Response to Referee Dr. Huisheng Bian

(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 authors investigated source attribution to the Pacific Ocean using a global chemical transport model MOZART-4 by tagging BC tracer to 13 source regions around the globe. They further quantified the aging timescales of those tagged BC tracers by constraining simulation with aircraft measurements from five HIPPO missions. This is a scientifically interesting study. Publications of AeroCom and other works have demonstrated that many global models currently overestimate BC in free and upper troposphere. This study points out a direction to solve this common problem in modeling global BC field. The paper is well written. I recommend publishing the paper on ACP after the authors make some minor modifications.

We really appreciate the thoughtful and valuable comments from Dr. Bian. It substantially helps to improve our manuscript by addressing these issues.

General Remarks: The authors summarized BC aging timescales associated to 13 source regions. The conclusion is instructive but may be not robust. The authors showed an improved BC simulation by MOZART-4 at current condition with this varying BC aging timescale. But can these aging timescales still be valuable if BC emission and atmospheric oxidant fields are changed in the future in MOZART-4? Can other models apply these adjusted BC aging timescales with some cautions? It may be more useful that the authors explore the key factors that control BC aging, such as emission types, oxidant fields, etc, and parameterize BC aging timescale based on these key factors.

This is an important remark. We agree with the reviewer that investigating the key factors that control aging processes, and developing aging schemes according to these factors will be more useful for future modeling studies. Following the reviewer's comments, we further analyze the spatiotemporal pattern of the optimized aging timescales and re-evaluate the parameterization of BC aging scheme developed by Liu et al (2011) , in which aging rate $k_a = \beta \cdot [OH] + \gamma$, is controlled by OH concentration (molecules m^{-3}) and two parameters β and γ . After conducting a number of sensitivity tests based on our optimized aging timescales, we find a larger value of parameters β (2.4×10^{-11}) and γ (1×10^{-6}) can fit well the HIPPO observations. The description of this scheme is added in Section 3:

"…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)."

We also evaluated the model with OH-dependent scheme. Please see Figure S1 and S2 in supplementary material or below:

