We thank Dr. Jing Ming and the anonymous referee for their valuable comments to the manuscript and their constructive suggestions for improving the presentation quality. Below, we explain how the comments and suggestions are addressed (responses in blue color) and make note of the changes we have made to the discussion paper (in italic).

Referee #2 (Dr. Jing Ming)

1. The paper should not properly be entitled as "Carbonaceous aerosols recorded in a Southeastern Tibetan glacier..." for the main dataset used in this work has been pre-published by Xu et al. (2009) in PNAS and the primary aims are not to reintroduce the variations of carbonaceous aerosols recorded in the ice core. It is suggested to be changed to "Modelling of carbonaceous aerosols for their sources and forcing based on an ice core in the Zuoqiubu glacier".

Response: We agree with the referee that the ice core data set used in the present study has been originally published by Xu et al. (2009) to elucidate the important influences of black carbon on the melting of Tibetan glaciers. Although the long-term variations and seasonal dependence of BC and OC have been shown in the literature, contributions from specific source regions and potential changes in combustion sources have not been analyzed, and the increasing importance of OC-in-snow radiative forcing has not been raised. Undoubtedly these issues are also important to address.

In the present study, we calculated the source contributions to BC deposition in the southeastern Tibetan glacier using the state-of-the-science global aerosol-climate model (CAM5) with an explicit source tagging technique, which suggests a dominant contribution of South Asia emissions to the annual mean deposition of carbonaceous aerosols in the southeastern Tibetan Plateau. Moreover, we did further in-depth analysis to the changes in combustion sources based on the ice-core recorded temporal variations of OC to BC ratio, and revealed the increasing contributions of biomass burning and/or coal combustion versus oil to carbonaceous aerosols. In addition, the influence of carbonaceous particles on radiative fluxes at the glacier surface has been estimated using the SNICAR model.

Considering the referee's concern on the main focus of this study, we have changed the title to "Carbonaceous Aerosols Recorded in a Southeastern Tibetan Glacier: Analysis of Temporal Variations and Model Estimates of Sources and Radiative Forcing".

2. This work introduced a new concept, OC's forcing, which has not been widely recognized by the societies. The introduction of a new concept must be previously supported by measurement, as we all know. The OC's forcing is just like an aerolite in the whole paper. The authors should firstly list some literatures that clarified OC has the characteristic of radiation absorption in snow as well as it does in atmosphere claimed by Bond and Bergstrom (2006) and Kirchstetter et al. (2004). The online SNICAR model only simulates the reductions of snow albedo caused by BC and dust, but cannot have the ability to simulate the forcing of OC. The mass absorption cross-section of OC in the atmosphere cannot be directly used for it in snow.

Response: Following the referee's suggestion, we have added a paragraph to mainly introduce the role of OC acting as a light-absorbing impurity in snow/ice as follows:

BC is often the most important light-absorbing impurity in surface snow because of its strong absorption of solar radiation. Effect of BC in snow on surface albedo reduction and resultant positive radiative forcing have been widely addressed and reported (e.g., Warren and Wiscombe, 1980; Clarke and Noone, 1985; Hansen and Nazarenko, 2004; Hadley and Kirchstetter, 2012; Flanner et al., 2007; 2009; McConnell et al., 2007; Ming et al., 2008; Kaspari et al., 2011; Qian et al., 2011). In contrast, the impact of OC in snow has not been widely assessed because of its relatively weak light-absorption over the entire spectrum compared to BC, and because of large uncertainties associated with OC light-absorbing properties and measurements of OC in snow. However, there have been increasing interests in light-absorbing OC (a.k.a. brown carbon) and its radiative effect in the atmosphere (e.g., Kirchstetter et al., 2004; Andreae and Gelencsér, 2006; Hoffer et al., 2006; Moosmüller et al., 2009; Yang et al., 2009; Lack and Cappa, 2010; Cheng et al., 2011). Hoffer et al. (2006) estimated that humus-like substances as part of OC from biomass burning contribute ~7% to the absorption over the entire spectrum, which is not negligible. Yang et al. (2009) highlighted that as the contribution to absorption from BC decreases towards the ultra violet wavelengths, absorption due to brown carbon and dust becomes more significant, and they reported that at an observation site near Beijing brown carbon contributes over 10% to total absorption at mid-visible wavelengths. Thus the contribution of OC in snow to the surface albedo reduction is likely to be important, which has also been considered in recent climate

modeling studies (see Qian et al., 2014 for a review).

BC's atmospheric mass absorption cross-section (MAC) is generally adopted in radiative forcing estimation at snow surface. In the present study, we use BC and OC's atmospheric MAC with respective value of 7.5 m^2 g^{-1} and 0.06 m^2 g^{-1} at 550 nm to calculate the radiative forcing in snow. Although MAC of OC shows a different spectral dependence from that of BC (Barnard et al., 2008), for simplicity we use the same spectral dependence of MAC for OC in the SNICAR model calculation. Therefore, the absolute value of radiative forcing derived from OC may has larger uncertainties than that of BC, but the increasing trend in the contribution of OC since 1990 is robust. We have made a note of this in the manuscript as follows:

Note that the mass absorption cross-section of OC is highly spectral dependent (Kirchstetter et al., 2004; Hoffer et al., 2005; Barnard et al., 2008; Yang et al., 2009). It increases greatly towards shorter wavelengths. Consequently, the absorption of OC may be largely biased. It is also important to note that we didn't consider variations in chemical compounds of OC, or the changes of OC during sample filtration. Although the estimation of OC radiative forcing herein is rather crude, the increasing trend should be robust.

3. Source attribution of CAs using CAM5 model may be an innovative highlight of this work. However, the authors missed introducing many details, which may confuse readers. For example, in the method description, the authors did not introduce the initial weather field that drives the model including the meteorological parameters and their temporal and spatial resolution. The performance of the model in the highly elevated Tibet region is not well known. If my understanding is right, the authors used an inventory of CAs in 2000 to calculate the deposition of BC and OC on a large-scale region including the Zuoqiupu glacier. Does that mean the whole history of CAs depositing in the glacier can be retrieved through only one-year emissions, or just the one-year CAs' concentrations recorded in the ice core in 2000? However, the comparison between the results of measurements and modelling is missing in the paper, which might be extremely concerned by readers.

Response: We agree with the referee that source attribution of BC at this Southeastern Tibetan Plateau site using the CAM5 model is novel. Our original plan was to just use

this modeling tool to guide the more in-depth analysis of ice-core measurements for the contrast between monsoon and inter-monsoon means and the long-term trend. The evaluation of the model performance in simulating BC in the entire Tibetan Plateau and some sub-regions is documented in a separate paper. The referee's understanding is correct that BC deposited to a single model grid (about 200km by 200km, in which the Zuoqiupu glacier locates) is assumed to be uniformly distributed and compared to measurements. The referee also raised a good point on the BC emission inventory. The IPCC AR5 year-2000 emissions are supposed to be representative for the present-day global scenario (for one decade). However, uncertainties in emissions are very large, up to a factor of 2-4 for global mean, and could be even larger for some specific regions like South Asia and East Asia. This is part of the reason why we did not design the model experiment to simulate the whole historical record of BC in the ice core, but rather a 10-year period to demonstrate the impact of meteorology (and associated transport and removal) on the seasonal dependence of BC deposition in the target region and the lack of longer-term trend of deposition without considering the temporal variation of emissions. To address the referee's concerns, we have added more details about the method description and model configuration, and made note of the limitations of coarse grid global models in this kind of model-observation comparison.

4. The presentation of this paper is very difficult to understand. Sometimes I have to guess the meaning that the drafting author really meant to explain. I strongly suggest the authors would find some language specialists related to this study to improve the presentation largely.

Response: Thanks for the suggestion. The writing has been improved by native English speakers.

Minor comments (not including the language errors)

1. Line 2 in Page 19720. How "high" is the temporal resolution. . .? The resolution of the ice core record is not introduced in the paper. The word "high" here is some like a tree without a root.

Response: The annual accumulation of snow/ice was around 2 meters at the drill site, and the oxygen isotope samples were cut at 10 cm intervals, resulting in 18 ice samples per year on average. To reduce uncertainties in using the filter-based method, about 9 liquid samples per year were analyzed for carbonaceous aerosols. This is the way that we

defined it a "high" temporal resolution record. We have now made it clear in the manuscript as follows:

The annual accumulation of snow/ice at the drill site was around 2 meters on average. The oxygen isotope ($\delta^{18}O$) samples were cut at 10 cm internals, and BC and OC samples at 10-25 cm, resulting in 18 and 9 samples per year on average, respectively. Thus this ice core provided a high temporal-resolution of $\delta^{18}O$, and BC and OC concentrations.

2. Line 10 in Page 19720. ". . .followed by East Asia (14% and 21%, respectively)". I don't understand what "14% and 21%" mean.

Response: 14% and 21% are the contribution of East Asia emissions to the southeastern Tibetan Plateau during non-monsoon season and on an annual basis, respectively. This was written in the context of the sentences ahead of it. To avoid confusion, this sentence has been revised as follows:

The model results show that South Asia has the largest contribution to the present-day (1996-2005) mean BC deposition at the ice core drilling site during the non-monsoon season (October to May) (81%) and all year round (74%), followed by East Asia (14% to the non-monsoon mean and 21% to the annual mean).

3. Line 14 in Page 19720. Should point out that South Asia as a main contributor "in the annual mean", because in different seasons main contributor changes.

Response: The text has been revised.

4. Line 17 in Page 19720. Be careful to state the forcing of OC.

Response: Following the referee's comments, we have made some of the statements regarding the OC radiative forcing more precise in the main text.

5. Line 17 in Page 19720. ". . .and organic carbon (OC), which also absorbs in the near infrared, but more weakly than BC". Here there should be a reference.

Response: We have added some references on this in the main text.

6. Line 2 in Page 19721. "Jacobson, 2001" should be changed to a more representative literature, e.g., the most recently released IPCC report.

Response: The reference has been added.

