Reply to Reviewer#1

Thank you very much for your comments and suggestions on our paper. They were very useful in revising the manuscript.

General Comments

In order to state the purpose and the conclusion of this study clearly, we have changed the last paragraph in Introduction and the last two paragraphs in Conclusion as follows:

Last paragraph in Introduction:

'In this study, we investigated the oxidative ratios of pollution events observed at HAT. Source regions of the individual pollution events were identified via back trajectory analysis, and the observed oxidative ratios were compared with the oxidative ratios calculated from the reported compositions of the fossil fuel types at their trajectory origins. Furthermore, in order to examine the relative impact of fossil fuel types used and variations in the regional emission to the observed O_2 and CO_2 changes at HAT, we employed an atmospheric transport model FLEXPART that uses the Lagrangian particle dispersion scheme (Stohl et al., 1998). Based on the comparison between the observation and the model result, we are able to speculate on the relative contributions to the observed oxidative ratios at HAT made by the emission types and the regional spatial variation of the emission itself on the continental Asia.'

Last two paragraphs in Conclusion:

'In order to examine how the regional FFBC emissions affect the oxidative ratios of the atmospheric pollution events observed at HAT, we simulated the observed atmospheric CO_2 and O_2 changes using the atmospheric transport model FLEXPART and the coupled model driven by O_2 and CO_2 fluxes from FFBC, TB, and the ocean. Analysis of the relative contributions of individual CO_2 fluxes to the atmospheric variations revealed that most of the peaks associated with the pollution events at HAT can be attributed to the FFBC CO_2 emissions. With the assigned fluxes, the transport models were able to reconstruct with a good agreement the average oxidative ratio of the pollution events assigned to China. Additional sensitivity analysis showed that the oxidative ratio of pollution events originating in China reflects about 90% of the change

in OR_{FFBC} for China. These results suggest that the observed OR at HAT could be used to detect changes in the composition of fossil fuel types used in China in future.

On the other hand, the model simulations underestimated the average OR for the Japan/Korea pollution events in comparison with the OR_{FFBC} for Japan/Korea region. This is because the emissions from China make substantial contributions to the Japan/Korea pollution events in the model. This transport problem could be solved by using higher spatially and temporally resolved meteorological fields in the model simulation than those used in this study, which has 1°×1° grid resolution. Using higher spatially resolved FFBC flux maps might also improve the agreement between the observed and simulated oxidative ratios for the Japan/Korea pollution events.'

As the reviewer mentioned, we measured O_2 and CO_2 mixing ratio (or mole fraction), not concentrations. So, we have changed the expression, 'concentration', to 'mixing ratio'.

In response to the reviewer's comments, we have adopted the term 'oxidative ratio' in our text. To define the term 'oxidative ratio', we have added the sentence 'In order to express the quantitative coupling between O_2 and CO_2, the oxidative ratio is defined as the $-O_2:CO_2$ molar exchange ratios: $OR=-\Delta O_2[mol]/\Delta CO_2[mol]$.' after the first sentence of 1st paragraph in Introduction (page 15632, line 23). To clarify that the $-\Delta O_2/\Delta CO_2$ ratio based on the atmospheric measurements are also referred as oxidative ratio, we have changed 'the $-\Delta O_2/\Delta CO_2$ changing ratios for such pollution events' (page 15633, line 12) to 'the oxidative ratios calculated as $-\Delta O_2/\Delta CO_2$ regression slopes for such short-term O_2 and CO_2 variations have recently been used'. Additionally, we have added the sentence 'From the linear regression slope, we obtain the oxidative ratio for the pollution event as $OR=-\Delta O_2/\Delta CO_2$' after the 5th sentence of the 2nd paragraph in Section 2.2.

To distinguish oxidative ratio based on the atmospheric observation from that based on the fossil fuel burning and cement production emission inventory, we have used OR_{FFBC}. We have used FFBC instead of FFB&C for fossil fuel and cement production.

Specific comments

Page 15632, line 17-19: In response to the reviewer's comment, we have changed relevant sentence to 'A sensitivity test suggests that the simulated atmospheric oxidative ratios at HAT is especially sensitive to changes in Chinese fuel mix.'

Page 15633, line 7 – 13: The atmospheric $-\Delta O_2/\Delta CO_2$ changing ratios were used to detect not only pollution events but also biomass burning and land biotic exchange as described after the relevant sentences. Thus inappropriate wording 'such pollution events' has been changed to 'such short-term O_2 and CO_2 variations'

Page 15633, line 13 – 23: In response to the reviewer's comment, we have added the observed oxidative ratios for the readers to understand the range of their variations. In addition, we have cited the following studies in the text: Sirignano et al. (ACP, 2010), van der Laan-Luijkx et al. (ACP, 2010), and Steinback et al (ACP, 2011). Consequently, the relevant sentences have been changed to 'For example, Stephens et al. (2003) conducted continuous in situ measurements of O_2 and CO_2 on research cruises in the equatorial Pacific and Southern Oceans and concluded that some short-term variations with OR=~1.4 were caused by the combustion of liquid fossil fuels. Examining the short-term atmospheric O_2 and CO_2 variations over a forest canopy at the WLEF tall-tower research site in Northern Wisconsin, Stephens et al. (2007) found that the ORs during winter range from 1.41 to 1.53, which is close to the average OR of 1.45 derived from the US fuel mix estimate in 2000. In addition, Sirignano et al. (2010) and van der Laan-Luijkx et al. (2010) investigated the influence of the Dutch fossil fuel mix (OR=1.69±0.06) , which has high natural gas share, on the atmospheric observation at Lutjewad and computed the observable OR of 1.49 with significant seasonality: lower in summer and higher in winter. To investigate the spatiotemporal variations in the ORs based on the atmospheric O_2 and CO_2 observations, Steinbach et al. (2011) developed a global data set of CO_2 emissions and O_2 uptake associated with the fossil fuel burning.

