

**The authors thank Dr. J. W. Taylor for his time to review our manuscript and particularly for his valuable comments and suggestions that have significantly improved the manuscript. We have made most of the changes suggested by the reviewer and have outlined these in detail below.**

#### *General comments*

*The manuscript describes black carbon and optical property measurements from a field site in the Tibetan Plateau, taken over a period of just under two weeks. They divide up their data into “clean days” and “pollution episode” and compare and contrast the BC mass loadings/properties, absorption and calculated radiative forcing, as well as attempting to identify source regions for the pollution episode. This is an important topic, and certainly deserves investigation.*

*Overall, the manuscript is well structured and reasonably well written, though it needs some thorough proofreading (including the title). The experimental data are high quality, and I enjoyed reading the paper. Having said that, there are some parts that I think need to be reworked, as some of the figures are not the best way of displaying the information the authors are trying to convey. In particular, the section linking mixing state to optical properties needs to be improved. I recommend this manuscript be published subject to the following revisions.*

**Response:** Again, we appreciate the reviewer’s careful and valuable comments and have made most of the changes suggested by the reviewer including the title, text and figures.

#### *Specific comments*

*Abstract- The main point I think you have shown, but need to highlight better is that the main factor that determines how much light is absorbed by BC is simply the concentration of BC. Comparing the pollution episode to the clean days, BC increased by 400% but  $MAC_{BC}$  only changed by ~15% according to figure 6. This point should be highlighted in the abstract and conclusions.*

**Response:** We reworked on Figure 6 as suggested by the reviewer and highlighted the new results in the abstract. It now reads “The rBC mass absorption cross section ( $MAC_{rBC}$ ) at  $\lambda = 532$  nm was slightly larger in clean days ( $14.9 \text{ m}^2 \text{ g}^{-1}$ ) than during pollution episode ( $9.3 \text{ m}^2 \text{ g}^{-1}$ ), likely due to the effects of brown carbon and the uncertainty of the  $MAC_{rBC}$  calculation. The  $MAC_{rBC}$  was positively correlated with number fraction of coated rBC during pollution episode with an increasing rate of  $0.18 (\text{m}^2 \text{ g}^{-1}) \%^{-1}$ .....Compared to rBC mass concentration, rBC mixing state is more important in determining absorption during pollution episode, estimated from the same percentagewise increment of either rBC mass concentration or the number fraction of coated rBC.”

*P14143 L4 metres above mean sea level*

**Response:** Change made.

*P14144 L2-6 for completeness you should also mention the semi-direct effect*

**Response:** In the revised manuscript, we added the following “BC also shows semi-direct effect through interaction with cloud processes (Koch and Del Genio, 2010).”

*P14146 L20&22 refractory BC mass*

**Response:** We now use refractory BC (rBC) instead of BC for SP2 related measurements and results.

*P14147 L7 Write a sentence saying what your calibration material was and what the absolute uncertainty is on the BC measurement (this is important for your  $MAC_{BC}$  calculation).*

**Response:** In the revised manuscript, we added the following “The incandescence signal was calibrated using a standard fullerene soot sample (Lot F12S011, Alpha Aesar, Inc., Ward Hill, Massachusetts). The total uncertainty in the rBC mass determination was ~25%. More details about the SP2 calibration and uncertainty can be found in our previous work (Wanget al., 2014a)”.

*L18 give some examples of atmospheric processes that can cause BC to be internally-mixed*

**Response:** Following the reviewer’s suggestion, we added the following “This number fraction is higher for more aged rBC particles due to the formation of coating from atmospheric physical and chemical processes including coagulation, condensation, and heterogeneous reactions (Liu et al., 2013; Browne et al., 2015).”

*L9-14 You cite Wang et al. (2014) for details of the mixing state measurement. They say that only particles <275nm VED were used due to saturation. You should also use a lower diameter limit as well. For small particles the scattering measurement is likely to be too noisy to reliably measure the time delay. Taylor et al. (2015) discuss the instrument’s limitation when considering leading-edge scattering, but similar arguments apply when using the time delay technique.*

**Response:** Following the reviewer’s suggestion, we added the following “Because the scattering measurement is rather noisy for small particles and become saturation for large particles, the mixing state was studied for rBC core between ~70 and ~275 nm VED, which constitute the majority of rBC particle numbers (Wang et al., 2014a). The

limitation of SP2 instrument is discussed in Taylor et al. (2015) when considering leading-edge scattering.”

