, atmospheric constituent data assimilation systems have recently been developed and employed to assimilate observed concentrations of chemical constituents into CTMs or atmospheric transport models by using variational methods [e.g., Elbernt et al., 1997; Khattatov et al., 2000; Elbern and Schmidt, 2001; Wargan et al., 2005; Chai et al., 2006] and ensemble-based approaches [e.g., Arellano et al., 2007; Constantinescu et al., 2007a, 2007b, 2007c]. Although such systems provide a useful framework for analyses of constituent variabilities and atmospheric environments [Rood, 2005; Lahoz et al., 2007], recent data assimilation systems have not yet fully succeeded in simulating constituent distribution because of the limitations of forecast models and data assimilation schemes [e.g., Geer et al., 2006; Coy et al., 2007; Errera et al., 2008]. The EnKF system is a potential data assimilation system employed for assimilating chemical constituent concentration because of its flow-dependent analysis and easy implementation of complex systems. Although several studies have demonstrated that EnKF works well for chemical constituent data assimilation [e.g., Arellano et al., 2007; Constantinescu et al., 2007c], further efforts are required to examine the ability of EnKF to assimilate chemical constituents with different spatiotemporal scales.

[7] This study evaluates the performance of the LETKF in the analysis of atmospheric circulation and long-lived tracer distributions, and it provides first-time results showing how LETKF data assimilation improves the long-lived tracer analysis. Long-lived tracers are useful for demonstrating the performance of atmospheric transport simulations. The LETKF system is applied to assimilate both meteorological fields and long-lived tracer concentrations into a GCM and atmospheric transport model, and the performance is evaluated from a perfect model experiment (i.e., without model and observation errors). Furthermore, this study demonstrates the influence of temperature bias and surface flux errors on the analysis of atmospheric circulation and the performance of tracer data assimilation. This paper is organized as follows. Section 2 describes the model and LETKF scheme. Section 3 demonstrates the performance of LETKF meteorological assimilation on the tracer transport calculation. Section 4 addresses the ability of the LETKF system to assimilate long-lived tracer concentrations. Section 5 presents the discussion and summary.

2. Description of the Model and Assimilation Technique

2.1. Frontier Research Center for Global Change Atmospheric Transport Model

[8] For the forecast models, we employed the Center for Climate System Research/National Institute for Environmental Studies/Frontier Research Center for Global Change (CCSR/NIES/FRCGC) atmospheric general circulation model (AGCM) version 5.7b [Numaguti et al., 1995] and an online atmospheric transport model (embedded in the AGCM) developed at FRCGC [Miyazaki et al., 2008]. These models had T42 truncation in the horizontal and 32 levels from the surface to 7 hPa in the vertical (total grid number is 262144 (= 128 × 64 × 32)). The global distribution of carbon dioxide (CO₂) was calculated from the transport model with a time interval of 10 min, by using the meteorological fields obtained from the GCM. Carbon dioxide, which is an inert gas in the atmosphere (except photodissociation in the upper atmosphere), can be regarded as an atmospheric passive tracer in the troposphere and stratosphere. The grid-scale advection of CO₂ was calculated explicitly by using the simulated wind fields and the transport scheme at each model grid point on the basis of a fourth-order flux-form advection
scheme. We considered subgrid transport processes by moist convection and vertical diffusion in which a parameterized convective transport represented the updraft and downdraft of CO$_2$ by cumulus convection, and turbulent mixing was dependent on the vertical diffusion coefficient estimated using a turbulent closer. The surface flux data obtained for CO$_2$ were based on the results of the TransCom3 experiment [Gurney et al., 2003], which consisted of anthropogenic emissions, atmosphere-biosphere exchange flux, and atmosphere-ocean exchange flux. A detailed description of the FRCGC transport model is provided by Miyazaki et al. [2008].

### 2.2. Local Ensemble Transformed Kalman Filter

[9] Data assimilation is a technique for combining observational information with models in order to obtain an optimal representation of the state of the atmosphere. The EnKF system is one of the recently developed advanced data assimilation techniques that are based on the representation of the probability density of the state estimate by a finite number of system states [Evensen, 1994, 2003]. In the EnKF system, the state update is computed in the low-dimensional space, and the forecast error covariance is advanced by using the model itself, rather than by estimating the error as a constant covariance matrix. The EnKF system is employed to transform a background ensemble ($x^b_{n,i}$; $i = 1, \ldots, k$) into an appropriate analysis ensemble ($x^a_{n,i}$; $i = 1, \ldots, k$), where $x$ represents the analysis variable; $b$, the background state; $a$, the analysis state; $i$, the number of ensemble members; $k$, the ensemble size; and $n$, the time.

In this study, we adopt a LETKF [Ott et al., 2004; Hunt et al., 2007] system, which has conceptual and computational advantages over the original EnKF. Following Hunt et al. [2007], the LETKF data assimilation process is introduced below. First, in the forecast step, a background ensemble $x^b_{n,i}$ is globally obtained from the evolution of each ensemble member as in the EnKF, according to the model $M$,

$$x^b_{n,i} = M(x^b_{n-1,i}).$$  \hspace{1cm} (1)

In the analysis step, the background ensemble mean and background ensemble perturbations are estimated from the ensemble forecast.

$$\bar{x}^b = \frac{1}{k} \sum_{i=1}^{k} x^b_i,$$  \hspace{1cm} (2)

$$x^b = x^b_i - \bar{x}^b.$$  \hspace{1cm} (3)

An ensemble of background observation vectors $y^b_i$ and an ensemble of background perturbations in the observation space $Y^b$ are obtained as follows:

$$y^b_i = H(x^b_i),$$  \hspace{1cm} (4)

$$y^b_i - \bar{y}^b.$$  \hspace{1cm} (5)

where $H$ is the observational operator. In the LETKF, the local analysis error covariance in the ensemble space is expressed by

$$\tilde{P}^a = \left[ (k-1)I + (y^b)^T R^{-1} y^b \right]^{-1},$$  \hspace{1cm} (6)

where $R$ denotes the observation error covariance. The analysis weights $\tilde{w}^a$ and perturbation analysis matrices of weights $W^a$ in the ensemble space are expressed by

$$\tilde{w}^a = \tilde{P}^a(y^b)^T R^{-1} (y^a - \bar{y}^a),$$  \hspace{1cm} (7)

$$W^a = [(k-1)\tilde{P}^a]^{1/2},$$  \hspace{1cm} (8)

where $y^a$ represents the vector of observations. The analysis perturbations $\tilde{w}^a$ are closely related to the background perturbations (E. Ott et al., Exploiting local low dimensionality of the atmospheric dynamics for efficient ensemble Kalman filtering, 2002, arXiv:physics/0203058v3), and can be regarded as a matrix of weights. Finally, the new analysis mean and ensemble analyses in the model space are obtained as follows:

$$\bar{x}^a = \bar{x}^b + X^b \tilde{w}^a,$$  \hspace{1cm} (9)

$$x^a_i = \bar{x}^b + X^b w^a_i,$$  \hspace{1cm} (10)

where an ensemble of analyses is generated centered on the best estimate of the state and representative of the uncertainty in the analysis. The analysis ensemble in the model space, $x^a_i$, is obtained from the linear combination of the background mean and background ensemble perturbations (equation (10)), in which the weight, $w^a_i$, is estimated by adding $\tilde{w}^a$ to each of the columns of $W^a$.