7. Paragraph 2 in Page 19721. Ming et al. (2013) in Adv. Water. Resources suggest BC deposited in Himalayan and High Asian glaciers cannot significantly affect their energy balances, which is a very minority but different viewpoint from most literatures listed here, which should not be neglect here.

Response: Thanks for pointing this out. We have now made the point and added the reference.

8. Line 21 in Page 19721. There should be "burning" after "biomass".

Response: The text has been corrected.

9. Paragraph 1 in Section 3.1. When heavy pollution occurred in South Asia, the aerosol monitoring in the Tibetan hinterland can also detect the signal of high BC concentration (See Zhao et al., 2013, "Observation of carbonaceous aerosols during 2006-2009 in Nyainqentanglha Mountains and the implications for glaciers" in Environmental Science and Pollution Research).

Response: Thanks for pointing this out. We have noted this in the paper and added the reference.

10. Line 11 in Page 19725. The "sink" should be "deposition".

Response: The text has been revised.

11. Paragraph 1 in Page 19725. This paragraph should be moved to Section 2 (method).

Response: We have moved the first half of the paragraph, which describes the division of BC source regions, to Section 2. As a result of this change, the original Figure 2 and Figure 3 have to be switched. The second half of the paragraph describing the different circulation patterns during monsoon and non-monsoon months does not fit well in the methodology. We decide to keep it in original Section 3.2 where circulation patterns are analyzed for BC transport.

12. Line 28 in Page 19725. I don't understand the relationship between Wang et al. (2014) and this work. Obviously, Wang et al. did an Arctic work.

Response: Wang et al. (2014) was the first study that described the BC source tagging capability in the CAM5 model and applied this method to identifying sources of BC in the Arctic. The present study uses the same modeling tool and experimental setup as in Wang et al. (2014) but focuses on the Southern Tibetan Plateau. To avoid any confusion, we have removed the reference at this particular location.

13. Paragraph 1 in Page 19728. The sentence "More recent . . . South Asia" is confusing.

Response: We have rewritten the sentence as follows.

BC and OC emissions during 1996-2010 from Lu et al. (2011) are also illustrated in Figure 6 to extend the emission data to cover the entire time period that the ice core data span. We note that the emission data from Lu et al. (2011) are only for India, which is the largest energy consumer and carbonaceous aerosol-emitting country in South Asia.

14. Paragraph 1 in Page 19729. The OC/BC can be used to separate the dominant sources of CAs. It is my understanding that if the ratio is higher than 60, biomass burning should be the primary source of CAs. However, the neglected OC can influence the reality of the ratio and thus miss judging the burning sources, although it cannot alter the trend of OC/BC.

Response: We agree that higher OC/BC ratio indicates larger contribution of biomass burning to carbonaceous aerosols. We use the OC/BC ratio results from Cao et al. (2005) to quantitatively support the statement that biomass burning produces the highest OC/BC ratio, followed by coal combustion. However, the value of OC/BC ratio varies with fuel types and combustion processes, especially for biomass burning (Yanju Chen, 2014, Yu Zhao, 2014, personal communication). We also agree that the underestimation of OC in the ice samples is very likely to result in lower values of OC/BC than in the atmosphere but it is less likely to alter the trend of OC/BC ratio in the ice core record, and we have noted this in the paper.

15. Last sentence of paragraph 1 in Page 19729. Improved combustion technology not only reduce the emission of BC, but that of OC, which can result in the unclear varying of OC/BC.

Response: We agree, and the sentence has been rewritten as follows:

Improved combustion technologies may have reduced both BC and OC emissions from the combustion of the same amount of fuels, but the influence on OC/BC ratio is unclear. Presumably improved combustion technologies after 1990 in South and East Asia did not dominate the OC/BC ratio.

16. Line 7 in Page 19731. There should be "ng g^{-1}" after "4.4".

Response: Added.

17. Line 10 in Page 19731. The forcing of BC increases from 0.75 W m^{-2} in 1956-1979 to 1.95 W m^{-2} in 2006, which is comparable to the result of East Rongbuk glacier conducted by Ming et al. (2008) and believing to be true.

Response: Thanks for pointing this out. We have added a comparison in this regard, noting the agreement between our result and some others including Ming et al. (2008):

Our estimate of mean BC forcing is lower than the estimated Eurasian radiative forcing (2.7 W m^{-2}) in spring (Flanner et al., 2009), but it's comparable to that in the East Rongbuk glacier over Himalayas, which was in the range of 1-2 W m^{-2} (Ming et al., 2008). Kaspari et al. (2009) reported a three-fold increase in radiative forcing from BC in snow over Himalayas after 1975, which is consistent with the increasing trend in our results.

18. Summary and conclusions. This part should be shortened.

Response: We meant to include a summary of motivation, methodology, results and conclusions in this section, which is just a different style of presentation. The entire section is not too long, so we decide to keep it as it is.

19. Figure 3. The wind field in the surface and of 500 hPa in the Tibetan Plateau area is very doubtful. The mean elevation of the TP is as high as 4000 m above sea level. In meteorology, wind field in this area is usually blanked in these geopotential heights.

Response: The original Figure 3 (now Figure 2) illustrates the wind fields at 500 hPa (top panel), which is around 5500 m above sea level, and at the surface (bottom panel). As the areas in Tibetan Plateau having surface pressure lower than 500 hPa are very small and

scattered, the wind fields at 1.9° × 2.5° resolution (at which small-area high elevations are smoothed) would not be blank over the Tibetan Plateau. As for the surface winds, they always follow the terrain but could be at different elevations spatially depending on the topography.

Referee #3

General Remarks

1. The authors need to go through the paper to clarify the concepts of seasonal variation of BC and OC and the seasonal variation of the ratio of OC/BC. The two seasonal variations should be discussed separately owing to different reasons behind them. For example, the authors listed potential reasons on page 19725 lines 9-14 to explain the seasonal dependence of the relationship between OC and BC shown in Fig.2. However, the explanations help only to explain the seasonal variation of BC and OC, not the ratio of OC/BC.

Response: We totally agree with the referee that the two seasonal variations are very likely due to different reasons. We have followed the suggestion to discuss them separately, and significantly rewritten the relevant section.

2. In additional to the change of emission and atmospheric removal, the change of atmospheric circulation pattern during monsoon and non-monsoon seasons also contributes to the carbonaceous aerosols at studied site. It is good that the authors describe the atmospheric circulation pattern during two seasons on page 19726 lines 12-26. It would be even better if the authors could show the corresponding circulation patterns on a figure, such as Fig 3.

Response: Following the suggestion, we have added the corresponding circulation patterns on Figure 2 (the original Figure 3).

3. Fig. 5 indicates a rapid increase of the ratio of OC/BC in the Zuoqiupu ice core after 1990. However, this increase is not observed from the regional emissions on the figure compiled by Bond et al., 2007 and Lu et al., 2011. Therefore, the conclusion of "Because

of the stronger increasing trend in OC than BC during 1990-2006, the contribution of OC to the total radiative forcing cannot be neglected" (page 19731 lines 17-18) needs to be further checked to its regional representation.

Response: (We believe that this comment is about Figure 6 rather than Figure 5.) We agree that there is a discrepancy between the OC/BC ratio of deposition and the OC/BC ratio of emissions in terms of the increasing trend. Although South Asia has been identified as the primary contributor to the BC deposition at the sampling site, changes in total emissions in South Asia still cannot be directly translated to changes in the deposition. In the model simulation for source attribution and in the plot of emission trends (Figure 6), we used total emissions in the entire South Asia. However, we believe that spatial distributions within major sources regions, including South Asia and East Asia, can also significantly affect the deposition over the sampling site. On the other hand, there are many known and unknown uncertainties in the emission inventories for OC and BC, and the higher order quantity, OC/BC ratio, could be even more uncertain. Therefore, our conclusion in that regard is purely based on the ice core recorded trends of BC and OC deposition fluxes, which indicates an increasing contribution of OC to the carbon mass and radiative forcing. We made it clear in the paper that this conclusion is only for the glacier site rather than South Asia.

Specific Comments

1. Page 19721 lines 5-6: Is this correct for the sentence "During the cold and dry winter monsoon seasons. . ."? The monsoon season should be June-Sept as indicated in the abstract.

Response: We meant the South Asian winter monsoon as opposed to the summer monsoon, but we agree that it is a little confusing to use both of them in the same paper. We have removed "monsoon" here.

2. Page 19721 line 23-24: The emissions of biofuel consumption and biomass burning are typically categorized separately.

Response: According to the referee's comments, we have revised the text as follows:

However, as reliable biomass consumption data are hard to obtain, estimates of BC and OC emissions from biomass burning are ambiguous and incomplete.

3. Page 19722 line 11 and 14: It would be better to use "source types" instead of "combustion sources" here.

Response: The text has been revised.

4. Page 19722 line 15: Is it robust to estimate regional RF impact with only Zuoqiupu glacier ice core data?

Response: We totally understand the referee's concern here. BC and OC concentrations and, therefore, the resultant radiative forcing vary spatially and temporally, especially at places having sharp changes in elevation and/or in close proximity of emission sources. Without doing a comprehensive survey over many more sites, we cannot conclude on how large area the Zuoqiupu glacier ice-core data can represent. Nonetheless, according to a comment by the other referee, the magnitude of our BC forcing estimates is very close to that by Ming et al. (2008) for a different glacier. The magnitude of BC-in-snow forcing is also comparable to the springtime Eurasian forcing estimated by Flanner et al. (2009). Meanwhile, a similar increasing trend of radiative forcing from BC in snow was shown in Himalayas region by Kaspari et al. (2009). We have added such comparisons with these previous studies to the paper, and made a note in the paper saying that the estimated forcing is only for the Zuoqiupu glacier. Please also refer to our response to the comment (#17) by the other referee.

5. Page 19723 lines 6-7: Are there any differences between EC and BC in terms of their concentrations? Do the authors need to convert the measured EC to BC concentration in order to compare with model simulation?