Page 15634, line 3: In response to the reviewer's comment, we have added the explanation of the national oxidative ratio and a pie chart showing fossil fuel shares. Consequently, we have added the sentences 'Actually, there is significant difference in the national average oxidative ratios for the emissions from fossil fuel burning and cement production (FFBC) among China ($OR_{FFBC}=1.11 \pm 0.03$), Japan ($OR_{FFBC}=1.37 \pm 0.02$) and Korea ($OR_{FFBC}=1.31 \pm 0.02$). These oxidative ratios are calculated using the fuel mix of the respective countries taken from CDIAC inventories for the year 2006 (Boden et al., 2010) and the oxidative ratios for the 3 main fuel types (Keeling, 1988) and the cement manufacturing ($OR_{cement}=0$). It should be noted that the higher percentage of the CO_2 emissions from coal and cement production for China results in a significantly lower oxidative ratio in comparison with Japan and Korea (Fig. 1).' after the last sentence of the 2nd last paragraph in Section 1.

In accordance with above change, we have deleted the 5th, 6th, 7th 8th and 9th sentences ('For comparison, we also calculate …it does not involve O_2 consumption.') of the 1st paragraph in Section 3.1 and added the sentences 'These averages of the observed oxidative ratios show excellent agreements with the fossil fuel oxidative ratios for China (OR_{FFBC}=1.11 ± 0.03) and Japan/Korea (OR_{FFBC}=1.34 ± 0.02), which are calculated from 2006 national fossil carbon inventories provided by CDIAC database. These values are summarized in Table 1 and are plotted in Figure 6.' after the 4th sentence in Section 3.1.A pie chart has been added as Figure 1.

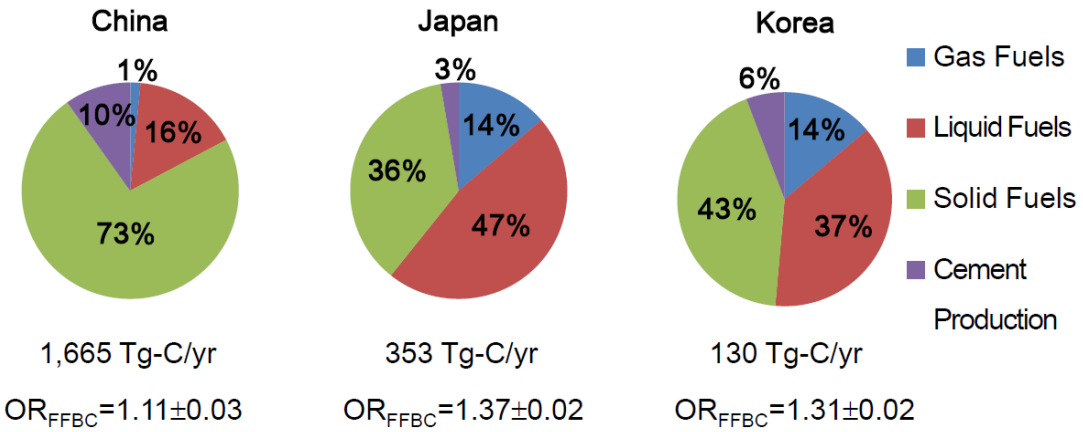

Figure 1. Compositions of fuel mix used in China, Japan and Korea in 2006 (Boden et al., 2010, Keeling, 1988).

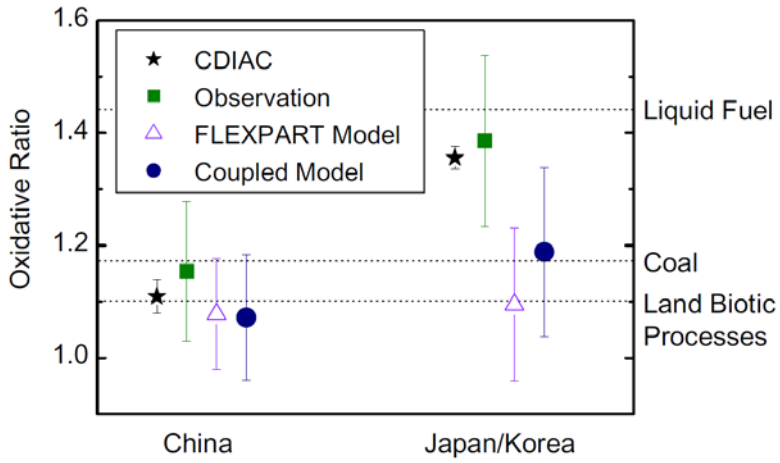

Figure 6. Oxidative ratios for the fossil carbon emissions from China and Japan/Korea, and average oxidative ratios for the China and Japan/Korea pollution events based on the observations and model simulations using FLEXPART and the coupled model. The national emission inventories of the fossil carbon from the CDIAC database are used to calculate the oxidative ratios for FFBC emissions. The vertical bar of CDIAC is the same as Table 1. The standard deviations are shown as vertical bars for observation, FLEXPART Model and Coupled Model. Dotted black lines show estimated oxidative ratios for land biotic processes (Severinghaus, 1995), coal, and liquid fuel burning (Keeling, 1998).

