Point-to-Point Response for "Sensitivity of atmospheric aerosols to precipitation characteristics"

We thank both referees for their very helpful comments. We have carried out further analyses and also revised the manuscript following the referees' comments. We have made itemized responses to all the comments as described below. The referees' comments are repeated below in the blue and italicized text and our responses are in normal font.

Response to Referee #2

This is a potentially interesting paper on changes in wet deposition due to precipitation intensity and amount changes. However, there are several issues that need to be addressed before the paper can be accepted: 1. The methods is not complete, and more details on the experiments and a better descirption of the cases (in a table) need to completed. More details below.

Thank you for pointing this out. We have added a new table (Table 1) in the MS to summarize and better describe the various cases done in this study. We have also provided more clarification throughout the MS as detailed below in response to specific comments and questions.

Table 1. Series of sensitivity model simulations carried out in this study.

Model simulations	Objective	Case names
Constant precipitation frequency (Fig. 1a)	To study the sensitivity of BC lifetime to precipitation intensity	f1i0.25, f1i0.5, f1i1, f1i2, and f1i4
Constant precipitation intensity (Fig. 1b)	To study the sensitivity of BC lifetime to precipitation frequency	f0.1i1, f0.25i1, f0.5i1, f0.75i1, and f1i1
Constant precipitation amount (Fig. 1c)	To compare the sensitivity of BC lifetime to precipitation intensity and precipitation frequency	f0.1i10, f0.25i4, f0.5i2, f0.75i1.33, and f1i1
Hygroscopicity of aerosols (100% vs 20% BC in fresh emissions are assumed to be hydrophilic)	To examine the impacts on wet deposition from the parameterization on the hygroscopicity of aerosols	f1i1 and f0.75i1.33

Aerosol size (BC aerosols are assumed to be in coarse mode vs accumulation mode)	To examine the impacts on wet scavenging from the parameterization on the size of aerosols	f1i1 and f0.75i1.33
Contour of BC lifetime (Fig. 2, 4-6)	To plot BC lifetime as a function of the precipitation intensity and frequency	f0.25i0.5, f0.25i1, f0.25i1.33, f0.25i2, f0.25i4, f0.5i0.5, f0.5i1, f0.5i1.33, f0.5i2, f0.5i4, f0.75i0.5, f0.75i1, f0.75i1.33, f0.75i2, f0.75i4, f1i0.5, f1i1, f1i1.33, f1i2, and f1i4

2. The trend in the precipitation using either the TRIMM or the reanalyses is unlikely to be robust or good enough for this analysis. The Renalysis are well know to have trouble with the moisture budget, while the TRIMM time series is too short.

Indeed, although the TRMM and the reanalysis datasets used in this study represent some of the best meteorological datasets available, each of them has their own shortcomings - the observational datasets are more reliable, but only cover shorter time periods; while the reanalysis datasets cover longer periods, but are less reliable. That's why we decided to combine multiple datasets in this study, including TRMM, NCEP, NCEP2, and MERRA, to give us a better idea about the potential trends of precipitation characteristics. We have provided discussions about the choices of datasets, like –

"TRMM (3B42v7) performances better than the previous version of satellite products (3B42v6), though there are still problems in detecting precipitation events with low precipitation rates [*Maggioni et al.*, 2016]. "

"We regrid the TRMM dataset from 0.25x0.25 to 2.5x2.5 (°lon x °lat) to reduce the computational cost and the relative errors at small precipitation rates [*Huffman et al.*, 2007; Gehne *et al.*, 2016]."

"Since the TRMM data only cover a relatively short period, we make similar analyses with three reanalysis datasets (NCEP, NCEP2, and MERRA) to cover a longer time period (2001-2010 vs. 1981-1990) (Fig. 5)."

"These variations across different data sources reflect the significant uncertainties associated with these datasets, as reported earlier [e.g., *Trenberth and Christian*, 1998; *Trenberth et al.*, 2011; Gehne *et al.*, 2016]."

"Our results are also affected by the limitations from the meteorology datasets. Although the TRMM and the reanalysis datasets used in this study represent some of the best meteorological datasets available, each of them has their own shortcomings - the

observational datasets are more reliable, but only cover a relatively short time period of 14 years; the reanalysis datasets cover longer periods, but are less reliable due to known issues such as the bias in moisture budget [e.g., *Trenberth and Christian*, 1998; *Trenberth et al.*, 2011; *Gehne et al.*, 2016]."

I wonder if the authors don't want to just do a correlation between the annual average precipitation in different regions and the wet deposition lifetime in that region, and see if there isn't a robust signal in that. Then you can still make some statements about how different regions are likely to move, based on climate projections or longer term precipitation trends. I would bet, if your results are robust, that you will get a good relationship just with annual averages (or seasonal), and then you can more safely extrapolate into the future.

We presume by "annual average precipitation" the review refers to the annual total precipitation amount (i.e. average intensity x frequency). Then indeed, we believe that we would see a good correlation between the annual average precipitation and BC lifetime. However, a major point of our study here is to show that it's not just the annual average precipitation but also the patterns (say frequent drizzles vs occasional heavy rain events) would matter for BC scavenging. That is, due to the different sensitivities associated with precipitation intensity and frequency, the same annual average precipitation may lead to very different scavenging efficiency and consequently BC lifetime.

More details:

"Our study, based on the GEOS-Chem model simulation, shows that the removal efficiency and hence the atmospheric lifetime of aerosols have significantly higher sensitivities to precipitation frequencies than to precipitation intensities, indicating that the same amount of precipitation may lead to different removal efficiencies of atmospheric aerosols." Please make it clear that this is dependent on the way that you have included wet deposition, but that we don't really know the right answer.

We have modified the first part to "Our sensitivity model simulations, through some simplified perturbations to precipitation in the GEOS-Chem model, show that …"

"We first analyze changes in the precipitations between two 7-yr periods (2008-2014 vs. 2001-2007)" This is a very short time scale to talk about: is this going to be interpretable? Please go into the details of statistical significance and interannual variability and what your goal is with such short time scale differences.

We have added clarification in the MS –

"The changes in the average precipitation intensities and frequencies between the periods of 2008-2014 and 2001-2007 for each region are shown as ratios in Fig. 4, with the width and height of the blocks indicting the standard errors of the calculated percentage changes in precipitation frequency and intensity, respectively. Although these TRMM data only cover 14 years, the standard errors as shown in Figure 4 indicate that the changes in precipitation intensity and frequency over most regions are statistically significant."

61:"model does not simulate meteorology;": it does simulate meteorology, but it does not do this prognostically, it just forces it from data.

We have modified the text from "model does not simulate meteorology" to "model does not simulate meteorology prognostically".

67: Change:"in details by" to in detail by

The text in MS has been modified from "in details by" to "in detail by".

68: "The efficiency of wet scavenging is very sensitive to the hydrophilicity of BC." Please make clear that this is a result from your study.

We have clarified this part to - "In GEOS-Chem simulation, the BC aerosols are classified into two types based on their hygroscopicity (hydrophobic vs hydrophilic) and wet scavenging is more efficient for hydrophilic BC."

96: f0.5i2: I appreciate that you tried to make your case names make sense, but they are still unintelligible, so you probably want a table describing all your cases, and try to NOT use your case name, but rather use English whenever possible.

We have added a new table (Table 1) in the MS to describe all the perturbation tests(also shown above). We feel the concise name such as f0.1i1 works well when citing the specific case; the long English case name such as "a case with 0.1 times of base precipitation frequency and base precipitation intensity" appears too wordy and can easily break the reading flow. We hope the descriptions in table would help making the short names clearer.

"Our result also agrees with the lifetime of 5.8 ± 1.8 days simulated by the GEOS Chem

We have compared to more models in AEROCOM. -- "For 13 models in AeroCom, the lifetimes of BC from anthropogenic fossil fuel and biofuel sources are simulated to be from 3.5 to 17.1 days, with 5.9 days as the median value [*Samset et al.*, 2014]."