Response: Measurement data on EC and BC concentrations are method specific. EC represents thermally refractory carbon in the sample, while BC denotes the extent of light-absorption by the sample as measured using optical methods. Thus they can have large differences, for example, when the sample contains other light-absorbing material that is likely to be attributed to BC. This is not an issue in model simulations where BC is

actually defined more like the measured EC. We didn't need to convert the measured EC to BC, and we use the term "BC" to be consistent with that in the model and in most previous studies.

6. Page 19723 line 8-9: Why can the ice-core measurements only account for the water-insoluble part of OC from aerosol emissions?

Response: This is because the filter-based method we used for sample analysis cannot capture OC dissolved in the liquid samples (after the ice samples melt). Most of water-soluble OC can move through the filter. We have noted this more clearly in the paper.

7. Page 19723 lines 23-25: Could you elaborate on the approach of "offline mode"? Do you mean the simulation used an approach typically used for chemistry transport model (CTM)? How does this approach provide dynamic feedback between cloud/precipitation and aerosol?

Response: This approach is similar to that typically used for chemical transport models (CTMs) in the sense that model meteorological fields (e.g., winds, temperature and pressure) are constrained with reanalysis products, which are supposed to be closer to reality. Therefore, there is no need to consider dynamical feedback between cloud/precipitation and aerosol. However, the microphysical interactions (e.g., impact of aerosol on cloud/precipitation processes, and the feedback from cloud/precipitation on aerosol wet scavenging and removal) are predicted in our model with more sophisticated schemes than in many CTMs. We believe this approach is more suitable for the present study than the conventional free-running approach.

8. Page 19725 lines 2-3: The numbers in text are opposite of the numbers shown in Fig. 2.

Response: Thanks for catching this mistake. It has been corrected in the text.

9. Page 19725 lines 9-11: Other potential reasons, such as the change of transport path and strength, may also have an impact.

Response: We agree, and this has been added to the text. We also added more discussions on the potential reasons, in response to the general comment #1.

10. Page 19726 lines 6-9: This could be simply because the time span of non-monsoon season (Oct-May) is much longer than that of monsoon season (Jun-Sept).

Response: The BC deposition fluxes in Figure 5 are seasonal averages (i.e., mass per unit area and time), not accumulations during the time periods. The seasonal variation of BC and OC deposition fluxes was calculated in a similar way. Both model results and measurements show higher deposition in non-monsoon season than in monsoon season.

11. Page 19727 lines 20-21: Why did BC and OC fluxes drop between mid-1990 to 2002?

Response: This is an interesting point, but we don't have a plausible explanation for it. It was likely due to the decreased energy consumptions in source regions. As shown in Figure 6, the coal consumption, which was the most important fuel in South Asia, stayed at a relative stable level during this period. The correlation between coal consumption in the primary source region and the response in BC and OC deposition fluxes at the sampling site could potentially indicate a causal connection, but it may also possibly be a coincidence. After all, changes in the spatial distributions of emissions in the major source regions could also affect the source-receptor relationship.

12. Page 19728 lines 14-15: What about after 2003 during which the OC/BC ratio in ice core still increased but the OC/BC emission ratio decreased as shown in Lu et al., 2011?

Response: Please also refer to our response to the general comment (#3) on this. There are many potential reasons for this discrepancy, among which we believe uncertainties in the total emissions and the spatial distribution of emissions within the major source regions likely play a key role.

13. Page 19729 lines 7-8: Why would including water-soluble OC lead to increased bias?

Response: As the filter-based method used in the analysis of liquid samples (melted from ice samples) cannot capture water-soluble OC, the OC herein has been underestimated. As a result, the OC/BC ratio was very likely underestimated, which is what we meant by the "low" bias. To avoid such confusion, we have changed the phrase "low bias" to "been underestimated".

14. Page 19729 lines 11-14: Is it possible that there is a positive trend of influence from South Asia?

Response: The model simulation with fixed emissions but varying meteorological conditions shows that meteorology alone (and associated transport and removal processes) didn't cause a discernable trend during the 10 years (1996-2005) in BC deposition (Figure 5) and OC/BC ratio (figure not shown). Thus the increasing trend of BC and OC fluxes is very likely due to changes in emissions from major source regions, which is consistent with the trends in South Asia emissions. Along the same line of reasoning, OC/BC ratio is likely due to the relatively more contribution of coal and/or biomass than oil to the total emissions from South Asia. The evidence seems to support a positive trend of influence from South Asia, but a concrete conclusion cannot be drawn because of the disagreement with OC/BC ratio of emissions.

15. Page 19729 line20 (also see Figure 6): According to the authors' explanation in section 3.4 first paragraph, the more coal combustion relative to oil consumption, the higher OC/BC would be. However, Fig. 6 shows that despite the increase of coal combustion being much slower than the increase of oil consumption during 1996-2002, corresponding OC/BC increased dramatically.

Response: First of all we note that we made a mistake in the units of oil consumption in the original Figure 6 (lower-right panel), which were supposed to be in "thousand barrels daily". We have now revised the plot using same units of "million tonnes" for both oil consumption data and coal consumption data. This change however does not affect any of our interpretation of the results.

The increasing trend of OC/BC ratio in Figure 6 could be due to an increased contribution of coal and/or biomass compared to oil in terms of source type, as emissions from coal and biomass combustion both produce higher OC/BC ratio than from oil (Cao et al., 2005). IEA (2009) reported that coal, biomass, and oil accounted for 41%, 27%, and 24%, respectively, of the primary energy demand in 2007 in India. Although the consumption of biomass was lower than coal, the OC/BC ratio for biomass burning emissions is much higher than from coal burning (60.3 vs. 12.0). BC emission factor is also higher for biomass burning (varying from 0.48 ± 0.18 g kg^{-1} for savanna and grassland burning to 1.5 g kg^{-1} for charcoal burning) than coal (0.2 g kg^{-1} for most combustion conditions) and oil combustions (0.3 g kg^{-1} on average, varying from 0.08 g kg^{-1} for heavy fuel oil to 0.66 g kg^{-1} for diesel) (Andreae and Merlet, 2001; Bond et al., 2004, 2007). The consumption of biomass, which is also mainly influenced by population, has potentially increased with the accelerated growth of population in South Asia. Thus, although the increase of coal combustion was slower than oil consumption during 1996-2002, the expansion of biomass usage may have determined the increase of OC/BC ratio. Similar points have been discussed in the third paragraph of section 3.4, and in addition, we made note of some of the above discussions.

16. Page 19730 lines 1-15: How does this knowledge help to interpret the measured and simulated data?

Response: As mentioned in the response to the comment (#15) above, this paragraph is supposed to address the role of biomass burning as an energy resource in South Asia. The possible growing emissions from biomass burning may have been another important contributor to the increasing trend of OC/BC ratio, but we don't have the long-term consumption data like those of coal and oil.

17. Page 19730 lines 26-27: Could the authors explain what the MAC scaling factor within spectral broadband is and how to get these factors for BC and OC?

Response: The mass absorption cross-section (MAC) is a wavelength-dependent parameter that is generally adopted to derive optical properties of snow impurities and calculate their radiative forcing in snow. In the online SNICAR model

(http://snow.engin.umich.edu), MAC is normalized within spectral broadband with a mass absorption cross-section of 7.5 m^2 g^{-1} at 550 nm for BC. There is another input parameter, called the MAC scaling factor, which can be used to adjust/rescale the wavelength-dependent MAC for BC in each broadband. A MAC of 7.5 m^2 g^{-1} at 550 nm is commonly used for uncoated BC particles (Bond and Bergstrom, 2006; Flanner et al., 2007), so we use the MAC scaling factor of 1 (i.e., 7.5/7.5). For OC, we adopted the MAC value of 0.6 m^2 g^{-1} at 550 nm that is typical for light-absorbing OC measured in Asia and from biomass burning emissions (e.g., Kirchstetter et al., 2004; Yang et al., 2009). The MAC of OC does show different spectral dependence from that of BC (Barnard et al., 2008), but for simplicity we just use the same spectral dependence of MAC for OC in the SNICAR model calculation, which makes the MAC scaling factor to be 0.08 (i.e., 0.6/7.5). We made a note in the paper that with such assumptions the estimated magnitude of OC radiative forcing may have large uncertainties but the trend in OC forcing is more robust.

18. Page 19732 line 6-7: This site is located in complex terrain, affected by both South Asia and East Asia. There may be a need for further investigation of its regional representation in terms of concentration and deposition before using the ice core measured carbonaceous aerosols to infer regional radiative forcing.

Response: We agree that further investigation is needed to understand the spatial variations of BC and OC concentrations in snow/ice and their influences on the regional radiative forcing. Here we have made it clear that the estimated radiative forcing is only for the Zuoqiupu glacier.

19. Page 19732 lines 14-15: Again, we cannot exclude changes in atmospheric transport path and strength as potential reasons.

Response: Agreed and the text has been revised accordingly.

20. Table 1: Adding BC emissions over these regions would help to explain the impact per unit emission in source regions.

Response: Following the referee's suggestion, BC emissions over the four source regions have been added to Table 1.

21. Technique corrections:

Page 19723 lines 18-19: "wet scavenging" and "removal by precipitation" are redundant.

Response: They are different processes (i.e., nucleation scavenging and wet deposition) in the model, but for simplicity in the description we have changed to "wet removal by precipitation".

Page 19731 lines 5-6: This repeats page 19730 lines 20-23.

Response: Following the referee's suggestion, we have deleted the repeated sentence.

Figure 2: Add the time period of the ice core measurements.

Response: The time period has been added in the caption of Figure 2.

Figures 3 and 4: Change location of ice core from color pink to black.

Response: The color has been changed in revised figures.

Figure 6: The dashed lines on subfigures are extended beyond 1979.

Response: The dashed lines are meant to be reference lines to show the increasing trend of OC/BC ratio, BC, and OC after 1980.