Page 15634, line 21-24: In response to the reviewer's comment, we have removed the details of the measurements. Consequently, the first sentence of the relevant paragraph has been changed to 'The air is sampled from an inlet at the height of 36.5 m on a tower (46.5 m above sea level) and then introduced into the O_2/N_2 measurement system including a gas chromatograph equipped with a thermal conductivity detector (GC/TCD).'

Page 15635/15636: In response to the reviewer's comment, we have redrawn Figure 1 (Fig. 2 in the revised manuscript) so that the oxygen changes are plotted as changes in the mixing ratio in ppm. According to this change, we have changed the wording 'O_2/N_2 ratio' to 'O_2 mixing ratio' in the last paragraph in Section 2.1 and the first paragraph in Section 2.2.

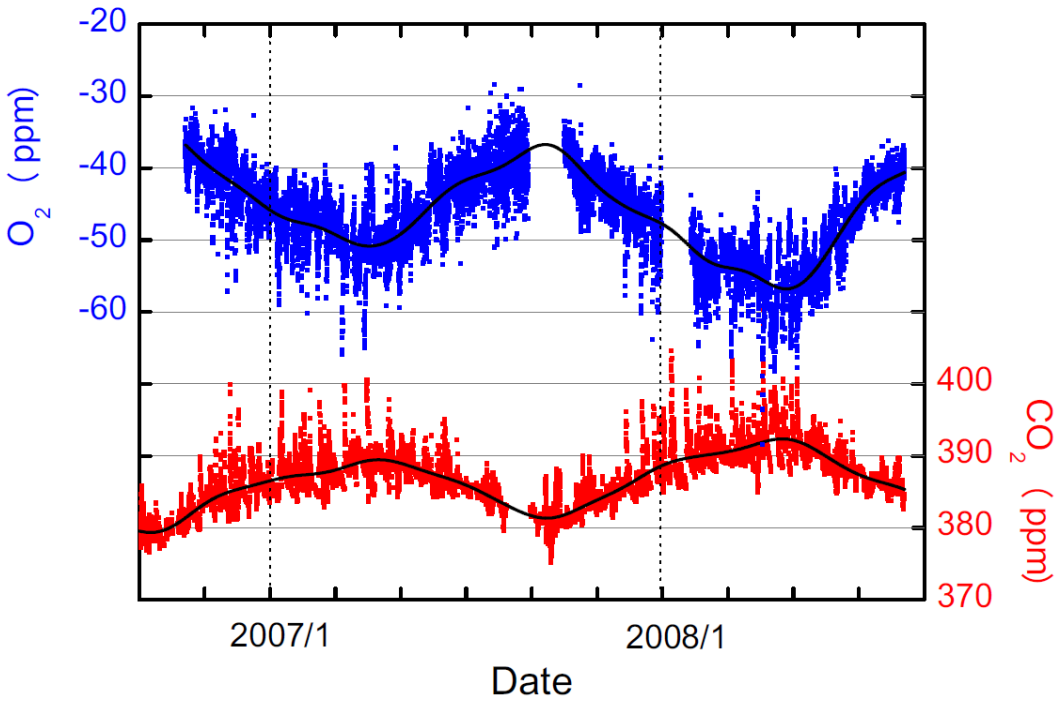

Figure 2. Time series of atmospheric (blue) O_2 and (red) CO_2 mixing ratios observed at HAT for whole period of this study. Each dot represents hourly average. The smooth curve fits to the data are shown as solid lines.

Page 15636, line 4-15: I have added, 'As an example,' at the beginning of the paragraph.

Page 15636, line 9-10: As the reviewer mentioned, we determined the start and end times of the peaks manually. In addition, in most cases, the correlation slopes are not sensitive to the choice of the start and end points of the pollution peaks. To clearly state these, we have changed the relevant sentence to 'It should be noted that the start and end times of each pollution event peak are determined manually and the following correlation analysis are not sensitive to the choice of the start and end time.'

Page 15637, line 2: To state what the flux categories exactly mean, '… individual flux categories to the pollution events, …' has been changed to '… individual flux categories such as FFBC fluxes, land biotic fluxes, and oceanic exchanges to the pollution events, ..'