*L28 Please give more details on the calibration of the PAX. What concentration of NO<sub>2</sub> did you use? How well do you know the concentration? Did you do it in the dark to prevent photolysis? What absorption cross-section did you use? Specifically, you need to say the absolute accuracy of the measurement, as it is important for the MAC<sub>BC</sub> calculation.*

**Response:** The PAX can also provide the light extinction coefficient independently using the laser power. NO<sub>2</sub> was used to produce extinction to establish a correction factor for the absorption calibration. Generally, one ppb of NO<sub>2</sub> can produce absorption ( $\lambda=532$  nm) of  $\sim 0.395$  Mm<sup>-1</sup> at standard temperature and pressure (0 °C and 1013.25 hPa). It is not necessary to know the exact concentration of NO<sub>2</sub> when calibrating the PAX. It only needs to ensure that there is enough NO<sub>2</sub> to produce light absorption gradient. The photolysis of NO<sub>2</sub> is prevented by using black conductive silicone tube. The calibration result is shown in Figure R1 in this Response. The uncertainty of the PAX is estimated to be  $\sim 10\%$  in this study. In the revised manuscript, we added the following “The PAX can provide the light extinction coefficient independently using the laser power. NO<sub>2</sub> was used to produce an absorption reading of  $\sim 500$ – $30000$  Mm<sup>-1</sup>. A correction factor was then established from the relationship between the calculated light extinction coefficient using laser power and the measured light absorption. The uncertainty of the PAX is estimated to be  $\sim 10\%$ .”

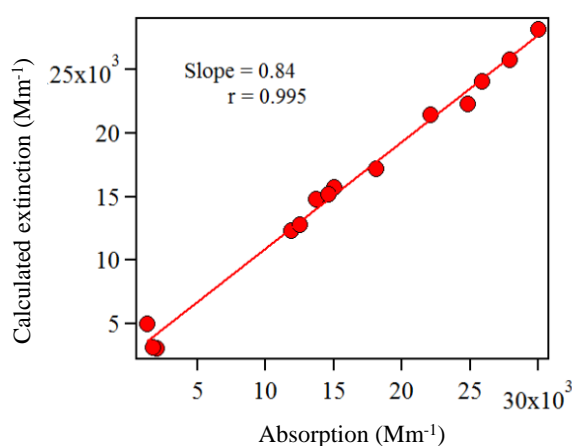


Figure R1. Correlation between the calculated extinction coefficient using laser power and the measured absorption.

*Figure 2 I can see what you are trying to do here but I’m not sure it’s appropriate in this case. This type of plot is useful when the diurnal variation is stronger than the day-to-day variation, and/or if you have a long dataset. It looks like there has been some artificial smoothing of the data- it should just be a stripe for each day. I think*

you'd be much better off with just a time series of BC concentration and coated fraction. You could also add in  $MAC_{BC}$  and  $BABs$  as well. And then highlight the pollution episode.

**Response:** Following the reviewer's suggestion, we provide time series of rBC concentration, number fraction of coated rBC,  $MAC_{rBC}$  and absorption in Figure 2 in the revised manuscript (also see below Figure R2).

