Response to Anonymous Referee #1

We thank the reviewer for the valuable and helpful comments. We believe that addressing the issues raised by the reviewer will considerably improve the manuscript. Please see our reply to each comment below.

This study uses springtime HIPPO flights across the Pacific to test the GEOS-Chem simulation of BC and aging/transport processes. The manuscript is straight-forward and clearly presented. I have a few major comments which should be addressed prior to publication.

1. Page 509, lines 25-29: You mention the importance of dry deposition in the Introduction but never discuss the dry deposition flux simulated with GEOS-Chem in the text. Is the dry deposition of BC high or low compared to other studies? Could near-field dry deposition processes impact your simulation?

In GEOS-Chem simulations, dry deposition is implemented following a standard resistance-in-series scheme as described by Wang et al. (1998) and is less important to wet deposition for fine aerosols in a global scale (Park et al., 2005). In GEOS-Chem, the global annual mean dry deposition velocity for BC is 0.1 cm s^{-1}, which is typical of current models (Wang et al., 2011). Figure R1 shows the dry deposition flux and velocity of BC during the period of our simulations. To reduce the influence of dry deposition on our simulation, we focus on an observation region between 2 km and 6 km. In addition, to assess the importance of dry deposition on our simulations, we conduct a sensitivity test by turning off dry deposition and compare it with BC simulations using standard GEOS-Chem model. Figure R2 shows GEOS-Chem simulated BC vertical profiles in our observation domain with and without dry deposition. BC concentrations have similar pattern but larger values when turning off dry deposition, while the differences are less than 20%. As a result, dry deposition does not have a large impact in this study. To make this clear, we have added a description on dry deposition in GEOS-Chem simulation in Section 2.2:

"*Dry deposition is implemented following a standard resistance-in-series scheme as described by Wang et al. (1998). Dry deposition is not important for fine aerosols (Park et al., 2005) and is small compared to wet deposition in BC simulation (Park et al., 2003). In GEOS-Chem, the global annual mean dry deposition velocity for BC is 0.1 cm* s^{-1}*, which is typical of current models (Wang et al., 2011).*"

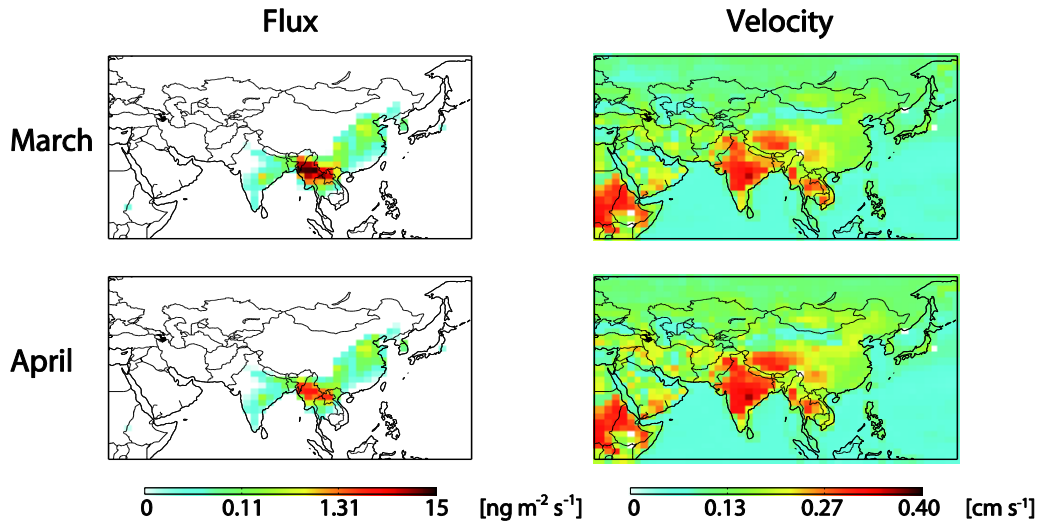

Figure R1. Dry deposition flux (left) and velocity (right) of BC in GEOS-Chem during 1 March–1 April 2010 (top) and 15 March–15 April 2010 (bottom).