Page 15637, line 21ff:
In order to clarify how to prepare FFBC CO_2 flux map, we have changed the ambiguous 5 sentences 'The CO_2 flux from fossil …top 20 countries were used.' to 'The CO_2 flux from FFBC was calculated using the fossil98 flux data that was employed in the TransCom model simulation (Law et al., 2008). The fossil98 flux dataset was based on the EDGARV32 CO_2 emission map with the spatial resolution of 1°×1° for the year 1990 (Olivier and Berdowski, 2001), which was then scaled to the emission level of 1998 using CDIAC totals. We used the individual national total emissions for the years 2006, 2007, and 2008 from the CDIAC database to scale the fossil98 CO_2 emission map for the top 20 CO_2 emitting countries. The emissions of the rest of the world is scaled to match the global total CO_2 emission of the year. Note that the FFBC CO_2 fluxes for the year 2008 were estimated by extrapolating the rate of increase from 2006 to 2007 for the top 20 countries.'

As the reviewer pointed out, ideally we should have used up-to-date emission maps because the emissions from China have shown rapid increase during the recent decade. However, it is our view that the influence resulting from the differences in the sub-country scale flux distributions on the observed oxidative ratio at HAT is likely relatively small because the individual country total emissions are scaled to match the CDIAC inventories. We have tried running FLEXPART with CO_2 and O_2 fluxes based on the EDGAR4.0 1x1 CO_2 emission flux map in 2005. The fluxes of the top 20 countries were scaled to match the CDIAC national totals for the year 2006, 2007 and 2008. The O_2 fluxes were calculated using the oxidative ratios in 2008. The average ORs for the China event stayed at 1.08 and the average OR for Japan/Korea events did not change very much and the change was from 1.09 to 1.12.Therefore, we consider our flux map to be of sufficient accuracy for reconstructing the oxidative ratios of source regions.

Page 15638, line 7ff: Following the reviewer's comment, we have already added the explanation how to calculate the national oxidative ratios from the fuel mix inventories at the last paragraph in Introduction. To clearly state what values were used to calculate the O_2 flux, we have changed the sentences "The national $-O_2$:CO_2 ratios of 1.11, 1.37, and 1.31 for China … to produce the O_2 fluxes." to "The national FFBC oxidative ratios of 1.11, 1.37 (1.38 in 2007 and 2008), and 1.31 for China, Japan, and Korea, respectively, are used to calculate the O_2 fluxes for the 3 years of 2006, 2007, and 2008. For the remaining countries, a single oxidative ratio, 1.45, based on the average fuel

compositions of these remaining countries is used for the O_2 flux map for the 3 years."

Page 15639, line 11-15 and 18-22: As the reviewer suggested, it has been moved to the 3rd paragraph in Introduction.

Page 15639, line 25-26: As the reviewer suggested, it is difficult to distinguish between the contribution of fossil fuel emissions and that of biospheric emissions for the pollution events assigned to the China origin. Therefore, we have deleted "and that land biotic and oceanic fluxes contribute little to the observed O_2 and CO_2 variations in these pollution events" from the last sentence in Section 3.1.

Page 15640, first paragraph: There are several pollution events between May and September, but all of the events are assigned to other regions and not to China or Japan/Korea. To state this clearly, we have added the sentence 'There is no pollution event between May and September in the figure because all the events during the period are assigned to other origins.' after the first sentence in Section 3.1.

As shown in Fig. 7 (Fig. 8 in the revised manuscript), most of the sharp peaks are derived from the fossil fuel emissions and the influences of the biospheric and oceanic emissions are relatively small. The phase difference may come from the coarse time and spatial resolution of the meteorological field and the air mass change at the observation point is captured with time gaps. In general, continental air masses seldom arrive at HAT during summer because of the East Asian monsoon. Therefore, discussion of the contribution from biospheric and oceanic emissions to the elevated peaks during summer is not within the scope of this paper.