[11] The LETKF solves the analysis equation in a local volume centered on each grid point; a localization technique is employed to remove sampling errors caused by the limited ensemble size. In a standard setting, we used a local patch grid size of $3 \times 3 \times 3 (= x \times y \times z)$. The localization treats the local patches surrounding every grid point, independently. By neglecting the correlation between distant points, the localization removes the effect of remote observations and reduces the ensemble size required to maintain the same error levels [Szunyogh et al., 2005]. We employed an inverse Gaussian function for the observation error covariance in order to dampen the spurious remote correlations that develop because of the small ensemble size. Most calculations in the LETKF are done in parallel, which reduces the computational cost.

[12] Covariance inflation is applied to the forecast error covariance in order to increase the spread of the ensemble to avoid filter divergence; this is because in nonlinear systems, LETKF generally underestimates the forecast error covariance since it neglects nonlinearity, sampling errors, and model errors. The sensitivity of the meteorological and tracer
concentration analyses to the covariance inflation will be discussed in sections 3.3 and 4.4, respectively.

3. Results: Performance of the Atmospheric Circulation Analysis System

3.1. Description of Perfect Model Experiment

By assuming that forecast models provide a perfect representation of the atmosphere (i.e., perfect model experiment), the sensitivity of the atmospheric transport calculation to the quality of meteorological analysis is investigated in this section. The perfect model experiment provides a useful framework for the analysis of the performance of the data assimilation scheme. Szunyogh et al. [2005] described the value of the perfect model experiment. They stated that in a chaotic system, uncertainties in the initial conditions are frequently amplified, and forecast errors occur and grow exclusively because of the uncertainties in the initial conditions and the sensitivity of the model solutions to such uncertainties. The data assimilation acts to remove the growing component of the errors from the background by using observational noise, and spreading the information to unobserved locations. Under the assumption of the perfect model scenario, the actual background error and observational error statistics can be determined precisely, so that the perfect model experiment allows us to demonstrate the performance of the data assimilation scheme without model and observational errors. The perfect model experiment results tell us how much error reductions can be expected through data assimilation cycle.

In the perfect model experiments presented in this section, artificial meteorological observation data obtained from a reference GCM simulation by adding noise were assimilated into the GCM, and the analyzed meteorological fields were employed to drive the atmospheric transport model (the CO₂ concentration was not assimilated). In the experiments, the analyzed meteorological fields were provided to the atmospheric transport model at every time step of the GCM, so that transport calculations were performed with a very short time interval (10 min) to reduce the time truncation error in a transport process (i.e., online transport model calculation). In contrast, most of the current transport models do not include any calculations of meteorological fields, but use meteorological reanalysis data for transport calculations (i.e., off-line transport model calculation). The results presented in this section demonstrate the performance of the LETKF meteorological assimilation on online transport model calculation. However, it can also provide insight into off-line transport model calculations (except the reduced time truncation error), i.e., how the LETKF meteorological reanalysis data improve off-line transport calculation.

A time series of a reference solution (or true state) for meteorological and CO₂ fields was generated by the simulation (without any assimilation) of the GCM and atmospheric transport model (embedded in the GCM), respectively. The reference solution was employed to obtain artificial observational data and initial conditions for ensemble simulations. Artificial observational data were obtained from the true state with the addition of zero-mean Gaussian random noise as observational

The background error covariance for the initial assimilation cycle was obtained from the lagged average forecast [Hoffman and Kalnay, 1983], as follows. First, the time lagged meteorological fields were obtained on 15 November 2003 (at a randomly selected time during the period from 1 to 30 November 2003) for each ensemble member from the reference simulation. The CO₂ concentration continuously increased with time because of time-constant anthropogenic sources, so that the CO₂ distribution was set to be the same for all ensemble members (obtained from the reference simulation conduction on 15 November 2003) in order to perform simulations without any bias. Second, by using the meteorological and CO₂ fields obtained on 15 November 2003, ensemble simulations from 15 November 2003 to 11 December 2003 were conducted for each ensemble member. We confirmed that the ensemble spreads of wind and CO₂ concentration did not significantly vary after the spinup simulations (see blue lines in Figure 2). The simulated meteorological and CO₂ fields on 11 December 2003 were used in the initial assimilation cycle. By using the above-mentioned settings, we could randomly generate meteorological and CO₂ fields for the initial assimilation cycle without any bias. Figure 1 depicts the CO₂ and zonal wind fields for the initial assimilation cycle on 11 December 2003 at 700 hPa. The initial CO₂ ensemble spread was large in regions where the wind spread and the surface source/sink of CO₂ were large (i.e., northern extratropics).

In the perfect model experiments, horizontal (zonal and meridional) wind, temperature, surface pressure, and specific humidity obtained from the above-mentioned GCM reference with the addition of zero-mean Gaussian random noise were assimilated into the GCM. Standard deviations of the observational error were set to 2 m/s for the horizontal wind, 2 K for the temperature, 4 hPa for the surface pressure, and, 2 × 10⁻⁴ kg/kg for the specific humidity. A standard perfect model experiment was conducted with 50 ensemble members, covariance inflation of 8%, and local patch grid size of 3 × 3 × 3 (= x × y × z) by using sparse and dense observation networks. In the sparse network, it was assumed that observation stations were located at 10% of the model grid points (selected as 1 station out of every 4 grid points on the horizontal plane), and the observations were analyzed every 6 h. The dense network, observation points were located at every model grid point (total number of observation stations = 262144 (128 × 64 × 32)), and the observations were analyzed every 6 h. Note that both in the dense or sparse observation networks, the number of observations was considerably larger than that in the real world. It was confirmed that the nudging technique (see section 3.2) did not significantly improve the analysis with a very sparse observation network (i.e., realistic observation network), while the LETKF effectively improved the analysis even with a very sparse observation network (figure not shown). Thus, in comparison with the conventional nudging technique, the use of a very sparse observation network tended to cause overestimation of the value of the LETKF system. Consequently, we employed the sparse and dense networks to demonstrate the advantage of the LETKF analysis over the nudging analysis, although these networks were not realistic.
3.2. Comparison With a Nudging Technique

[18] The LETKF meteorological assimilation results were compared with the reference solution and assimilation results obtained by employing a conventional assimilation technique based on a simple Newton relaxation technique (i.e., nudging technique [Hoke and Anthes, 1976; Jeuken et al., 1996]). The nudging force relaxes the model state ($x^b$) toward the observations ($y^o$) and gradually corrects the model fields during assimilation periods to obtain the analysis ($x^a$),

$$x^a = x^b + \frac{y^o - x^b}{\tau}; \quad (11)$$

where $\tau$ indicates the nudging relation time. The nudging technique produces accurate analysis for a long assimilation window [Stensrud and Bao, 1992; Telford et al., 2008], and it has been used to assimilate meteorological analysis into GCMs in order to simulate the observed constituent variability with CTMs [e.g., Akiyoshi et al., 2002; van Aalst et al., 2004]. However, the external force due to the nudging might considerably disturb the dynamic balance in the analysis field, similar to the case of the 3D-VAR analysis, and contrary to the dynamically balanced analysis conducted using the 4D-VAR nKF systems. It is noted that 3D-VAR is a considerably more advanced scheme than nudging; thus, the comparison with a nudging scheme tended to overestimate the value of the LETKF scheme compared to that obtained with a 3D-VAR scheme.