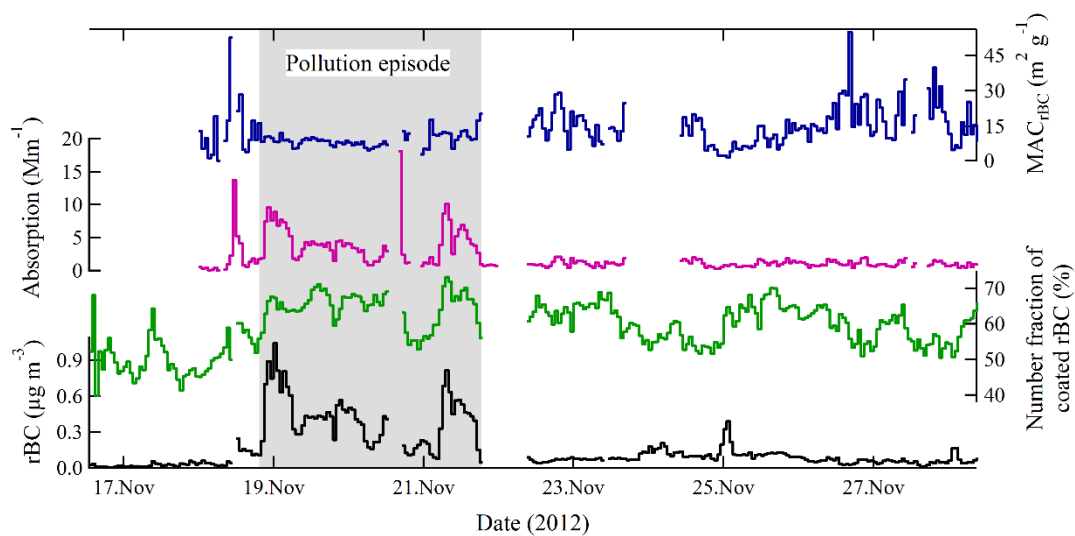


Figure R2. Time series of the rBC mass concentration, number fraction of coated rBC, light absorption at  $\lambda = 532$  nm, and mass absorption cross section of rBC ( $MAC_{rBC}$ ) during the entire campaign period. The pollution episode is highlighted with grey background.

*P14148 L10 I think you are using too much precision when reporting some of your results. For example,  $59.3 \pm 6.9\%$  would be better as  $59 \pm 7$ . You've done this in many parts of the manuscript (e.g. table 1).*

**Response:** We made changes throughout the manuscript.

*L11 Please rephrase this sentence*

**Response:** This sentence now reads "The mean number fraction of coated rBC is found to be  $59 \pm 7\%$  (range 40–73%), suggesting the majority of aged rBC particles in wintertime in the Qinghai Lake region."

*L12 I don't see the significance of 30% of the values being higher than the mean, that just means you have a non-gaussian distribution of BC concentration.*

**Response:** Following the reviewer's suggestion, we revised Figure 2 in the manuscript and the 30% of values being higher than the mean can be seen from the

time series of rBC mass concentration in Figure 2. Also, the rBC concentration is Gaussian distribution as shown in Figure R3 in this Response.

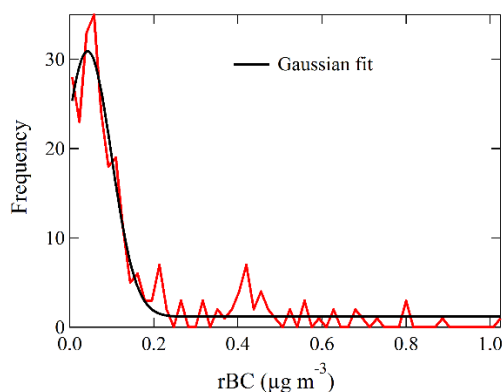


Figure R3. Frequency distribution of rBC mass concentration during the entire campaign period.

*L13-14 I'm not sure how you get from a large variation coefficient to a large BC burden. Doesn't the measured BC concentration show that?*

**Response:** The variation coefficient reflects the degree of data dispersion. The larger variation coefficient of rBC values means that there are some data much larger than average. In the revised manuscript, the large rBC burden can be seen from the time serial of rBC concentration in Figure 2. Now this sentence reads “It is found that ~25% of the rBC values are higher than the 75th value, and the variation coefficient (defined by SD/mean) of rBC values reaches as high as 120%, suggesting a large rBC burden even at the free tropospheric altitude.”

*Figure 3 I know the y axis is arbitrary units but it would make it easier for the reader if you make the pollution episode larger than the “all days” distribution, as the concentrations were higher than average.*

**Response:** Following the reviewer's suggestion, we revised Figure 3 in the manuscript. This Figure is shown below as Figure R4.

