## Response to Reviewers

The authors would like to thank both reviewers for their careful and insightful comments, questions and suggestions. As a result of these reviews we have reanalyzed the data and refined some of the analysis to the degree that we feel that the revised manuscript is a more clear contribution much improved from the original submission Our responses below are given in bold italics.

### Response to Reviewer 1

Explicitly state the wavelength of the PAX used here.

> ***The revision explicitly describes the PAX used in our study as a 870 nm version of this instrument.***

The potential effect of wet scavenging is mostly ignored. Only on line 26 of page 12557 there is a brief mention of precipitation. Using the data available (I would guess that rain gauges data are also available for the area for example), it should be possible to see if wet scavenging (either nucleation or collision and coalescence) might be an important factor influencing seasonal variability of some of the pollutants, especially with regard to particulate. It would be interesting to see at least some mention of this possible source of variability.

> ***The reviewer raises and important point about the significance of wet removal by precipitation. In addition to adding a figure that shows the almost daily rain in Mexico City, we have added a paragraph that discusses the importance of precipitation for not only removal of particles by inertial scavenging, but all the impact of evaporating rain in humidifying and cooling the atmosphere with the subsequent effect on aqueous processes.***

The authors conclude that the emission reduction strategies did not bring much change in eBC over the last several years. I believe that might be the case; however, the comparison is done using different measurement techniques, the PAX for this study vs. the PSAP for example, for some of the past studies. It is possible that filter-based artifacts might make this comparison less robust. In addition, even assuming eBC indeed remained unchanged, I would guess that the number of vehicles might have substantially increased over the years, so a flat eBC might still be compatible with a modest positive reduction of per-vehicle emissions. This not to disagree with the authors that a more stringent control on vehicle emission (especially diesel) should indeed be sought.

> ***In the revised version of the manuscript, we make note of the relevance of no decrease in BC within the expected uncertainties and have also added in the discussion and summary information on how the number of cars has almost doubled in 15 years, increasing by approximately 5% per year. Given that the current mitigation***

***strategies do little to reduce particle emissions, it is actually surprising that there has not been an increase in BC, so maybe actually some of the mitigation efforts are slowing the increase.***

Section 2.2:
Line 15, page 12545: Moosemuller should read Moosmüller.

***Corrected as noted.***

2. Line 19, page 12545: Does that means that there were 4 PAXs with 4 different wavelengths at the site? I think not, so be explicit that the wavelength used here was 870 nm and consider not mentioning the other wavelengths. In addition, what is the laser power?

***This has been modified to clarify that only the 870 nm version was operated at the measurement site.***

3. Do the authors know what the potential line losses in the instrument might be? In other terms what is the transmission 50% size-cut?

***As discussed already in the text, a PM2.5 cyclone is used at the inlet, i.e. a 50% cut-size of 2.5 µm. We have now added that the length of 3/8“ conductive tubing is 3 m. Using the Baron and Willike Aerocalc, at a flowrate of 1 lpm, the transmission efficiency of particles < 2.5 µm is close to 100%***

4. What is the typical Q of the resonator?

***The resonator quality factor (Q) recorded for the PAX operated during this period was 27.2.***

5. Line 6 through 9, page 12548: does that means that the PAX used in this experiment did not have Helmholtz filters or did it? It is not clear to me.

***The revised manuscript clarifies that because of the low noise environment where the PAX was installed, no filters were necessary.***

6. A MAC of 4.74 m2g-1 is used and the Bond and Bergstrom 2006 paper is cited as a source. If I recall correctly that paper discusses the MAC for shorter wavelengths. Is the 1/lambda dependence used here to extrapolate the value at 870 nm? Please clarify.

***The reviewer is correct. Bond and Bergstrom recommended 7.5 m2 g-1 at 550 nm. The value at 870 is wavelength corrected. This is clarified in the revised text.***

7. Line 6 through 2 and line 16, page 12549: Beers should read Beer's from August Beer.

***Corrected***

8. The truncation angle of 4% mentioned in the paper is estimated how and for what particle size? In addition, extinction measurements also suffer from similar issues related to the collection angle of the detector used for the extinction measurement and the divergence of the light source. The extinction measurement used here could be discussed a little bit more.

***The truncation angle has not been measured directly but is estimated directly from the optical geometry (Nakayama et al., 2015). This is now briefly mentioned in the revision, as well as changing 4° to 6°. The extinction derived from the sum of the absorption and scattering coefficients will be impacted by the truncation angle limitation, but in this particular study, the extinction is not use; however, we do now mention that this is a source of uncertainty for scattering and extinction coefficients, as well as he SSA that will also be impacted.***

9. Line 18 through 21, page 12549: Considering the estimate of 4% truncation angle above, does this mean that this uncertainty should be less than 4%?