[19] In the nudging experiment, horizontal wind and temperature data were assimilated into the GCM in every model time step, with an optimized relaxation time of 0.5 (0.35) days for wind fields and 1 (0.5) day for temperature in the sparse (dense) network, by using linearly interpolated observational data. The observation data used in the nudging experiment were the same as those used the LETKF experiment. Although surface pressure and specific humidity data were not assimilated into the GCM in the nudging experiment, we found that the assimilation of these data did not significantly improve the analysis of meteorological conditions and atmospheric circulation in the nudging experiment to some extent (figure not shown). Ten ensemble simulations were conducted for the nudging experiment by using initial conditions identical to those used in the LETKF experiments. By using the same initial conditions, free-running ensemble model simulations were also conducted without any assimilation.

3.2.1. Temporal Evolution of Analysis Errors

[20] Figure 2 compares the temporal evolution of root mean square (RMS) errors (difference between the true state and assimilation or free-running results) of zonal wind and CO$_2$ mixing ratio (Figures 1c and 1d, in ppm) at 0000 UTC on 11 December 2003 at 700 hPa.
CO₂ concentrations and the two assimilation schemes and free-running model simulations by using the dense observation network. Without any assimilation, global mean RMS errors related to zonal wind and CO₂ mixing ratio were approximately 10 m/s and 0.9 ppm, respectively, at 700 hPa. The errors, caused by the temporal growth of uncertainties in initial conditions, could be fatal considering that the mean amplitude of observed CO₂ variations on the order of less than several ppm/d in the free troposphere. We confirmed that the initial errors in CO₂ distribution affected the CO₂ forecast error significantly as compared to those in meteorological fields under the perfect model scenario (figure not shown), because of strong nonlinearity (error growth) in the dynamical system.

Assimilation of meteorological data significantly reduced both the wind analysis and the CO₂ forecast errors. The comparison between the two assimilation schemes showed a greater improvement in the case of the LETKF meteorological analysis than in the case of nudging. The global mean RMS error related to zonal wind estimated using the dense observation network was approximately 0.43 m/s and 0.76 m/s in the LETKF analysis and the

Figure 2. Temporal evolution of global RMS errors related to the (a) zonal wind and (b) CO₂ mixing ratio calculated with the LETKF meteorological analysis (black line), nudging meteorological analysis (red lines), and free-running model simulation (without any assimilation, blue lines) at 700 hPa, using the dense observation network (observations are located at every model grid point).
nudging analysis, respectively. In the LETKF analysis, the ensemble spread (not shown) and analysis error of wind quickly tended to be less than the observational error within a couple of days after the initial assimilation cycle. Because the CO$_2$ concentration was not assimilated in these cases, the CO$_2$ forecast error showed a lower convergence speed as compared to that estimated by the wind analysis, with settled global mean CO$_2$ RMS errors of approximately 0.05 ppm and 0.13 ppm in the LETKF and nudging analyses, respectively.

When either the dense or the sparse observation network was used, the LETKF meteorological analysis effectively improved transport simulation (Figures 2 and 3). In comparison to the LETKF analysis, the nudging analysis degraded further with a decrease in the number of assimilated observations. The settled levels of zonal wind and CO$_2$ RMS errors increased to more than twice the levels when the sparse observation network was employed instead of the dense one in the nudging analysis. Thus, it is concluded that the nudging analysis requires a strong force as a result of a large number of observations in order to effectively reduce the background error. The LETKF analysis was less sensitive to the number of assimilated observations. It must be noted that the nudging assimilation results depended on the relaxation time ($\tau$). A shorter relation time generally improved the nudging analysis.

![Figure 3](image_url)

Figure 3. Same as Figure 2 but with the sparse observation network (observations are regularly carried out at 25% of model grid point).
whereas the use of a significantly short relaxation time had an unfavorable effect on the analysis owing to strong nonphysical external force due to assimilation (optimized values were used in the simulations).

[23] We found that significant improvement by the LETKF meteorological analysis occurred around strong wind shear regions, owing to flow-dependent error corrections (Figure 4). The LETKF analysis efficiently corrected the background wind error in regions where the wind shear and the instability in the atmosphere were large, thus allowing the simulation of rapid tracer variations. In contrast, the nudging analysis showed large wind analysis errors and the difficulty in representing rapid tracer variations around strong wind shear regions (e.g., over the eastern Pacific in Figures 4f and 4g).

3.2.2. Transport Characteristics

[24] Figure 5 shows the meridional cross section of errors that occurred during the analysis of meteorological parameters (which relate to atmospheric transports) and CO₂ concentration obtained from the LETKF and nudging meteorological analyses for the dense observation network. Both meteorological assimilation results showed the maximum CO₂ RMS error in the tropical and northern extratropical troposphere (Figures 5b and 5c), but they exhibited a larger error in the case of the nudging analysis. In the tropical troposphere, the atmospheric thermal structure significantly influences tracer transports. The comparison of RMS errors within the Brunt-Vaisala frequency showed large errors in the analysis of atmospheric stability in the tropical upper troposphere in the nudging analysis (Figures 5h and 5i). The incorrect variations in the stability and wind fields probably lead to excessive vertical and horizontal dispersions of tracers in the nudging analysis. In the northern extratropics, large CO₂ forecast errors obtained with the nudging analysis mainly arose from upward and poleward transports of surface high-CO₂ air associated with synoptic-scale disturbances, as partly observed in the case of large RMS errors in the wind and mass stream function (Figures 5f and 5i). The results obtained from the comparisons imply that an improvement in the tracer simulation is possible through the utilization of better balancing assimilation schemes (i.e., LETKF) for meteorological analysis. It must be noted that the tracer simulation results obtained with the LETKF meteorological analysis were worse around the Arctic stratosphere than those obtained in other regions and by the nudging analysis (Figures 5b and 5c), probably because of a significant nonlinearity of the Arctic polar night jet.

[25] To identify transport processes that cause CO₂ forecast errors, a zonal mean tracer continuity equation based on the mass-weighted isentropic zonal means [Miyazaki and Iwasaki, 2005] was analyzed by using the assimilation outputs. This analysis framework allowed an accurate representation of meridional tracer transports by using mean-meridional circulation (i.e., mean transport) and large-scale eddy mixing (i.e., eddy transport). Figure 6 shows that both the mean and the eddy transports were better analyzed with the LETKF analysis than with the nudging analysis, and an apparent difference between the two analyses was observed in the case of the eddy transport. The eddy transport error dominantly caused large transport simulation errors with the nudging analysis, particularly in the upper troposphere for the sparse observation network.