References:

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Carbonaceous Aerosols Recorded in a Southeastern Tibetan Glacier: Analysis of Temporal Variations and Model Estimates of Sources and Radiative Forcing

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Abstract. High temporal resolution measurements of black carbon (BC) and organic carbon (OC) covering the time period of 1956-2006 in an ice core over the southeastern Tibetan Plateau show a distinct seasonal dependence of BC and OC with higher respective concentrations but lower OC/BC ratio in the non-monsoon season than during the summer monsoon. We use a global aerosol-climate model, in which BC emitted from different source regions can be explicitly tracked, to quantify BC source-receptor relationships between four Asian source regions and the southeastern Tibetan Plateau as a receptor. The model results show that South Asia has the largest contribution to the present-day (1996-2005) mean BC deposition at the ice core drilling site during the non-monsoon season (October to May) (81%) and all year round (74%), followed by East Asia (14% to the non-monsoon mean and 21% to the annual mean). The ice-core record also indicates stable and relatively low BC and OC deposition fluxes from late 1950s to 1980, followed by an overall increase to recent years. This trend is consistent with the BC and OC emission inventories and the fuel consumption of South Asia (as the primary contributor to annual mean BC deposition). Moreover, the increasing trend of OC/BC ratio since the early 1990s indicates a growing contribution of coal combustion and/or biomass burning to the emissions. The estimated radiative forcing induced by BC and OC impurities in snow has increased since 1980, suggesting an increasing potential influence of carbonaceous aerosols on the Tibetan glacier melting and the availability of water resources in the surrounding regions. Our study indicates that more attention to OC is merited because of its non-negligible light absorption and the recent rapid increases evident in the ice core record.

Keywords

Carbonaceous aerosol, Tibetan glacier, Emissions, Radiative forcing

1. Introduction

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Carbonaceous aerosol, released from fossil fuel, biofuel and/or biomass combustion, contains both black carbon (BC, a.k.a. elemental carbon, EC), a strong light absorber, and organic carbon (OC), which also absorbs in the near infrared, but more weakly than BC (Kirchstetter et al., 2004; Bond et al., 2006). Often mixed with other aerosol species, BC impacts human health, crop yields and regional climate (Auffhammer et al., 2006; Tie et al., 2009), and is believed to be the second strongest climate warming forcing agent after carbon dioxide (Jacobson, 2001; IPCC, 2013).

Because of their high population density and relatively low combustion efficiency, developing countries in South and East Asia such as India and China are hotspots of carbonaceous aerosol emissions (Ramanathan and Carmichael, 2008). During the cold and dry winter season, haze (heavily loaded with carbonaceous aerosols) builds up over South Asia, and exerts profound influences on regional radiative forcing (Ramanathan et al., 2007; Ramanathan and Carmichael, 2008), hydrologic cycles (Menon et al., 2002; Ramanathan et al., 2005), and likely Himalaya-Tibetan glacier melting that could be accelerated by the absorption of sunlight induced by BC in the air and deposited on the ice and snow surfaces (Ramanathan et al., 2007; Hansen and Nazarenko, 2004; Ming et al., 2013).

Due to the lack of long-term observations of emissions and concentrations of atmospheric carbonaceous aerosols, it is difficult to evaluate the effects of BC and OC on historical regional climate and environment before the satellite era. Some studies have evaluated historical anthropogenic emissions based on the consumption of fossil fuels and biofuels (Novakov et al., 2003; Ito and Penner, 2005; Bond et al., 2007; Fernandes et al., 2007). While fossil fuel is the major energy source in the urban areas of South Asia and East Asia, biomass combustion, such as fuel wood, agricultural residue and dung cake, is prevalent in rural areas (Revelle, 1976; Venkataraman et al., 2010; Street and Waldhoff, 1998). Biomass burning has been considered as the major source of black carbon emissions (Reddy and Venkataraman,

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2002; Venkataraman et al., 2005). However, as reliable biomass consumption data are hard to obtain, estimates of BC and OC emissions from biomass burning are ambiguous and incomplete.

Measurements of carbonaceous aerosol concentrations in glacier ice are an ideal means to reconstruct historical emissions and reveal long-term trends of anthropogenic aerosol impacts on local climate. Greenland ice core measurements were previously used to reconstruct the North American BC emission history and its effects on surface radiative forcing back to the 1880s (McConnell et al., 2007). Himalayan ice cores retrieved from the Tibetan Plateau have revealed the mixed historical emissions from South Asia, Central Asia and the Middle East and also been used to evaluate radiative forcing from BC in snow (Ming et al., 2008; Kaspari et al., 2011). Using the Snow, Ice, and Aerosol Radiative (SNICAR) model, Flanner et al. (2007) estimated an instantaneous regional forcing of exceeding 20 W m^{-2} by BC in snow/glaciers over the Tibetan Plateau during the spring season.

By using five ice core records, Xu et al. (2009a) elucidated an important contribution of BC to the retreat of Tibetan glaciers in addition to greenhouse gases. Due to the short atmospheric lifetime of carbonaceous aerosols compared to greenhouse gases, emission reductions may be an effective way to mitigate their warming effects. Thus it is particularly important to identify the source regions and the source types of carbonaceous aerosols observed in Tibetan glaciers. Xu et al. (2009a) suggested that BC deposited on Tibetan Plateau was broadly from Europe and Asia. However, they didn't perform in-depth analysis on emissions from more specific source regions and the source types. In this study, we use the ice core retrieved from the southeastern Tibetan Plateau, also known as the Zuoqiupu ice core in Xu et al. (2009a), to reconstruct the history of atmospheric deposition of carbonaceous aerosols in this glacier, and to characterize emissions and source-receptor relationships with the help of a global climate model in which BC emitted from different source regions can be explicitly tracked. We also estimate the

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respective contributions from BC and OC to radiative forcing in the Zuoqiupu glacier using the ice core measurements and the SNICAR model.

2. Methods

2.1 Measurements of carbonaceous aerosols in ice core

Zuoqiupu glacier is in the southeastern Kangri Karpo Mountains, located at the southeastern margin of the Tibetan Plateau (Figure 1). In 2007, an ice core of 97 meters in depth (9.5 cm in diameter) was retrieved within the accumulation zone of Zuoqiupu glacier at 96.92°E, 29.21°N, 5600 m a.s.l. The ice core was kept frozen and transported to laboratory facilities at the Institute of Tibetan Plateau Research (Lhasa branch) for analysis. The annual accumulation of snow/ice at the drill site was around 2 meters on average. The oxygen isotope ($\delta^{18}O$) samples were cut at 10 cm internals, and BC and OC samples at 10-25 cm, resulting in 18 and 9 samples per year on average, respectively. Thus this ice core provided a high temporal-resolution of $\delta^{18}O$, and BC and OC concentrations. BC and OC concentrations were measured by using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer following the IMPROVE TOR protocol (Chow et al. 1993; Chow and Watson 2002; Cao et al. 2008). Note that according to the thermal/optical measurement method, the analytical result is technically called "EC". Herein we use "BC" to be consistent with the notation in our model simulations and in the literature. The reported OC concentrations from the ice-core measurements can only account for water-insoluble part of OC in the ice samples because most of the water-soluble part cannot be captured by the filter-based method applied to liquid samples (melted from the ice). Further details on the analysis methods, ice core dating and calculation of BC and OC seasonal deposition fluxes can be found in Xu et al. (2009a).

2.2 Model and experimental setup

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We use the Community Atmosphere Model version 5 (CAM5; Neale et al., 2012) to help understand the emissions, transport and dry/wet deposition of carbonaceous aerosols in the atmosphere. In the default aerosol scheme of CAM5, BC and primary OC are emitted into an accumulation size mode, where they immediately mix with co-existing hygroscopic species such as sulfate and sea salt (Liu et al., 2012). Hygroscopic aerosol particles in the accumulation mode are subject to wet removal by precipitation. Recent model improvements to the representation of aerosol transport and wet removal in CAM5 by Wang et al. (2013) have substantially improved the global distribution of aerosols, particularly, over remote regions away from major sources. To minimize the model biases in simulating meteorological conditions and, particularly, circulations that are critical to aerosol transport, we configure the CAM5 model to run in an offline mode (Ma et al., 2013) with wind, temperature, surface fluxes and pressure fields constrained by observations. However, cloud/precipitation fields and interactions between aerosol and clouds are allowed to evolve freely. A source tagging technique has been recently implemented in the CAM5 model to allow for explicitly tracking aerosols emitted from individual source regions and, therefore, assists in quantitatively characterizing source-receptor relationships (Wang et al., 2014). This tagging technique along with the CAM5 model is used in the present study to do source attribution for carbonaceous aerosols deposited to the Zuoqiupu glacier.

We conducted an 11-year (1995-2005) CAM5 simulation at horizontal grid spacing of 1.9° × 2.5° and 56 vertical levels, with prescribed sea surface temperatures and sea ice distribution. Reanalysis products from NASA Modern Era Retrospective-Analysis for Research and Applications (MERRA) (Rienecker et al., 2011) are used to constrain the meteorological fields of CAM5. For aerosols (including OC, BC and other important species), we use the year-2000 monthly mean emissions described by Lamarque et al. (2010) that have been used in many global climate models for present-day climate simulations, included in the fifth

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assessment report (AR5) by the Intergovernmental Panel on Climate Change (IPCC). The monthly mean emissions are repeatedly used for each year in the 11-year simulation. Note that we do not intend to design the model experiment to simulate the whole historical record of BC in the ice core, but rather for a period of time to demonstrate the impact of meteorology (and associated transport and removal of aerosols) on the seasonal dependence of BC deposition in the target region and the lack of longer-term trend in deposition without considering the temporal variation of emissions.

As the ice core drill site was located at a remote and elevated area over the southeastern Tibetan Plateau, where local emissions are minimal. Deposition of carbonaceous aerosols is most likely contributed by the non-local major emission sources (e.g., distributions of mean BC emissions during non-monsoon and monsoon seasons shown in Figure 2) in South Asia and East Asia. These two regions, along with Southeast Asia and Central Asia, are identified as the potential source contributors. Thus BC emissions from the four regions and the rest of the world are explicitly tracked in the CAM5 simulation.