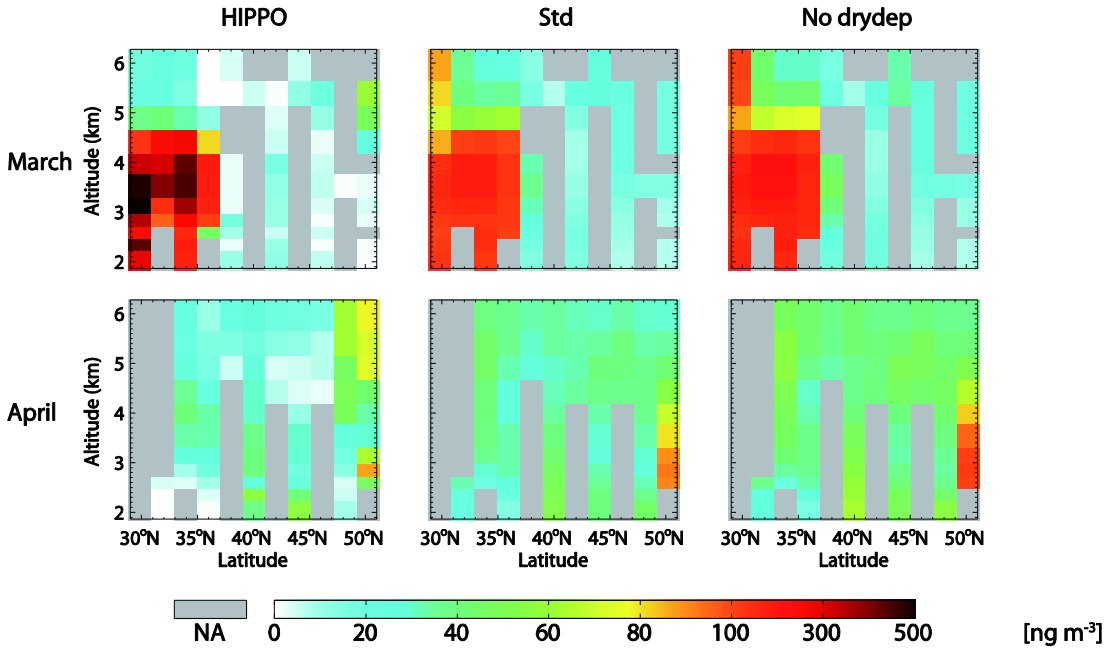

Figure R2. BC vertical profiles in observational domain (150–160 °W) on 29 March 2010 (top) and 13 April 2010 (bottom). HIPPO aircraft observations (left) and GEOS-Chem model estimates (sampled along aircraft track) with (center) and without (right) BC dry deposition are shown.

2. Page 517: The high BC concentrations aloft in April are surprising, given that the authors rule out biomass burning. Could you speculate as to the cause? Is this evidence of lofting or drier export? Is there a mechanism that you could use in the model to reproduce this signature at 6km?

Some studies have discussed the vertical transport and export efficiency of BC particles using observations in the free troposphere (Oshima et al., 2013;Park et al., 2005). Oshima et al. (2013) identified that the uplifting of BC in association with migratory cyclones over northeastern China and the subsequent BC transport by the midlatitude tropospheric westerlies provided the major pathway for BC export from East Asia to the free troposphere over the western Pacific during springtime in the A-FORCE period. The high BC concentrations aloft in April measurements during HIPPO-3 may be caused by similar mechanism and uncertainties in the calculations of vertical transport of BC in the model account for some of the model biases. Figure R3 compares GEOS-Chem model simulated CO concentrations over the North Pacific with observations in April measurements during HIPPO-3. Observed CO concentrations have a similar pattern as BC near 50 ℃N, which model is unable to reproduce. As CO cannot be removed by wet deposition, this further indicates that large uncertainties remain in modeling the vertical transport processes.