[26] A wave decomposition analysis conducted by using Fourier transform showed that the nudging analysis distinctly overestimated zonal CO₂ variations (i.e., eddy components) on planetary and synoptic scales (zonal wave number ranging from 1 to 7) in the middle and upper troposphere (Figure 7b). These large-scale motions mostly controlled the eddy tracer transport in the troposphere and lower stratosphere (figure not shown), and thereby the nudging analysis faced considerable problems in the tracer simulation because of excessive large-scale dispersions. The excessive eddy dispersion has been commonly found in the 3D-VAR analysis [e.g., Tan et al., 2004], which is probably attributed to flow-independent analysis increment of the nudging and 3D-VAR analysis, although 3D-VAR is a more advanced scheme than nudging. In contrast, LETKF analyzes large-scale wave phenomena (Figure 7a) well and causes fewer eddy transport errors.

[27] It should be noted that the LETKF analysis showed relatively large errors in small-scale CO₂ disturbances (e.g., with zonal wave number greater than approximately 15) rather than large-scale disturbances (Figure 7a), although the errors contributed less to the total CO₂ variations. The LETKF analysis underestimated the small-scale zonal CO₂ disturbances. It was also found that the representation of the small-scale variations strongly depended on the local patch size in the LETKF analysis, and became apparently prominent with a decrease in the number of assimilated observations (figure not shown). Increasing the horizontal local length degraded the small-scale variations, but it slightly improved the large-scale variations (Figure 8).

3.3. Sensitivity to Assimilation Parameters

[28] The sensitivities of the LETKF meteorological assimilation results and CO₂ forecast errors to LETKF meteorological assimilation parameters (i.e., ensemble size, observation density, local length, and covariance inflation) were investigated using the sparse observation network. Table 1 provides a comparison between the global mean RMS errors related to zonal wind and CO₂ mixing ratio among perfect model experiments with different parameter values for the LETKF meteorological assimilation. The CO₂ concentration was not assimilated for all cases. The global mean RMS error in the CO₂ forecast was approximately proportional to that of wind analysis among the experiments, confirming that the quality of the meteorological analysis controlled the performance of the transport simulation in a perfect scenario. Note that the observation network used in the sensitivity experi-

**Figure 4.** Northern Hemispheric distributions of (a, b, and c) meridional wind (in m/s) and (d, e, and f) CO₂ mixing ratio (in ppm) obtained from the reference solution (Figures 4a and 4d) and the errors (minus the reference solution) calculated with the LETKF meteorological analysis (Figures 4b and 4e) and nudging meteorological analysis (Figures 4c and 4f) at 1200 UTC on 29 December 2003 at 400 hPa. (g) Isopleths of the CO₂ mixing ratio of 384.4 ppm calculated from the reference solution (black), LETKF meteorological analysis (red line), and nudging meteorological analysis (green line).
Figure 5

- (a) CO2 RMSE: TRUE (ppm)
- (b) CO2 RMSE: LETKF (ppm)
- (c) CO2 RMSE: NUDG (ppm)
- (d) U TRUE (m/s)
- (e) U RMSE: LETKF (m/s)
- (f) N^2 RMSE: LETKF (%)
- (g) N^2 RMSE: NUDG (%)
- (h) Stream-func. RMSE: NUDG (10^9 kg/s)
- (i) Stream-func. RMSE: LETKF (10^9 kg/s)
- (j) Stream-func. TRUE (10^9 kg/s)
ments was not realistic (see section 3.1). The sensitivity obtained here may differ depending on the difference in observation networks.

The ensemble size is an important factor that affects the capture of background error patterns and improvement in the analysis quality. Larger ensemble sizes allow accurate sampling of the probability distribution of background errors, whereas small ensemble sizes result in small spreads and underestimation of model errors. The underestimation of the forecast error covariance reduces the influence of observations and can cause filter divergence [e.g., Houtekamer and Mitchell, 1998]. Global mean RMS errors, both in the wind analysis and CO$_2$ forecasts, decreased by approximately 30% when the ensemble size increased from 20 to 50, but they did not vary significantly when the ensemble size was increased from 50 to 60. Thus, ensemble sizes greater than 50 were preferred for meteorological assimilation in order to obtain high-quality transport simulations (but the computational cost increased with increasing ensemble size).

The meteorological assimilation results were highly sensitive to the local patch size. Global mean RMS errors in both the wind analysis and CO$_2$ forecasts, increased by approximately 30% when the ensemble size increased from 20 to 50, but they did not vary significantly when the ensemble size was increased from 50 to 60. Thus, ensemble sizes greater than 50 were preferred for meteorological assimilation in order to obtain high-quality transport simulations (but the computational cost increased with increasing ensemble size).

Assimilation results are also shown for different observation networks. Large analysis errors occurred when the number of observations was insufficient to reduce the background error. By decreasing the number of assimilated meteorological observations, from 25 to 10.8% of all the grid points (a total 28,224 (= $42 \times 21 \times 32$) points), the

**Figure 5.** Latitude-pressure distribution of the (a, b, and c) zonal mean CO$_2$ mixing ratio (in ppm), (d, e, and f) zonal mean zonal wind (in m s$^{-1}$), (g, h, and i) square of Brunt-Vaisala frequency (in $10^{-4}$ s$^{-2}$), and (j, k, and l) mass stream function (in $10^9$ kg s$^{-1}$), obtained from the reference solution (Figures 5a, 5d, 5g, and 5i) and the RMS errors calculated using the LETKF meteorological analysis (Figures 5b, 5e, 5h, and 5j) and nudging meteorological analysis (Figures 5c, 5f, 5i, and 5l), averaged during 12 January 2004 by using the dense observation network.

**Figure 6.** Vertical profiles of global mean RMS errors of CO$_2$ time tendency (in ppm/month) by the mean-meridional transport (solid lines) and eddy transport processes (dashed lines) calculated with the LETKF meteorological analysis (thick lines) and nudging meteorological analysis (thin lines), averaged during 4 and 14 January 2004 by using the (left) sparse and (right) dense observation networks.
wind analysis error increased by approximately 65%. However, even when the meteorological observations were considered to be only 10.8% of all the grid points, the global mean wind analysis error tended to remain at a level smaller than the observational error. The accuracy of the LETKF analysis was slightly affected until a critically low density is attained, whereas the performance of the static schemes (e.g., nudging, 3D-Var) degraded dramatically with a decrease in observation density (see section 3.2.1). The result implies that the LETKF effectively improves the meteorological analysis even with a very sparse (i.e., realistic) observation network. However, a better tracer transport simulation requires the wind analyses produced with a larger amount of meteorological observation data.