We have added more description about the sensitivity test on hygroscopicity in the text –

"We first examine the possible impacts on our results from the parameterization on the hygroscopicity of aerosols. With the default parameterization in GEOS-Chem, 20% of the fresh BC emissions are assumed to be hydrophilic. We set up sensitivity runs with another parameterization, where all BC are assumed to be hydrophilic. With these two different parameterization schemes, we examine the changes in the BC lifetime between two scenarios (f1i1 vs. f0.75i1.33) respectively. "

We have also included brief description on this in Table 1, showing that one case with "20% BC in fresh emissions are assumed to be hydrophilic (default)" and the other case with "100% BC in fresh emissions are assumed to be hydrophilic".

We have described the wet deposition algorithm for both accumulate and coarse mode: "The washout rate constant (k) is affected by the particle size and the form of precipitation. For washout by rain with precipitation rate P (mm/h), $k = 1.1 \times 10^{-3}P^{0.61}$ for accumulation mode (aerosols with diameters between 0.04 µm and 2.5 µm) and $k = 0.92P^{0.79}$ for coarse mode (aerosols with diameter between 2.5 µm to 16 µm); for washout by snow with precipitation rate P, $k = 2.8 \times 10^{-2}P^{0.96}$ for accumulation

mode and $k = 1.57P^{0.96}$ for coarse mode [*Feng,* 2007, 2009]. The coefficients for accumulation-mode are used in calculating k for fine particles including BC in GEOS-Chem. "

"We find that during these 14 years, the average precipitation intensity has increased over most regions, but the average precipitation 180 frequency has decreased over more than one third of the total regions including western North America (nwNA and swNA), southern South America (sSA), western Europe (wEU), southern Africa (sAF), and southwestern Asia (swAS)." This is a very short time period to argue for increases or decreases: this could just be interannual variability. Do you really want to argue for increases or decrsease? If so, show a statistically significant difference, etc. I would argue a better way to do it, is just use annual averages, which will allow you to use more data (as described above).

We have updated this part to "The changes in the average precipitation intensities and frequencies between the periods of 2008-2014 and 2001-2007 for each region are shown as ratios in Fig. 4, with the width and height of the blocks indicting the standard errors of the calculated percentage changes in precipitation frequency and intensity, respectively. Although these TRMM data only cover 14 years, the standard errors as shown in Figure 4 indicate that the changes in precipitation intensity and frequency over most regions are statistically significant."

Our results indicate that though the results based on 14 years of data from TRMM (Fig. 4) show a much larger standard error than the results based on 30 years of data from other datasets (Fig. 5), there are still significant tendencies of increase and decrease in the results of 14 years for most regions.

"By combining these precipitation changes for various regions as shown in Fig. 4 with the relationship between precipitation characteristic and the BC lifetime as illustrated in Fig. 2, we can analyze the long-term changes in the atmospheric aerosol lifetimes driven by precipitation changes." Because I don't believe you have a long enough time series, I don't believe you can extend it, unfortunately. Please think about doing this analysis in a much more robust manner (with error bars showing the trends are important enough, believeable, etc) or just pull this section out of the paper.

We used the size of red block in Fig. 4 and Fig. 5 as the error bar to show the standard error. To clarify this point, we have added more discussion in MS – "The changes in the average precipitation intensities and frequencies between the periods of 2008-2014 and 2001-2007 for each region are shown as ratios in Fig. 4, with the width and height of the blocks indicting the standard errors of the calculated percentage changes in precipitation frequency and intensity, respectively. Although these TRMM data only cover 14 years,

the standard errors as shown in Figure 4 indicate that the changes in precipitation intensity and frequency over most regions are statistically significant."

"Since the TRMM data only cover a relatively short period, we make similar analyses with three reanalysis datasets (NCEP, NCEP2, and MERRA) to cover a longer time period (2001-2010 vs. 1981-1990) (Fig. 5). We find that, similar to the TRMM data, all the 195 three reanalysis datasets show increasing trends for precipitation intensity over most regions but more divergent trends for precipitation frequency in the past decades." Here you might have enough data to talk about this, but still very short time period. Again, show the standard deviations, show that they are signficiant, suppor tiwth other studies that try to show trends across such very short time periods in such a highly variable value (precipitation).

We used the size of red block in Fig. 4 and Fig. 5 as the error bar to show the standard error.

Also, there are significant problems with the moisture budgets in the reanalyses: are you sure you even want to do this? Might be better to just use climate model output because of the problems with inconsistencies in the data (please see all the papers by Kevin Trenberth showing the very large warts in the moisture budgets for all the reanalyses; not just one paper).

Good point. We have added more discussion in MS to acknowledge the limitation of the reanalysis datasets.–

"Our results are also affected by the limitations from the meteorology datasets. Although the TRMM and the reanalysis datasets used in this study represent some of the best meteorological datasets available, each of them has their own shortcomings - the observational datasets are more reliable, but only cover a relatively short time period of 14 years; the reanalysis datasets cover longer periods, but are less reliable due to known issues such as the bias in moisture budget [e.g., *Trenberth and Christian*, 1998; *Trenberth et al.*, 2011; *Gehne et al.*, 2016]."

By combining multiple datasets including TRMM and the reanalyses data, we are hoping that the analyses can still offer us some insights on some likely trends in precipitation.

Please notice that the argument that the frequency of precipitation is more useful than intensity for understanding wet deposition lifetime changes in context of changes in aerosol lifetime for the LGM/current was used in the Mahowald et al., 2011 paper in Quaternary Science Reviews, which might help your arguments here.

We have added the citation in MS. "*Mahowald et al.* [2011] also discussed the importance of precipitation frequency in wet deposition based on simulations showing large removal rate of dust in precipitation events."

Mahowald, N., et al.: Model insight into glacial–interglacial paleodust records. Quat. Sci. Rev. 30.7-8, 832-854, 2011.

Response to Referee #3

Hou et al. systematically investigate the effect of precipitation frequency and intensity on aerosol scavenging using a coarse resolution global model with a rather simplistic description of aerosol scavenging. The topic is especially interesting since the changes in precipitation characteristics (e.g. more extreme precipitation and possibly less drizzle) constitute an important contributor to the climate change signal. While the finding that the change of the black carbon lifetime in a changing climate might be dominated by changes in precipitation frequency and not in precipitation amount does not seem overly surprising in the light of the cited literature, this study nevertheless seems very interesting and useful to me, especially since to my knowledge this study represents the first attempt to investigate the topic in such a focused and systematic fashion. The study nicely explains why an increase in total precipitation amount does not necessarily lead to a decrease in aerosol lifetime (independent of changes in spatial patterns that may for example impact some regions with high emissions more than others). In my opinion the manuscript serves to highlight a rather interesting and important topic and in spite of some limitations it can serve as a very good base for further studies. I recommend to publish the manuscript subject to minor revisions.

Specific comments/suggestions

1. l. 86ff: I think that (a) scaling the precipitation by a uniform factor for each grid box and (b) using a stochastic function where precipitation is turned off regardless of whether it is heavy or light precipitation may in principle lead to different outcomes compared to what might be expected from climate change (in which e.g. strong precipitation intensity may be enhanced while weak precipitation may decrease or remain unchanged and models also suggest very distinct spatial patterns) and I am not sure that the results from these very idealized sensitivity tests can be used to deduce a quantitatively correct answer for the climate change signal. I suggest to discuss this point. Also, as far as possible, I would appreciate if the authors could put their estimate of the change in aerosol lifetime into the context of other estimates from the literature, e.g. in the conclusion section in l. 241, although most of the existing literature estimates will not be directly comparable since they look at different regions and times. For example, Fang et al. (2011) estimate a change in lifetime for their SAt tracer. I also wonder if it would make sense to construct

Very good points. We have added more discussion in the MS – "We should note that there are some caveats for our idealized sensitivity simulations. The way we reduce precipitation frequency in the model (based on a stochastic function as discussed in section 2) can be very different from climate-driven precipitation change in the real world. The globally uniform scaling factors applied to precipitation intensity do not account for the spatial variations. As a consequence, the sensitivities of BC lifetime to precipitation changes over a specific region may be different from those shown in Fig. 2."