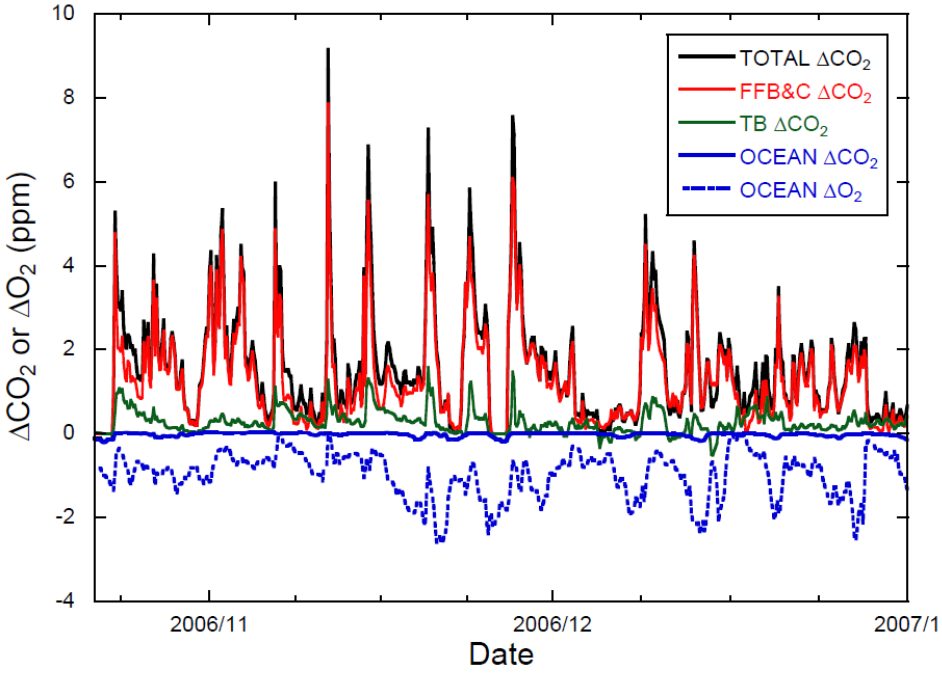

Figure 8. Components of the ΔCO_2 variation at HAT calculated by FLEXPART. Black line is the total amount of CO_2 from the following three components, red line is the amount of CO_2 from fossil fuel burning and cement production (FFBC), green line is from terrestrial bioshere (TB), and blue line is from the ocean. Oceanic ΔO_2 component is also plotted as broken blue line.

Page 15640, second paragraph: We think the reviewer's question is in regard to the content of the third paragraph. We have checked the CO_2 release and oxygen uptake from the Fossil Fuel Emission Estimate (COFFEE) dataset developed by Steinbach et al. (ACP, 2011), which includes temporal flux changes. We have found that the temporal variations in the OR_{FF} for the fossil fuel emissions from China during the period from the late autumn to early spring is small, at most about 0.02. Therefore, it is our view we that the temporal changes in the OR_{FFBC} are not responsible for the disagreement in the oxidative ratio between the observation and simulation for individual events. We think, as the reviewer has indicated, that the heterogeneity of the actual OR_{FFBC} distribution on a sub-country scale might partially contribute to the differences between the observed and simulated oxidative ratios for the individual pollution events. However, it is highly possible that the disagreements are mainly caused by transport errors.

Page 15641, line 4-5:

During this time of the year (mainly winter), the mid latitude regions of the western North Pacific generally absorb atmospheric O_2. Because the absorbing flux of O_2 is larger than that of CO_2, the air masses influenced by the air-sea gas exchanges have more depleted O_2 mixing ratio than CO_2. When the air masses arriving at HAT are transported from the continental region, the simulated oceanic O_2 component sometimes shows positive peak-like changes depending on the changes in the influence of the air-sea gas exchanges. The time series of the simulated oceanic O_2 component are depicted in the redrawn Fig. 7 (Fig. 8 in the revised manuscript). These positive peaks of the oceanic O_2 component attenuate the O_2 depletion associated with the continental pollution events, resulting in a reduction of the $-\Delta O_2/\Delta CO_2$ ratios. Consequently, we have changed the relevant sentence 'This is mostly due to … closer to 1.10' to 'During this time of the year, the mid-latitude regions of the western North Pacific ocean act as a sink for the atmospheric CO_2 and O_2, and the absorbing fluxes of O_2 are much larger than those of CO_2. The time series of the simulated oceanic O_2 component, shown in Fig. 8, sometimes show positive peak-like changes when the air masses are transported from the continental side because of the reduction in the contribution from the air-sea gas exchange. Such O_2 peaks could reduce the apparent magnitude of the O_2 depletion associated with the continental pollution events, resulting in the lower-than-biospheric oxidative ratios. Indeed, we find that removing the oceanic CO_2 and O_2 components in model simulations brings those averages closer to 1.10.'

P15641, line 20-23:

In response to the reviewer's comment, we have changed the relevant sentence and the following sentence, 'That is due to … at this time of year). In addition, the predominant … with little spatial variability.' to 'That is probably explained by the fact that the distribution of the FFBC CO_2 emissions is localized in highly populated areas while that of biospheric emissions is rather homogeneous. In addition, the localized FFBC CO_2 fluxes are generally one order and two orders of magnitude larger than the TB CO_2 and oceanic fluxes, respectively (CO_2 is absorbed slightly by the ocean in the marginal region of the East Asia at this time of the year). The spatial distributions of the FFBC flux for 2006 and the TB and oceanic CO_2 fluxes for January are shown in Fig. 9.'