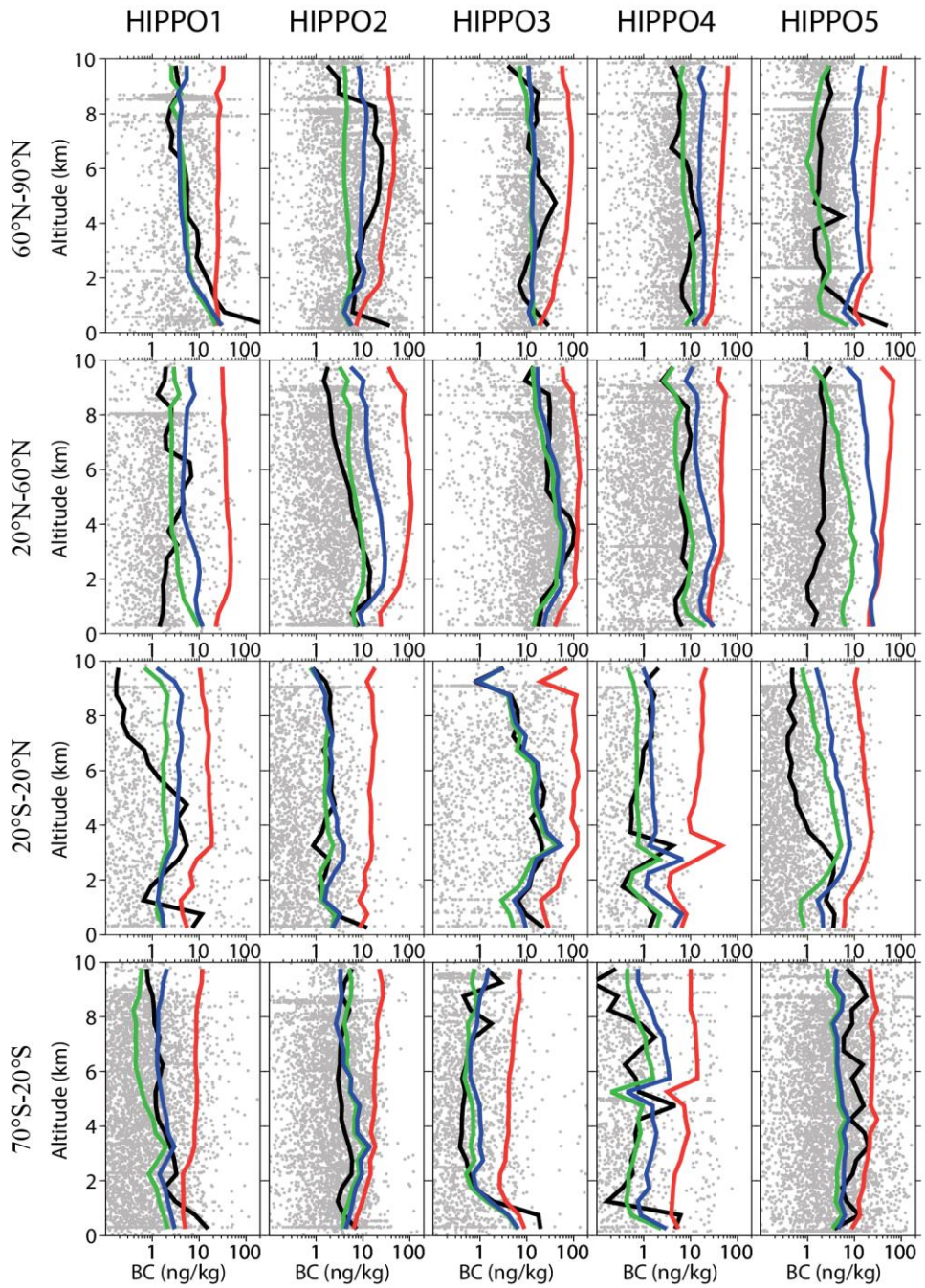


Figure S1. Vertical profiles of simulated and observed BC mass mixing ratios over 0.5 km altitude bins along the flight tracks of HIPPO 1-5 over the central Pacific Ocean (130 °W-160 °E). Data are shown separately as averaged over 70 °S-20 °S, 20 °S-20 °N,20 °N-60 °N, and 60 °N-90 °N. The black, red, green, and blue lines are mean values of BC mixing ratios from observations, default model, improved model with optimized region-specific aging timescale, and improved model using OH-dependent aging scheme respectively. The gray dots represent measured BC concentrations.