In addition, MODIS daily fire data (https://firms.modaps.eosdis.nasa.gov/firemap/) suggests that there were some fires in northeast Eurasia in April, 2010. The biomass burning BC may also result in the high BC concentrations at higher altitudes near 50 ℃N in April. We use monthly biomass burning emissions in this study, which causes some biases. We have added discussion on this issue in Section 4. Now we have:

"*There is larger disagreement between model simulations and observations in April, as the model cannot resolve the spatial variation of observed BC concentrations. Model generally over-estimates BC at 40-45 ℃N. Another major bias is that there is an over-estimate in model simulations at lower altitudes and an under-estimate at higher altitudes near 50 ℃N, mainly because there are large uncertainties in the calculations of vertical transport of BC (Koffi et al., 2012;Oshima et al., 2013). Oshima et al. (2013) identified that the uplifting of BC in association with migratory cyclones over northeastern China and the subsequent BC transport by the mid-latitude westerlies provided the major pathway for BC export from EA to the free troposphere over the western Pacific during the springtime A-FORCE period. Here the observed high BC concentrations aloft in April may be caused by the same mechanism. Similar model biases are found when we compare GEOS-Chem model simulated CO concentrations over the North Pacific with observations in April measurements during HIPPO-3 (not shown), which further indicates that large uncertainties remain in modeling the vertical transport processes.*"

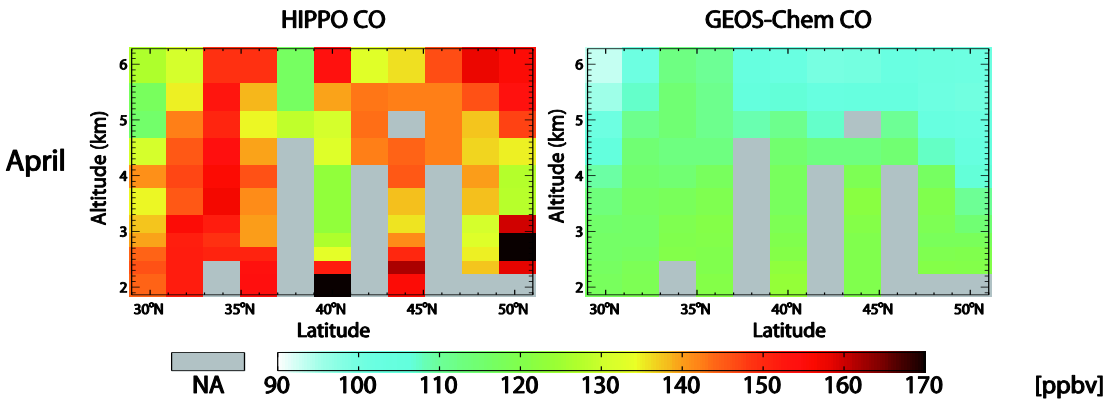

Figure R3. HIPPO observed (left) and GEOS-Chem simulated (right) CO vertical profiles in the observation domain on 13 April 2010.

3. I'm a little unclear as to what we learn from the analysis of Section 5.2. Why did the authors assume N=5? How do the results change if you change N?

In Section 5.2, we perform full CTM simulations for different BC aging times and get the total BC transported to the relevant geographical region for each run. We then use our idealized model to fit the transported BC as a function of BC emission, the time of precipitation events that remove BC en-route, and BC aging time. Our goal is to give a sense of the average time between BC emissions and removal by each precipitation during HIPPO-3, and thus to give some implications for the general precipitation pattern that significantly influences the HIPPO-3 obs. For tracers of 1- and 2-week lifetimes (e.g, BC), the average transpacific transport time from East Asia to the western North America in spring is 2–3 weeks (Liu and Mauzerall, 2005). Our idealized model indicates that most precipitation events occur in 0-3 days after BC aerosols are emitted. Thus we conclude that there is a prevalence of precipitation events near the East Asian source region during HIPPO-3, and hence that BC aging rate near source regions is important in BC simulations.