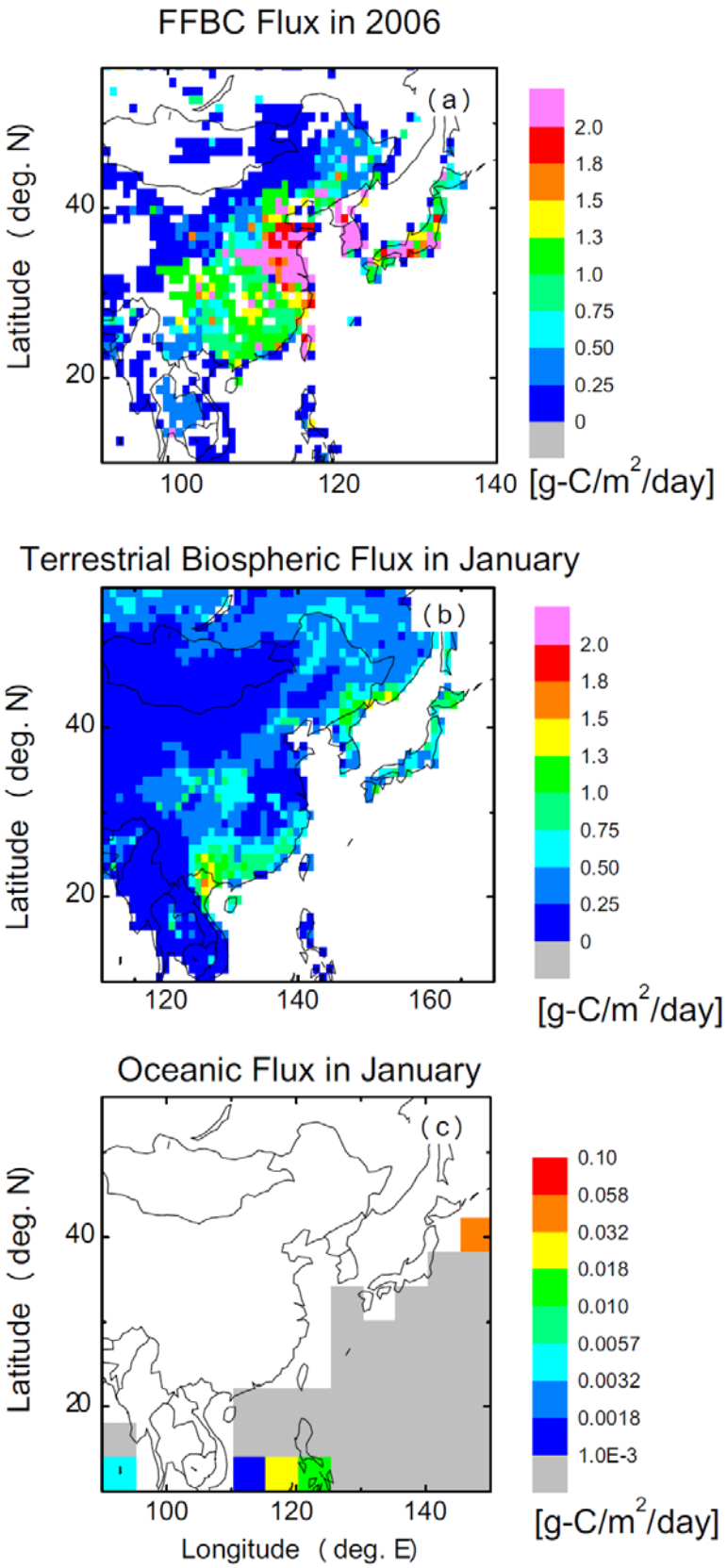
FFBC Flux in 2006
(a)
Latitude (deg. N)
40
20
100
120
140
2.0
1.8
1.5
1.3
1.0
0.75
0.50
0.25
0
[g-C/m2/day]
Terrestrial Biospheric Flux in January
(b)
Latitude (deg. N)
40
20
120
140
160
2.0
1.8
1.5
1.3
1.0
0.75
0.50
0.25
0
[g-C/m2/day]
Oceanic Flux in January
(c)
Latitude (deg. N)
40
20
100
120
140
Longitude (deg. E)
0.10
0.058
0.032
0.018
0.010
0.0057
0.0032
0.0018
1.0E-3
[g-C/m2/day]

Figure 9. The spatial distributions of (a) FFBC flux for 2006, (b) TB flux for January, and (c) oceanic CO_2 flux for January. These fluxes are used in the model simulation of this study.

Some additional thoughts on Figure 7 (Fig. 8 in the revised manuscript):

1. It is not just a coincidence. Most of the biospheric signals are correlated with the fossil fuel events. However, the peak shapes are different in details between biospheric and fossil fuel signals, reflecting the differences in the flux distributions. In addition, the contributions of the TB fluxes on the simulated oxidative ratios for the pollution events are negligible.

2&3. Probably, the unclear plots confuse you. We have redrawn the figure so as to clarify the individual lines. In addition, we have added the O_2 component derived from oceanic O_2 fluxes in Fig. 7 (Fig. 8 in the revised manuscript). The oceanic O_2 plots would help to explain how the oceanic O_2 signals affect the observed oxidative ratios for some pollution events.

Page 15643, line 12ff:

1. In Fig. 8 (Fig. 10 in the revised manuscript), we did not depict the footprint on the ocean because oceanic CO_2 fluxes are generally negative (absorbing flux) and the magnitude of the oceanic fluxes are much smaller than the continental fluxes. As the reviewer mentioned, the footprint in Fig. 8b looks rather localized, but this is due to the fact that the footprints on the ocean are omitted. Therefore, we have added the distribution of the dispersed particles calculated by FLEXPART in the figures. These distributions clearly show that the dispersed particles are not localized. In general, the simulated oxidative ratios for the Korea/Japan pollution events are lower than the observation because of the substantial contribution from the emissions from China to the Korea/Japan events in the model simulation. From these results, we simply concluded that the particles in FLEXPART seem to spread more than in reality.