In addition, we have constructed contour plots similar to Fig.2 but for various regions. It appears these regional plots are very close to the global one (Figure 6), so we decided to keep the global contour in Fig. 2-4 but add more analysis and discussion about the regional plots in the MS - "To partly address this issue, we have constructed some regional contour plots similar to that in Fig. 2 but based on sensitivities of BC lifetime for those specific regions (Fig. 6). Comparison of these regional contours with the global one indicate some differences in the sensitivity of BC to precipitation changes, but generally less than 3%."

We have added the comparison of our results and literature that "Our results are consistent with *Kloster et al.* [2010] and *Fang et al.* [2011] who reported increasing atmospheric aerosol burden due to climate change, although their results are based on future climate change."

Figure 6: Compare the contours calculated on the global and regional scale: a). global; b). southeast North America (seNA); c). northeast Asia (neAS). The contours indicate the atmospheric lifetimes of the black carbon aerosols from the interpolation of 20 cases and show the potential changes of BC lifetimes from the base BC lifetime (in the control run) driven by the changes of precipitation intensity and frequency. The contour calculated on the global scale is the same with Fig. 2. seNA and neAS are two most extreme cases among all regions, with the smallest and largest sensitivities between BC lifetimes and precipitation changes.

We have shown the standard errors for the calculated changes in precipitation characteristics in Fig. 4 with the size of red block, which reflects the magnitude of the interannual variability in these precipitation fields. Covering a shorter time period, TRMM (Fig. 4) shows much larger standard errors than other datasets (Fig. 5). Based on the TRMM data, it appears the average changes in precipitation characteristics over this 14-year period are significantly larger than the interannual variabilities. To clarify this point, we have added more discussion in MS – "The changes in the average precipitation intensities and frequencies between the periods of 2008-2014 and 2001-2007 for each region are shown as ratios in Fig. 4, with the width and height of the blocks indicting the standard errors of the calculated percentage changes in precipitation frequency and intensity, respectively. Although these TRMM data only cover 14 years, the standard errors as shown in Figure 4 indicate that the changes in precipitation intensity and frequency over most regions are statistically significant."

Good point. We have added clarification in the MS – "In addition, to clearly demonstrate that the BC lifetime has different sensitivities to precipitation intensity and frequency, our sensitivity simulations cover a wide range of precipitation intensity and frequency. Some of these applied perturbations are significantly larger than those induced by climate change, especially at large (such as regional or global) scales. Therefore, simple interpolation of some results from this study in examining the effects from climate change may introduce some uncertainties."

Other specific comments/suggestions/questions:

1. In the introduction, there are a few cases (Salzmann et al., line 28; Trenberth et al., 2007, line 29; Trenberth et al., 2011, line 31; Dawson et al., 2007, line 36; Fang et al., 2011, line 40) in which it might be nice to know what the cited findings are based on (e.g. observations, regional/global model, theoretical arguments, combination of modeling and observations, models constrained by observations, etc).

We have added more information about the findings we cited. –

"*Salzmann* [2016] found that the global mean precipitation did not change significantly since 1850 with climate models, while *Trenberth et al.* [2007] reported that the total precipitation amount increased over land north of 30°N in the past century and decreased in the tropical region after the 1970s based on observational data. *Trenberth* [2011] also noted that theoretically a warmer climate could lead to less frequent but more intense precipitation. "

"For example, *Dawson et al.* [2007] found a strong sensitivity of the concentrations of the PM2.5 (particulate matter with diameter less than 2.5 µm) to precipitation intensity over a large domain of the eastern US with perturbation tests. Only a few studies focused on precipitation frequency."

"*Fang et al.* [2011] projected with the Geophysical Fluid Dynamics Laboratory chemistry-climate model (AM3) that wet deposition has a stronger spatial correlation with precipitation frequency than intensity over the US in January, although they concluded that frequency has a minor effect on wet deposition in the context of climate change."

2. l. 59: in addition to the URL, please also cite at least one paper that describes GEOS-Chem, even if it not exactly the version that is used here.

We have added a citation [*Bey et al.*, 2001] to describe GEOS-Chem.

Bey, I., et al.: Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation. J. of Geophys. Res. Atmos., 106.D19, 23073-23095, 2001.

3. l. 72: unit of P?

We have added the unit of P, which is mm/h.

4. l. 78: did the authors check whether the results are sensitive to this definition?

We picked this definition because it has been widely used in the literature and haven't explored other definitions. This is an interesting point and we may revisit this issue when we carry out relevant analyses in the future. Since we use data from 4 different datasets, re-processing and analyzing all the data would take a long time.

5. l. 115f: did the authors check whether the result is sensitive to this?

We did not check the sensitivity to grid resolution with TRMM data but based on the literature studies [e.g. *Huffman et al*., 2007; *Gehne et al*., 2016], the 2x2.5 resolution would help reduce the relative errors at small precipitation rates. Also the 2x2.5 resolution works well for the continental scale we are looking at in this study.

6. l. 157 and lines 165ff: good points that are nicely explained.

Thank you.

7. l. 178: are those the standard deviations of the yearly means?

No, we calculated the standard errors of the percentage changes using all the data points directly. The temporal resolution for the data varies across datasets; e.g. the TRMM data are 3-hr averages, while the NCEP data are 6-hr averages.

8. l. 240 ff: "precipitation changes" is used here and also further below. It would be better to be more specific regarding whether this is mostly frequency or intensity.

We have changed "precipitation change" to more specific "the changes of precipitation intensity and frequency".

Good point. We have removed this sentence to avoid the possible confusion.

Thank you very much for catching these. We have implemented all of these corrections in the MS.

We have increased the size of the labels as well as the resolutions of figures and hope the high-resolution figures will carry over through the file uploading process.

Sensitivity of atmospheric aerosol scavenging to precipitation intensity and frequency in the context of global climate change

(Marked-up version showing the changes made to the manuscript)

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Abstract. Wet deposition driven by precipitation is an important sink for atmospheric aerosols and soluble gases. We investigate the sensitivity of atmospheric aerosol lifetimes to precipitation intensity and frequency in the context of global climate change. ~~Our sensitivity model simulations, through some simplified perturbations to precipitation in the GEOS-Chem model, shows that~~Our study, based on the GEOS-Chem model simulation, shows that the removal efficiency and hence the atmospheric lifetime of aerosols have significantly higher sensitivities to precipitation frequencies than to precipitation intensities, indicating that the same amount of precipitation may lead to different removal efficiencies of atmospheric aerosols. Combining the long-term trends of precipitation patterns for various regions with the sensitivities of atmospheric aerosol~~s~~ lifetimes to various precipitation characteristics allows us to examine the potential impacts of precipitation changes on atmospheric aerosols. Analyses based on an observational dataset show that precipitation frequency in some regions have decreased in the past 14 years, which might increase the atmospheric aerosol lifetimes in those regions. Similar analyses based on multiple reanalysis meteorological datasets indicate that the changes of precipitation intensity and frequency~~the precipitation changes~~ over the past 30 years can lead to perturbations in the atmospheric aerosol lifetimes by 10% or higher at the regional scale.

1 Introduction

Wet scavenging is a major removal process for aerosols and ~~other atmospheric elements~~soluble trace gases [*Atlas and Giam*, 1988; *Radke et al.*, 1980]. Global climate change implies significant perturbations of precipitation, which can directly affect the wet scavenging process. *Salzmann* [2016] ~~found~~found that the global mean precipitation did not change significantly ~~in the past~~since 1850 with climate models, while *Trenberth et al.* [2007] reported that the total precipitation amount increased over

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land north of 30°N in the past century and decreased in the tropical region after the 1970s based on observational data. *Trenberth* [2011] also noted that theoretically a warmer climate could lead to less frequent but more intense precipitation.