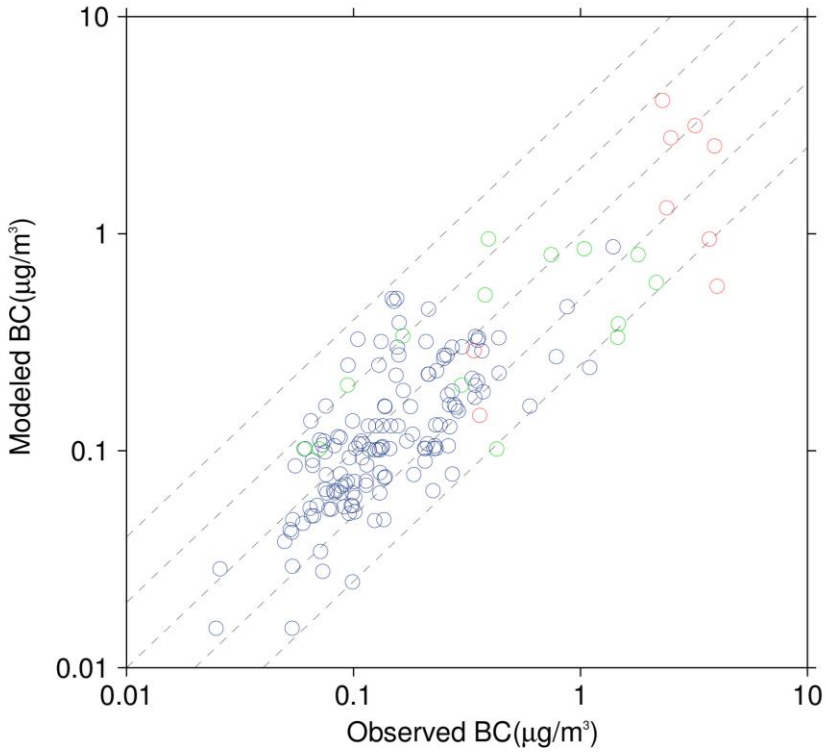


Figure S2. Modeled (employing OH-dependent aging scheme) versus observed surface annual mean concentration of BC at sites in IMPROVE (blue), EMEP (green), and China (red). Dash lines are 1 : 4, 1 : 2, 1 : 1, 2 : 1, and 4 : 1 ratio lines. BC observations in China are attained from Zhang et al. (2008).

Specific comments:

1. Page 16946 line 22-24: How fast is the aging rate so that the lifetime of BC is dominated by factors that control its local deposition?

Good question. In the model, hydrophobic BC cannot be removed by precipitation, while aging enables hydrophobic BC to convert to hydrophilic BC, and be removed by precipitation. Therefore, when aging is slow, most BC stays hydrophobic near its source, and is not affected significantly by local precipitation. On the contrary, when aging is fast, BC is more susceptible to local precipitation, and might be removed shortly after emitted. To make this sentence clearer, we revise this sentence in abstract:

"The lifetime of BC is more determined by factors that control local deposition rates (e.g. precipitation) when aging is fast versus slow."

2. Page 16946 line 27 – page 16947 line 1: This sentence repeats the first sentence of abstract.

We revised this sentence in the abstract:

"Our findings suggest that the aging timescale of BC varies significantly by region and season, and can strongly influence the contribution of source regions to BC burdens around the globe. Therefore, improving parameterizations of the aging process for BC is important for enhancing the predictive skill of global models. "

3. Page 16949 line 4: How thick must coating be for a hydrophobic BC to be named as a hydrophilic BC?

Kuwata et al (2009) estimate the critical condensed mass for hydrophobic BC converting to hydrophilic is 0.18 fg (supersaturation = 0.9%) and 0.07 fg (supersaturation = 1.3%) for 100-nm particles. Their results are based on their measurements in Tokyo, where they found the compounds of coating are mainly organic compounds. Besides thickness of coating, solubility of of the coating material is another important factor. Thus, the thickness of coatings to convert hydrophobic BC into hydrophilic BC may depend on the components of coatings as well as supersaturation.

4. Page 16954 line 20: It may be good to clarify the terms of "aging", "aging rate", and "aging timescale" used in the paper.

Good suggestion. We clarified the definitions of aging timescales and rate in the revised Introduction. Please see our revisions in the fourth paragraph of Section 1 or below:
"…Exponential timescale for this aging process to occur, the so-called "aging timescale", which is the reciprocal of aging rate, therefore highly influences the timing of cloud formation and wet deposition, and thus is of great research interest (Liu et al., 2011)…"

We also clarified the definitions of aging in Section 2.4:
"…Note that while we define aging timescale as that for converting BC from hydrophobic to hydrophilic, some other studies use this term to describe the change from thinly to thickly coated BC (Moteki et al., 2007;Saikawa et al., 2009)."

5. Page 16954 line 25-28: Why does biomass burning BC have a larger fraction of coated particles and thicker coatings than urban BC? This seems to conflict with the discussion in section 2.1 (page 16949 line 14-24) that indicates that urban pollutions (i.e. sulfate, nitrate, ozone, and nitrogen oxide) are primary components in coating. Also the short aging timescale of East Asia BC summarized by the authors seems to not support this statement either.

It is shown by observations that biomass burning BC has a larger fraction of coated particles and thicker coatings than urban BC as emitted (Schwarz et al., 2008). However, both thickness and solubility of coatings determine whether BC-containing particles can become cloud condensation nuclei (CCN). Thus, BC needs to be coated by sufficient soluble compounds (e.g. sulfate, nitrate, or organics), so as to be converted to CCN. Despite that previous studies find that biomass burning BC has a larger fraction of coated particles and thicker coatings, the solubility of the coatings on biomass burning emitted BC could be low. On the other hand, although coatings of

urban BC are thinner, they could be sulfate or nitrate, which are extremely soluble (Johnson et al., 2005;Cheng et al., 2012). Future observations on the evolution of hygroscopicity of BC-containing particles from different source regions are needed. In the revised manuscript, we modified the paragraph in Section 3 to make it clearer:

"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."