3. Results and Discussion

3.1 Seasonal dependence of carbonaceous aerosols

BC and OC concentrations in the Zuoqiupu ice core both exhibit statistically significant seasonal variations at the 0.05 level corresponding to the stable oxygen isotope variability, which shows high values during the winter and low values during the summer (Xu et al., 2009a). As shown in Figure 3, concentrations of BC and OC have distinct differences between the summer monsoon and non-monsoon seasons. Seasonally varying emissions and meteorological conditions that determine the transport pathways of BC and OC emitted from major sources, removal during the transport, and local precipitation rate can cause the seasonal variations of BC and

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OC in ice at the sampling site. The seasonal dependence of BC and OC in ice core is consistent with available observations of atmospheric aerosols in the south slope of the Himalayas and the southeastern Tibetan Plateau, where the high concentration of carbonaceous aerosols during the cold and dry season was suggested to associate with the South Asian haze (Cong et al., 2009; Marinoni et al., 2010; Kaspari et al., 2011; Zhao et al., 2013a; Zhao et al., 2013b). The consistency between the seasonal dependence of airborne BC and OC concentrations and the seasonal variation of ice-core measurements indicates that seasonal differences in local precipitation rate is less likely to be the determining factor. Our model results (details discussed in the section 3.2) suggest that the seasonal dependence of BC deposition flux in the target region could be mainly due to meteorological conditions (and associated transport pathways and wet removal processes). The small seasonal contrasts in BC emissions from the major source regions (see Table 1) that are used in the model simulation do not seem to be able to explain the large seasonal difference in BC deposition, although the BC emissions are known to have large uncertainties.

Our further analysis shows that the ratio of OC to BC also has clear seasonal dependence. In Figure 3, the slope of the fitted line to measured OC versus BC concentrations during monsoon season is ~6.3, which is twice the slope for non-monsoon season (~3.2). The analysis of covariance (ANCOVA) for slope differences of single linear regressions of OC against BC between monsoon and non-monsoon seasons indicates that the seasonal dependence of the relationship between the concentrations of OC and BC is significant (at the 0.05 significance level). This also agrees with measurements derived from the ice core drilled from the Palong-Zanbu No. 4 Glacier (Xu et al., 2009b) and in atmospheric samples collected from Lulang, southeastern Tibetan Plateau (Zhao et al., 2013b). The seasonal dependence of the OC/BC ratio can possibly be derived from the seasonal sources of carbonaceous particles, circulation strength, transport pathways, and/or atmospheric deposition processes. Compared to the respective BC and OC

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concentrations, the seasonal dependence of OC/BC ratio is less straightforward to understand. Circulation patterns together with wet removal processes still determine the transport pathways of emissions from major BC and OC source regions to the sampling site, which however are less likely to change OC/BC ratio from certain sources. Therefore, it is more plausible due to seasonally dependent contributions from source regions and/or emission sectors (including fuel types, quantity, and combustion conditions). Cao et al. (2005) found that the average OC/BC ratios measured from plumes of residential biomass burning and coal combustion are substantially higher than from vehicle exhaust. Higher OC/BC ratio during summer monsoon might indicate more contributions from biomass and/or coal burning than fossil fuel combustion.

3.2 Source attribution

To quantitatively attribute the source of BC at the drilling site (as a receptor region), we use the CAM5 model with the BC source tagging capability to conduct an 11-year simulation, with the last 10 years (1996-2005) used for analysis. The surrounding area is divided into four source regions (see Table 1 and Figure 4): South Asia, East Asia, Southeast Asia and Central Asia. BC emissions from each of the four regions and the rest of the world are explicitly tracked, so that the fractional contributions by emissions from the individual source regions to BC deposition at the receptor region can be explicitly calculated. Figure 4 shows the spatial distribution of fractional contribution from the four source regions. BC deposition at the drilling site (indicated by the black box in Figure 4), which has a consistent seasonal dependence (i.e., more during the non-monsoon season; Figure 5) with ice core measurements, is predominately (over 95%) from South Asia and East Asia. The seasonal dependence of BC deposition is also consistent with a recent regional climate modeling study on BC deposition on the Himalayan snow cover from 1998 to 2008 (Ménégoz et al., 2014).

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The 10-year (1996-2005) average wind fields (at the surface and 500 hPa from MERRA reanalysis datasets), as shown in Figure 2, indicate distinct circulation patterns during summer monsoon (June-September) and non-monsoon (October-May) season, which in part determine the seasonal dependence of transport of aerosols emitted from the different major sources. During the non-monsoon season, strong westerly dominates the transport from west to east at all levels. Emissions from northern India and central Asia can have influence on BC in the direct downwind receptor region over southeastern Tibetan Plateau. During the summer monsoon season, the westerly moves northward and the monsoon flow from Bay of Bengal at the surface and middle levels (e.g., 500hPa), coupled with the monsoon from Indochina peninsula and South China Sea, exert influence on BC in the receptor area. The strong monsoon precipitation removes BC from the atmosphere during the transport. The high Himalayas can partly block the further transport of emissions from South Asia to Tibetan Plateau, although small local topographical features such as the Yarlung Tsangpo River valley can provide a gate for the pollution to enter the inner Tibetan Plateau (Cao et al., 2010). Elevated emissions from the west (or northern part of South Asia) can take the pathways at middle and upper levels but they have minimal contribution to deposition. Therefore, BC emissions from East Asia play a relatively more important role affecting deposition at the Zuoqiupu site during the monsoon season.

The fractional contributions to 10-year mean BC deposition at the drilling site from the four tagged regions are summarized in Table 1. Results show that South Asia is the dominant contributor (~81%) during the non-monsoon season with ~14% from East Asia, while the contribution of East Asia (~56%) is larger than that of South Asia (~39%) during the monsoon season. For the annual mean BC deposition, South Asia (~75%) is the biggest contributor, followed by East Asia (~21%). Emissions from the central Asia and Southeast Asia regions have much smaller contributions (<3%) for all seasons. These results agree well with the short-term

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source attribution study by Lu et al. (2012) using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model.

For comparison, seasonal and annual mean BC emissions from the individual tagged source regions are also included in Table 1. Apparently, the contrast in strengths of regional emissions alone cannot explain their relative contributions to BC deposition at the sampling site, and the small seasonal variations in emissions are unlikely the cause of seasonal dependence of source attribution. Note that the BC emission inventory (Lamarque et al., 2010) used in CAM5 doesn't consider seasonal variations in anthropogenic emissions, which is likely to have introduced biases in the quantitative model estimates of seasonal dependence of contributions, but the relative importance of source regions should be robust.

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3.3 Interannual variations and long-term trend

Based on annual snow accumulation and BC and OC concentrations derived from the ice record, the annual BC and OC deposition fluxes can be estimated, which are then used to examine the interannual variations and long-term trend in the fluxes and the ratio of OC/BC, as well as the relationship with emissions from the major contributor. As illustrated in Figure 6, from late 1950s to 1980, the BC and OC fluxes in the Zuoqiupu ice core are relatively low and stable in comparison to those after 1980. During the period 1956 to 1979, average fluxes are 9.1 and 28.7 mg m^{-2} a^{-1} for BC and OC, respectively. Both BC and OC fluxes began to show increasing trends from early 1980s. These trends continued in the early 1990s but started to drop in the mid-1990s, reaching a minimum in 2002 followed by a rapid increase. In 2006, BC and OC fluxes are 19.2 and 93.9 mg m^{-2} a^{-1}, respectively, which are two and three times the respective average fluxes before 1980. The five-year average OC/BC flux ratio is steady before 1990; however, it shows a continual increase afterwards and has been higher than the average value (3.2) for the period of 1956-1979 since mid-1990s (Figure 6). The 10-year CAM5 model

simulation, in which annual emissions are fixed but meteorological conditions vary, shows no increasing trend in BC and OC deposition fluxes (BC deposition shown in Figure 5), indicating that the increasing trend seen in the observations was not due to changes in meteorology.

As shown in the CAM5 model simulation, the annual mean atmospheric deposition of BC over southeastern Tibetan Plateau is mostly contributed by emissions from South Asia, particularly, in the non-monsoon season. The BC and OC deposition fluxes derived from the ice-core measurements may reflect changes in South Asian emissions to some extent. The temporal variations of BC and OC deposition fluxes (see Figure 6) are compared with the primary BC and OC emissions from fossil fuel and biofuel combustion in South Asia during 1955-2000 (Bond et al., 2007). BC and OC emissions during 1996-2010 from Lu et al. (2011) are also illustrated in Figure 6 to extend the emission data to cover the entire time period that the ice core data span. Note that the emission data from Lu et al. (2011) are only for India, which is the largest energy consumer and carbonaceous aerosol-emitting country in South Asia. There are differences between the emissions of Bond et al. and Lu et al. during the overlap time period (1996-2000). However, good agreements on the increasing trend can be found in the respective deposition fluxes and emissions of BC and OC (Figure 6). The OC/BC emission ratio also shows an increasing trend from the late 1990s to 2003, which is consistent with that of OC/BC ratio in the ice core record. The annual mean aerosol index over industrial and populated cities in the northern part of India increased from 1982-1993 and more significantly from 2000-2003 (Sarka et al., 2006). This trend is similar to that of carbonaceous aerosols in the ice core record, and it might indicate a causal relationship between BC and OC over southeastern Tibetan Plateau and emissions from north part of South Asia.

