

We thank referees for their insightful comments and useful references. The feedback from the reviewer was quite useful for improving the manuscript. The reviewer's specific comments (shown in italics) are addressed below.

Answers for Referee #2 comments

Comment 1

It should be investigated in more detail what causes the adverse behavior of the coupled model at Samoa, where its performance in terms of the variance ratio is degraded relative to the Eulerian model, and at Barrow, where its performance in terms of the correlation is degraded.

Answer 1

We add following sentences to Results and discussion.

“The benefits of coupled transport modeling at regional and global scales for locations downwind of highly heterogeneous surface fluxes can be fully realized by using emission inventories at 10 km resolution (Olivier et al., 2008; Gurney et al., 2009) or 1 km resolution (Kannari et al., 2007; Oda and Maksyutov, 2010) and for coastal background monitoring sites influenced by contrast between air arriving from land and ocean (Ramonet and Monfray, 1996). Hence, a coupled model can calculate tracer-gas concentrations at point of interest anywhere on the globe with high-spatial-resolution surface fluxes, requiring less computation time than conventional Eulerian models. The coupled model's advantage in computational efficiency can be reduced in case of simulation very large volume observational data like in case of space observations.”

Comment 2

The more detailed investigation of model-model differences should attempt to identify the causes, e.g. there could be differences in advection (different wind fields are used), convective redistribution associated with cloud transport, and turbulent mixing / diffusion within the boundary layer, which is certainly different in the two models.

Answer 2

The inconsistency between models is an important issue but it is often acceptable in many nested modeling setups as the optimal parameterizations are different in different resolution models. In our case we can not deny the inconsistency between component models but the improvement in simulations justifies the use of a model combination suggested in this study.

Comment 3

The abstract should contain more quantitative results from the comparison between the two models and the observations. Also the discussion or conclusions should contain some statement on how the

coupled model can or will be used for inverse modelling.

Answer 3

We add sentences on model-observation correlation increase due to introduction of the new method.

We add following sentences to Abstract.

“Compared with the Eulerian model alone, the coupled model yields improved agreement between modeled and observed CO₂ concentrations. In the area where the CO₂ concentration variability is dominated by fossil fuel signal spikes the correlation between model and observations increases by 0.05 to 0.1 from the original values of 0.5 to 0.6.”

We add following sentences to Conclusion.

“In the case of Hateruma, the model-observation correlation of the coupled model exceeds that of NIES TM by more than 0.1. The advantage of the coupled model is visible in reproducing pollutant events observed in winter season at Hateruma therefore it would be safe to say that the coupled model improves results for cites influences by contaminated air.”

To explain benefit of using the coupled model for inverse modelling, we add following sentences to Introduction.

“Also, a global coupled model setup allows one to implement a single stage inversion and data assimilation schemes as opposed to two-stage approaches proposed for nested model setups by Rödenbeck et al. (2009) and Peylin et al. (2005). By combining an adjoint of the Eulerian model with a Lagrangian model a cost-efficient high resolution surface flux data-assimilation system can be constructed, for which only a single run for every observation of the high resolution Lagrangian model is sufficient, such as in Stohl et al. (2010), while many iterations with lower resolution Eulerian part are still needed for convergence (Chevallier et al., 2007).”

Comment 4

Will the adjoint of the Eulerian model be coupled with the footprint or influence information from the backward LPDM simulation? Those technical issues should be at least mentioned.

Answer 4

Authors thank the reviewer for suggestion. The added the discussion of the benefits and prospects of the coupled model setup for [variational and ensemble] data assimilation applications. (see answer to the Comment 3)

Comment 5 for P 2052, L 4

add "the" between "by" and "near field"

Comment 6 for P 2053, L 22

replace "resolutions" by "resolution"

Answer 5, 6

We revise our paper according to the comments.

Comment 7 for 2053, L25

It remains unclear what is meant by "fully realized". A resolution of 10 km might be appropriate or not, but this depends for example on the heterogeneity of the spatial distribution of fluxes in the vicinity of the observational site.

Answer 7

To clarify our statement, we add following sentences to Introduction.

“The benefits of coupled transport modeling at regional and global scales for locations downwind of highly heterogeneous surface fluxes can be fully realized by using emission inventories at 10 km resolution (Olivier et al., 2008; Gurney et al., 2009) or 1 km resolution (Kannari et al., 2007; Oda and Maksyutov, 2010) and for coastal background monitoring sites influenced by contrast between air arriving from land and ocean (Ramonet and Monfray, 1996).”