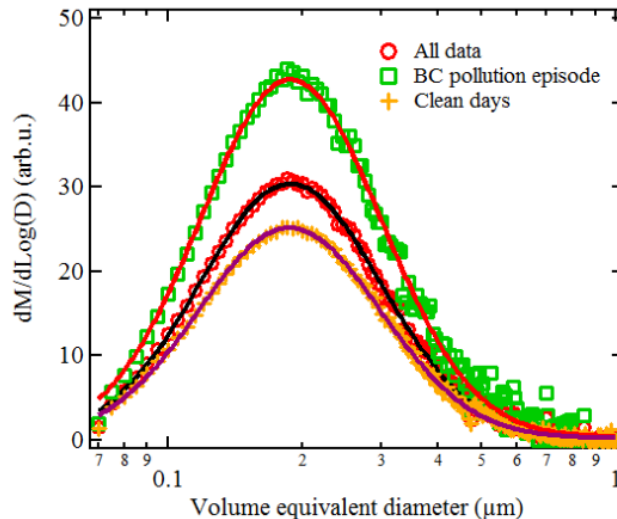


Figure R4. Mass size distribution of rBC in volume equivalent diameter during different sampling periods at Qinghai Lake. The solid lines represent lognormal fit. “M” and “D” in vertical label represent rBC mass and void free diameter (assuming  $2 \text{ g cm}^{-3}$  density), respectively.

*P14149 L1-8 You are correct that coagulation is slow and BC size distributions don't tend to change much. That means that the measured BC are likely to be from similar types of sources. Fossil fuel and biomass burning tend to have different size distributions. See (Kondo et al., 2011; Sahu et al., 2012; Taylor et al., 2014), it would be useful to compare to these studies and probably back up your assertion that the BC is from biomass burning.*

**Response:** We agree with the reviewer. In the revised manuscript, now it reads “Therefore, the similarity in VED size distribution for rBC core between clean and pollution episode indicates that the measured rBC particles are likely from biomass burning emissions, given that fossil fuel and biomass burning tend to have different rBC size distributions and that the peak diameter measured in this study is similar to the reported rBC peak diameter from biomass burning plumes (range  $\sim 187\text{-}193 \text{ nm}$ , see Kondo et al., 2011; Sahu et al., 2012; Taylor et al., 2014).”

*L11 The HYSPLIT URL has a typo in it. Also you should see this page [http://www.arl.noaa.gov/HYSPLIT\\_pubs.php](http://www.arl.noaa.gov/HYSPLIT_pubs.php) for a better reference for HYSPLIT (i.e. one or more of the papers on that page). You should also state what meteorology data you used, and what method was used for vertical motion.*

**Response:** It now reads “To examine the contribution of regional rBC transport, five-day back trajectories were calculated using the hybrid single-particle Lagrangian integrated trajectories (HYSPLIT) model (<http://ready.arl.noaa.gov/HYSPLIT.php>). The HYSPLIT model was driven with full vertical dynamics using gridded meteorological data (Global Data Assimilation System, GDAS1).”

*L12-14 I don't agree with this as the reason for using 5-day trajectories. It's more that 5 days (or less) is about as long as you might typically trust a trajectory. BC lifetime varies- on average it's around a week but some plumes last longer and some not as long. It will depend on the meteorology.*

**Response:** We agree with the reviewer that the BC lifetime varies and it is around a week on average. To be more precise, we removed “The five-day period...in the order of one week” in the revised manuscript.

*Some of this section (e.g. the AOD and MODIS data) reads as if you are saying that if you follow back a trajectory 5 days then you will find the source of the BC. This is possible, but you might also pass through a strong source region 1 day back, in which case most of the BC would have come from there. I think you are probably right that the pollution episode came from fires in North India but that really comes from the PSCF part of this section rather than the trajectories alone. So, I think you should move the part about AOD and MODIS after the discussion of the PSCF model. You ran some trajectories, you used the PSCF model to try work out where/when the sources might be, then you used AOD and MODIS data to back up your findings.*

**Response:** Following the reviewer's suggestion, we moved the parts about AOD and MODIS after the discussion of the PSCF model to back up our findings.

*P14150 L10-20 You should clarify how you did your data averaging- you ran trajectories every 6 hours (with a timestep of 1 hour?) but get 1s data from the SP2- did you average to the timestep of the trajectories?*

**Response:** The SP2 data were averaged to 1 hr in order to match the timestep of the trajectories. We clarified this point in the revised manuscript.