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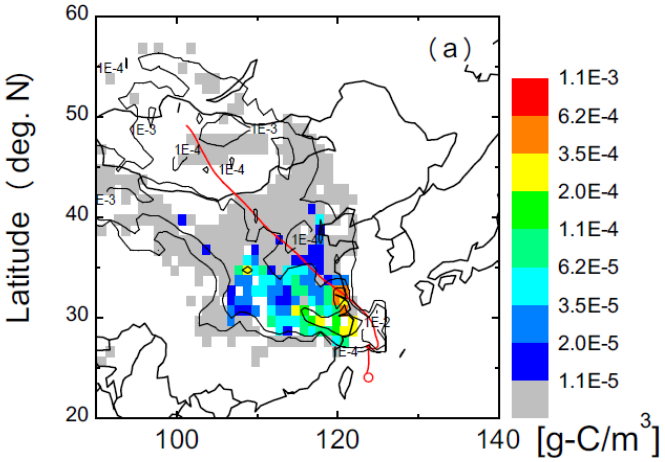

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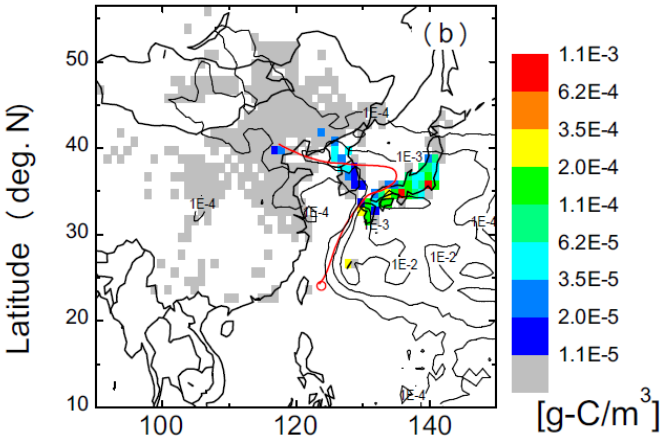

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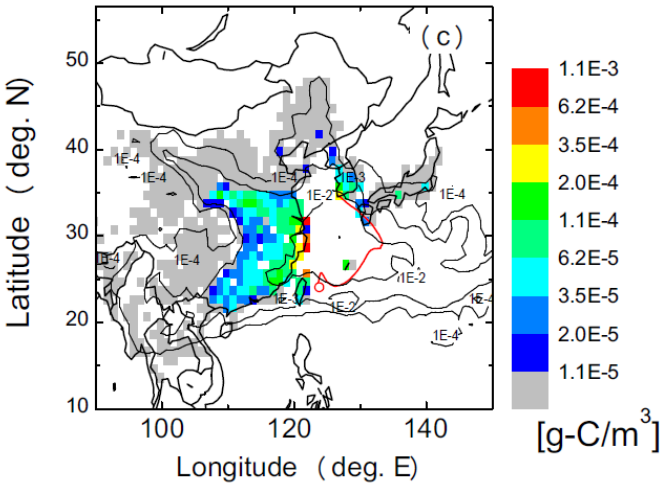

Figure 10. Footprint for HAT calculated by FLEXPART for the cases in which air mass origins are assigned to (a) China, and (b,c) Japan/Korea by back trajectory analysis. (b) and (c) are the cases with small and large Chinese influence, respectively. Color scale is logarithmic. The distributions of the dispersed particles are also shown as thin-black contours, which represent the fraction of the number of particles occurs in each grid cell below the mixing layer height of 1000m in the duration of backward simulation, 8 days. The red lines represent 5-day back trajectories calculated by METEX.

2. We have changed the misleading wording 'both problems' to 'the problem'.

3. We agree with the reviewer's comment that the model-observation mismatch is mainly attributable to transport problems. Accordingly, we have changed the sentence to read 'Another approach is to use more spatially resolved fluxes.' to 'More spatially and temporally resolved fluxes might help to improve the agreement between simulated and observed individual events.'

4. Besides the altitude at which the particles absorb the flux, we changed the number of particles to release, the range of initial positions of the particles, and time step at which the particles absorb the flux, but the results were not improved. At this point, except the spatiotemporal resolution of meteorological fields and input fluxes, we cannot think of any other possible changes we can make in the model setup. Therefore, we have deleted the last sentence in Discussion.

Page 15644, line 3ff:
In response to the reviewer's comments, we have changed the relevant paragraph as shown in the reply to the reviewer's General comments.

Technical comments/suggested text edits

In response to the reviewer's comment that some sentences are quite long, we have asked a native English speaker to correct our manuscript.

Page 15635, line 24: The relevant sentences have been changed as suggested.
Page 15636, line 2-3: The wording 'in the following' has been added to the relevant sentence.

Page 15636, line 4: ‘as follows.’ has been changed to ‘as follows:’

Page 15636, line 14: ‘in the followings’ has been changed to ‘in the following’.

Page 15636, line 19: ‘close with’ has been changed to ‘close to’.

Page 15636, line 23 (probably typo of 17)**:** To be consistent through the manuscript, ‘METEX (METeorological …/metex/)’ has been changed to ‘METeorological data Explorer (METEX, http://db.cger.nies.go.jp/metex/)’.

Page 15636, line 16 and 28 (as well as later on page 15643, line 23): Following the reviewer’s suggestion, ‘other’ has been changed to ‘Other Origins’.