30   The impacts of long-term changes in precipitation characteristics on air quality have not been well studied. Most previous studies focused on the correlation between air pollution and the total precipitation amount or precipitation intensity [*Cape et al.*, 2012; *Pye et al.*, 2009; *Tai et al.*, 2012]. For example, *Dawson et al.* [2007] found a strong sensitivity of the concentrations of PM$_{2.5}$ (particulate matter with diameter less than 2.5 μm) concentrations to precipitation intensity over a large domain of the eastern US

35   with perturbation tests. Only a few studies focused on precipitation frequency. *Jacob and Winner* [2009] noted that precipitation frequency could be more important than precipitation intensity for air quality, because the wet scavenging process due to precipitation is very efficient [*Balkanski et al.*, 1993]. *Fang et al.* [2011] projected with the Geophysical Fluid Dynamics Laboratory chemistry-climate model (AM3) that wet deposition has a stronger spatial correlation with precipitation frequency than intensity over the

40   US in January, although they concluded that frequency has a minor effect on wet deposition in the context of climate change. *Mahowald et al.* [2011] also mentiondiscussed the importance of precipitation frequency in wet deposition based on simulations with showing large removal rate of dust in precipitation events.

In this study, we first use GEOS-Chem, a global 3-D chemical transport model (CTM), to examine the

45   sensitivities of atmospheric aerosol lifetimes to various precipitation characteristics, including the precipitation intensity, frequency, and total amount. By isolating these precipitation characteristics from other meteorological fields through a suite of perturbation simulations, we are able to better understand the sensitivities of atmospheric aerosols to various precipitation characteristics. We focus on black carbon (BC) as a proxy for atmospheric aerosols to examine the impacts of changes in precipitation

50   characteristics. BC is nearly inert in the atmosphere [*Ramanathan and Carmichael*, 2008], making it a good tracer for studying the transport and deposition of atmospheric species. We also analyze the long-term trends of the precipitation characteristics over various regions around the world, based on the observational and reanalysis meteorological datasets for the past decades. We then combine the long-term trends in the precipitation patterns for various regions with the sensitivities of BC to

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55 precipitation characteristics to ~~quantity~~ quantify their potential impacts on atmospheric aerosols in the context of global climate change.

2 Methods

We utilize a global 3-D chemical transport model (CTM), GEOS-Chem *[Bey et al., 2001]* version

9-02-01 ~~[Bey et al., 2001]~~ (www.geos-chem.org), to carry out a suite of perturbation tests to examine the

60 sensitivities of atmospheric aerosols to precipitation characteristics. As a chemical transport model, the GEOS-Chem model does not simulate meteorology prognostically; instead it is driven by assimilated meteorological data from the Goddard Earth Observing System (GEOS) of NASA GMAO. We use the GEOS-5 meteorological dataset in this study. We conduct global simulations with a horizontal resolution of 4° latitude by 5° longitude and 47 vertical layers. All the model simulations in this study run from 1

65 July 2005 to 1 January 2007, i.e., for one and half years, with the first half year serving as the model spin-up.—

The wet deposition scheme in GEOS-Chem includes scavenging in convective updrafts, in-cloud scavenging (rainout), and below-cloud scavenging (washout), which were described in detail~~s~~ by *Liu et al.* [2001] and *Wang* et al. [2011]. In GEOS-Chem simulation, the BC aerosols are classified into two

70 types based on their hygroscopicity (hydrophobic vs hydrophilic) and wet scavenging is ~~only~~more efficient~~ly~~ when~~for~~ hydrophilic BC.~~The efficiency of wet scavenging is very sensitive to the~~ ~~hydrophilicity of BC in the scheme of GEOS-Chem~~ . GEOS-Chem assumes the ratio between hydrophobic BC ~~(BCPO)~~ and hydrophilic BC ~~(BCPI)~~ to be 4:1 in fresh emissions and ~~BCPO~~ hydrophobic BC converts to ~~BCPI~~ hydrophilic one with an e-folding lifetime of 1.15 days.

75 The washout rate constant (k~~k~~) is affected by the particle size and the form of precipitation. For washout by rain with precipitation rate P~~P~~ –(mm/h)~~(mm h⁻¹)~~, $k = 1.1 \times 10^{-3}P^{0.61}$ for accumulation mode (aerosols with diameters between 0.04 μm and 2.5 μm) and $k = 0.92P^{0.79}$ for coarse mode (aerosols with diameter between 2.5 μm to 16 μm); for washout by snow with precipitation rate P~~P~~, $k = 2.8 \times 10^{-2}P^{0.96}$ for accumulation mode and $k = 1.57P^{0.96}$ for coarse mode [*Feng, 2007, 2009*]. The

80 coefficients for accumulation-mode are used in calculating k for fine particles including BC in GEOS-Chem.

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Our study focuses on three precipitation characteristics: the precipitation intensity, frequency, and total amount. We define precipitation events as the data points with ""significant" (we use precipitation rate more than 1 mm/day as the criterion in this study) precipitation. Precipitation intensity is the average precipitation rate on precipitation events, with a unit of mm/day. Precipitation frequency is the fraction of precipitation events during the study period (i.e., the probability of any given data points with more than 1 mm/day precipitation rate), which is dimensionless. Total precipitation amount is defined as the average amount of precipitation rate during the study period, with a unit of mm/day. Assuming that precipitation is negligible on data points with no "precipitation events", we would have

$$total\ precipitation\ amount \cong precipitation\ intensity \cdot precipitation\ frequency \qquad (1)$$

For sensitivity tests focused on precipitation intensity, we scale the base GEOS-5 precipitation values from the control run by a uniform factor for each grid box. For the sensitivity tests focused on precipitation frequency, we use a stochastic function to turn off the precipitation at a given data point. For example, in a simulation where we reduce the precipitation frequency by 25%, for a data point (i,j,t)~~(i, j, t)~~, we modify the initial precipitation rate $P_0(i,j,t)$~~(i, j, t)~~ to

$$P(i,j,t) = \begin{cases} P_0(i,j,t); & R(i,j,t) \geq 0.25 \\ 0; & R(i,j,t) < 0.25 \end{cases} \qquad (2)$$

where R~~R~~ is a random function with a range of (0, 1). In this way, we decrease the precipitation frequency of each grid box to 75% of its base value across the whole study domain and keep the base spatiotemporal precipitation patterns over each specific region.

For convenience in identifying and describing all the ~~simulation~~ sensitivity tests, we name them after their precipitation frequency and intensity scaling factors. For instance, the case f0.5i2 represents the simulation with half the base precipitation frequency and twice the base precipitation intensity, while the case f1i1 indicates the control simulation with a base frequency and intensity. We carry out more than 20 sensitivity model simulations to cover various precipitation intensities and frequencies as shown in Table 1.

105 Table 1. The designs of perturbation testsSeries of sensitivity model simulations carried out in this study.

TestModel runs	Objectiveon	Case names
Constant precipitation frequency (Fig. 1a)	To study the sensitivity of BC lifetime to precipitation intensity	f1i0.25, f1i0.5, f1i1, f1i2, and f1i4
Constant precipitation intensity (Fig. 1b)	To study the sensitivity of BC lifetime to precipitation intensity	f0.1i1, f0.25i1, f0.5i1, f0.75i1, and f1i1
Constant precipitation amount (Fig. 1c)	To compare the sensitivity of BC lifetime to precipitation intensity and precipitation frequency	f0.1i10, f0.25i4, f0.5i2, f0.75i1.33, and f1i1
Hygroscopicity of aerosols (100% vs 20% BC in fresh emissions are assumed to be hydrophilic)	To examine the impacts on wet deposition from the parameterization on the hygroscopicity of aerosols	f1i1 and f0.75i1.33 f1i1 and f0.75i1.33
Aerosols size (BC aerosols are assumed to be in coarse mode vs accumulation mode)	To examine the impacts on wet scavenging from aerosol size	f1i1 and f0.75i1.33 f1i1 and f0.75i1.33
Contour of BC lifetime (Fig. 2, 4-6)	BC lifetime as a function of the precipitation intensity and frequency	f0.25i0.5, f0.25i1, f0.25i1.33, f0.25i2, f0.25i4, f0.5i0.5, f0.5i1, f0.5i1.33, f0.5i2, f0.5i4, f0.75i0.5, f0.75i1, f0.75i1.33, f0.75i2, f0.75i4, f1i0.5, f1i1, f1i1.33, f1i2, and f1i4

The abundance of atmospheric aerosols is determined by both the aerosol emission rates and their atmospheric residence times, i.e., their lifetimes. The average atmospheric lifetimes of aerosols are

110 calculated as

$$lifetime = \frac{burden}{removal\ rate} = \frac{burden}{dry\ deposition\ rate + wet\ deposition\ rate} \quad (3)$$

Therefore, more efficient wet scavenging would lead to shorter atmospheric aerosol lifetimes.