*My other main comment about this section is that it's useful to identify where the pollution episode originated from, but most of the BC you measured was during the clean periods as the pollution episode only lasted 2 days. If you use a lower threshold with the PSCF, can you say anything about the BC sources during the clean days? These might be more important in the long-run.*

**Response:** As already clarified in the manuscript, although PSCF model can be used to determine the potential source regions, a limitation of this model is that grid cells can have the same PSCF value when sample concentrations at the receptor site are either only slightly higher or extremely higher than the criterion. Therefore, if one uses a lower threshold, it will lead to difficulties in distinguishing moderate sources from strong ones. Also the average rBC concentration in clean days was low (86 ng m<sup>-3</sup>) and we are more interested in the sources of high rBC loadings because the latter may lead to much significant effect on the Qinghai-Tibetan region.

*Section 3.3 and Figures 6 and 7*

*In this section you have tried to address two questions. 1- what is the  $MAC_{BC}$  of the BC during the clean and pollution days and how much is this linked to mixing state? 2- What is the main factor that determines absorption (i.e. BC concentration or mixing state)? You have focused on the first and touched upon the second in lines 16-23 on P14152, but the second question is arguably the most important.*

**Response:** We improved this section as suggested, please see below.

*The way you've plotted figure 6, the slopes of the lines are determined mostly by the points in the top/right of the plot. For the pollution episode the points follow a straight line reasonably well but for the clean days some of the lower down points are quite far away from the line. If you calculated  $MAC_{BC}$  as a function of time, I think you would have some periods during the clean days where the calculated values would be quite different to the straight line fit. In fact, if you take the mean or median values of BC and BAbs from table 1 and use them to calculate  $MAC_{BC}$ , then you would come to the conclusion that  $MAC_{BC}$  was higher during the clean periods, which is opposite to what you have found using the straight line fits!*

**Response:** Thanks for the reviewer to point out this. We reworked on Figure 6 as suggested. Please see below for discussion in more detail.

*Figure 7 looks quite strange- why is there a neat straight line cutting off the bottom right of the graph? Was there really no data in that section? Without the data in the bottom right corner it's difficult from this plot to work out whether BC concentration or mixing state are more important for determining absorption.*

**Response:** The data in Figure 7 were processed to matrix, and there was no data in the bottom right of the graph. To make this graph clearer we revised it (see below the Figure R5 in the Response) and also clarified whether BC concentration or mixing state is more important for determining absorption. It reads "To further investigate whether rBC concentration or mixing state is more important for determining absorption, the increases in light absorption are compared based on the same percentage-wise increment of either rBC mass concentration or the number fraction of coated rBC. According to the regression function in Figure 8b and the correlation between absorption and rBC mass (Absorption =  $-0.38 + 10.17[rBC]$ ,  $r = 0.92$ ), the increase of light absorption is larger for number fraction of coated rBC (e.g,  $\Delta$  light absorption =  $1.8 \text{ Mm}^{-1}$ ) than for the rBC mass (e.g,  $\Delta$  light absorption =  $0.5 \text{ Mm}^{-1}$ ), suggesting that, compared to rBC mass concentration, rBC mixing state is more important in determining absorption during pollution episode."



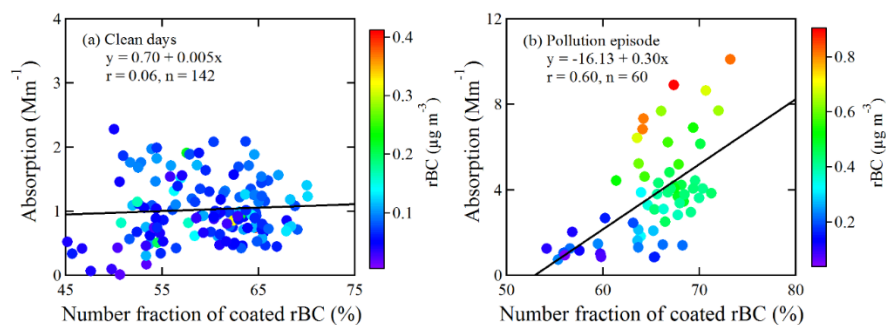


Figure R5. Light absorption as a function of number fraction of coated rBC during (a) clean days and (b) pollution episode. Data points are color coded for rBC mass concentration.