3.4 Emission source analyses

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BC and OC in the atmosphere are co-emitted from a variety of natural and anthropogenic sources, including combustion of fossil fuel, biofuel and/or biomass burning. In general, open biomass burning typically produces more abundant OC (i.e., larger OC/BC ratio) compared to fossil fuel combustion due to a lower process temperature (Ducret and Cachier, 1992). The OC/BC ratio has often been used to discriminate fossil fuel combustion and biomass burning emissions in the atmosphere and in precipitation (Novakov et al., 2000; Stone et al., 2007; Ducret and Cachier, 1992; Xu et al., 2009b). For example, Cao et al. (2005) collected particulate matter samples from the plumes of residential biomass burning, coal combustion, and motor-vehicle exhaust sources, and analyzed OC and BC with DRI Thermal/Optical Carbon Analyzer (Model 2001). They reported average OC/BC ratios of 60.3, 12.0, and 4.1 for biomass burning, coal-combustion and vehicle exhaust, respectively. The increasing OC/BC ratios based on the ice core measurements since the early 1990s (Figure 6) suggest an expanded coal consumption and/or usage of biomass fuel, although the ratios might have been underestimated because water-soluble OC was not captured in the sample analyses. However, such bias would have occurred to all the samples and had little impact on the trend, unless including water-soluble OC could dominate the temporal variation of OC/BC ratio. Otherwise, our results indicate that the relative contribution of coal combustion and biomass burning to the carbonaceous particles deposited into the ice core in southeastern Tibetan Plateau has been increasing faster than the contribution of fossil fuel combustion since early 1990s. Improved combustion technologies may have reduced both BC and OC emissions from the combustion of the same amount of fuels, but the influence on OC/BC ratio is unclear. Presumably improved combustion technologies after 1990 in South and East Asia did not dominate the OC/BC ratio.

The temporal variations of BC and OC in the Zuoqiupu ice core, along with the source attribution analysis of the CAM5 model results, suggest an increasing trend

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in emissions and altered emission sources in South Asia during the late 20th century. Coal has been the primary energy source in South Asia. For example, in India coal accounted for 41% of the total primary energy demand in 2007, followed by biomass (27%) and oil (24%) (IEA, 2009). The consumption data of coal and crude oil in South Asia (BP Group, 2009) is compared with the BC and OC fluxes in Figure 6 (bottom right). Coal consumption had an increasing trend from 1965 to 2008, particularly in the two time periods 1980-1995 and 2003-2008 after a level off during 1996-2002. This trend is consistent with the variations of BC and OC deposition fluxes in the Zuoqiupu ice core. The correlations between coal consumption and BC ($R^2 = 0.43$, $p < 0.001$) and OC ($R^2 = 0.62$, $p < 0.001$) in the ice core are both statistically significant. The oil consumption had a comparable increasing trend as coal before it slowed down during 2000-2006.

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Biomass is the second largest energy resource in South Asia, and it is essential in rural areas. In India, 70% of the population lives in rural areas, and depends substantially on solid fuels (i.e., firewood, animal dung, and agriculture residues) for cooking and heating (Heltberg et al., 2000). Even in urban areas, biomass contributes to 27% of the household cooking fuel (Venkataraman et al., 2010). Although the consumption of biomass is lower than coal, the OC/BC emission ratio for biomass burning is much higher than from coal combustion (60.3 vs. 12.0) (Cao et al., 2005). BC emission factor for biomass burning (varying from 0.48 ± 0.18 g kg^{-1} for savanna and grassland burning to 1.5 g kg^{-1} for charcoal burning) is also generally higher than that for coal (0.2 g kg^{-1} for most combustion conditions) and oil combustion (0.3 g kg^{-1} on average, varying from 0.08 g kg^{-1} for heavy fuel oil to 0.66 g kg^{-1} for diesel) (Andreae and Merlet, 2001; Bond et al., 2004, 2007). Therefore, it is very likely that the OC/BC ratio of atmospheric carbonaceous aerosols and in the ice-core samples (Figure 6) was dominated by biomass burning emissions. Previous studies have concluded that carbonaceous aerosol emissions from biomass burning are the largest source in South Asia (Venkataraman et al.,

2005; Gustafsson et al., 2009). A general increase in energy-intensive life-styles associated with the accelerated growth of population and economy put pressure on energy resources, and induced energy transitions and use of non-sustainable biomass in South Asia (Sathaye and Tyler, 1991; Pachauri, 2004; Fernandes et al., 2007). For instance, biofuel consumption in South Asia increased by 21% per decade on average during 1950-2000 (Bond et al., 2007; Fernandes et al., 2007). In addition, fuel wood, a more desirable biofuel option, contributed 68% in 1978 to total energy demand by rural populations in India, and increased to 78% in 2000 (Fernandes et al., 2007).

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3.5 Radiative forcing induced by carbonaceous aerosols in Tibetan Glaciers

BC is often the most important light-absorbing impurity in surface snow because of its strong absorption of solar radiation. Effect of BC in snow on surface albedo reduction and resultant positive radiative forcing have been widely addressed and reported (e.g., Warren and Wiscombe, 1980; Clarke and Noone, 1985; Hansen and Nazarenko, 2004; Hadley and Kirchstetter, 2012; Flanner et al., 2007; 2009; McConnell et al., 2007; Ming et al., 2008; Kaspari et al., 2011; Qian et al., 2011, 2014a,b). In contrast, the impact of OC in snow has not been widely assessed because of its relatively weak light-absorption over the entire spectrum compared to BC, and because of large uncertainties associated with OC light-absorbing properties and measurements of OC in snow. However, there have been increasing interests in light-absorbing OC (a.k.a. brown carbon) and its radiative effect in the atmosphere (e.g., Kirchstetter et al., 2004; Andreae and Gelencsér, 2006; Hoffer et al., 2006; Moosmüller et al., 2009; Yang et al., 2009; Lack and Cappa, 2010; Cheng et al., 2011). Hoffer et al. (2006) estimated that humus-like substances as part of OC from biomass burning contribute ~7% to the absorption over the entire spectrum, which is not negligible. Yang et al. (2009) highlighted that as the contribution to absorption from BC decreases towards the ultra violet wavelengths, absorption due

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to brown carbon and dust becomes more significant, and they reported that at an observation site near Beijing brown carbon contributes over 10% to total absorption at mid-visible wavelengths. Thus the contribution of OC in snow to the surface albedo reduction is likely to be important, which has also been considered in recent climate modeling studies (Qian et al., 2014b).

In this study, we use the SNICAR-online model (available at http://snow.engin.umich.edu/; Flanner et al., 2007) to estimate radiative forcing induced by the observed BC and OC as if they were present in snow. Detailed description of the SNICAR model has been documented by Flanner and Zender (2005, 2006) and Flanner et al. (2007). Here we only briefly describe the setup of input parameters required for running the SNICAR model. A mass absorption cross-section (MAC) of 7.5 m^2 g^{-1} at 550 nm for uncoated BC particle and 0.6 m^2 g^{-1} for OC (Bond and Bergstrom, 2006; Kirchstetter et al., 2004; Yang et al., 2009) is assumed, and thus one of the input parameters for the online SNICAR model, MAC scaling factor, should be 1 for BC and 0.08 for OC. According to the previous studies (Cuffey and Paterson, 2010; Wiscombe and Warren 1980) and measurements in Qiyi glacier and Zuoqiupu glacier, an effective radius of 100 μm with density of 60 kg m^{-3} for new snow, and the effective radius of 400 μm with density of 400 kg m^{-3} for aged snow are adopted for the forcing calculation. As we focus on the estimation of radiative forcing by carbonaceous particles, other impurity contents, such as dust and volcanic ash, are set to be zero. The annual mean BC and OC concentrations during 1956-1979 was 4.4 ng g^{-1} and 13.8 ng g^{-1}, respectively, and they increased to 12.5 and 61.3 ng g^{-1} in 2006. As a consequence, the annual mean radiative forcing induced by BC (OC) in snow increases from 0.75 (0.20) W m^{-2} to 1.95 (0.84) W m^{-2}. Our estimate of mean BC forcing is lower than the estimated Eurasian radiative forcing (2.7 W m^{-2}) in spring (Flanner et al., 2009), but it's comparable to that in the East Rongbuk glacier over Himalayas, which was in the range of 1-2 W m^{-2} (Ming et al., 2008). Kaspari et al. (2009) reported a

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three-fold increase in radiative forcing from BC in snow over Himalayas after 1975, which is consistent with the increasing trend in our results. Although BC concentration is one order of magnitude lower than OC, radiative forcing of BC is about two times larger than OC due to its much stronger absorption of solar radiation. Note that the MAC value of OC is highly spectral dependent (Kirchstetter et al., 2004; Hoffer et al., 2006; Barnard et al., 2008; Yang et al., 2009). It increases greatly towards shorter wavelengths. Consequently, the absorption of OC may be biased. It is also important to note that we didn't consider variations in chemical compounds of OC, or the changes of OC during sample filtration. Although the estimation of OC radiative forcing herein is rather crude, the increasing trend should be robust.

BC and OC concentrations in the ice core increased rapidly since 1980, and the induced radiative forcing rose as a consequence. According to the estimates using the SNICAR model, the average BC radiative forcing had increased 43% after 1980, and OC radiative forcing had an increase of 70%. These numbers are by no means accurate, but the stronger increasing trend in the ice core recorded OC than BC during 1990-2006 (Figure 6) suggests that the contribution of OC to the total radiative forcing in the glacier induced by snow/ice impurities deserves more attention.

4. Summary and Conclusions

Light-absorbing carbonaceous aerosols can induce significant warming in the atmosphere and in snow and glaciers, which likely accelerates the melting of glaciers over Himalayas and Tibetan Plateau. Ice-core measurement of carbonaceous aerosols is a useful mechanism for evaluating historical emission inventories and revealing long-term changes in anthropogenic aerosols and their impacts on regional climate. In this study, we analyze carbonaceous aerosols recorded in an ice core (97 meters in depth and 9.5 cm in diameter) retrieved from the Zuoqiupu glacier

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(96.92°E, 29.21°N, 5600 m above sea level) in the southeastern Tibetan Plateau for their seasonal dependence and long-term trend. The glacier has a unique geographical location that is in close proximity to major Asian emission sources. With the help of a global climate model (CAM5) in which black carbon (BC) emitted from different source regions can be explicitly tracked, we are able to characterize BC source-receptor relationships between four Asian source regions (i.e., South Asia, East Asia, Southeast Asia and Central Asia) and the Zuoqiupu glacier area as a receptor. We also estimate the radiative forcing in snow due to BC and OC using the ice core measurements and an offline snow-ice-aerosol-radiation model (called SNICAR).