***See response above.***

10. This type of calibration procedure seems like has been discussed previously in the literature that probably could be cited.

***Yes, it has by Arnott et al. as well as in Nakayama et al. We add these references in the revised manuscript.***

11. If the uncertainty on Babs is 20% then it would seem that the uncertainty on eBC should be definitely larger than 20% and the MAC can introduce substantial error as well; therefore, the estimate of eBC uncertainty of 20-30% seems a bit low. How was this range estimated?

***The reviewer is correct, given the uncertainty in the MAC of at least ±50%, the propagated error would be approximately ±55%. We have corrected this in the revised text.***

Section 3.1: 1.
I am not sure I understand the sentence "(with the exception of the SSA that is the average, not the average maxima)". This becomes a bit clearer later on in the same page.

***The Table has been changed to list the minimum rather than the maximum value for the SSA with an explanation now given in the text.***

2. Line 1, page 12553: I believe there should be no period and lower “t” after (2008).

***We couldn’t find this typo to which the reviewer is referring but we did find a misspelling of Stephens in a section further on.***

3. Lines 1 to 7, page 12553: Stephens et al. 2008 is cited 3 times, maybe once would be sufficient.

***Modified as recommended.***

4. Lines 10, page 12553: “displays” should be “display” probably, or otherwise “show” should be “shows” in line 24 for consistency.

***We had what the reviewer suggests originally but the ACP editor changed it.***

5. “A decrease in SSA indicates that there is proportionally more light absorption than scattering”. I think this is a confusing sentence as it seems almost to suggest that the scattering coefficient is > than the absorption coefficient in this case, meaning that the SSA would be <0.5. Obviously this is not the intent of the authors. The adverb “proportionally” seems to mitigate this issue, but I still think the sentence could be clarified.

***The revised text now reads: “A decrease in the SSA can be a result of decreases in $B_{scat}$ or increases in $B_{abs}$; however, since $B_{scat}$ is seen to be relatively insensitive to seasonal changes, the changes with SSA are primarily a result of the seasonal sensitivity of $B_{abs}$, i.e. changes in the eBC***

6. Lines 5, page 12554: This implies that no other aerosol but BC is responsible for absorption, this is probably a very good assumption at 870 nm, but maybe it should be mentioned.

***The following sentence has been added: “It should be noted that other aerosols like certain organics, as well as dust, will also absorb light, but at shorter wavelengths than the 870 nm used by the PAX. Hence, the majority of the absorption measured in this study is by BC.***

Section 3.2:

1. Line 12, page 12554: see comment 4 in section 3.1 for “Figure 6a-c illustrates” vs. “. . .illustrate” for consistency.

***See response to comment 4. Apparently the editor disagrees with the reviewer and us.***

2. Line 2, page 12555: I do not think “virtual shift” is the most appropriate terminology here, because most of the human activities in the city are probably dictated by the “standardized” time and less by the “natural” time, while the changes in incoming solar radiation are obviously driven mostly by the “natural” time; therefore, there is in my opinion a “real shift”. . .

> ***We agree with the reviewer and had struggled originally when writing the manuscript to describe this shift. The knowledgeable reader should be able to understand our meaning so we have modified the text to read: “The shift in the time of the peaks between the cold-dry and other seasons is due to the shift in Mexico from daylight savings time (DST) the first Sunday in November to standard time then back to DST the first Sunday in April; however, the measurement time base does not shift with changes in DST.”***

3. “. . .the average eBC to CO ratio on workdays was 3.5 µgm-3 of eBC to 1.0 ppm of CO. This compared to the Sunday ratio that is 2.4 µgm-3 of eBC;. . .” does the 2.4 µgm-3 also refer to 1.0 ppm CO? If these are ratios of eBC to CO then the units should be µgm-3/ppm(?)

> ***Correct. This has been revised to “…2.4 µg $m^{-3}$ of eBC to 1 ppm of CO.***”

4. Line 27, page 12558: Minor comment: the eBC and Babs are directly and uniquely related by the MAC chosen in this paper so eBC is not really a proxy for Babs, it is exactly proportional to eBC.

> ***The text has been modified to the following: “These trends can be best understood by comparing the trends in $PM_{2.5}$, that is correlated with Bscat, and eBC and observing …”.***

5. Line 3, page 12561: “These” what? “These observations”? Or “These conclusions”? Or “These interpretations”?...

> ***Modified to “these interpretations”***

Table 1: Is there a reason to use the world “Maxima” plural of “Maximum” while all the other parameters are singular, e.g., “Average” vs. “Averages”?