Comment 8 for P 2053, L 29

The computational time required for backward LPDMs depends on the number of locations for which mixing ratios are simulated. In case of space based observations a Eulerian approach might well require less computational time. This should be discussed in the paper.

Answer 8

The comment is correct. We add following sentences to Introduction.

“Hence, a coupled model can calculate tracer-gas concentrations at point of interest anywhere on the globe with high-spatial-resolution surface fluxes, requiring less computation time than conventional Eulerian models. The coupled model’s advantage in computational efficiency can be reduced in case of simulation very large volume observational data like in case of space observations.”

Comment 9 for P 2055, L 7

The results of those experiments should be included in a table, as this would indicate how sensitive the coupled model is to the choice of the duration of the Lagrangian simulation.

Answer 9

To explain sensitivity of the duration of the Lagrangian simulation to the coupled model and to the reason why we employ 7-day duration, we add following sentences to Materials and methods.

“To check sensitivity of the duration of the Lagrangian simulation to the coupled model, we calculated CO₂ concentrations at Hateruma for one year using double backward transport period, which is 14 days. Compared 14-day simulation with 7-day simulation, there was no large difference in peak shape and amplitude of concentrations between two, while shorter duration leads to decreasing peak amplitudes. The differences were within 2 ppm and the averaged absolute value of the differences was 0.52 ppm. This mismatch can be considered modestly small in comparison with observation error. Hence, the present study employs 7 days as backward transport span.”

Comment 10 for P 2055, L 24

what are those transformations? May be rewrite ": : : transformations as given in the equations below", if this is meant.

Answer 10

The “transformations” means chemical reaction of gases in atmosphere. We remove mention of the transformations as our study is limited to non reactive gas case. Although we understand the extension to chemically reactive gases case is possible.

Comment 11 for P 2056, L 3

both, $m(r)$ and $C(r)$ are time dependent, this should be taken into account in the equations.

Comment 12 for P 2057, L24

Again, $m(r)$ should also depend on time.

Answer 11, 12

As referee remarks, variables, $m(r)$ and $C(r)$ depend on time. We revise equations which include these variables as follows;

$$\Delta C = \frac{1}{M_o} \int_{T_b}^{T_e} dt \int dx \int dy \int dz m(\mathbf{r}, t) \frac{\delta C(\mathbf{r}, t)}{\delta t} \quad \text{Eq.2}$$

“... and $m(\mathbf{r}, t)$ is the particle density field ...” page 2056, line 6

$$m(\mathbf{r}, t) = \sum_{i=1}^N \mu_i \cdot \delta(\mathbf{r} - \mathbf{r}_i) \quad \text{Eq. 3}$$

$$C_{init}(T_e) = \frac{1}{M_o} \int dx \int dy \int dz m(\mathbf{r}, T_e) C_{3D}(x, y, z, T_e) \quad \text{Eq. 6}$$

“... Eq. (3) for particle concentration $m(\mathbf{r}, T_e)$ both ...” page 2057, line 4

“... the global particle concentration field $m(\mathbf{r}, T_e)$ in the ...” page 2057 line 5

Comment 13 for P 2057, L 10

This of course depends on the details of the "crude representation of C₃-D". So either those are specified here, or the discussion on seasonal variations should be dropped.

Answer 13

We change the discussion to be based on the results of Stohl et al. 2009.

Comment 14 for P 2057, L 22

Given the strong time dependence of biospheric fluxes on sub-diurnal time scales that influence the observations, it would be better to not limit the interpolation to a simple linear one, but instead use e.g. radiation to interpolate from daily to e.g. hourly fluxes. The implications of limiting the resolution to daily fluxes should at least be discussed.

Answer 14

Study by Patra et al 2008 found synoptic scale variability is not changed much with replacing hourly fluxes by daily, leading to the conclusion that major contribution to synoptic scale variability is made by fluxes with longer than hourly time scale.

Comment 15 for P 2057, L27

The spatial and temporal resolution of the forcing meteorology should be given.

Answer 15

To give spatial and temporal resolution of the forcing meteorology, we revise a paragraph which describes meteorological data used in this study in Materials and methods.

“FLEXPART is forced with the 6-hourly analysis data of the Global Forecast System (GFS) model of NOAA/NCEP (National Centers for Environmental Prediction). GFS data has $1.0^\circ \times 1.0^\circ$ horizontal resolution and 26 pressure levels. NIES TM is driven by the 12-hourly reanalysis data of the NCEP/NCAR Reanalysis Project (Kalnay et al., 1996) and is run with a horizontal resolution of $2.5^\circ \times 2.5^\circ$, with 15 sigma levels.”