As mentioned in Section 2.4, we assume BC is removed by several independent precipitation events during transpacific transport, while in reality, it is more likely that we have continuous precipitations. Since our target is to analyze the general precipitation pattern, N should not have a large impact on the results. We simply assume five precipitation events and find that three of them occur shortly after BC aerosols are emitted, leading to the conclusion that there is rapid wet scavenging near BC source regions. Figure R4 shows the fitting results for $N = 6$ and Table R1 shows the parameters. Four out of six precipitation events occur within four days after BC aerosols are emitted with relatively high removal efficiency, which proves that N does not largely affect our conclusion. We have added more discussions in Section 5.2:

"*The timing of precipitation experienced by the Asian outflow is another factor affecting the transpacific transport. In order to further analyze the characteristics of*

wet removal of BC during HIPPO-3, we use Eq. (3) to fit the amount of BC transported to the North Pacific (C) in the sensitivity runs as a function of the BC aging timescale (τ). We assume five major precipitation events during the transpacific transport (i.e., N = 5) and find that the assumption of N does not have a large impact on our conclusions when N > 5. We also assume 80% of BC emitted is hydrophobic (i.e., α = 0.8), which is the same as the assumption in GEOS-Chem. Figure 9 shows the fitting results in March and April missions, where y is the total BC emission contributions and x is the inverse of the e-folding time (1/τ). When assuming transpacific transport of BC is determined by the aging rate, the precipitation time, and the fraction of BC removed by precipitation, our idealized BC transport model has satisfied fitting precision (R^2 = 0.999) in both missions and can show some implications for general precipitation and wet removal patterns during HIPPO-3."

Table R1. Parameters derived from the fit of BC contributions in the sensitivity runs in March and April emissions based on Eq. (3) when assuming N = 6.

Parameter	T_1 (d)	T_2 (d)	T_3 (d)	T_4 (d)	T_5 (d)	T_6 (d)
March	0.08	0.38	1.3	3.3	8.2	19.6
April	0.08	0.42	1.5	3.6	9.0	20.9
Parameter	R_1	R_2	R_3	R_4	R_5	R_6
March	5.6%	15.8%	32.5%	42.8%	22.0%	12.7%
April	2.1%	8.0%	28.9%	38.0%	25.6%	14.3%

