Response to Anonymous Referee # 3

(Note: Reviewer comments are listed in grey, and responses to reviewer comments are in black. Pasted text from the new version of the paper is in italics.)

The study Zhang et al., "Long range transport of black carbon to the Pacific Ocean and its dependence on aging timescale", used a chemical transport model (MOZART-4) to test the sensitivity of BC simulations to prescribed aging timescales. The authors tagged the emission sources to study source-receptor relationships. They also used observations to calibrate the aging timescale parameters, and discussed the difference between the default and the improved model. Overall I find this study interesting and scientifically important. The manuscript is well-prepared that it is straightforward to follow and concise. Therefore, I recommend the paper to be published on ACP. I only have a few minor comments for the authors to consider if they think they can further improve the paper:

We thank the helpful comments and suggestions indicated by the reviewer. We believe addressing his suggestions greatly improve the quality of our paper. Please see our response to each comment below.

1. Page 16950, line 22: The citation Emmons et al., 2010 should be placed right after MOZART-4, instead of after NCAR.

Thanks for correcting this. We have revised the sentence as the follow:

"In this research, we use the Model for Ozone and Related Chemical Tracers, version 4 (MOZART-4) (Emmons et al., 2010), a global chemical transport model developed at the National Center for Atmospheric Research (NCAR). "

2. Page 16950, line 24: Please provide the citation for MATCH.

Thanks for pointing this out. We added the following citation in the revised manuscript (see Section 2.1).

"Built on the framework of the Model of Atmospheric Chemistry and Transport (MATCH) (Rasch et al., 1997) with a series of updates…"

Full citation:

Rasch, P. J., Mahowald, N. M., and Eaton, B. E.: Representations of transport, convection, and the hydrologic cycle in chemical transport models: Implications for the modeling of short-lived and soluble species, Journal of Geophysical Research-Atmospheres, 102, 28127-28138, Doi 10.1029/97jd02087, 1997.

3. Page 16951, line 13: This model is run at rather coarse resolution. I wonder if the conclusion will change with higher model resolution. It might be interesting to cite a few papers and a few sentences to acknowledge changing model resolutions might affect aerosol-cloud interactions, and change the sensitivity test of aging timescale here.

Thanks for bringing up this important issue. We agree with the reviewer that changing resolution in climate models might change the optimized aging timescales. We have added

discussions and cited more papers on resolution and aerosol-cloud interactions in the Caveat section:

"For example, as model resolution increases, aerosol-cloud interactions in climate models can be better resolved, which can improve the simulation of BC transport (Ma et al., 2013;Ma et al., 2014). Therefore, the optimized aging timescales might change if models with different cloud schemes or spatial resolutions are used."

Citations:
Ma, P. L., Rasch, P. J., Fast, J. D., Easter, R. C., Gustafson, W. I., Liu, X., Ghan, S. J., and Singh, B.: Assessing the CAM5 physics suite in the WRF-Chem model: implementation, resolution sensitivity, and a first evaluation for a regional case study, Geosci. Model Dev., 7, 755-778, 10.5194/gmd-7-755-2014, 2014.
Ma, P. L., Rasch, P. J., Wang, H. L., Zhang, K., Easter, R. C., Tilmes, S., Fast, J. D., Liu, X. H., Yoon, J. H., and Lamarque, J. F.: The role of circulation features on black carbon transport into the Arctic in the Community Atmosphere Model version 5 (CAM5), Journal of Geophysical Research-Atmospheres, 118, 4657-4669, 10.1002/jgrd.50411, 2013.

4. Page 16951, line 16: I am confused. Is MACCity emissions used for IPCC-AR5 simulations? What about the Lamarque et al. (2010) emissions?

Thanks for catching this mistake. According to the website http://accent.aero.jussieu.fr/MACC_metadata.php, the suggested documentation should be Lamarque et al., ACP, 2010. Also, "In support of the fifth IPCC-AR5 (Intergovernmental Panel for Climate Change Assessment Report 5), the ACCMIP (Atmospheric Chemistry and Climate - Model Intercomparison Project) historical emissions dataset has been developed (Lamarque et al, 2010), on a decadal basis (from 1850 to 2000 for the historical dataset), as well as emissions scenarios called RCPs (Representation Concentration Pathways, Van Vuuren et al., 2010). As part of two projects funded by the European Commission, MACC and CityZen, the ACCMIP and the RCPs emissions dataset have been adapted and extended on a yearly basis for the period 1990-2010. For anthropogenic emissions, emission data were interpolated on a yearly basis between the base years 1990, 2000, 2005 and 2010. For the years 2005 and 2010, the RCP 8.5 emissions scenario was chosen (4 emissions scenarios were developed in support of the IPCC-AR5: RCP 2.6, RCP 4.5, RCP 6, RCP 8.5).For biomass burning emissions, the ACCMIP dataset was extended as well on a monthly basis. This 'extension' of the ACCMIP and RCPs emission dataset for the MACC and CityZEN projects is referred to as MACCity (MACC/CityZen) emission dataset." We modified the sentence to make it more precise:
"Anthropogenic emissions are based on the MACCity emission inventory (http://www.pole-ether.fr/eccad), which is extended from the database used for IPCC Coupled Model Intercomparison Project (Lamarque et al., 2010)."

