Response to Anonymous Referee #4

The authors present an interesting investigation into how BC aerosols are transported over long distances in the GEOS-CHEM model, to end up in the region covered by the HIPPO-3 campaign. Further, they test the effects of reduced BC lifetime on the modeled BC vertical profiles, and compare to observations.
The manuscript is concise and well presented, and should proceed to publication in ACP. However I have a few requests for clarifications of the methods, as it is not at present obvious how, in particular, the idealized transport model used to study BC lifetime was employed.

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 responses to each comment below.

Major comments:
I find the idealized transport model presented in sec. 2.4, and used for analysis of the impact of different e-folding times in sec. 5.2, to be too briefly described. As I understand it, the authors have performed full CTM simulations with different e-folding times, and extract the transported BC volume into the relevant geographical region for each. Then, the idealized transport model is used to extract information on the prevalence of precipitation events that remove BC en-route. The authors conclude that there is a prevalence of precipitation events shortly after emission, and hence that ageing rates near source regions is crucial for correct modelling. To make this statement, I believe the authors first need to validate the output of the idealized model against existing weather data, e.g. the precipitation in the meteorological fields used to drive GEOS-CHEM.

We think the reviewer understands the idealized transport model correctly. 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 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 idealized model simplifies the evolution of BC, during which BC is emitted from source regions, becomes hydrophilic through aging, removed by several key precipitation events, and the remaining BC is transported to the observational domain. In Eq. (3),

$$C = Ee^{-\frac{T_N}{\tau}} + \sum_{n=1}^{N-1} Ee^{-\frac{T_n}{\tau}}\left(1 - e^{-\frac{T_{n+1}-T_n}{\tau}}\right)(1-R_{n+1})\cdots(1-R_N)$$

$$+ E\left(1 - e^{-\frac{T_1}{\tau}}\right)(1-R_1)\cdots(1-R_N) \quad (3)$$

C is the amount of BC transported to the observational domain obtained from full CTM simulations. We assume the air mass sampled during HIPPO-3 may generally

experience *N* individual precipitation events. While in reality, precipitation along western Pacific may occur continuously. In addition, the time for trans-Pacific transport of BC from different source regions to mid Pacific is different. Therefore the goal of our idealized model is to give a sense of the average time between BC emissions and removal by the n^{th} precipitation during HIPPO-3. For tracers of 1- and 2-week lifetimes (e.g, BC), the average transpacific transport time from East Asia to the mid North Pacific in spring is 1.5–2 weeks (Liu and Mauzerall, 2005). Our idealized model indicates that most precipitation events occur in 0-3 days after BC aerosols are emitted. We have added more discussion in Section 2.4:

"*In this study we assume that the BC sampled by HIPPO-3 may experience a number of (N) precipitation events after it is released from EA and SE, while in reality it is more likely that there is continuous precipitation during mid-latitude transpacific transport. We also put all BC emissions from different source regions at different times into one single variable (E), while the time for transpacific transport of BC emitted at different times and locations is different. As a result, all the variables derived from the equation are averages of our simulation periods, and the goal of our idealized model is to analyze the general precipitation pattern which determines transpacific transport of BC during HIPPO-3.*"

Since our idealized model is used to estimate the average time between BC emissions and precipitation events during HIPPO-3, it is difficult to directly validate the results against existing weather data. GEOS-Chem CTM is driven by assimilated meteorological data from the Goddard Earth Observing System (GEOS-5) of the NASA Global Modeling and Assimilation Office (GMAO). GEOS-5 data have $0.5° \times 0.667°$ horizontal resolution, which is degraded to $2° \times 2.5°$ for input to GEOS-Chem. In our study we conduct two separate one-month simulations for 1 March–1 April and 15 March–15 April 2010, and here we show the total precipitation at the ground during 1 March–15 April 2010 in Figure R1. We can see that there are substantial precipitations near the East Asia source region as well as the western Pacific. This may indicate that results from our idealized model are reasonable when compared to the meteorological data.

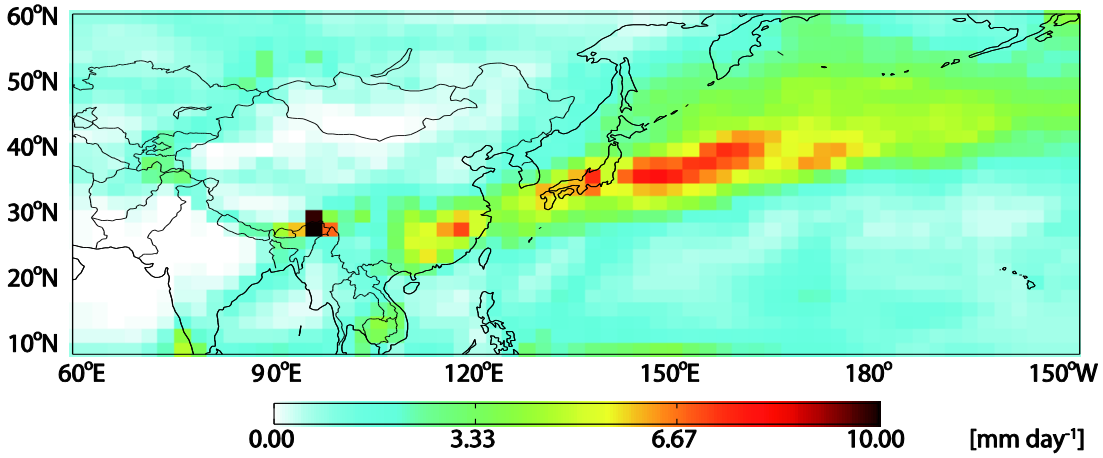

Figure R1. Total precipitation at the ground during 1 March–15 April 2010 used for input to GEOS-Chem. Data are from GEOS-5 of the NASA GMAO.