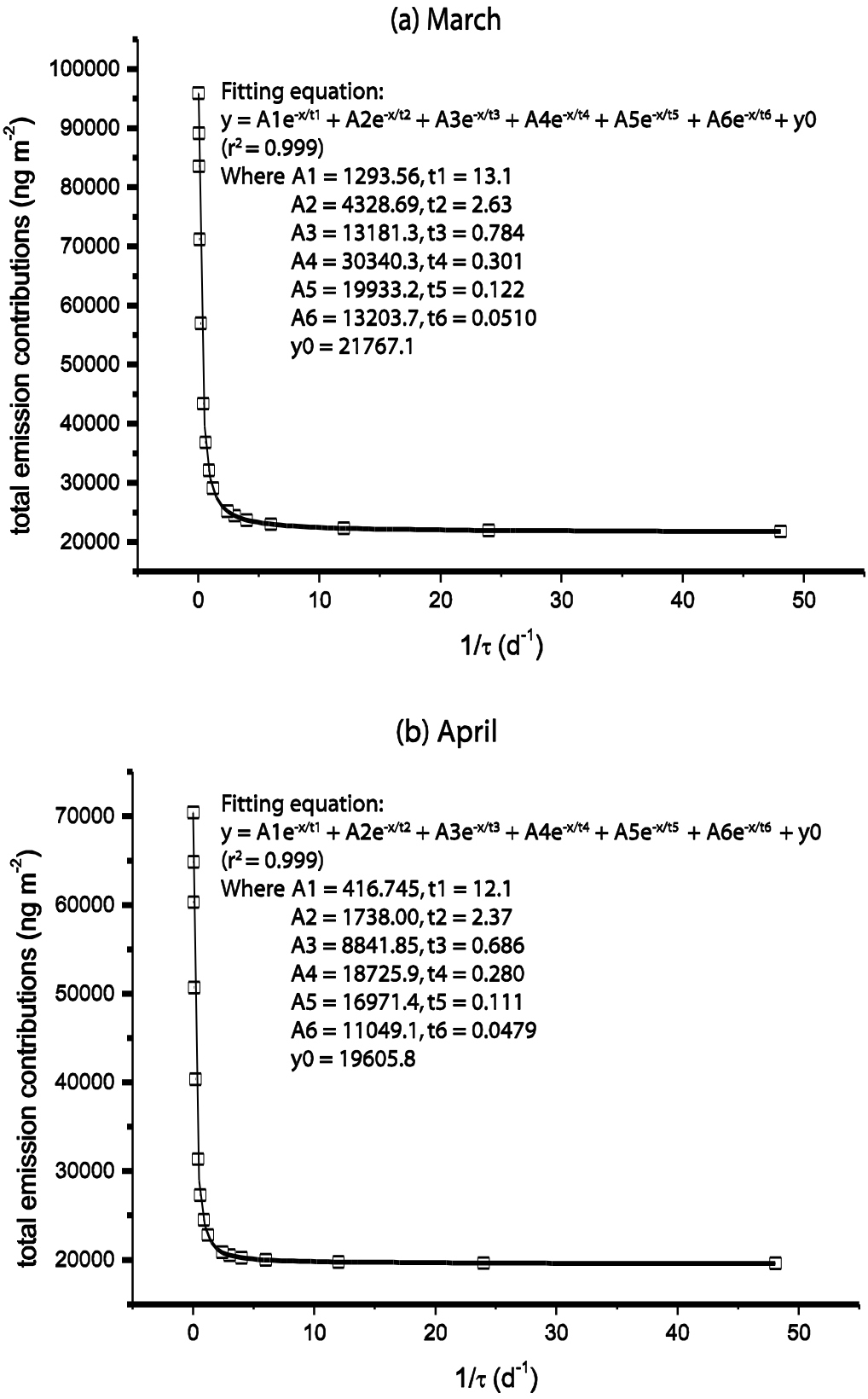

Figure R4. Result of fitting transpacific transport of BC with different e-folding aging times to idealized transport model (Eq. 3) in (a) March 2010 and (b) April 2010 when N = 6.

Figure 5 shows that changing the aging rate does not significantly improve the model ability to capture the vertical profile (just shifts it). Did the authors consider if there might be errors in the cloud top heights or vertical distribution of precipitation? Or rain rate? Or type of event? Your paper shows that neither modifications to the emissions nor aging rate can substantially improve the model simulation, so I'm left with the question of why this simulation performs so poorly compared to these observations.

Figure 5 shows that for the regions where most BC comes from anthropogenic sources, using a smaller aging time improves the model, while for the regions where BC is dominated by biomass burning, using a larger aging time is better. Hence, changing the aging rate of BC from all sources uniformly does not significantly improve the model performance.

It is true that in our study, neither modifications to the emissions nor the aging rate can significantly improve the model. As mentioned by the reviewer, there are errors in other factors controlling transpacific transport of BC. We assume a fixed aging time of BC, while some chemical or physical factors may cause the BC aging rate to vary. For example, Huang et al. (2013) implemented a new detailed aging scheme for carbonaceous aerosols in GEOS-Chem to account for both the chemical oxidation and the physical condensation-coagulation effects, resulting in large spatial and temporal variations in the aging time and an improvement in model simulations for the remote areas in the Northern Hemisphere. Wet deposition, which is the main BC sink, is one of the most important of model biases in the remote troposphere. Wang et al. (2014) compared GEOS-Chem model simulated BC with HIPPO observations and found that BC wet scavenging may be much more efficient than what is implemented in models. Treatment of cloud processes in models, which has large uncertainties, is important for BC removal and thus for BC simulations, especially at high altitudes. Fan et al. (2012) proposed that a latitude-independent information on ice formation processes in the atmosphere is important to BC removal in models. We have added discussion on this issue in Section 5.1 (see below) and our future work may focus on other factors controlling long-range transport of BC.

"*It should be noted that in our study, neither modifications to the emissions (Figure 2) nor the fixed aging rate (Figure 5) are able to significantly improve the GEOS-Chem model simulation of BC over the North Pacific during HIPPO-3, mainly because there are large uncertainties remaining in other factors controlling transpacific transport of BC. We assume a fixed aging time of BC, while some chemical or physical factors may cause the BC aging rate to vary. Huang et al. (2013) implemented a new detailed aging scheme for carbonaceous aerosols in GEOS-Chem to account for both the chemical oxidation and the physical condensation-coagulation effects, resulting in large spatial and temporal variations in the aging time and an improvement in model simulations for the remote areas in the Northern Hemisphere. Wet deposition, which is the main BC sink, is one of the most important of model biases in the remote*

troposphere. Wang et al. (2014) compared GEOS-Chem model simulated BC with HIPPO observations and found that BC wet scavenging may be much more efficient than what is implemented in models. Treatment of cloud processes in models, which has large uncertainties, is important for BC removal and thus for BC simulations, especially at high altitudes. Fan et al. (2012) proposed that a latitude-independent information on ice formation processes in the atmosphere is important to BC removal in models. Here we focus on the fixed aging rate of BC in model simulations, but the processes mentioned above should be addressed in future work."