5. Page 16957, line 2: I think it might read better if you move the discussion from line 8 (mixing in Asia and North America is quick) to line 2, following "(less than half a day)".

We took the reviewer's suggestion and revised this paragraph as below:

"Optimized aging timescales for each source region and season are shown in Table 1. Ranges of plausible values for each optimized aging timescale based on perturbation simulations are also summarized in Table S1 in the supplementary materials. As shown in Table 1, values differ significantly by source region and season. The aging timescale of BC from East Asia, North America, India, and Southeast Asia is in most cases relatively short (i.e., less than half a day). The optimized BC timescales reported here for East Asia and North America are consistent with observations in these regions, which show that BC is quickly mixed with hydrophilic species. For instance, observations over an urban region of Japan find that the timescale for BC to become internally mixed is 12 hours, with coatings made of primarily sulfate and soluble organic carbon (Moteki et al., 2007). In Beijing and Mexico City, urban BC is observed to become internally mixed with sulfate in a few hours (Johnson et al., 2005;Cheng et al., 2012). Over Southeast Asia, BC emissions are mainly anthropogenic in origin (with a fast aging rate), except during spring when large-scale biomass burning activities generate tremendous amounts of BC. The optimized springtime BC aging timescale for Southeast Asia is around 2 days, consistent with the findings of Shen et al. (2014). On the contrary, the optimized aging rate is relatively slow in the high-latitude regions (Canada, the former Soviet Union and in particular Europe) in all seasons except summer (June-July-August, JJA), which can be explained by slower photochemistry in high latitudes under low sunlight in non-summer months. Since measurements on BC aging timescale are scarce and limited to few places, more observations are needed to measure the hygroscopicity of BC-containing particles in different continents covering both source and downwind areas.

The seasonality of aging timescale reported here is largely consistent with Liu et al. (2011), who develop a parameterization for BC aging rate as a function of OH radical concentration. In this study, we further improve the parameterization of Liu et al. (2011) by finding best-fit values for constants that best match HIPPO observations with reference to our BC aging timescales. After conducting additional sensitivity simulations, we find that a set of parameters (i.e., β=2.4×10^{-11}, and γ=1×10^{-6} in Equation (4) in Liu et al. (2011)) when employed in MOZART-4 can fit well the HIPPO observations as well as ground observations (see Figure S1 and S2 in supplementary material)."

6. Regarding Figure 4, I notice that even with the improved model in many cases the model still under-estimate the BC concentration by an order of magnitude, but there is no discussion on this feature. Please elaborate.

In the revised manuscript, we further expanded the discussion on the differences between the modelled and observed BC concentrations. Please see the revised text in Section 3 or below:

"In a few cases, relatively large differences between the improved model and observations remain. These differences could be attributed to any number of factors (e.g., emissions, transport, cloud/precipitation, aging process, wet removal efficiency, etc.). For example, models could misrepresent BC wet deposition, originating from biases in precipitation. As shown in Figures S4 in the supplementary materials, though MOZART-4 generally captures well the spatial extent of precipitation during all HIPPO campaigns, biases occasionally

appear when comparing to the NCEP reanalysis over the western Pacific. As another example, the model uses a monthly biomass burning emission inventory. This means that modeled emissions lack daily variation in biomass burning activities that could be important where biomass burning emissions dominate BC loading. Underestimates in BC mixing ratio may be partially due to abrupt emissions events that are not captured by the model. Lastly, since this study assumes that BC aging timescale in all the southern hemispheric continents is the same, we do not account for variability in BC aging rates from these regions that may exist in reality."

7. Page 16959, line 12: Since the term "Pacific Ocean" has been used many times previously in the text, the acronym "PO" is out of place and not needed.

We follow the reviewer's suggestions and removed this acronym in the revised manuscript.

8. Page 16961, line 15: Perhaps replace "dT/dtau" with "S=dT/dtau", and discuss S later in this paragraph. 9. Page 16962, line 1: Please elaborate why the theoretical value of the slope is 0.8.

Good suggestion. We have substituted dT/dtau by s in the revised manuscript.

9. Page 16962, line 1: Please elaborate why the theoretical value of the slope is 0.8.

According to equation 7, theoretically, s = $\frac{(1-\alpha)K_{\mathrm{W}}}{(K_{\mathrm{D}}+K_{\mathrm{W}})(1+K_{\mathrm{D}}\tau)^2}$. If dry deposition coefficient (K_{D}) is negligible, then s = 1 - α. α = 0.2 in the model, so s = 0.8. We revised the text a bit to make it more understandable:

"In MOZART-4, α is assumed to be 0.2 for all emission sources. So if K_D is negligible, the theoretical slope of the T- τ curve is 1-0.2=0.8, which is very close to the curve for untagged BC (black line) in Figure 6. "

References:

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