The covariance inflation reduced the wind analysis error, because it attempted to remove the underestimation of forecast error covariance. Larger analysis errors occurred when covariance inflation was not allied than when it was applied, because the underestimation of forecast error covari-

### Table 1. Global Mean RMS Errors Related to the Zonal Wind and CO₂ Mixing Ratio at 700 hPa

<table>
<thead>
<tr>
<th>Setting</th>
<th>U RMSE (m/s)</th>
<th>CO₂ RMSE (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>STD</td>
<td>0.71</td>
<td>0.082</td>
</tr>
<tr>
<td>m = 20</td>
<td>1.06</td>
<td>0.110</td>
</tr>
<tr>
<td>m = 30</td>
<td>0.97</td>
<td>0.107</td>
</tr>
<tr>
<td>m = 40</td>
<td>0.92</td>
<td>0.102</td>
</tr>
<tr>
<td>m = 60</td>
<td>0.71</td>
<td>0.082</td>
</tr>
<tr>
<td>l = 1 x 3</td>
<td>0.89</td>
<td>0.094</td>
</tr>
<tr>
<td>l = 5 x 3</td>
<td>0.95</td>
<td>0.109</td>
</tr>
<tr>
<td>l = 7 x 3</td>
<td>1.28</td>
<td>0.144</td>
</tr>
<tr>
<td>l = 3 x 3</td>
<td>0.87</td>
<td>0.118</td>
</tr>
<tr>
<td>l = 3 x 5</td>
<td>0.68</td>
<td>0.075</td>
</tr>
<tr>
<td>l = 3 x 7</td>
<td>0.68</td>
<td>0.072</td>
</tr>
<tr>
<td>l = 3 x 9</td>
<td>0.69</td>
<td>0.073</td>
</tr>
<tr>
<td>o = 42 x 21 x 32 (10.8%)</td>
<td>1.17</td>
<td>0.134</td>
</tr>
<tr>
<td>o = 128 x 64 x 32 (100%)</td>
<td>0.46</td>
<td>0.054</td>
</tr>
<tr>
<td>ci = 0%</td>
<td>0.84</td>
<td>0.085</td>
</tr>
<tr>
<td>ci = 4%</td>
<td>0.71</td>
<td>0.078</td>
</tr>
<tr>
<td>ci = 12%</td>
<td>0.76</td>
<td>0.094</td>
</tr>
<tr>
<td>ci = 16%</td>
<td>0.82</td>
<td>0.110</td>
</tr>
</tbody>
</table>

*Averaged during 28 and 30 December 2003, obtained with the LETKF meteorological analysis with different assimilation parameters (ensemble size (m), local patch grid size (l), number of observations (o), and covariance inflation (ci, in percent)), using the sparse observation network. In the standard experiment (STD), the values of the parameters are m = 50, l = 3 x 3 x 3, o = 64 x 32 x 32 (25% of model grid point), and ci = 8%.

Figure 7. RMS errors in the monthly and zonal mean amplitudes of the zonal CO₂ disturbances (shaded, in percent) obtained (a) with the LETKF meteorological analysis and (b) by nudging meteorological analysis using the dense observation network as a function of zonal wave number and pressure, averaged during 4 and 14 January 2004, between 74° S and 74° N. The black contour line shows the mean difference (bias; the assimilation experiment minus the reference simulation) of 0. (c) The difference in the RMS error between the two experiments: Figure 7a values minus Figure 7b values.

Figure 8. Same as Figure 7 but for the difference in the RMS error of the zonal CO₂ disturbance between the LETKF experiments with different local patch sizes for the horizontal direction (l = 3 x 3 x 3 – l = 5 x 5 x 3) using the dense observation network.
Table 2. Global Mean RMS Errors of CO$_2$ Mixing Ratio at 530 hPa and of Temperature at 260 hPa

<table>
<thead>
<tr>
<th>Setting</th>
<th>CO$_2$ RMSE (ppm)</th>
<th>T RMSE (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LETKF perfect</td>
<td>0.04</td>
<td>0.25</td>
</tr>
<tr>
<td>LETKF T-bias</td>
<td>0.08</td>
<td>1.96</td>
</tr>
<tr>
<td>Nudging perfect ($\tau = 1$ day)</td>
<td>0.10</td>
<td>0.41</td>
</tr>
<tr>
<td>Nudging T-bias ($\tau = 0.5$ days)</td>
<td>0.23</td>
<td>1.00</td>
</tr>
<tr>
<td>Nudging T-bias ($\tau = 1$ day)</td>
<td>0.18</td>
<td>1.27</td>
</tr>
<tr>
<td>Nudging T-bias ($\tau = 3$ days)</td>
<td>0.13</td>
<td>1.76</td>
</tr>
<tr>
<td>Nudging T-bias ($\tau = 5$ days)</td>
<td>0.12</td>
<td>2.00</td>
</tr>
</tbody>
</table>

$^a$Averaged during 28 and 30 December 2003, obtained with the LETKF and nudging meteorological analyses by using the dense observation network. The results are shown for the perfect case and imperfect case (with the biased temperature data), and for different temperature relaxation time ($\tau$) for the nudging analysis.

3.4. Sensitivity to Systematic Temperature Bias

[33] The performance of data assimilation is strongly affected by model bias [e.g., Dee, 2005; Polavarapu et al., 2005]. Most of the current GCMs are known to typically have a systematic cold temperature bias in the upper troposphere and stratosphere [Pawson et al., 2000; Austin et al., 2003]. Although recent improvements in radiation schemes have succeeded in reducing most of the temperature bias [e.g., Cagnazzo et al., 2007], some GCMs may still have such a temperature bias. To clarify the influence of the cold temperature model bias on the atmospheric circulation analysis and transport simulation, several assimilation experiments were conducted by considering a possible temperature bias. To simplify the setting, instead of considering a cold temperature bias in the model, we considered a warm temperature bias in the observation. Although the impact of considering a cold model bias and a warm observation bias on assimilation results can differ (since the consideration of a cold model bias changes the dynamical and thermodynamic balance of a GCM solution), the difference is considered to be significantly smaller than the impact. In order to obtain temperature observation data with a warm bias, we estimated a monthly and zonal mean temperature difference between the CCSR/NIES AGCM climatology and NCEP/AMIP-II reanalysis [Kanamitsu et al., 2002]. A simplified distribution of the estimated temperature bias was added to the true state. The obtained temperature bias maximized at approximately 100 hPa in the tropics and 250 hPa in the extratropics with a maximum value of approximately 5 K, but it remained very small above the middle stratosphere and below the middle troposphere.

Table 2 compares the RMS errors related to the zonal wind and the CO$_2$ mixing ratio of the perfect case and imperfect case (with bias temperature data) by using the LETKF and nudging schemes with the dense observation network. Assimilation of the biased temperature data seemed to increase the CO$_2$ forecast error by approximately 0.04 ppm in the LETKF analysis at 700 hPa. Because of the absence of a temperature-dependent source/sink term in CO$_2$ tendencies in the simulation, such deterioration of the tracer simulation occurred through changes in circulation fields. We confirmed that the deformation of the mean meridional circulation occurred in the imperfect experiment, thus degrading the tracer transport simulation result in the troposphere and lower stratosphere (figure not shown). Simultaneously, in the imperfect case, a large temperature bias remained in the LETKF analysis, although approximately half of the temperature bias was removed in the case of the data assimilation cycle.