BC and OC concentrations in small segments of the Zuoqiupu ice core were measured using a thermal-optical method. Ice core dating based on significant seasonal variations of oxygen isotope ratios ($\delta^{18}O$) was used to construct the time series of BC and OC concentrations, which turned out to span the time period of 1956–2006. Not only do the concentrations of OC and BC in the ice core exhibit significant differences between the summer monsoon and non-monsoon seasons, which is likely due to changes in transport pathways and wet removal, but also the ratio of OC to BC shows a clear seasonal dependence that might be due to seasonal change in contributions from source regions and/or emission sectors. The CAM5 results show a similar seasonal dependence of BC and OC deposition to the glacier.

The MERRA reanalysis products used to drive the CAM5 model simulation show distinct circulation patterns during summer monsoon (June-September) and non-monsoon (October-May) seasons. Both the circulation patterns (and associated aerosol transport and wet removal) and seasonal variation of emissions in major source regions influence the seasonal deposition of aerosol at the Zuoqiupu site. The CAM5 simulation with tagged BC regional sources shows that South Asia is the dominant contributor (81%) to the 10-year mean BC deposition at the Zuoqiupu site during the non-monsoon season with 14% from East Asia, while the contribution of

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East Asia (56%) is larger than that of South Asia (39%) during the monsoon season. For the annual mean BC deposition, South Asia (75%) is the biggest contributor, followed by East Asia (21%).

The annual mean BC and OC deposition fluxes into the ice core are also estimated to explore the interannual variations and long-term trends. Results show stable and relatively low BC and OC fluxes from late 1950s to 1979, followed by a steady increase through the mid-1990s. A more rapid increase occurred after the minimum in 2002. The BC and OC deposition fluxes in 2006 were two and three times the respective average before 1980.

The overall increasing trend in deposition fluxes since 1980 is consistent with the BC and OC emissions in South Asia as the major contributor. Moreover, the increasing trend of OC/BC ratio since early 1990s indicates a growth of the contribution of coal combustion and/or biomass burning to the carbonaceous aerosol emissions in the major contributing source regions, which is consistent with the trends in the consumption of coal, oil and biomass in South Asia.

Our offline calculation using the SNICAR model shows a significant increase of radiative forcing induced by the observed BC and OC in snow after 1980, which has implications for the Tibetan glacier melting and availability of water resources in the surrounding regions. More attention to OC is merited because of its non-negligible light absorption and the recent rapid increases evident in the ice core record.

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Table 1. Source regions (South Asia, East Asia, Southeast Asia, and Central Asia) and corresponding BC emissions (Tg a^{-1}) and fractional contributions (%) to BC deposition flux at the Zuoqiupu site in monsoon (June-September), non-monsoon (October-May), and all months during 1996-2005.

Source Regions	Latitude	Longitude	Monsoon		Non-monsoon		Annual	
			Contribution	Emission	Contribution	Emission	Contribution	Emission
South Asia	5-35°N	50-95°E	38.51	0.65	81.26	0.74	74.48	0.71
East Asia	15-50°N	95-150°E	56.24	1.75	13.91	1.90	20.66	1.85
Southeast Asia	0-15°N	95-130°E	0.05	0.28	0.16	0.33	0.15	0.31
Central Asia	35-50°N	50-95°E	2.62	0.11	0.86	0.09	1.14	0.10

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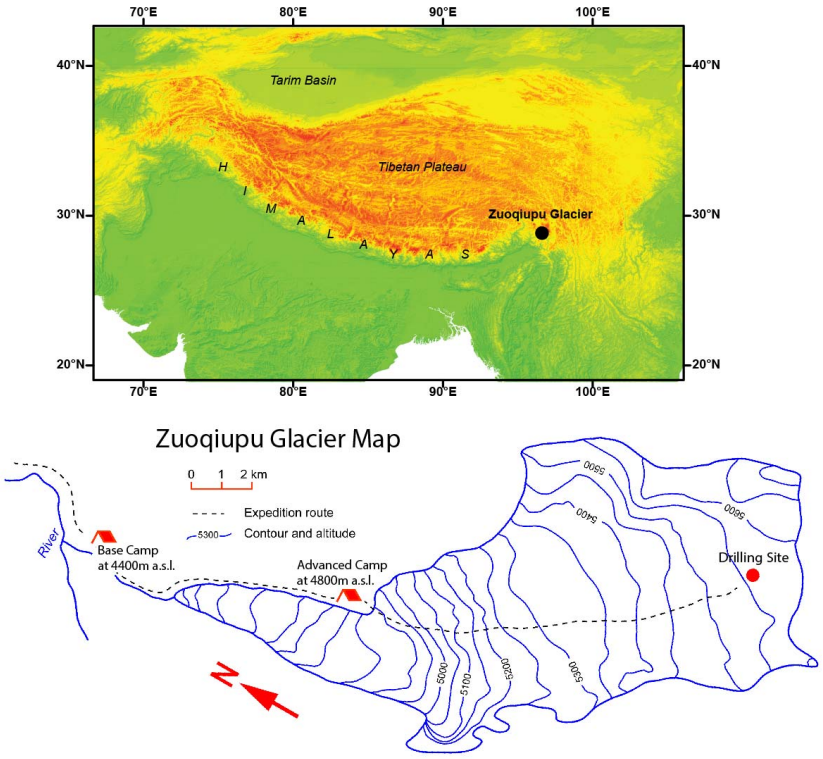

Figure 1. Site location of Zuoqiupu Glacier (top): black circle represents the location of Zuoqiupu Glacier, and warm colors indicate high elevations over the Tibetan Plateau. Detailed elevation contours of the Zuoqiupu Glacier are shown in the bottom panel. Red circle marks the ice core drill site.

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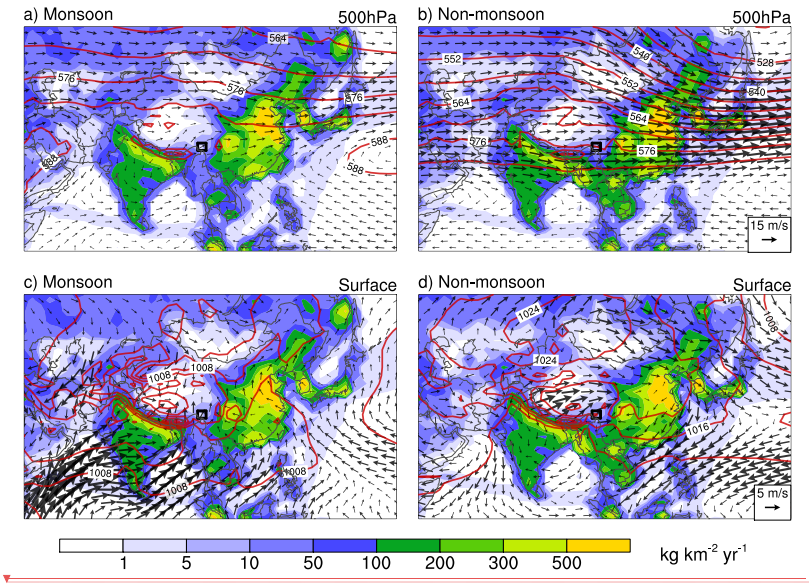

Figure 2. 10-year (1996-2005) mean wind vectors (denoted by arrows) at 500hPa (a, b) and the surface (c, d) during summer monsoon (June-September; a, c) and non-monsoon season (October-May; b, d) from MERRA reanalysis datasets used to drive the CAM5 simulation. 500 hPa Geopotential height (units: 10 m) contours with an interval of 60 m and mean sea-level pressure (units: hPa) contours with an interval of 4 hPa are superimposed on panels (a, b) and (c, d), respectively. The background colors show mean BC emission rates based on the IPCC present-day scenario for the corresponding months. The small black box marks the model grid-cell in which the ice core drill site resides.

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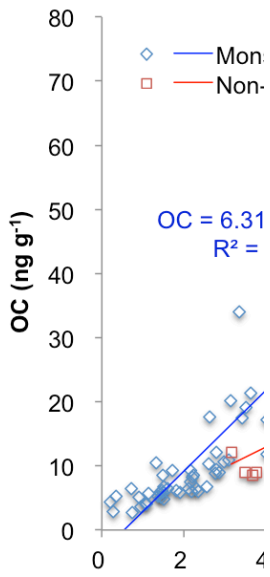

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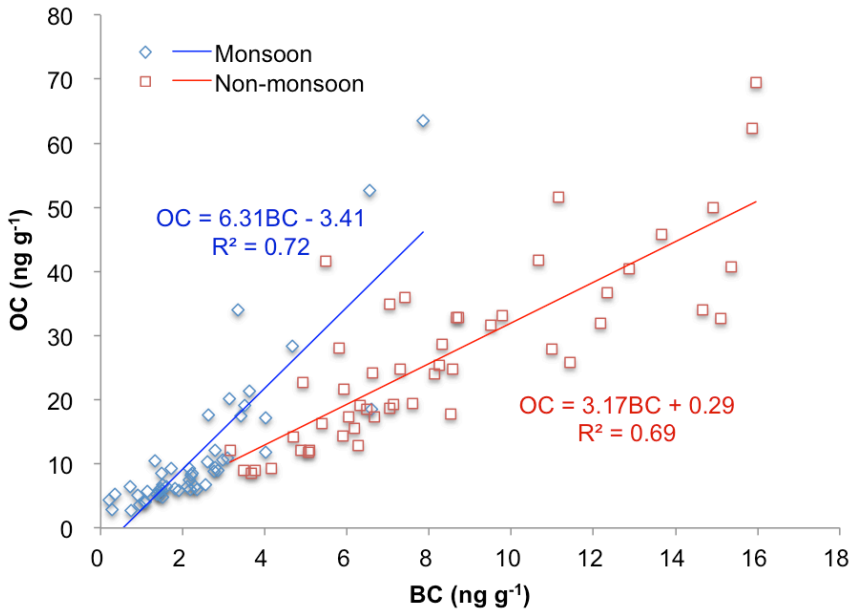

Figure 3. Scatter plots for yearly monsoon and non-monsoon mean OC and BC concentrations during 1956-2006, obtained from the ice core measurements, and corresponding linear regressions.