The model, they state, is equivalent to assuming that all BC is hydrophobic at emission time, and that all removal happens through precipitation. Can these assumptions be put in to the full GEOS-CHEM for a set of test runs as validation of the idealized model? Adding such documentation and tests, and an expansion on the presentation of the CTM runs done with altered e-folding time, would generally lift the discussion of a topic that is presently seeing a lot of interest.

GEOS-Chem assumes a fixed ratio (80%) of BC emitted is hydrophobic. In the revised manuscript, we follow the reviewer's suggestion and add the fraction of hydrophobic BC (α) in our idealized model. The new Eq. (3) is

$$C = \alpha E e^{-\frac{T_N}{\tau}} + \sum_{n=1}^{N-1} \alpha E e^{-\frac{T_n}{\tau}} \left(1 - e^{-\frac{T_{n+1}-T_n}{\tau}}\right)(1 - R_{n+1}) \cdots (1 - R_N)$$

$$+ \left[(1-\alpha)E + \alpha E \left(1 - e^{-\frac{T_1}{\tau}}\right)\right](1 - R_1) \cdots (1 - R_N) \quad (3)$$

where C is the amount of BC transported to a certain domain, E is BC emission, N is the number of precipitation events, T_n is the time interval between BC emissions and the nth precipitation, τ is the e-folding time of BC aging, and R_n is the fraction of BC removed by the nth precipitation. The new equation is very similar to the original one, except for an additional constant before BC emission. Therefore our results do not change much when considering emissions of both hydrophobic and hydrophilic BC. We use the new Eq. (3) to fit the amount of BC transported to the North Pacific as a function of the BC aging timescale, which yields the same results as Figure 9 shows, and derive the precipitation time (T_n) and the fractional BC removal (R_n) based on the parameters:

$$t_n = \frac{1}{T_n}, n = 1, 2, \cdots, N$$

$$A_n = \alpha E R_n (1 - R_N) \cdots (1 - R_{n+1}), \qquad n = 1, 2, \cdots, N-1$$
$$A_N = \alpha E R_N$$
$$y_0 = E(1 - R_N) \cdots (1 - R_1) \tag{4}$$

Table R1 shows the estimated timing of precipitation events and the corresponding ($T_{1\text{-}N}$) BC removal efficiency ($R_{1\text{-}N}$). The precipitation time is the same as that resulted from the original equations, and there is only little difference in the removal efficiency. We have modified Eqs. (3) and (4) and the descriptions of the equations in the revised manuscript.

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

Parameter	T_1 (d)	T_2 (d)	T_3 (d)	T_4 (d)	T_5 (d)
March	0.09	0.52	2.0	5.5	17.5
April	0.13	0.79	2.5	7.0	19.3
Parameter	R_1	R_2	R_3	R_4	R_5
March	9.1%	25.7%	49.2%	38.2%	18.0%
April	4.5%	20.6%	46.6%	34.8%	19.0%

It is true that we ignore dry deposition in our idealized model. However, we think this should not have a large impact on our results. In GEOS-Chem simulations, dry deposition for BC is not very important compared to wet deposition. We conduct a sensitivity test by turning off the dry deposition of BC in GEOS-Chem to estimate its effect on BC simulation. Figure R2 shows model simulated BC concentrations with and without dry deposition, and the difference is less than 20%. Thus, our fitting result (Figure 9) will not change substantially and dry deposition will only have a relatively small impact on BC removal efficiency by precipitation ($R_{1\text{-}N}$). We believe our idealized model can give some implications for the general precipitation pattern for BC wet removal along trans-Pacific transport during HIPPO-3. It indicates that there is a prevalence of precipitation events shortly after BC emission. Therefore, a slower or faster aging rate near source regions will significantly affect the amount of BC exported to the remote Pacific.