[35] In the nudging analysis, the influence of the bias data assimilation was highly sensitive to the relaxation time for temperature nudging, $\tau$. When $\tau$ was set to be 1 day, the CO$_2$ analysis error increased up to 0.08 ppm because of the assimilation of the biased temperature data. The tracer analysis error decreased with an increase in $\tau$, in the imperfect case (but was less sensitive to $\tau$, in the perfect case). Shorter $\tau$ added a stronger external force to the thermodynamic equation, and the dynamics did not have sufficient time to adjust. In other words, spurious heating by assimilation degraded atmospheric circulation through thermal wind balance and wave-mean flow interactions [Miyazaki et al., 2005]. We confirmed that the nudging analysis with $\tau$, shorter than approximately 3 days degraded the mean-meridional circulation analysis and CO$_2$ forecast to a greater extent than the LETKF analysis. Therefore, higher values of $\tau$ are desirable to prevent degradation of atmospheric circulation in the presence of the temperature bias, while the temperature bias remains if higher values of $\tau$ are employed. The existence of temperature error affects the chemical processes in CTMs. Consequently, although the LETKF generally provided a better atmospheric circulation analysis and tracer simulation than the nudging in the imperfect model experiment, both the nudging and LETKF meteorological analyses had difficulties in simulating chemical and transport processes in the presence of a temperature bias. Hence, the use of high-quality GCMs and observations or bias correction schemes [e.g., Dee, 2005] is necessary to obtain realistic tracer estimates with model and observation bias. Monge-Sanz et al. [2007] observed that the use of bias correction and balance operator improves the transport calculation in the new ECMWF reanalysis (i.e., ERA-Interim) associated with an analysis increment.

4. Results: Performance of the Tracer Concentration Assimilation System

4.1. Description of Perfect Model Experiment

[36] This section discusses the ability of LETKF to assimilate the concentration of long-lived tracers into atmospheric transport models. The impact of the concentration assimilation was assessed by using perfect model experiments in which we assumed that the atmospheric transport model provided a perfect representation of CO$_2$ distributions. For this purpose, a reference solution obtained from an atmospheric transport model simulation (driven by the reference GCM solution; see section 3.1) was used for
generating the true state, and CO$_2$ observation data were obtained by adding random noise as observational error to the true state. We assumed that the CO$_2$ observations are regularly located at 25% of the model grid points (selected as 1 station out of every 4 grid points on the horizontal plane but located on every model level), and the observations are recorded ever . The number of the assumed observations was larger than that in the current CO$_2$ observation network. However, the number of CO$_2$ observations has recently been rapidly increasing owing to development of satellite observation network. Thus, we adopted the assumed (dense) observation network to demonstrate the performance of the LETKF data assimilation in near future observation networks that include some satellite
data, although the uniform distribution assumption may be unrealistic. A standard deviation in the CO$_2$ observational error was set to 0.4 ppm. A standard experiment used 50 ensemble members, covariance inflation of 4%, and local patch grid size of $3 \times 3 \times 3$ (= $x \times y \times z$) for the CO$_2$ concentration assimilation. The initial conditions used were similar to those in the LETKF meteorological analysis, and meteorological fields for each ensemble simulations were obtained from the standard LETKF meteorological analysis by using the sparse observation network (see section 3.1).

4.2. Temporal Evolution of Analysis Errors

Figure 9 shows the spatial structures of the background error, ensemble spread, and analysis increment of CO$_2$ concentration, demonstrating the performance of the LETKF tracer concentration assimilation. Background errors in the CO$_2$ concentration were large in regions where CO$_2$ concentration rapidly varied mainly because of strong vertical transports. Ensemble spreads captured the background error structure of CO$_2$ concentration, and the LETKF concentration assimilation effectively corrected the background CO$_2$ error where the ensemble spread was large, reflecting an anisotropic (flow-dependent) analysis increment.

[37] Figure 10 shows the temporal evolution of the global mean CO$_2$ analysis error. The CO$_2$ analysis error rapidly settled because of the assimilation of the CO$_2$ concentration together with meteorological fields by LETKF (black line). In this case, the settled value of the CO$_2$ RMS error tended to be approximately 25% of the observational error within a couple of days. Although the use of the LETKF meteorological analysis could not prove the CO$_2$ forecast in the perfect model scenario (light blue line), the concentration assimilation effectively and rapidly reduced the tracer background error caused by initial distribution errors. However, the settled value of the CO$_2$ RMS error did not significantly vary with the concentration assimilation.

[39] The settled levels of the CO$_2$ analysis error obtained with the LETKF concentration assimilation results were found to significantly differ among experiments with different meteorological analysis (Figure 10, black and red lines). When the nudging meteorological analysis was used instead of the LETKF meteorological analysis, the settled level of CO$_2$ RMS error almost doubled. This is because of worse circulation fields in the nudging analysis than in the LETKF analysis (see section 3.2). The result implies that the quality of meteorological analysis have a considerable influence on the accuracy of the long-term tracer analysis in the perfect case, although the quality of concentration data also affects the tracer analysis. Without any meteorological assimilation, the CO$_2$ analysis error remained large even when the CO$_2$ concentration was assimilated (Figure 10, green line, with a global mean RMS error of approximately 0.35 ppm).

4.3. Sensitivity to Surface Flux Error

Surface flux is one of major uncertain sources of current atmospheric constituent simulations. Surface flux error can considerably degrade atmospheric constituent simulation, particularly for species with a long chemical lifetime and strong surface fluxes (e.g., CO$_2$ and CH$_4$). To investigate the influence of the surface flux error on the tracer analysis, we considered a more realistic case than the perfect scenario by using imperfect surface flux data. From the viewpoint of
assimilating real observations, it is very important to demonstrate the performance of the data assimilation system under an imperfect scenario. The imperfect surface flux data is obtained from a top-down estimation of a surface flux condition by using an inversion model [Patra et al., 2005]. The difference between the imperfect flux data (Figure 11b) and perfect flux data (Figure 11a, obtained with a bottom-up estimation; see section 2.1) may have the same order of magnitude to a possible error in the current surface flux data. The tracer assimilation results obtained for the perfect and imperfect scenarios were compared by using the LETKF meteorological analysis with the sparse observation network. [41] Figure 12 shows that CO₂ RMS errors increased with the use of imperfect flux data instead of the perfect flux data. Without CO₂ concentration assimilation (but with assimilated meteorological fields), the CO₂ RMS error calculated with the imperfect flux data increased with time and increased up to approximately 0.38 ppm on 21 December 2003 at 700 hPa (Figure 12, thin dashed line), which was approximately 0.29 ppm higher than that calculated with the perfect model (Figure 12, thin solid line). By assimilating CO₂ concentration along with meteorological fields, we could significantly reduce the influence of the surface flux error. In this case, the settled value of the global mean CO₂ RMS error was 0.02 ppm higher in the imperfect case than in the perfect case (Figure 12, thick solid and dashed lines). Thus, the LETKF tracer concentration assimilation provided a significant reduction in the tracer background error caused by uncertainties in the surface flux data (i.e., model error).