We then examine the long-term changes in precipitation characteristics for various regions around the

world in past decades. We first analyze changes in the precipitations between two 7-yr periods

(2008-2014 vs. 2001-2007) based on an observational dataset, the 3-Hour Realtime Tropical Rainfall

Measuring Multi-Satellite Precipitation Analysis version 7 (TRMM3B42v7, short for TRMM, https://pmm.nasa.gov/TRMM). TRMM (3B42v7) performances better than the previous version of satellite products (3B42v6), though there are still problems in detecting precipitation events with low precipitation rates [*Maggioni et al.*, 2016]. We then examine three reanalysis datasets with longer temporal coverage (2001-2010 vs 1981-1990): the National Centers for Environmental Prediction (NCEP) reanalysis dataset [*Kalnay et al.*, 1996], the NCEP-DOE AMIP-II (NCEP2) reanalysis dataset [*Kanamitsu et al.*, 2002], and NASA's Modern-Era Retrospective ~~Analysis~~ analysis for Research and Applications (MERRA) ~~database~~ dataset [*Rienecker et al.*, 2011]. These datasets have different resolutions and spatial coverage. TRMM only covers 60°N-60°S, while other datasets cover the whole globe. The resolutions (°lon x °lat x hour) for TRMM, NCEP, NCEP2, and MERRA are 0.25x0.25x3, 2.5x2.5x6, 2.5x2.5x6, 2.5x2x1, respectively. We regrid the TRMM dataset from 0.25x0.25 to 2.5x2.5 (°lon x °lat) to reduce the computational cost and the relative errors at small precipitation rates [*Huffman et al.*, 2007; *Gehne et al.*, 2016]. By combining the resulting sensitivities of BC lifetimes to precipitation characteristics with the results of the long-term trends in precipitation characteristics, we then estimate the impacts of long-term changes in precipitation characteristics on the atmospheric lifetime of BC.

3 Results

The global annual mean lifetime of BC is calculated at 5.29 days in our control simulation (Fig. 1). This value is similar to the results of a previous study, which stated that the lifetime of BC would be around one week [*Ramanathan and Carmichael*, 2008]. Our result also agrees with the lifetime of 5.8 ± 1.8 days simulated by the GEOS-Chem model [*Park et al.*, 2005] and the 5.4 days result simulated by the ECHAM5-HAM model [*Stier et al.*, 2005]. For 13 models in AeroCom, the lifetimes of BC from anthropogenic fossil fuel and biofuel sources are simulated to be from 3.5 to 17.1 days, with 5.9 days as the median value [*Samset et al.*, 2014].

We first compare the results of the control run with other simulations with the same precipitation frequency (f1i0.25, f1i0.5, f1i1, f1i2, and f1i4) to examine the sensitivity of BC lifetime to precipitation intensity (Fig. 1a). We find that an increase in precipitation intensity leads to decreases in both the BC lifetime and the sensitivity of the BC lifetime to precipitation intensity. That is, the impact of

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precipitation intensity on BC aerosols is saturated when the intensity is very high, which is consistent with a previous study [*Fang et al.*, 2011]. We then compare the control run with other simulations with the same precipitation intensity (f0.1i1, f0.25i1, f0.5i1, f0.75i1, and f1i1) to study the sensitivities of the BC lifetime to precipitation frequency (Fig. 1b). Again, the BC lifetime responds non-linearly to the changes in precipitation frequency, and the sensitivity decreases with increases in precipitation frequency.

When we compare the simulations with a common precipitation amount (f0.1i10, f0.25i4, f0.5i2, f0.75i1.33, and f1i1), we find that the BC lifetime increases with increasing precipitation intensity (Fig. 1c). For example, case f0.1i10 has an annual average BC lifetime of 7.86 days, which is much longer than the 5.29 days of the control simulation (case f1i1). This indicates that the sensitivity of the BC lifetime to precipitation frequency is stronger than that to the precipitation intensity.

The calculated efficiency of wet scavenging can be affected by model parameterizations. We first examine the possible impacts on our results from the parameterization on the hygroscopicity of aerosols. With the default parameterization in GEOS-Chem, 20% of the fresh BC emissions are assumed to be hydrophilic. We use this as the control run and then set up sensitivity runs We set up sensitivity runs with another parameterization, where all BC are assumed to be hydrophilic. With these two different parameterization schemes, we examine We compare the changes in the BC lifetime between two scenarios (f1i1 vs. f0.75i1.33) respectively with alternative parameterization schemes. We find that with the default setting in GEOS-Chem (20% BC in fresh emissions are hydrophilic), the atmospheric lifetime of BC under the f0.75i1.33 scenario is slightly higher than the f1i1 scenario by 0.4%. In comparison, Iif all the BC emission is assumed to be hydrophilic, the BC lifetime under the f0.75i1.33 scenario would be 3.6% higher. This implies that for hydrophilic aerosols, the sensitivity to precipitation frequency would be even higher.

We also evaluate the impacts on wet scavenging from aerosol size with sensitivity simulations. If we assume the aerosols to be in coarse mode, we find that thisit would lead to more efficient scavenging and consequently much shorter lifetime (compared to the default setting in GEOS-Chem that all BC aerosols are in accumulation mode). However, there are no significant effects on the relative sensitivities to precipitation frequency vs. intensity – the percentage change in BC lifetime between the f1i1 and

170   f0.75i1.33 scenarios is very similar to the cases with parameterization for accumulation mode (0.3% vs. 0.4%). This indicates that the relative sensitivity of the BC lifetime to precipitation frequency and precipitation intensity is not significantly affected by the parameterization of particle size in the wet scavenging scheme in GEOS-Chem. It is worth noting that our model does not resolve the size of precipitation droplet, which can also affect the efficiency of wet scavenging.

175   The stronger sensitivity of the BC lifetime to precipitation frequency than that to intensity implies that an increase in the total precipitation amount does not necessarily lead to a decrease in the BC lifetime. This is better illustrated in Fig 2, which shows the BC lifetime as a function of the precipitation intensity and frequency based on 20 cases (f0.25, f0.5, f0.75, f1 versus i0.5, i1, i1.33, i2, i4). Compared with the control scenario (i.e., f1i1, the base precipitation intensity and frequency, as labeled by the black star),

180   any point in the area between the two solid curves (the green one shows a constant total precipitation amount and the red one shows a constant BC lifetime) would have a higher total precipitation amount and a longer BC lifetime. This indicates that, even with an increased total precipitation, the BC lifetime (and hence the atmospheric concentrations of BC) can still increase if the precipitation frequency decreases significantly. This feature may help explain the decrease of the wet deposition flux found in wetter future

185   climate simulations, despite their slightly increased total precipitation amounts (Xu and Lamarque, personal communication; manuscript under review)[*Xu et al.*, 2018].

The lifetime contour plot in Fig. 2 can be employed as a simple tool to help us understand the impacts of long-term changes in precipitation on atmospheric aerosols, so we also investigate the long-term trends in the precipitation characteristics over the past decades for various regions around the world. In

190   considering the spatial variations of precipitation patterns and their long-term trends, we divide the global continental regions into multiple subcontinental areas to better resolve the spatial variations (Fig. 3). We first carry out an analysis based on precipitation data from the TRMM dataset. The changes in the average precipitation intensities and frequencies between the periods of 2008-2014 and 2001-2007 for each region are shown as ratios in Fig. 4, with. tThe width and height of the blocks in Fig. 4

195   indicateindicting the standard deviations errors of the calculated percentage changes ofin precipitation frequency and intensity, respectively. Although these TRMM data only cover 14 years, Tthe standard errors as shown in Figure 4 indicate that though covering a relatively short time period, most

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~~percentage~~the changes ~~of~~in precipitation intensity and frequency ~~over most regions~~ ~~in TRMM dataset~~are statistically significant. We find that during these 14 years, the average precipitation intensity has increased over most regions, but the average precipitation frequency has decreased over more than one third of the total regions including western North America (nwNA and swNA), southern South America (sSA), western Europe (wEU), southern Africa (sAF), and southwestern Asia (swAS). Based on the TRMM dataset, we find that almost all (5 out of 6) of the regions with decreasing precipitation frequency are expected to experience longer atmospheric aerosol lifetimes.