MINOR

1. The HIPPO flight tracks are not shown in any figure. These should be included somewhere so that the reader has a better idea of the domain of the observations.

We have added the HIPPO flight tracks in Figure 1.

2. Abstract, line 15: uncertainties in removal as well as transport?

We have changed "transport" to "removal as well as transport".

3. Page 512, line 23: Please specify if the biomass burning emissions used are for 2010.

Yes, and we have added "in 2010" after "GFED3 inventory".

4. Page 512/513: The adjoint model is based on an older version of GEOS-Chem. What inconsistencies does this introduce in the analysis?

As mentioned in Section 2.2, the version of the adjoint model we used in this study is v34, which is based on v8-02-01 of GEOS-Chem but updated according to v9-01-03. The developers do their best to keep the adjoint model up-to-date. Although it isn't always in sync with the most recent version of the forward model (GEOS-Chem), for black carbon, the simulations of the adjoint model and the forward model are very much the same.

5. Page 516, line 12 and line 22: The domain of Figure 1 extends beyond Asia (includes the Middle East). If the percentages listed in the figure are for the entire domain, they should not be cited in the text as representing "from Asia" or "combustion in China".

Yes the percentages listed are the ratio of the contributions of each BC source to all BC sources for the entire domain. We have calculated the percentages for BC sources in Asia only (60 °E-150 °E), as shown in Figure R5. Since the colorbar is on the log

scale, the contribution of BC from Asia is much larger than that from the rest area in the domain, therefore the numbers do not change much. We have changed "Asia" to "Asia and Middle East" in the revised manuscript.

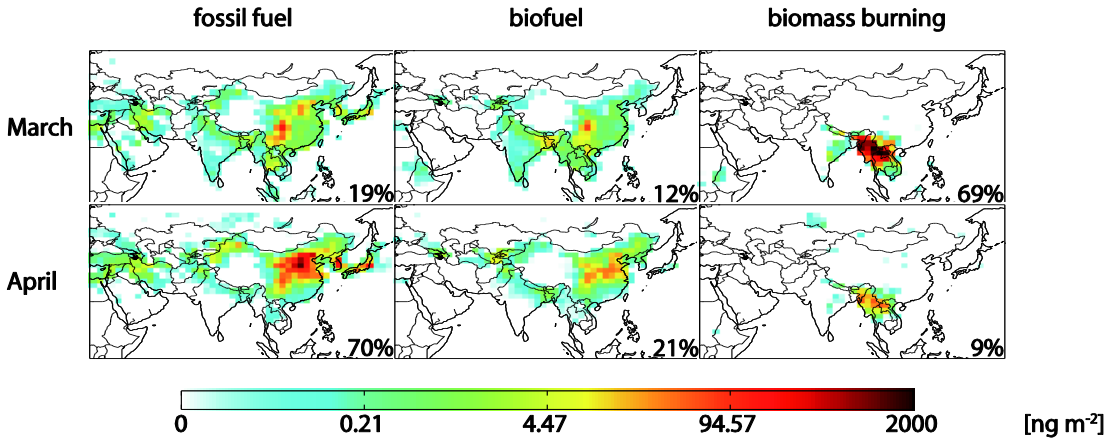

Figure R5. Adjoint model sensitivities of BC concentrations in the HIPPO-3 observational domain over the North Pacific in March and April missions with respect to BC emissions from different sources. The percentages listed on each plot indicate the relative contribution of specific Asian BC sources in each month.

6. Page 516, line 17: How different are March and April in the GFED inventory used?

Figure R6 shows BC emissions from biomass burning in the GFED inventory used in our study. Asian BC biomass burning emissions in March are larger than those in April by a factor of 6. We have added "*In this study, BC emissions from biomass burning in SE based on GFED3 inventory in March are larger than those in April by a factor of 6.*" after "biomass burning in SE peaks in March".