[42] As an exception, even when the CO₂ concentration was assimilated, the CO₂ analysis error remained for both the imperfect and the perfect cases at low levels, although the concentration assimilation reduced approximately half of the analysis errors (figure not shown). The surface flux error continuously increased the tracer background errors and overcame the correction by the concentration assimila-

![Figure 11. Spatial distribution of surface CO₂ fluxes (in 10⁻¹⁹ kg-CO₂ m⁻² s⁻¹) obtained from the (a) bottom-up estimation (the perfect surface flux data) and (b) top-down estimation (the imperfect surface flux data) in December 2003.](image1)

![Figure 12. Temporal evolution of global mean RMS errors of CO₂ mixing ratio calculated from the perfect model experiments (solid lines) and from the imperfect scenario using imperfect surface flux data (dashed lines) at 700 hPa. The results are shown for the simulation with the LETKF meteorological analysis (without any CO₂ assimilation, thin lines) and for the CO₂ assimilation result with the LETKF meteorological analysis (thick lines).](image2)
Table 3. Same as in Table 1, but Obtained With the CO2 Concentration Assimilation Results With Different Assimilation Parameters, Using the LETKF Meteorological Analysis With the Sparse Observation Network

<table>
<thead>
<tr>
<th>Setting</th>
<th>CO2 RMSE (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>STD</td>
<td>0.078</td>
</tr>
<tr>
<td>m = 20</td>
<td>0.135</td>
</tr>
<tr>
<td>m = 30</td>
<td>0.079</td>
</tr>
<tr>
<td>m = 40</td>
<td>0.079</td>
</tr>
<tr>
<td>o = 42 × 21 × 10 (3.4%)</td>
<td>0.095</td>
</tr>
<tr>
<td>o = 64 × 32 × 16 (12.5%)</td>
<td>0.085</td>
</tr>
<tr>
<td>o = 128 × 64 × 16 (50%)</td>
<td>0.076</td>
</tr>
<tr>
<td>o = 64 × 64 × 32 (50%)</td>
<td>0.074</td>
</tr>
<tr>
<td>o = 128 × 64 × 32 (100%)</td>
<td>0.070</td>
</tr>
<tr>
<td>l = 1 × 1 × 3</td>
<td>0.081</td>
</tr>
<tr>
<td>l = 5 × 5 × 3</td>
<td>0.079</td>
</tr>
<tr>
<td>l = 7 × 7 × 3</td>
<td>0.080</td>
</tr>
<tr>
<td>l = 3 × 3 × 1</td>
<td>0.083</td>
</tr>
<tr>
<td>l = 3 × 3 × 3</td>
<td>0.078</td>
</tr>
<tr>
<td>l = 3 × 3 × 7</td>
<td>0.077</td>
</tr>
<tr>
<td>l = 3 × 3 × 9</td>
<td>0.078</td>
</tr>
<tr>
<td>c1 = 0%</td>
<td>0.076</td>
</tr>
<tr>
<td>c1 = 8%</td>
<td>0.092</td>
</tr>
<tr>
<td>c1 = 12%</td>
<td>0.107</td>
</tr>
</tbody>
</table>

*4In the standard experiment (STD), the assimilation parameters are m = 50, l = 3 × 3 × 3, o = 64 × 32 × 32 (25% of model grid point), and c1 = 4%.

4.4. Sensitivity to Assimilation Parameters

[43] Table 3 shows the sensitivity of the CO2 analysis RMS error to several parameters for the CO2 concentration assimilation. Perfect model experiments were conducted with differing assimilation parameters for the CO2 concentration assimilation but with common meteorological fields obtained from the standard experiment (see section 3.1).

[44] CO2 assimilation experiments with different choices for the ensemble member demonstrated that increasing the number of ensemble members from 20 to 30 resulted in significant improvements in the CO2 analysis (approximately 40%). This reflects the increase in the capability of the ensemble to capture the space of uncertainty. In contrast, by increasing the ensemble size from 30 to 40 or to 50, the CO2 analysis did not vary significantly, confirming that setting the number of ensemble members to 30 was almost sufficient to capture the background error structure of the long-lived tracer fields. The sufficient ensemble size required to capture the background error structure was slightly smaller in the case of CO2 concentration assimilation than that in the case of the meteorological assimilation (see section 3.4).

[45] The accuracy of the CO2 analysis degraded as the density of the CO2 concentration observation was reduced. The RMS error related to the CO2 mixing ratio increased by approximately 22% when the observations were considered to be 3.4% of all the grid points as compared to the sparse observation network case (considered to be 25% of the grid points). Increasing the amount of observations effectively reduced the background error of the CO2 fields. When observations were located at all model grid points, the tracer errors were approximately 10% smaller than those in the case of the sparse observation network. The tracer analysis at 700 hPa was found to be slightly more sensitive to zonal observational density than vertical and meridional densities (not shown), possibly related to the large zonal CO2 variations associated with heterogeneous surface flux distributions.

[46] The concentration assimilation results were also affected by the local patch size for both horizontal and vertical directions. The optimal values of the local patch grid size were found to be 3 and 7 for the meridional and vertical directions at 700 hPa, respectively. Local patch sizes that were considerably small resulted in large tracer analysis errors because of the neglected influence of remote observation information. The optimal values may depend on the location and season reflecting circulation fields and surface flux distributions.

4.5. Covariance Between Meteorological Field and Tracer Distribution

[47] Restricting the application of covariance inflation allowed the best analysis for CO2 concentration, which was different from the meteorological analysis. The tracer analysis error slightly increased with the covariance inflation. Because of a weak nonlinearity in the atmospheric transport model, assimilation of a long-lived tracer may not require large covariance inflation. In contrast, data assimilation of the chemical constituent with active chemical reactions may need to introduce the covariance inflation because of the complex (nonlinear) chemical system.
2004. This reduction had a large positive impact on the tracer analysis in the free troposphere. The use of the background error covariance matrices is an effective way to improve the tracer analysis in a LETKF data assimilation system. The impact of considering the covariance became weaker with a decrease in the number of meteorological observations and an increase in the observation error (figure not shown). Consequently, I would suggest that high-quality and dense meteorological observations can improve the long-lived tracer analysis through two methods with a LETKF data assimilation cycle; i.e., (1) by improving transport simulation (i.e., circulation fields) and (2) by considering the covariance to the long-lived tracer distribution.

[50] As an exception, considering that the covariance had a considerably less impact near the ground surface, particularly within the PBL (figure not shown), at low levels, subgrid transport processes (i.e., convection and diffusion) and surface flux forces significantly varied the tracer distributions and possibly lead to a weak correlation between the tracer and (grid-scale) wind fields.