Since the TRMM data only cover a relatively short period, we make similar analyses with three reanalysis datasets (NCEP, NCEP2, and MERRA) to cover a longer time period (2001-2010 vs. 1981-1990) (Fig. 5). We find that, similar to the TRMM data, all the three reanalysis datasets show increasing trends for precipitation intensity over most regions but more divergent trends for precipitation frequency in the past decades. The NCEP data show that precipitation frequency has decreased over about two thirds of the total regions while NCEP2 and MERRA data show decreasing precipitation frequency over one third and half of the total regions, respectively. In addition, even when the different datasets indicate the same direction for the precipitation change over a specific region, the magnitude of the changes may vary significantly across datasets. For example, the derived changes in the average precipitation intensity over neNA (northeastern North America) based on NCEP, NCEP2, and MERRA data are +8%, +12%, and +3% respectively. These variations across different data sources reflect the significant uncertainties associated with these datasets, ~~partly driven by the bias of moisture budget in reanalysis datasets,~~ as reported earlier [e.g., *Trenberth and Christian*, 1998; *Trenberth et al.*, 2011; *Gehne et al.*, 2016].

On the other hand, ~~previous~~previous analysis on global land-average precipitation showed that various reanalysis datasets have similar trends and interannual variability with other gauge- and satellite-based datasets during 2001-2010, though the estimated trend of precipitation varies based on temporal and spatial scales [*Gehne et al.*, 2016]. In addition, our study focuses on the changes over continental regions, where the precipitation data in the reanalysis datasets are found to be more reliable than over the ocean regions [*Trenberth et al.*, 2011]. Therefore despite the uncertainties associated with each meteorological dataset, we can use Fig. 5 to estimate the expected changes in the atmospheric BC lifetimes for certain

regions, especially for those regions showing consistent trends across different datasets. Assuming the effects of precipitation on wet deposition is the only factor that affects the atmospheric BC aerosol lifetimes, all three datasets indicate that atmospheric BC aerosol lifetimes could have decreased in the northern regions of North America (neNA and nwNA), the northwestern and southern regions of South America (nwSA and sSA), South Africa (sAF), and North Oceania (nOC). All three meteorological datasets show increasing trends in aerosol lifetimes over southwestern North America (swNA), Middle Africa (mAF), and South Oceania (sOC), which imply increasing trends for the concentrations of particulate matter (PM) over these regions, driven by changes in precipitation. At the regional scale, precipitation changes over the past 30 years can easily lead to perturbations in atmospheric BC lifetimes by 10% or higher.

We should note that there are some caveats for our idealized sensitivity simulations. The way we reduce precipitation frequency in the model (based on a stochastic function as discussed in section 2) can be very different from climate-driven precipitation change in the real world. The globally uniform scaling factors applied to precipitation intensity do not account for the spatial variations. As a consequence, the sensitivities of BC lifetime to precipitation changes over a specific region may be different from those shown in Fig. 2. To partly address this issue, we have constructed some regional contour plots similar to that in Fig. 2 but for variousbased on sensitivities of BC lifetime for those specific regions (Fig. 6). Comparison of these regional contours with the global one indicate some differences in the sensitivity of BC to precipitation changes, but generally less than 3%. In addition, to show the changesclearly demonstrate that of the BC lifetime clear, wehas different sensitivities to precipitation intensity and frequency, choose relatively large factors of changes in our sensitivity simulations cover a wide range of precipitation intensity and frequency. Some of these applied perturbations are significantly larger than those induced by climate change, especially at large (such as regional or global) scales. Therefore, simple interpolation of some results from this study in examining the effects from climate change may introduce some uncertainties. when compare to the changes of precipitation in climate change, which may also introduce uncertainty in our prediction results.Our results are also affected by the limitations from the meteorology datasets. Although the TRMM and the reanalysis datasets used in this study represent some of the best meteorological datasets available, each

of them has their own shortcomings - the observational datasets are more reliable, but only cover a relatively short time period of 14 years; the reanalysis datasets cover longer periods, but are less reliable due to known issues such as the bias in moisture budget [e.g., *Trenberth and Christian*, 1998; *Trenberth et al.*, 2011; *Gehne et al.*, 2016].

4 Conclusions and Discussion

The efficiency of the wet scavenging of atmospheric aerosols is affected by not only the precipitation amount but also the precipitation patterns. Our results, based on sensitivity simulations with the GEOS-Chem model, show that the atmospheric lifetimes of BC are more sensitive to precipitation frequency than precipitation intensity, and as a consequence, increases in the total precipitation amount do not always lead to a more efficient wet scavenging of atmospheric aerosols. The sensitivities of the atmospheric lifetimes of aerosols to the precipitation characteristics derived from our model simulations offer a simple and convenient tools for us to better examine the implications of long-term changes in precipitation (including the total amounts and patterns) for atmospheric aerosols in various regions. We should note that there are some caveats for our idealized sensitivity simulations. The way we reduce precipitation frequency in the model (based on a stochastic function as discussed in section 2) can be very different from climate-driven precipitation change in the real world. The globally uniform scaling factors applied to precipitation intensity do not account for the spatial variations. As a consequence, the sensitivities of BC lifetime to precipitation changes over a specific region may be different from those shown in Fig. 2. To partly address this issue, we have constructed contour plots similar to that in Fig. 2 but for various regions (Fig. S1). Comparison of these regional contours with the global one indicate some differences in the sensitivity of BC to precipitation changes, but generally less than xx%.

Analysis of satellite data (TRMM) for the past 14 years (2001-2014) reveal that precipitation intensity have has increased in most regions. On the other hand, decreasing precipitation frequency are found in some regions such as western North America, southern South America, western Europe, southern Africa, and southwestern Asia. The decreases in precipitation frequency could lead to increases in atmospheric aerosol lifetimes over these regions. Our further analyses based on three meteorological datasets (NCEP, NCEP2, and MERRA) for the past decades (1981-2010) show increases in precipitation intensities over

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most continental regions, but significant decreases in precipitation frequency are identified over some regions. These changes in precipitation characteristics affect the wet deposition of aerosols and consequently the total burdens of aerosols and their atmospheric lifetimes. Despite the significant uncertainties associated with meteorological data, we find that the changes in~~of~~ precipitation intensity and frequency~~changes~~ over the past 30 years could have led to perturbations in the regional atmospheric aerosol lifetimes by 10% or higher. Our results ~~agree with the simulations of~~are consistent with *Kloster et al.* [2010] and *Fang et al.* [2011] who reported increasing~~that~~ atmospheric aerosol burden ~~increases~~ due to climate change, although their ~~findings~~results are based ~~focus~~ on ~~a~~ future climate change. We also find that all three meteorological databases are consistent to show that the changes ~~of~~in precipitation intensity and frequency~~precipitation changes~~ over the past decades have led to decreases in atmospheric aerosol lifetimes over the northern regions of North America, northwestern and southern regions of South America, South Africa, and North Oceania. They are also consistent in indicating increasing trends of atmospheric aerosol lifetimes in the southwestern region of North America, Middle Africa, and South Oceania. The increasing trends in atmospheric aerosol lifetimes over these regions driven by the changes ~~of~~in precipitation intensity and frequency~~precipitation changes~~ in the context of global climate change could pose challenges for the local PM air qualities. It should be noted that the results from this work can be affected by the parameterization in the GEOS-Chem model and have certain limitations. Our study does not account for the impacts of precipitation on wildfires which can emit massive amount of aerosols including BC [*Dawson et al.*, 2014]. ~~In addition, our study perturbation tests does not account for the feedbacks from the changes of aerosols on radiation and precipitation. It may be worthy to carry out some future work accounting for these indirect effects and feedbacks to further evaluate the impacts of long-term changes in precipitation on atmospheric aerosols.~~

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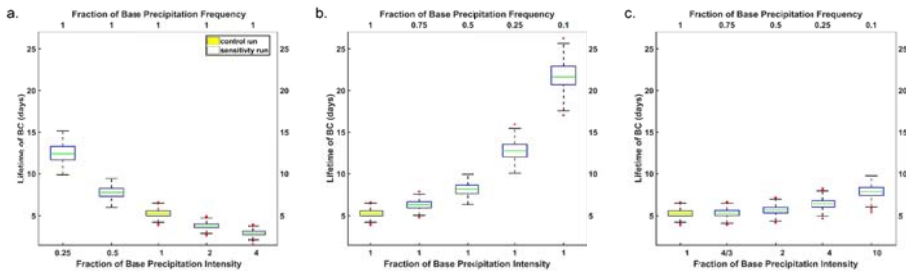
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Table 1. Series of sensitivity model simulations carried out in this study.