*I think using a different approach would answer these questions much more clearly. Based on figure 7 I think you should have enough signal/noise to calculate  $MAC_{rBC}$  as a function of time. You could add it to Figure 1 as well. Then instead of figure 6, you could add the mean/quartiles of  $MAC_{rBC}$  to table 1, and replace figure 6 with histograms showing the variation of  $MAC_{rBC}$  during the clean and polluted periods. That will tell you if  $MAC_{rBC}$  is different in each time period, and show the variation.*

**Response:** Thanks for the reviewer's suggestion. We added the time series of  $MAC_{rBC}$  in Figure 2 in the revised manuscript and the mean/quartiles of  $MAC_{rBC}$  in Table 1. The old Figure 6 was replaced with histograms as shown in Figure R6 in the Response. In the revised manuscript, it now reads "Figure 6a and b show histograms of the  $MAC_{rBC}$  values during clean days and pollution episode, respectively. The distribution of  $MAC_{rBC}$  in clean days tends to larger values than that during pollution episode, with an average value of  $14.9 \pm 8.9 \text{ m}^2 \text{ g}^{-1}$  for clean days and  $9.3 \pm 3.1 \text{ m}^2 \text{ g}^{-1}$  for pollution episode. These values are higher than the  $MAC_{rBC}$  of  $7.8 \text{ m}^2 \text{ g}^{-1}$  for uncoated rBC particles (interpolated to 532 nm from 550 nm assuming an Absorption Ångström Exponent of 1.0) suggested by Bond and Bergstrom (2006). It is interesting that the  $MAC_{rBC}$  in clean days is ~60% larger than that during pollution episode, the reason for which is not clear. A possible explanation involves the interference from brown carbon. Previous studies demonstrate that brown carbon, like black carbon, is an important light-absorbing aerosol composition in the atmosphere which can absorb light at visible wavelength (e.g.,  $\lambda = 532 \text{ nm}$ ) (Yang et al., 2009). In the rural areas of Qinghai, biofuels including yak and sheep dung, firewood, and crop residues account for ~80% of total household energy (Ping et al., 2011). Biofuel/biomass combustion emissions are considered as especially significant sources for brown carbon (Andreae and Gelencser, 2006). It may produce enough brown carbon (particularly during the smoldering combustion phase) influencing the light absorption when rBC loading is low. Thus, the  $MAC_{rBC}$  may be overestimated in clean days. In addition, the calculation method using the light absorption and rBC mass may also introduce uncertainty, especially when rBC concentration is low. The high  $MAC_{rBC}$  values always correspond to the very low rBC mass. The  $MAC_{rBC}$  calculation method can bring ~30% uncertainty estimated from the square root of uncertainties in the PAX

(10%) and SP2 (25%) measurements.”

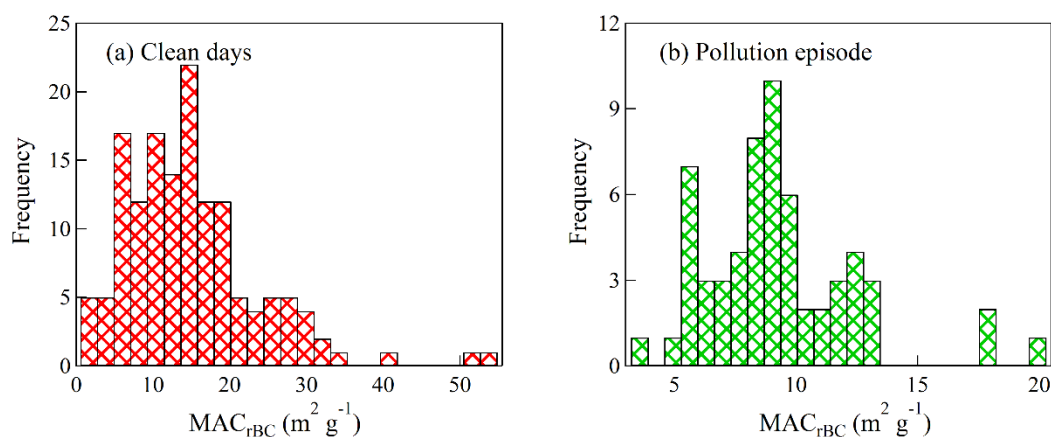


Figure R6. Frequency distributions of rBC mass absorption cross section ( $MAC_{rBC}$ ) during clean days and pollution episode.