Comment 16 for P 2058, L15

What is meant by "the offset values"? Is there a single number for the global average mixing ratio at the time of the start of the spin-up of the global Eulerian model, or was a site specific offset calculated from the average difference between observation and each model added? What was used as a spin-up time?

Answer 16

We add following sentences to Materials and methods.

“In the simulation of NIES TM, initial concentration of all grids are set to zero and two years after begin the simulation are used as a spin-up time.”

We revise a following sentence to Results and discussion.

“the offset values, which are obtained by subtracting average model-predicted concentration from average observed concentration, are added to the original model outputs.”

Comment 17 for P 2059, L3

May be replace "the fewer difference with" with "smaller differences to the". Also: A good statistics would be the standard deviation of model-observation differences for the coupled and the Eulerian model, this could be included either in the text or in the figure.

Answer 17

I calculate RMSE of the absolute value of the difference between deseasonalized model-predicted and observed CO₂ concentrations ($|\Delta\text{CO}_2|$) at Hateruma for 2002-2004 and show the value in the figure and change the range of x-axis from Jan.2003-Dec.2003 into Jan.2002-Dec.2004.

We add a following sentence to Results and discussion.

“The root mean square errors (RMSEs) of $|\Delta\text{CO}_2|$ are calculated for both case of NIES TM and the coupled model and the values of NIES TM and the coupled model are 2.21 and 1.81, respectively.”

Comment 18 for P 2059, L12

this sentence is unclear, may be the authors mean "... would increase when restricting the analysis to only the winter season"

Answer 18

In the case of Hateruma, wintertime pollution events are common because of the transport of emissions from the Asian continent as opposed to other two sites. Therefore, model-observation correlation and variance ratio are dominated by reproducibility of pollution events in winter. We add a following sentence to Results and discussion.

“when many sharp peaks appear in the observation due to contaminated air mass come from continental region by Asian monsoon in the case of Hateruma.”

Comment 19 for P 2059, L18

The authors should state if this difference is statistically significant. Note that a correlation coefficient of 0.5 means that only 25% of the observed variations are explained by the model.

Answer 19

We add a following sentence.

“The difference “0.1” of the correlation coefficient between modeled and observed concentration can be considered significant in comparison with usual range of correlation coefficient variability between transport models (Patra, et al., 2008) which is found to be in the range of 0.2 and best results rarely exceeding 0.8.”

Comment 20 for P 2060, L14

Given that daily fluxes have been used for biospheric fluxes, the simulated variations at sub-daily time scales are likely unrealistic. So the statement "the coupled model can resolve concentration variations at an hourly time scale or less" is not really supported.

Answer 20

In some sites like Hateruma in winter the persistent emitters like large cities upstream the observation station are contribution to observed short term spikes of several hours duration, so we meant that the coupled model is doing better in resolving those, even without using hourly resolving fluxes.

Comment 21 for P 2060, L18-23

This has not been shown in this paper. It strongly depends on the importance of mesoscale circulations (land-sea breeze, mountain-valley circulation) in the vicinity of observing sites. At least a reference needs to be given for such a statement.

Answer 21

Of course when the mesoscale circulation is involved the mesoscale resolving wind fields have to be used for simulation, so we add following a sentence to Conclusions.

“In case of the mesoscale circulations the proper wind data are required and large scale interpolated winds may not be sufficient.”

Added references are follows:

Chevallier, F., Breon F.-M., and Rayner P. J.: Contribution of the Orbiting Carbon Observatory to the estimation of CO₂ sources and sinks: Theoretical study in a variational data assimilation framework, J. Geophys. Res., 112, D09307, doi:10.1029/2006JD007375, 2007.

Peylin, P., Rayner, P. J., Bousquet, P., Carouge, C., Hourdin, F., Heinrich, P., Ciais, P., and AEROCARB contributors.: Daily CO₂ flux estimates over Europe from continuous atmospheric

measurements:1, inverse methodology, Atmos. Chem. Phys., 5, 3173–3186, 2005, <http://www.atmos-chem-phys.net/5/3173/2005/>.

Rödenbeck, C., Gerbig, C., Trusilova, K., and Heimann, M.: A two-step scheme for high-resolution regional atmospheric trace gas inversions based on independent models, Atmos. Chem. Phys., 9, 5331–5342, 2009, <http://www.atmos-chem-phys.net/9/5331/2009/>.

Ramonet, M., and Monfray P.: CO₂ Baseline concept in 3-D atmospheric transport models, Tellus, 48B, 502-520, 1996.