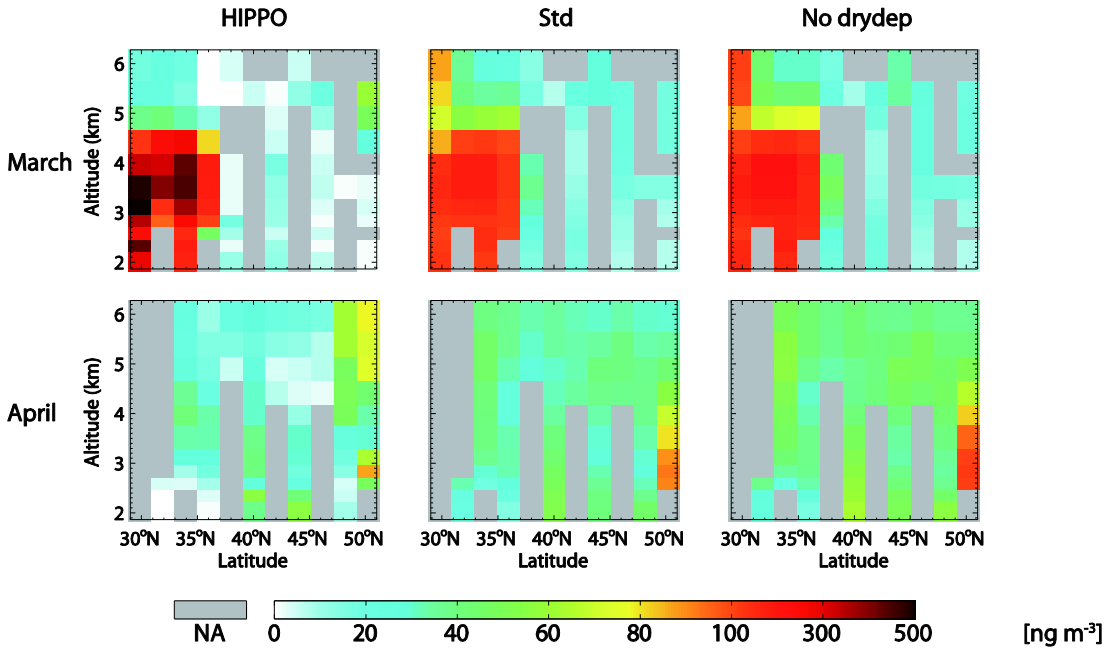

Figure R2. BC vertical profiles in observational domain (150–160 °W) on 29 March 2010 (top) and 13 April 2010 (bottom). HIPPO aircraft observations (left) and GEOS-Chem model estimates (sampled along aircraft track) with (center) and without (right) BC dry deposition are shown.

Separately, I find the source attribution plots in Figures 3 and 4 very relevant for ongoing discussions on the climate impact of BC. I encourage the authors to perform similar studies for the full range of HIPPO results, if possible. (I do realize it's beyond the scope of the present paper, but perhaps as a follow-up.)

Thanks for the suggestion. We will apply the methods to the full HIPPO data in our future work to see if the results have some general patterns and seasonal variations, and we would like to present our results in the follow-up paper.

Minor comments:
P508,l25: I suggest adding the recent paper by Schwarz et al. 2013, GRL, to the list here, as it presents the data later used in the present paper.

We have added Schwarz et al, 2013 to the references.

P521,l27: "Our idealized BC transport model has satisfied fitting precision in both missions." This statement needs to be better explained, and quantified.

We have rewritten the sentence as "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 give some implications for general precipitation patterns during HIPPO-3."

I also support the comments from the other reviewer regarding the need for a better description of biomass burning emissions used, and that showing the relevant regions and HIPPO flight tracks would improve the clarity of the manuscript.

We have clarified that the biomass burning emissions used in our study are for year 2010 and described the difference between March and April emissions (see Figure R3) in the revised manuscript ("*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.*"). We have also added HIPPO flight tracks in Figure 1.

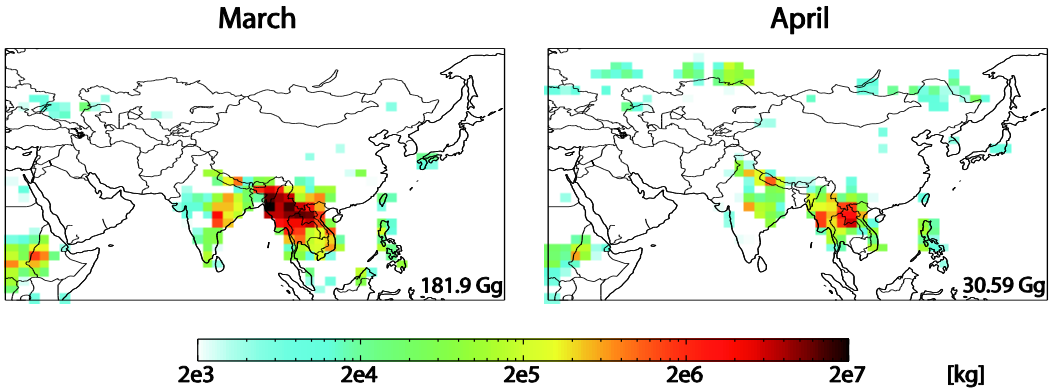

Figure R3. BC emissions from biomass burning during March and April, 2010 based on GFED3 inventory. The numbers listed on each plot indicates total monthly biomass burning BC emissions in Asia.

Reference

Liu, J. F., and Mauzerall, D. L.: Estimating the average time for inter-continental transport of air pollutants, Geophys Res Lett, 32, Artn L11814, Doi 10.1029/2005gl022619, 2005.