*I think the key plot to work out if  $MAC_{BC}$  is linked to mixing state is make a plot like figure 2 in Lan et al. (2013). Divide up  $MAC_{BC}$  into bins of coated number fraction and see if there is any relationship. That will tell you how much any changes in  $MAC_{BC}$  are linked (qualitatively) to mixing state. Then also say how much brown carbon could have affected your result.*

**Response:** Following the reviewer’s suggestion, we added a new figure (i.e., Figure 7 in the revised manuscript or Figure R7 in the Response) to discuss the relationship between  $MAC_{rBC}$  and number fraction of coated rBC. In the revised manuscript, we added the following “To further investigate the effect of rBC mixing state on  $MAC_{rBC}$ , the  $MAC_{rBC}$  values were plotted against the number fraction of coated rBC. As shown in Figure 7, the  $MAC_{rBC}$  was not correlated with the number fraction of coated rBC during clean days, but positive correlation was observed during pollution episode suggesting that the mixing state leads to the increase of the  $MAC_{rBC}$ . The slope of  $0.18 \text{ (m}^2 \text{ g}^{-1}) \%^{-1}$  obtained from the linear regression is arguably representative of the rate of the mixing state effect on the  $MAC_{rBC}$ .”

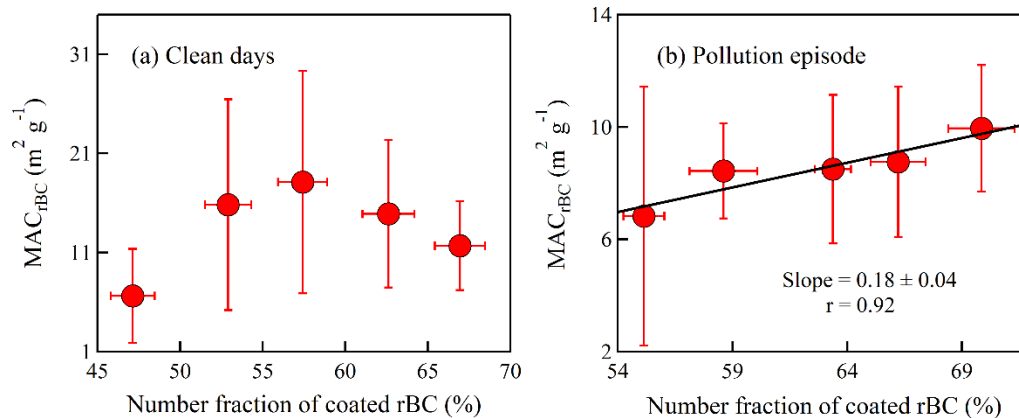


Figure R7. Mass absorption cross section of rBC ( $MAC_{rBC}$ ) versus number fraction of coated rBC during clean days and pollution episode. The error bars correspond to the standard deviations of  $MAC_{rBC}$  and number fraction of coated rBC.

Finally, to answer whether BC concentration or  $MAC_{BC}$  is more important for determining absorption, just look at the numbers in table 1 and see which changes most. I don't think this should be too much effort, and I think it would make the conclusions much clearer and more robust.

**Response:** As discussed above, following the review's suggestion we re-visited the data which leads to new conclusion. We added in the revised manuscript "To further investigate whether rBC concentration or mixing state is more important for determining absorption, the increases in light absorption are compared based on the same percentagewise increment of either rBC mass concentration or the number fraction of coated rBC. According to the regression function in Figure 8b and the correlation between absorption and rBC mass (Absorption =  $-0.38 + 10.17[rBC]$ ,  $r = 0.92$ ), the increase of light absorption is larger for number fraction of coated rBC (e.g,  $\Delta$  light absorption =  $1.8 Mm^{-1}$ ) than for the rBC mass (e.g,  $\Delta$  light absorption =  $0.5 Mm^{-1}$ ), suggesting that, compared to rBC mass concentration, rBC mixing state is more important in determining absorption during pollution episode."