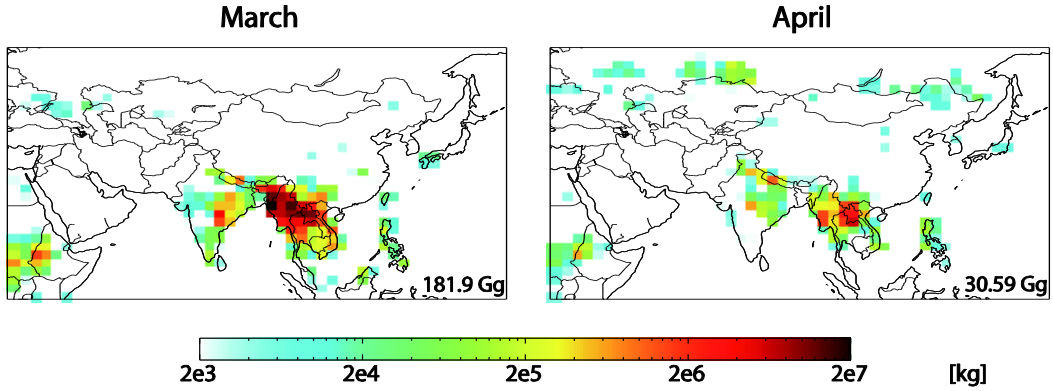

Figure R6. BC emissions from biomass burning during March and April, 2010 based on GFED3 inventory. The numbers listed on each plot indicates total monthly biomass burning BC emissions in Asia.

7. Page 518, lines 20-28: Your assumption of diagonal error covariances is also unrealistic and likely introduces substantial uncertainties in the inversion.

As discussed in the manuscript, using a diagonal error covariance matrix could have an impact on the inverse results. However in previous studies on adjoint inverse modeling, the assumption of a diagonal error covariance matrix is often made for simplicity and practical reasons (Henze et al., 2009;Kopacz et al., 2010;Kopacz et al., 2009;Wang et al., 2012). We always do not necessary have good information on what the off-diagonal elements should be, so estimating them just introduces another level of uncertainty. In practice, the penalty term of the cost function can be viewed more as a smoothness constrain on the inverse modeling solution rather than a rigorous statistical description. Also, as mentioned in Section 4, perturbation tests on the error covariance (in low resolution case, not shown) do not largely impact the results. As the target of our study is to analyze the factors causing model biases in BC simulation instead of providing an optimized BC emission inventory, we think that assuming diagonal error convariances is sufficient.

8. End of Section 5.1: What about co-emitted organics in biomass burning?

It is true that there are some organics co-emitted with biomass burning BC. There are many measurements of the coating of biomass burning BC, and the hygroscopicity of the coating materials is important in determining how much of BC can be treated as CCN and removed along transpacific transport (Petters et al., 2006). We have discussed more about this in the revised manuscript in Section 5.1:

"*Chamber studies and measurements have shown that chemical aging of biomass burning aerosols by atmospheric oxidants increase their hygroscopicity and hence their ability to active as CCN (Akagi et al., 2012;Petters et al., 2009). However, the coating thickness and the likelihood of removal of these aerosols has not been determined, and the fate of carbonaceous aerosols is actually controlled by the interaction with more hydrophilic species (Petters et al., 2006). Also, the oxidation aging of BC is largely affected by ozone and water vapor concentrations, resulting in long BC lifetimes in the tropical areas (Huang et al., 2013). The main pathway for hydrophobic-to-hydrophilic conversion of carbonaceous aerosols is still unclear and further observations (e.g., the hygroscopicity of BC coating materials) are needed to better understand the mechanism of the aging process.*"

9. Section 5.2: The description of Figure 8b is confusing

We have changed the description of Figure 8 to
"*Figure 1 Effect of BC aging on transpacific transport of BC. (a) Total emission contributions to BC over the North Pacific observational domain as a function of BC*

aging rate. (b) Sensitivity of total BC transported to the observational domain from all the sources to the rate of BC aging (i.e., the rate of change of emissions contributions in (a) with respect to the e-folding aging time τ)."

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