Sensitivity of atmospheric aerosol scavenging to precipitation

intensity and frequency in the context of global climate change

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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 study, based on the GEOS-Chem model

15 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 aerosols lifetimes to various precipitation characteristics allows us to examine the potential

20 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 precipitation changes over the past 30 years can lead to perturbations in the atmospheric aerosol lifetimes by 10% or higher at the regional scale.

25 **1 Introduction**

Wet scavenging is a major removal process for aerosols and other atmospheric elements [*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 that the global mean

precipitation did not change significantly in the past, while *Trenberth et al.* [2007] reported that the total

30   precipitation amount increased over land north of 30°N in the past century and decreased in the tropical

region after the 1970s. *Trenberth* [2011] also noted that a warmer climate could lead to less frequent but

more intense precipitation.

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

35   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) to precipitation intensity over a large domain of the eastern US. 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

40   due to precipitation is very efficient [*Balkanski et al.*, 1993]. *Fang et al.* [2011] projected 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.

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

55   precipitation characteristics to quantity 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 version 9-02-01
(www.geos-chem.org), to carry out a suite of perturbation tests to examine the sensitivities of
60   atmospheric aerosols to precipitation characteristics. As a chemical transport model, the GEOS-Chem
model does not simulate meteorology; 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 July 2005 to 1 January
65   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 details by *Liu et
al.* [2001] and *Wang* et al. [2011]. The efficiency of wet scavenging is very sensitive to the hydrophilicity
of BC. GEOS-Chem assumes the ratio between hydrophobic BC (BCPO) and hydrophilic BC (BCPI) to
70   be 4:1 in fresh emissions and BCPO converts to BCPI with an e-folding lifetime of 1.15 days.

The washout rate constant (k) is affected by the particle size and the form of precipitation. For washout
by rain with precipitation rate P, $k = 1.1 \times 10^{-3} P^{0.61}$ for accumulation mode (aerosols with diameters
between 0.04 μm and 2.5 μm) and $k = 0.92 P^{0.79}$ 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
75   mode and $k = 1.57 P^{0.96}$ for coarse mode [*Feng*, 2007, 2009]. The coefficients for accumulation-mode
are used in calculating k for fine particles including BC in GEOS-Chem.

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
80   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

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

85   For sensitivity tests focused on precipitation intensity, we scale the base GEOS5 precipitation values

from 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), we modify the

initial precipitation rate $P_o(i, j, t)$ to

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

90   where 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 tests, we name them after their

precipitation frequency and intensity scaling factors. For instance, the case f0.5i2 represents the

95   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.

The abundance of atmospheric aerosols is determined by both the aerosol emission rate and their

atmospheric residence times, i.e., their lifetimes. The average atmospheric lifetimes of aerosols are

100   calculated as

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

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

105   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

110   (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 (MERRA) database

[*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.

115   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.

120   **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

125   ECHAM5-HAM model [*Stier et al.*, 2005].

We first compare the results of 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 precipitation intensity

130   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.

135   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.

140   The efficiency of wet scavenging can be affected by model parameterization. We first examine the impacts on our results from the parameterization on the hygroscopicity of aerosols. We compare the changes in the BC lifetime between two scenarios (f1i1 vs. f0.75i1.33) 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

145   scenario by 0.4%. If 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 would lead to more efficient scavenging and

150   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 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

155   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.

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

160   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., the base precipitation intensity and frequency, as labeled by the black star), 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

165   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

climate simulations, despite their slightly increased total precipitation amounts (Xu and Lamarque,

personal communication; manuscript under review).

170   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

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.

175   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. The width and height of the blocks in Fig. 4 indicate the

standard deviations of precipitation frequency and intensity, respectively. 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).

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

185   long-term changes in the atmospheric aerosol lifetimes driven by precipitation changes. With given

perturbations in precipitation intensity and frequency for a specific region, the potential change in the

BC lifetime can be estimated based on the lifetime-contour plots in Fig. 4. The design of the sensitivity

tests with precipitation frequency only allows us to carry out "one-way" sensitivity tests (i.e. with

reduced precipitation frequency compared to the control run), as a consequence, all the contours in Fig.

190   2, Fig. 4, and Fig. 5 end at the level of base precipitation frequency. 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.

195   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

200   datasets indicate 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, as reported earlier [e.g., Trenberth *et al.*, 2011;

205   Gehne *et al.*, 2016].

On the other hand, 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

210   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

215   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

220 (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.

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

225 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 simple and convenient tools for us to better examine the implications of long-term changes in

230 precipitation (including the total amounts and patterns) for atmospheric aerosols in various regions.