Page 15637/15638: ‘made’ has been changed to ‘calculated’.

Page 15637, line 26: Following the reviewer’s suggestion, the citation of (Marland et al., 2007) has been changed to ‘Marland, G., Boden, T. A., and Andres, R. J.: Global, regional, and national CO_2 emissions, Carbon Dioxide Inf. Anal. Cent., Oak Ridge Natl. Lab., U. S. Dep. Of Energy, Oak Ridge, Tenn., http://cdiac.ornl.gov/trends/emis/tre/regn.html, last access: 3 Aug. 2008, 2007.’

Page 15639, line 4: ‘are shown in Fig. 4a, in which the ratios are collapsed into a single year’ has been changed to ‘are shown in Fig. 4a, plotted against time of the year’.

Page 15639, line 5: ‘both events’ has been changed to ‘both types of events’.

Page 15639, line 6-7: The relevant sentence ‘the range for China origin tends to be lower (1.0-1.4) than the latter (1.1-1.7)’ has been changed to ‘O_2/CO_2 ratios for events with origin from China tend to be lower (ranging from 1.0-1.4) than those with origin from Japan/Korea (1.1-1.7)’.

Page 15641, line 15-17: ‘This result suggests that the pollution events at HAT …’ has been changed to ‘This result suggests that the synoptic-scale events at HAT …’.

Page 15642, line 2 – 13: In response to the reviewer’s comment, we have added Table 2, in which the results of the sensitivity tests and their uncertainties are summarized. Following this change, we have modified the relevant paragraph to read as: ‘In order to confirm that the emissions from FFBC are the main contributor to the simulated ORs and not the emissions from TB, two sensitivity tests were performed. First, a FLEXPART model simulation was performed in which the land biotic oxidative ratio was changed from 1.1 to 1.0, and the ORs for the pollution events were then recalculated. Second, the national OR_{FFBC} for China was changed from 1.11 to 1.00, and then the ORs for the pollution events were recalculated after the FLEXPART model simulation. The results of the sensitivity tests are summarized in Table 2. The former experiment decreased the average OR for China events from 1.08 to 1.05, while the average OR for Japan/Korea events stayed at 1.09. The latter experiment decreased the average OR for China and Japan/Korea from 1.08 to 0.98 and 1.09 to 1.06, respectively.

These results confirm that the average OR for China events is much more sensitive to the Chinese national OR_{FFBC} than to the land biotic oxidative ratio.'

Table 2. Summary of the sensitivity tests for the FFBC and land biotic oxidative ratio in China

Air mass origin	FLEXPART		
	Original Results	OR_{FFBC}=1.0	OR_{TB}[a]=1.0
China	1.08±0.10 (14)	0.98±0.09(14)	1.05±0.09(14)
Japan/Korea	1.09±0.14 (13)	1.06±0.17(13)	1.09±0.14(13)
Other	1.15±0.16 (14)	1.18± 0.18(14)	1.13± 0.16(14)

[a]OR_{TB} refers to an oxidative ratio for land biotic processes.

Page 15642, line 20ff: In response to the reviewer's comment, we have changed the order of the explanations. We have also modified the redundant explanations in the relevant paragraph. Consequently, we have changed the relevant paragraph to read as:
'Figure 10 shows footprints as determined by the FLEXPART simulation for representative pollution events observed at HAT. Figures 10a, 10b, and 10c correspond to the periods of 04:00-22:00 (LT) on March 3, 2008, 02:00 on 6 November-09:00 on 7 November, 2007, and 01:00 on 14 March-14:00 on 15 March, 2008, respectively. The footprint [g-C m^{-3}] is defined as a product of the ratio of particle number in a grid cell in a pre-determined mixed layer height to the total particle number [no unit], the residence time [day], and the anthropogenic FFBC CO_2 flux [g-C m^{-2} day] divided by the pre-determined mixing layer height of 1000 m. Thus, those grid cells, which have large flux and particle numbers, have large contribution to the observed signal at HAT. In Fig. 10, the distributions of the dispersed particles are also shown as thin-black contours. In addition, the red curves in the figure show the backward trajectories for the individual pollution events. Based on the trajectory analysis, the pollution event shown in Fig. 10a is shown to originate in China, and is consistent with its footprint. Although the both pollution events shown in Figs. 10b and 10c are assigned origins in Japan/Korea, the distribution patterns of the footprints are significantly different. The former footprint (Fig. 10b) covers mainly Japan and Korea while the latter footprint (Fig. 10c) shows that the emissions from China contribute about 80% of the pollution event.'
Page 15642, line 23 and 25: 'grid' has been replaced by 'grid cell'.
Page 15642, line 21: This paragraph is rewritten and the expression is not used.
Page 15643, line 9ff: The relevant sentence has been changed to 'In the FLEXPART

simulation, pollution events categorized by the back trajectory analysis as Japan/Korea in origin almost always contains substantial contribution of Chinese fluxes, therefore their simulated average $\Delta O_2/\Delta CO_2$ value is closer to that of China.'

Page 15644, line 9: '… at HAT are attributed to FFBC …' has been changed to '… at HAT are indeed attributed to FFBC…'.