Model simulations	Objective	Case names
Constant precipitation frequency (Fig. 1a)	To study the sensitivity of BC lifetime to precipitation intensity	f1i0.25, f1i0.5, f1i1, f1i2, and f1i4
Constant precipitation intensity (Fig. 1b)	To study the sensitivity of BC lifetime to precipitation frequency	f0.1i1, f0.25i1, f0.5i1, f0.75i1, and f1i1
Constant precipitation amount (Fig. 1c)	To compare the sensitivity of BC lifetime to precipitation intensity and precipitation frequency	f0.1i10, f0.25i4, f0.5i2, f0.75i1.33, and f1i1
Hygroscopicity of aerosols (100% vs 20% BC in fresh emissions are assumed to be	To examine the impacts on wet deposition from the parameterization on the	f1i1 and f0.75i1.33

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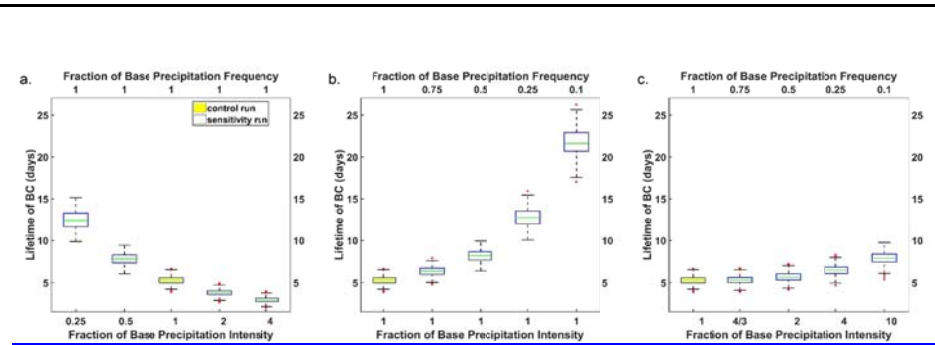| hydrophilic) | hygroscopicity of aerosols | |
| Aerosol size (BC aerosols are assumed to be in coarse mode vs accumulation mode) | To examine the impacts on wet scavenging from the parameterization on the size of aerosols | f1i1 and f0.75i1.33 |
| Contour of BC lifetime (Fig. 2, 4-6) | To plot BC lifetime as a function of the precipitation intensity and frequency | f0.25i0.5, f0.25i1, f0.25i1.33, f0.25i2, f0.25i4, f0.5i0.5, f0.5i1, f0.5i1.33, f0.5i2, f0.5i4, f0.75i0.5, f0.75i1, f0.75i1.33, f0.75i2, f0.75i4, f1i0.5, f1i1, f1i1.33, f1i2, and f1i4 |

Figure 1: Impacts of the precipitation characteristics on the atmospheric lifetime of BC under given a) constant precipitation frequency; b) constant precipitation intensity; and c) constant precipitation amount. The top x-axis reflects the precipitation frequency set in each perturbation test, shown as fractions of base precipitation frequency. Base precipitation frequency is the precipitation frequency used in the control case. Similarly, the bottom x-axis reflects the settings of precipitation intensity in the perturbation tests. The box plot shows the probability distribution of BC lifetime for each case, where the top and bottom edges of each box show the third and first quartiles, respectively; the green central bar shows the median; the whisker shows the range of the non-outliers that cover 99.3% of the data, assuming normally distributed data; and the red plus shows the outliers.
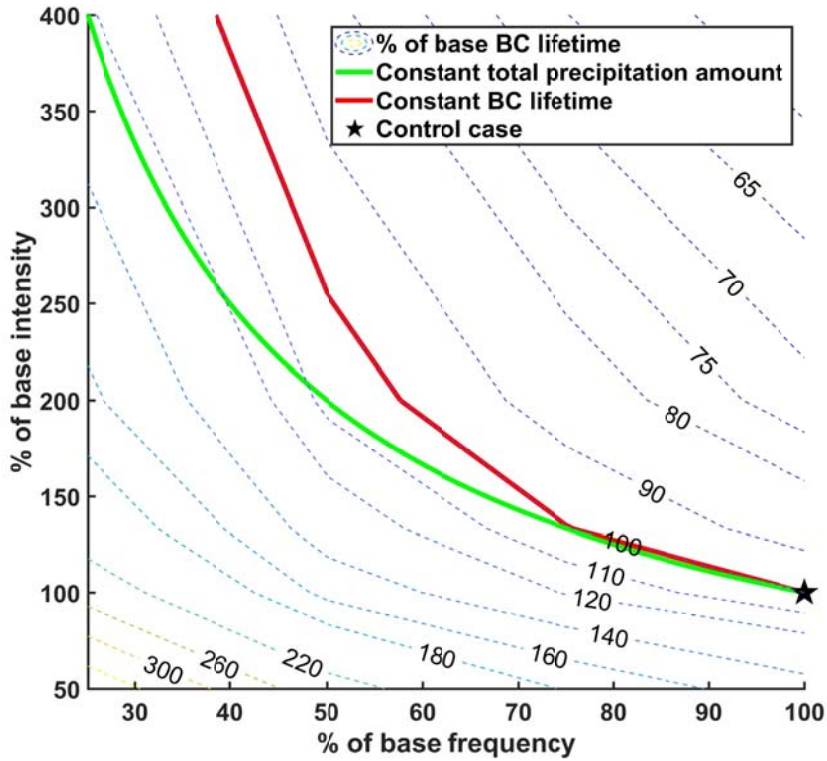
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Figure 2: Model calculated BC atmospheric lifetime as a function of precipitation intensity and frequency. The dashed contour lines indicate the atmospheric lifetimes of the black carbon aerosols from the interpolation of 20 cases, which show the potential changes of BC lifetimes from the base BC lifetime (in the control run) driven by the changes of precipitation intensity and frequency. The green solid line represents a

425 **total precipitation equal to that of the base simulation (control run). The red solid line indicates the conditions leading to atmospheric black carbon aerosol lifetimes that match the base simulation (control run).**

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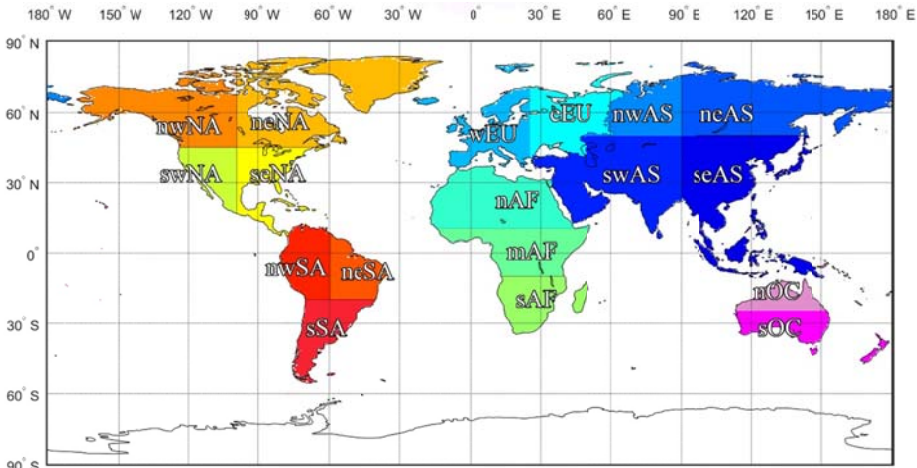
Figure 3: The definitions of the continental regions in this study. The uppercase letters in the region names