[51] In comparison with the tracer analysis, the analysis of wind fields was found to be less sensitive to the consideration of the covariance. The global mean RMS error related to zonal wind analysis varied by only less than 0.02 m/s in the troposphere, considering the covariance (figure not shown), although it was slightly affected by the abundance and quality of tracer observations. The result indicates that it is more difficult to correct the wind analysis from the inclusion of the covariance as compared to the tracer analysis, probably because of the complex dynamical balance of circulation fields. Further improvement in the wind analysis may be possible through the use of a sophisticated balance operator or the covariance between tracer and potential vorticity fields.

5. Conclusions and Discussion

[52] The performance of the LETKF data assimilation system used for the analysis of atmospheric circulation and long-lived tracer distribution was assessed by using a global atmospheric transport model driven by a GCM system. The LETKF system was applied to assimilate both meteorological fields and long-lived tracer concentrations into the GCM and atmospheric transport model in a perfect model scenario. The global distribution of CO2 concentrations was simulated and analyzed as an atmospheric long-lived tracer. However, the results obtained from the CO2 analysis are considered to be common for the analysis of other long-lived species (e.g., CH4, N2O, and CFCs).

[53] The quality of meteorological analysis strongly influenced the tracer simulation results. The LETKF meteorological analysis provided high-quality meteorological data to drive atmospheric transport simulations. It captured atmospheric transport properties, particularly those related to large-scale motions, and contributed toward an accurate tracer transport simulation, either with sparse or dense observation networks. In contrast, a conventional analysis that employed the nudging technique (which provides isotropic analysis increment similar to 3D-VAR, although 3D-VAR is a more advanced scheme than nudging) caused excessive eddy dispersions, particularly on planetary and synoptic scales, and it degraded the tracer simulation. Settled global mean CO2 RMS errors with a dense observation network were approximately 0.05 ppm and 0.13 ppm.
in the lower troposphere in the LETKF and nudging analyses, respectively. Significant improvement in the tracer simulation by the LETKF meteorological analysis was found around the region with strong wind shear, because flow-dependent LETKF analysis captured the rapid changes in circulation fields. However, the tracer transport simulation with the LETKF meteorological analysis underestimated the small-scale tracer variations (with the horizontal scale of less than several thousand kilometers). The representation of small-scale disturbances was found to be sensitive to local patch size in the LETKF analysis, and thereby it may be corrected by improving the treatment of the local patch.

[54] The influence of systematic bias of GCMs on the atmospheric circulation analysis and tracer transport simulation was investigated by considering a possible temperature bias in observation data. An important point to emphasize is that a cold temperature model bias (i.e., a warm observation bias) around the tropopause had a negative influence on the atmospheric circulation analysis and the tracer simulation, because spurious heating by the analysis increment upset the physical balance of the circulation fields. By considering a possible temperature bias with a maximum value of approximately 5 K near the tropopause, the CO₂ forecast error almost doubled. Thus, the successful tracer simulation with the LETKF meteorological analysis is possible through the use of bias correction schemes or high-quality GCMs and observations.

[55] The LETKF data assimilation can assimilate the tracer concentration into an atmospheric transport model, which directly reduces its background error. By assimilating tracer concentration with LETKF, the tracer analysis error tended to be approximately 25% of the observational error within a couple of days. However, in the perfect case, the settled level of the tracer analysis error did not significantly vary with the concentration assimilation, but it depended on the quality of the meteorological analysis. It should be emphasized that, the assimilation of tracer concentration significantly decreased the settled level of the tracer analysis error in the imperfect case with uncertainties in the surface flux data. In this case, the tracer background error grows because of uncertainties in the surface flux data. Consequently, the use of high-quality meteorological analysis is a must for performing a successful analysis for long-lived tracers, while the assimilation of tracer concentration plays a very important role in removing the tracer background error caused by the initial distribution and model errors.

[56] An interesting finding is that the coupling in the data assimilation system, which considers the covariance between meteorological fields and tracer distribution in a background error matrix of LETKF, can further improve the tracer analysis in the free troposphere and stratosphere. Consideration of the covariance reduced approximately 20% of the tracer analysis error in the free troposphere. Therefore, high-quality meteorological data can improve the tracer estimates in two ways in the LETKF data assimilation system, by improving the transport simulation (i.e., circulation fields) and by considering the covariance to the tracer distribution.

[57] Analyses of both meteorological fields and tracer distribution were found to be sensitive to parameters for LETKF data assimilation, such as ensemble size, covariance inflation, local patch size, and observation density. The sufficient ensemble size required to stabilize and obtain high-quality analyses was slightly smaller in the case of CO₂ concentration assimilation than in the case of the meteorological assimilation. The optimized value of the covariance inflation for the meteorological assimilation was larger than that for the tracer concentration assimilation for the same ensemble size in the perfect case, because of strong nonlinearity in the dynamical system. Localization was applied to remove the spurious remote correlations that developed because of the small ensemble size. The optimal values of the local patch size were almost similar for the meteorological and tracer concentration assimilations in the free troposphere. Because the atmospheric CO₂ concentration shows large horizontal variations in the lower troposphere, the tracer assimilation greatly improved with the horizontal density of the observation network.

[58] To summarize, we have clarified that (1) the high-quality LETKF meteorological analysis is capable of significantly improving atmospheric tracer simulations, (2) the LETKF data assimilation effectively reduces the tracer concentration background error caused by initial tracer distribution and surface flux errors, (3) consideration of the covariance between wind fields and tracer distribution in a background error matrix of LETKF is capable of further improving the tracer analysis, and (4) use of high-quality models or surface flux data is important to improve the tracer analysis in the LETKF data assimilation system. These new findings obtained from this study are of great value for future developments of high-quality reanalysis system for atmospheric constituents with advanced data assimilation techniques.

[59] The future direction of this study is focused toward the application of the developed LETKF data assimilation system to estimate surface fluxes of CO₂ with satellite observations (e.g., obtained from GOSAT). The 4D-LETKF data assimilation system allows the estimation of the surface fluxes without an adjoint model. Another aim of the study is the application of the developed system to assimilate chemically active species (e.g., O₃) into CTMs. The LETKF system is a potential assimilation scheme for such applications because it can easily be implemented in complex chemical systems. Furthermore, a foreseeable extension of this research would be to compare the performances between the LETKF and the variational data assimilation systems (i.e., 3D-VAR and 4D-VAR) with regard to the analysis of atmospheric circulation (e.g., the age of air) and chemical constituent data assimilation. The comparison with the nudging scheme presented in this study may tend to overestimate the value of the LETKF scheme compared to that obtained with variational schemes.

Acknowledgments. The data assimilation scheme is developed on the basis of the LETKF system [Ott et al., 2004] constructed by Miyoshi [2005] and the LETKF system [Hunt et al., 2007] constructed by Junjie Liu. I am grateful to Takemasa Miyoshi and three anonymous reviewers for their helpful discussions. This study was partly supported by the Japan Society for the Promotion of Science Grant-in-Aid for Young Scientists (B) 19740300, the Global Environment Research Fund (B-93) by the Ministry of the Environment, Japan, and by means of the Grants-in-Aid for Creative Scientific Research (2005/17G50203) of the Ministry of Education, Science, Sports and Culture, Japan.

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