Analysis of satellite data (TRMM) for the past 14 years (2001-2014) reveal that precipitation intensity

have 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

235 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

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

240 uncertainties associated with meteorological data, we find that the precipitation changes over the past 30

years could have led to perturbations in the regional atmospheric aerosol lifetimes by 10% or higher. We

also find that all three meteorological databases are consistent to show that 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

245 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 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

250   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 does not account for the feedbacks from 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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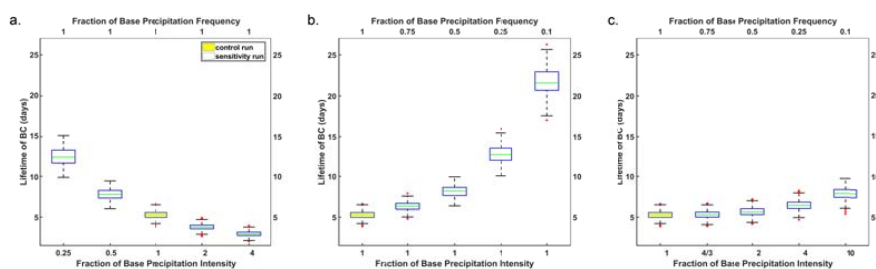

335

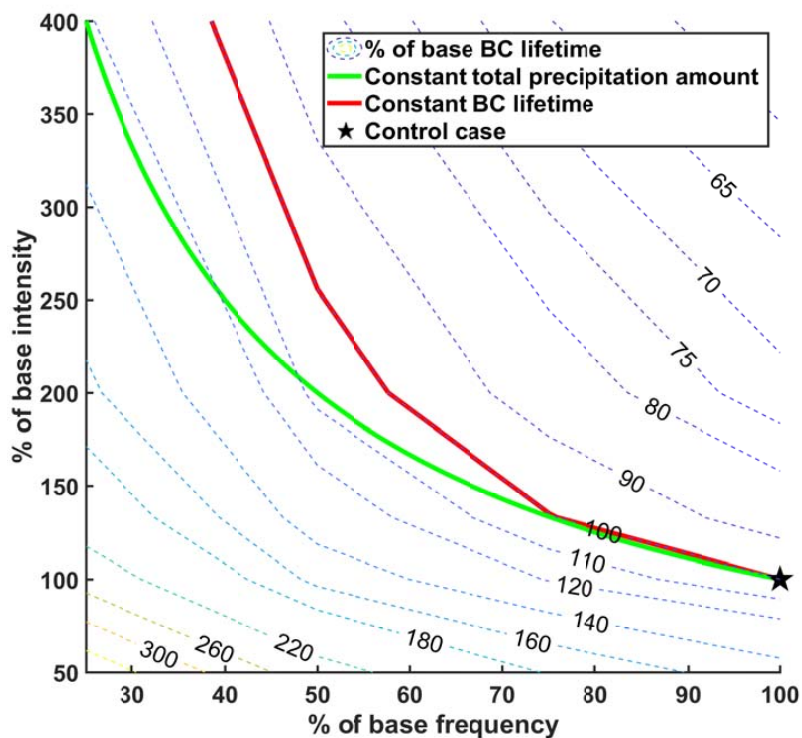
**Figure 1: Impacts of the precipitation characteristics on the atmospheric lifetime of BC under given a)
constant precipitation frequency; b) constant precipitation intensity; and c) constant precipitation amount.**
345   **The top x-axis reflects the precipitation frequency set in each perturbation test, shown as fractions of base
precipitation frequency. Base precipitation frequency is the precipitation frequency used in 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**
350   **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
355 **control run) driven by the changes of precipitation intensity and frequency. The green solid line represents a**
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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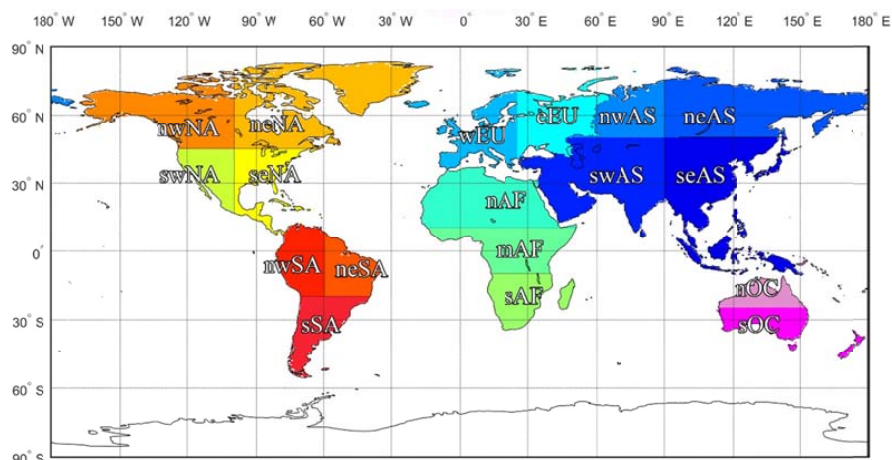
Figure 3: The definitions of the continental regions in this study. The uppercase letters in the region names