P14152 L25 I'm not sure how much the  $7.5 m^2/g$  from Bond and Bergstrom (2006) is set in stone. For example, Lan et al. (2013) measured values lower than this value. Also, it is critical that you calculate the absolute calibration error on the BC and BAbs measurements in order to quantitatively compare your calculated  $MAC_{BC}$  to any other value.

**Response:** The  $MAC_{BC}$  of  $7.5 \pm 1.2 m^2/g$  for fresh light-absorbing carbon suggested by Bond and Bergstrom (2006) was summarized from multiple studies. It's reasonable that different study shows varied  $MAC_{BC}$  values because it may increase due to coating or decrease due to particle coagulation and aggregate collapse. In the revised manuscript, we added the calibration errors of SP2 and PAX measurements.

*P14153 A point to mention is that although the BC concentration was 4x higher in the polluted days, the radiative forcing was only about 2x higher, meaning other aerosol components are also important.*

**Response:** Following the reviewer's suggestion, we added the following "It is worth to note that the rBC concentration during pollution episode was 4 times higher than that in clean days, but the DRF<sub>BC</sub> was only enhanced by a factor of two, suggesting the importance of other aerosol components which made negative contribution to DRF."

Reference:

Browne, E.C., Franklin, J.P., Canagaratna, M.R., Massoli, P., Kirchstetter, T.W., Worsnop, D.R., Wilson, K.R., Kroll, J.H.: Changes to the chemical composition of soot from heterogeneous oxidation reactions, *J. Phys. Chem. A*, 119, 1154–1163, 2015.

Koch, D. and Del Genio, A. D.: Black carbon semi-direct effects on cloud cover: review and synthesis, *Atmos. Chem. Phys.*, 10, 7685-7696, 2010.

Liu, D., Allan, J., Whitehead, J., Young, D., Flynn, M., Coe, H., McFiggans, G., Fleming, Z.L., Bandy, B.: Ambient black carbon particle hygroscopic properties controlled by mixing state and composition, *Atmos. Chem. Phys.*, 13, 2015-2029, 2013.

Ping, X., Jiang, Z., Li, C.: Status and future perspectives of energy consumption and its ecological impacts in the Qinghai-Tibet region, *Renew. Sustain Energy Rev.*, 15, 514–523, 2011.

Sahu, L.K., Kondo, Y., Moteki, N., Takegawa, N., Zhao, Y., Cubison, M. J., Jimenez, J. L., Vay, S., Diskin, G. S., Wisthaler, A., Mikoviny, T., Huey, L. G., Weinheimer, A. J. and Knapp, D. J.: Emission characteristics of black carbon in anthropogenic and biomass burning plumes over California during ARCTAS-CARB 2008, *J. Geophys. Res.*, 117(D16), D16302, doi:10.1029/2011JD017401, 2012.

Taylor, J. W., Allan, J. D., Allen, G., Coe, H., Williams, P. I., Flynn, M. J., Le Breton, M., Muller, J. B. A., Percival, C. J., Oram, D., Forster, G., Lee, J. D., Rickard, A. R., Parrington, M., and Palmer, P. I.: Size-dependent wet removal of black carbon in Canadian biomass burning plumes, *Atmos. Chem. Phys.*, 14, 13755-13771, 2014.

Taylor, J. W., Allan, J. D., Liu, D., Flynn, M., Weber, R., Zhang, X., Lefer, B. L., Grossberg, N., Flynn, J., Coe, H.: Assessment of the sensitivity of core/shell parameters derived using the single-particle soot photometer to density and refractive index, *Atmos. Meas. Tech.*, 8(4), 1701–1718, 2015.

Wan, X., Kang, S., Wang, Y., Xin, J., Liu, B., Guo, Y., Wen, T., Zhang, G., and Cong, Z.: Size distribution of carbonaceous aerosols at a high-altitude site on the central Tibetan Plateau (Nam Co Station, 4730 m a.s.l.), *Atmos. Res.*, 153, 155-164, 2015.

Wang, Q., Liu, S., Zhou, Y., Cao, J., Han, Y., Ni, H., Zhang, N., Huang, R.:

Characteristics of black carbon aerosol during the Chinese Lunar Year and weekdays in Xi'an, China, *Atmosphere*, 6, 195–208, 2015.