430 **represent the names of their continents: North America (NA), South America (SA), Europe (EU), Africa (AF), Asia (AS), and Oceania (OC). The lowercase letters in the region names represent the subregions inside the continent: north (n), south (s), west (w), east (e), middle (m), northwest (nw), northeast (ne), southwest (sw), and southeast (se).**
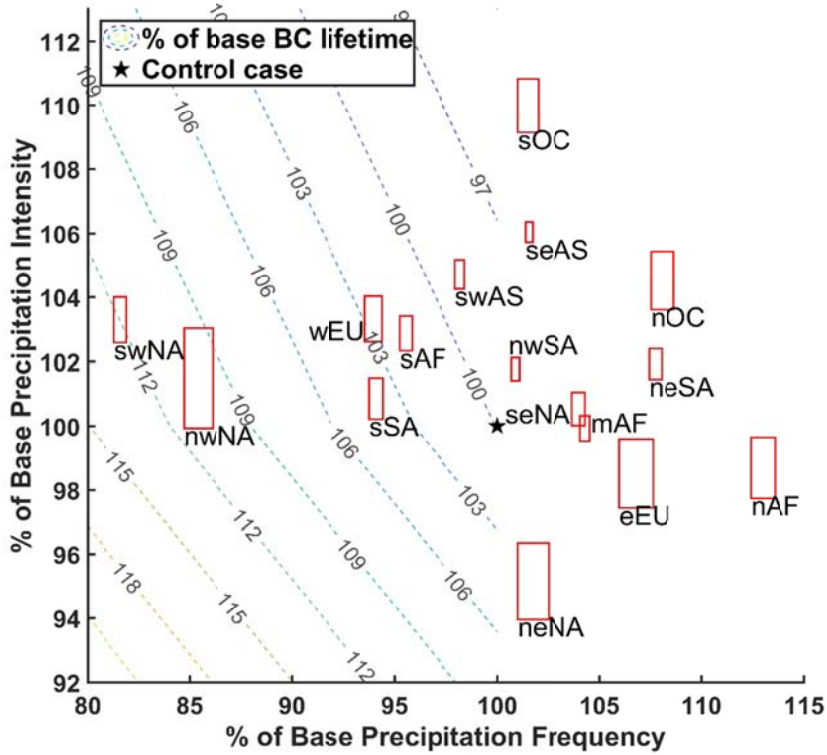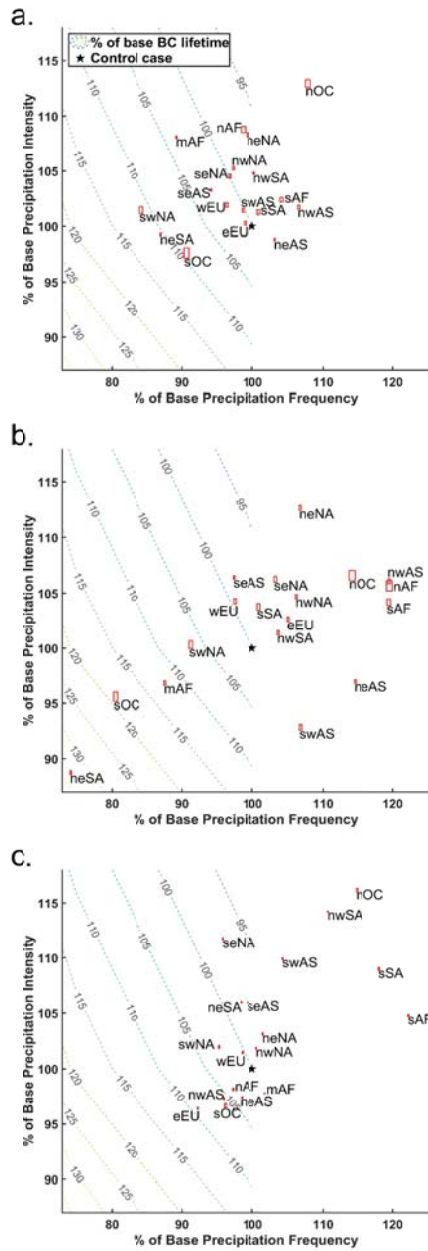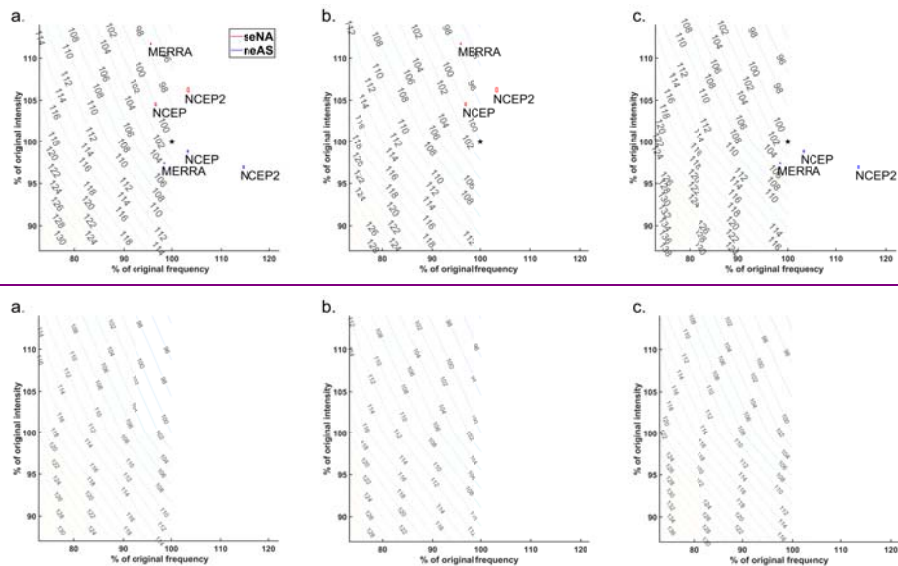
Figure 4: The potential change of atmospheric BC aerosol lifetime driven by the changes between the two periods (2008-2014 and 2001-2007) in precipitation characteristics based on meteorological datasets TRMM. The dashed contours are the same as in Fig. 2, which indicate the atmospheric lifetimes of the black carbon aerosols from the interpolation of 20 cases and show the potential changes of BC lifetimes from the base BC lifetime (in the control run) driven by the changes of precipitation intensity and frequency. Red blocks show the changes of precipitation intensities and frequencies, with the size of the block showing the standard error of the ~~mean~~ percentage changes.

Figure 5: The potential change of atmospheric BC aerosol lifetime driven by the changes between the two periods (2001-2010 and 1981-1990) in precipitation characteristics based on multiple meteorological datasets: a). NCEP; b). NCEP2; c). MERRA. The dashed contours are the same as in Fig. 2, which indicate the atmospheric lifetimes of the black carbon aerosols from the interpolation of 20 cases and show the potential changes of BC lifetimes from the base BC lifetime (in the control run) driven by the changes of precipitation intensity and frequency. Red blocks show the changes of precipitation intensities and frequencies, with the size of the block showing the standard error of the ~~mean~~ percentage changes.

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Figure 6: Compare the contours calculated on the global and regional scale: a). global; b). southeast North America (seNA); c). northeast Asia (neAS). The contours indicate the atmospheric lifetimes of the black carbon aerosols from the interpolation of 20 cases and show the potential changes of BC lifetimes from the base BC lifetime (in the control run) driven by the changes of precipitation intensity and frequency. The contour calculated on the global scale is the same with Fig. 2. seNA and neAS are two most extreme cases among all regions, with the smallest and largest sensitivities between BC lifetimes and precipitation changes.