We thank the reviewer for the positive and constructive comments on our manuscript. The feedback from the reviewer has improved the quality of the manuscript. The reviewer's specific comments (shown in italics) are addressed below.

Answers for Referee #1 comments

Comment 1 for Abstract and Conclusions

I consider that the abstract (also Conclusions) needs to be completed with some explicit quantification of the results. This may help to understand the merit of this model.

Answer 1

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."

Comment 2 for P-2056, L14

I consider the simulation results are sensitive to a value of Zs. I consider the suitable value is depending on atmospheric conditions (ex. stability). Please comment on this.

Answer 2

As the referee mentioned, the simulation results depend on thickness of near surface layer Zs and using optimal Zs is more desirable than using fixed Zs.

We add following explanation to Materials and methods.

"We select Zs to balance between exact approximation of the transport equation which is achieved with small Zs but requires very large number of particles to transport. So we select Zs as big as possible without compromising results for mixing in well mixed boundary layer in daytime conditions, assuming we will analyze observations of the stable tracers often sampled in daytime. So our choice of Zs = 300 m is a rough approximation of the minimum value of the daytime mixed layer height." Just for reference, I list figures below. The left figure shows concentrations at Hateruma using Zs = 300 m (black line) and Zs = PBL height. The right figure shows differences of concentration between Zs = 300 m and Zs = PBL. The differences between two are within from 0.2 ppm to -0.4 ppm.



Comment 3 for Results and discussion

In general, there is no discussion about the effect of vertical transport. I consider that the precision is relatively reduced in this model when an air parcel is from free atmosphere. Is it possible to calculate correlation coefficients in such a case or comment on this issue?

Answer 3

Although we didn't present any results for free troposphere, we know that LPDM based analysis has been used successfully in free troposphere too so one can expect improvements in correlations there as well, see for example (Pisso et al, ACPD, 2010).

Comment 4 for P-2059, L16

The considerable reason why the correlation coefficients are similar between two models in BRW is not shown. Please comment it.

Answer 4

We add following sentences to Results and discussion.

"The benefit of using the coupled model is not visible equally for each of the three stations selected to represent different meteorological conditions around the globe. It was already observed (see Fig.4 in Patra et al, (2008)) with Eulerian models that the correlation between model and continuous observation time series varies for station to station, and generally lower correlations are observed for remote locations with few strong emission sources leading to lower CO2 concentration variance and sparse observations available for correcting reanalysis winds leading to quality degradation of the transport simulation due to wind speed and direction errors. BRW appears to be more difficult to simulate with other Eulerian models (Patra et al, 2008). Also, as for the variance ratio, remote/marine sites such as SMO yield worse results in comparison to the sites positioned closer to strong sources (see

Fig.6(d) in Patra et al, (2008)). Therefore, it is not surprising the correlation and variance ratio are not drastically improved in this study by introducing more accurate transport method while using similar quality wind fields."

Comment 5 for P-2060, L23

I agree the merits of this combined model. However, there are some issues (vertical advection, transport and atmospheric stability) which we need to consideration in using this model in inverse model or data assimilation. Could you comment on this?

Answer 5

We add following sentences to Conclusions.

"Another important issue is the quality of the wind and vertical mixing data used in the forward simulation with both Eulerian and Lagrangian components. The misfit between the model and observation can be blamed on numerical diffusion in the grid based models, but when the Lagrangian model is used it seems that the major contributor to the misfit is the imperfection of the wind field and it is going to be a main factor limiting accuracy of the forward and inverse modeling."

Comment 6 for Tables and Figures

Fig.1: I consider it is not easy to distinguish color of lines between Observation and NIES TM. The Fig.2 has the same issue.

Fig.3: I consider it better to show some mismatch value (ex. RMSE) in the figure or table as they have some meaning to show model performance.

Answer 6

I change the thicknesses of lines in Fig.1 and Fig.2 to easy distinguish color of lines between observation and simulations.

I calculate RMSE of the absolute value of the difference between deseasonalized model-predicted and observed CO₂ concentrations ($|\Delta CO_2|$) 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 following a sentence to Results and discussion.

"The root mean square errors (RMSEs) of $|\Delta CO2|$ 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."