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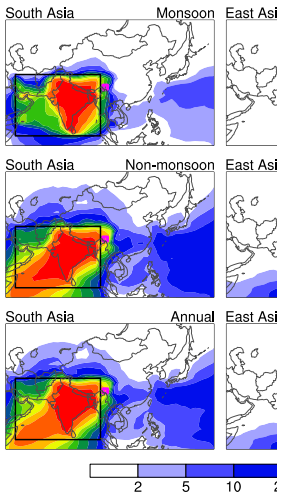

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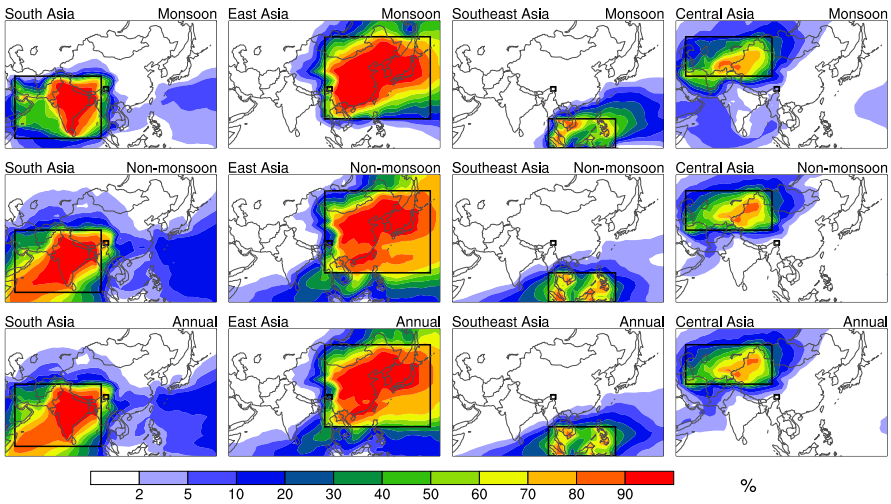

963 Figure 4. Spatial distributions of fractional contribution from the four source regions
964 (South Asia, East Asia, Southeast Asia, and Central Asia) to monsoon, non-monsoon, and
965 annual mean BC deposition fluxes during 1996-2005. The large black boxes indicate the
966 boundary of source regions, and the small black box marks the model grid-cell where the
967 Zuoqiupu drill site is located.
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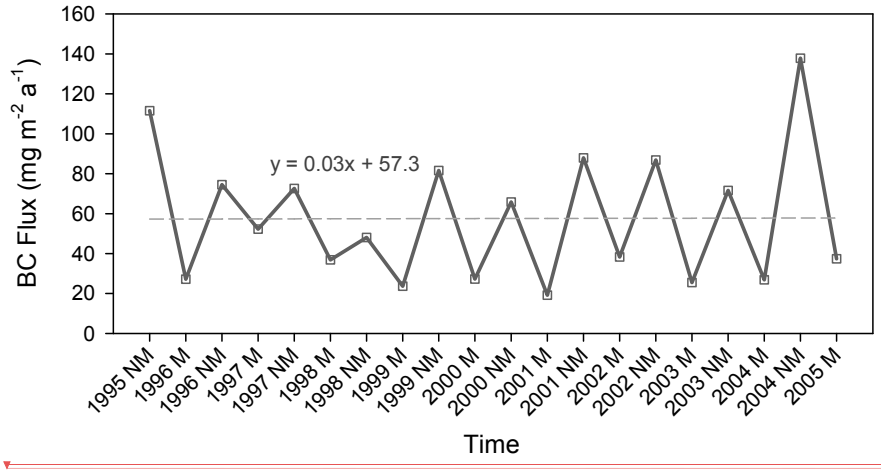

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976 Figure 5. Seasonal dependence ("NM" for non-monsoon and "M" for monsoon season),

977 of BC deposition flux at the Zuoqiupu site from 1995 to 2005 simulated in CAM5. The

978 dash line represents a linear regression of all data points.

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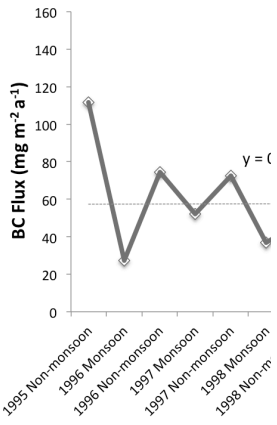

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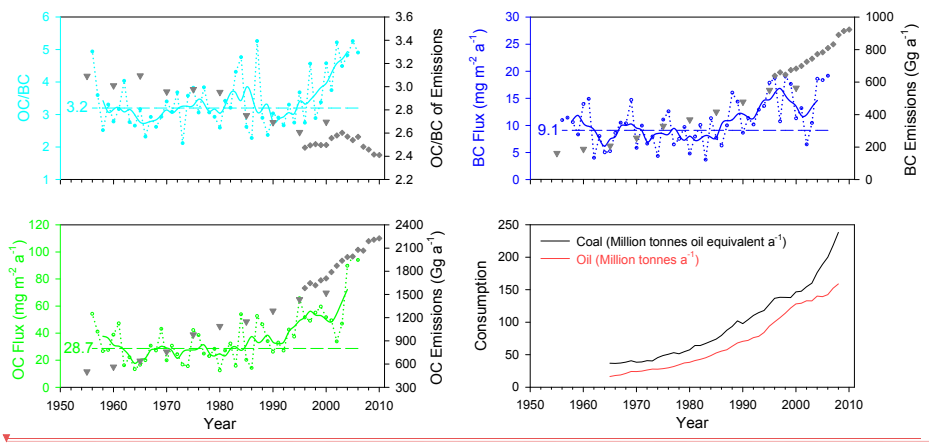

Figure 6 Time series of annual (dotted line with circles) and 5-year averaged (solid line) OC/BC ratios (top-left), BC (top-right) and OC deposition fluxes (bottom-left) in the Zuoqiupu ice core for the time period of 1956-2006. The average values of OC/BC ratio, BC and OC during 1956-1979 are marked by dashed lines. BC and OC emissions in South Asia (Bond et al., 2007) and corresponding OC/BC emission ratios are illustrated with gray triangles, and with gray diamonds for emissions in India (Lu et al., 2011). Coal and oil consumption data are shown in the bottom-right panel (BP Group, 2009).

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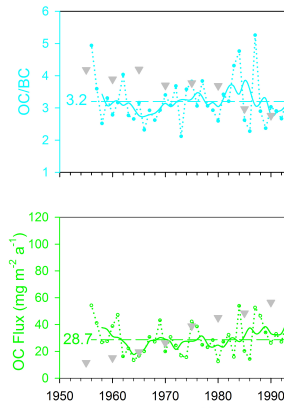

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Scatter plots for OC and BC concentrations and corresponding linear regression for the monsoon and non-monsoon seasons obtained from the ice core measurements

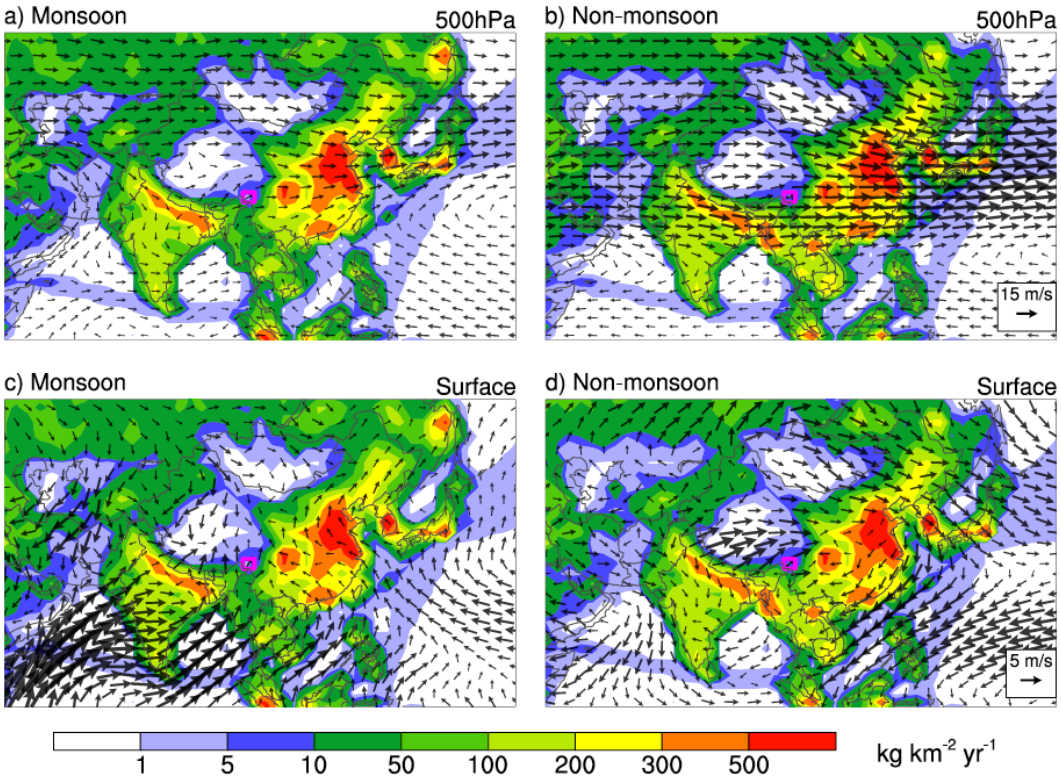
a) Monsoon
500hPa
b) Non-monsoon
500hPa
15 m/s
c) Monsoon
Surface
d) Non-monsoon
Surface
5 m/s
1
5
10
50
100
200
300
500
kg km-2 yr-1

Figure 3.