6. Page 16955 equation 3: How about longitude bins?

We mainly focus on the central Pacific (160 °E -130 °W), where BC is mostly contributed by long-range transport. So we average our results over this longitude band. The longitude range is described in section 2.3:

"…Note that we use only the HIPPO observations taken in the Central Pacific Ocean (160 °E - 130 °W) and ignore observations near source regions… "

7. Page 16956 line 27 and Page 16957 line 1-2: Why is the aging of these urban polluted regions vary fast? Is it faster than that of biomass burning dominated regions such as South America, Africa? I am confused since it is not consistent with the observation facts discussed in section 2.5 (see specific comments 5 above).

Thanks for bringing this question up. As we described in comment 5, though biomass burning BC may be more thickly coated comparing to what has been seen in urban regions, the solubility of coating materials between these two types of BC could be different. In the revised manuscript, we rewrote this paragraph in a clearer way. Please see the text below or our detailed response to comment 5.

"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."

8. Page 16958 line 12-14: How sensitive is the summarized BC aging timescale to a change in precipitation? In other words, what is the potential uncertainty of aging timescale in response to a potential bias in precipitation predicted by MOZART-4?

Good question. MOZART-4 is driven by reanalysis meteorology, and precipitation was found to have a better agreement with observations when water vapor concentrations was calculated online (the preferred mode of operation in MOZART-4, see Emmons et al., 2010). As shown in Figure S4 (see below), the pattern of precipitation in MOZART-4 is in general consistent with NCEP reanalysis, with moderate difference in magnitude.