360 **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).
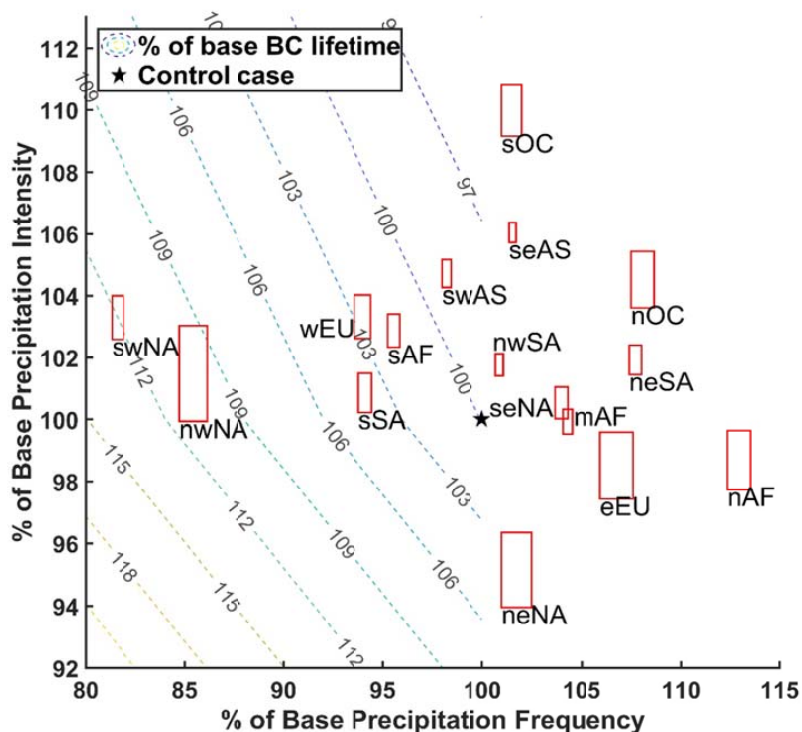
Figure 4: The potential change of atmospheric BC aerosol lifetime driven by the changes between the two periods (2008-2014 and 2001-2007) in precipitation characteristics based on meteorological datasets TRMM.
370 **The dashed contours are the same as in Fig. 2, which indicate the atmospheric lifetimes 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 changes.**
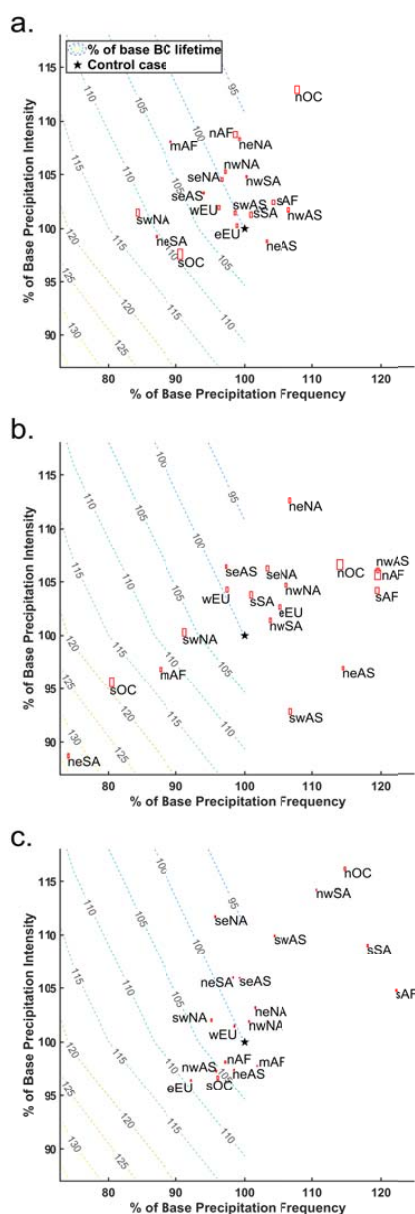
Figure 5: The potential change of atmospheric BC aerosol lifetime driven by the changes between the two periods (2001-2010 and 1981-1990) in precipitation characteristics based on multiple meteorological datasets: a). NCEP; b). NCEP2; c). MERRA. The dashed contours are the same as in Fig. 2, which indicate 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 changes.

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