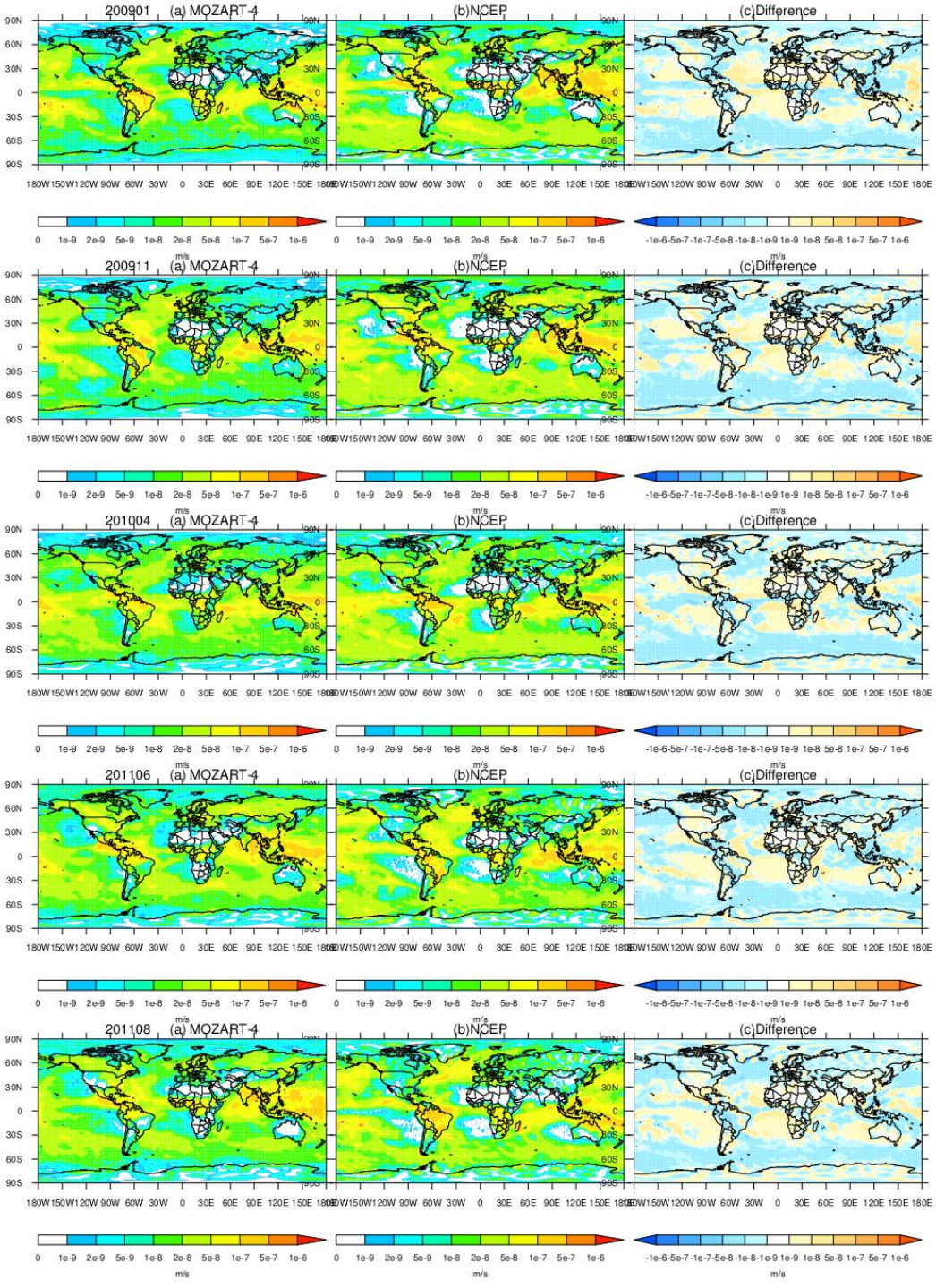


Figure S4. Average precipitation of MOZART-4 model, NCEP reanalysis meteorology, and their difference (MOZART-4 minus NCEP) during HIPPO I (January, 2009), HIPPO II (November, 2009), HIPPO III (April, 2010), HIPPO IV (June, 2011), and HIPPO V (August, 2011).

On the other hand, we agree with the reviewer that bias in precipitation might also influence our results of optimization. According to our wet deposition scheme (see Equation 2), wet removal of

BC increases as precipitation increases if cloud water content keeps the same. So the change in precipitation will influence the loadings of BC over the Pacific Ocean and other regions as well. We added discussion on the uncertainties related to precipitation in Caveat Section:

"…Firstly, we assume that model parameterizations of wet and dry deposition, precipitation, transport, and emissions are realistic, even though these processes also affect BC distributions and have uncertainties (Vignati et al., 2010;Fan et al., 2012). Consequently, the optimized aging timescales may partially counter biases in these processes (i.e. other than aging), and may vary according to the model used. 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…"

We also revised the explanations for the remaining bias in section 3:

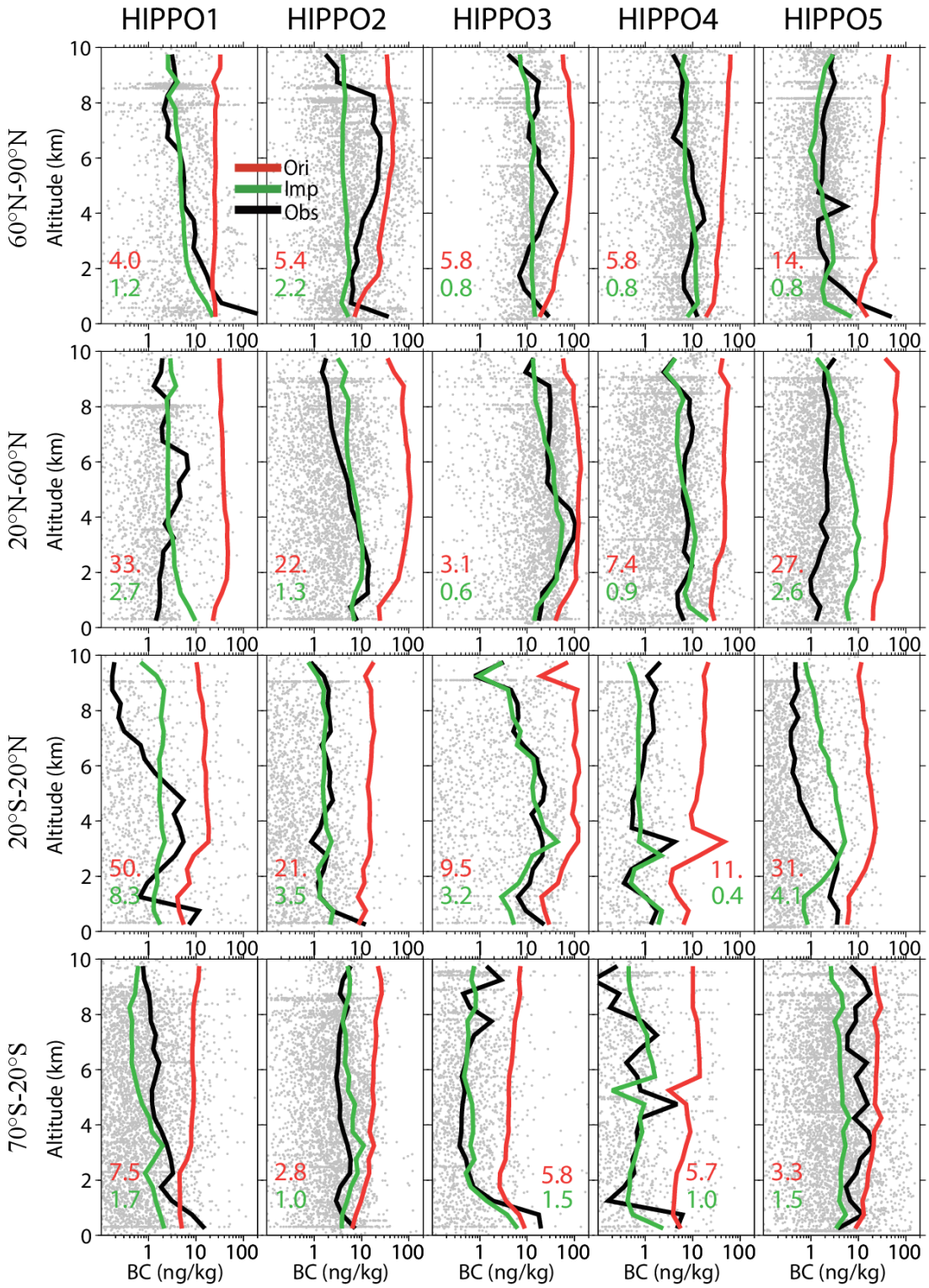
"In a few cases, relatively large differences between the improved model and observations remain. These differences could be attributed to any number of factors (e.g., emissions, transport, cloud/precipitation, aging process, wet removal efficiency, etc.). For example, models could misrepresent BC wet 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…"

9. Figure 5: It is interesting to know that Africa emission is not a dominant contributor to most of the Pacific Ocean since it is the largest contributor to global BC (Page 16960 line 10-11) and Africa is closest to the Pacific Ocean.

Although Africa is the largest contributor to global BC, due to wind directions and the long distance of Africa to the Pacific Ocean, Africa is not the largest contributor to the Pacific Ocean. On the other hand, as shown in Figure 5, Africa contributes the most to some areas in the Atlantic Ocean and the Indian Ocean. In the future, measurements over the Atlantic Ocean and Indian Ocean can be very helpful to clarify the contribution of African BC to the Pacific.

Technique corrections: 1. Figure 4: Please change color of improved model results (green line and number) so that it is more distinct from the grey dots that represent measured BC concentrations.

Thanks for this correction. We have changed the color, format, and position of dots and numbers to make them more readable in Figure 4, as shown below:


HIPPO1
HIPPO2
HIPPO3
HIPPO4
HIPPO5
60°N-90°N
20°N-60°N
20°S-20°N
70°S-20°S
Altitude (km)
BC (ng/kg)
Ori
Imp
Obs
4.0
1.2
5.4
2.2
5.8
0.8
5.8
0.8
14.
0.8
33.
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22.
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3.1
0.6
7.4
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27.
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50.
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9.5
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11.
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4.1
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3.3
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