> ***Good point. Change to “Maximum”, also changed in other parts of the text where appropriate.***

Figure 2: The text in the figure is a little bit blurry, it would be better to provide a higher resolution image. Is the Laser power monitor used for the extinction measurement during the calibration? If so, it might be good to write this explicitly in the text and in the caption.

***The new figure has new labels. The power monitor is used for the extinction measurement as the reviewer noted and we now have added that explicitly to the text.***

Figure 3: “absorption” should be capitalized in the y-axis for consistency. What does the “A” indicate on the top graph near the 1.5 value on the y-axis?

***Corrected as suggested. The “A” and “B” have been changed to (a) and (b) to label the two panels and correspond to the figure title.***

Figure 4 and 5: The font-size for the y-axis number as well as the x-axes labels is definitely too small and very difficult to read.

***Font sizes have been increased for better legibility.***

Figure 6: Add x-axis title for consistency with the other graphs in the following figures.

***For some reason the published version has chopped off the label of the figure we submitted. We will be sure to submit a figure that will fit this time.***

Figure 8: Y axis title on top graph should read PM2.5 not PM25. Bottom graph y-axis title, the closing bracket should not be a consistent size.

***Corrected as recommended***

**Response to Reviewer 2**

General comments:
The conclusions regarding the lack of change in BC concentrations over a 10+ year period seems to be stated a bit strongly, given uncertainties regarding differences in measurement artifacts by each method used (PSAP, PAX); combined with the short duration and inconsistent sampling strategy during which BC concentrations were characterized in the past. The main conclusions from the year-long measurements that the estimated eBC concentrations are alarmingly high, and the magnitude of eBC concentrations to traffic of the region (over all seasons regardless of different conditions of atmospheric mixing, and so on) is still valid, and the authors should take care to stress this analysis more than the decadal comparison, which appears to be written with more authority than the present study merits.

> ***In order to better explain why the BC concentrations have not changed with time, we now introduce statistics that show how the number of autos on the road have increased in the past 10 years. We also emphasize the large uncertainty in the measurements; however, our conclusions are supported by the data and our observations that a different mitigation strategy will be needed to diminish the BC concentrations are still valid.***

The rapid dispersion of eBC and PM2:5 is surprising; does this imply that the PAX measurement station is upwind of the bulk of traffic in Mexico City? This also suggests that averages of previous eBC measurements taken at different stations may not be comparable to the present site, if there is a strong spatial gradient in concentrations.

> ***The authors are not clear on what dispersion is being referred to by the reviewer here. If the reviewer is commenting on the afternoon dilution by the boundary layer growth, this is a tendency that is common to sites throughout the city. There are indeed strong spatial gradients between certains parts of the city, but studies of pollutants like CO, O3 and NOx from the RAMA monitoring stations show a very consistent diurnal variation. The winds in general are very light in Mexico City so that the terms "upwind" and "down wind" are not as relevant as in urban areas with stronger circulations.***

Can the authors comment on the general divergence in diurnal profiles BC, PM2:5, and SSA?

> ***The authors are not clear on the divergence to which the reviewer refers, unless it is the differences in diurnal patterns seen in in the eBC, PM2.5 and SSA. We go to great lengths in the manuscript to explain why these three parameters do not follow the same diurnal trends given the different processes that govern the behavior of BC versus particle mass.***

Given the debate about the "lensing effect" of scattering components of PM2:5 on the absorption of light by BC, is it surprising that the SSA decreases with the formation and condensation of photochemical oxidation products in the afternoon? For instance, Johnson et al. (2005) found that BC particles were rapidly coated by ammonium sulfate in the Mexico City (MC) urban environment, while Adachi et al. (2010) estimated the observed internal mixing configurations in particles from MC would have a weaker lensing effect than typical core-shell models.

Johnson, K. S., Zuberi, B., Molina, L. T., Molina, M. J., Iedema, M. J., Cowin, J. P., Gaspar, D. J., Wang, C., and Laskin, A.: Processing of soot in an urban environment: case study from the Mexico City Metropolitan Area, Atmos. Chem. Phys., 5, 3033-3043, doi:10.5194/acp-5-3033-2005, 2005.

Adachi, K., S. H. Chung, and P. R. Buseck (2010), Shapes of soot aerosol particles and implications for their effects on climate, J. Geophys. Res., 115, D15206, doi:10.1029/2009JD012868.

***In the revision we have discussed the lensing effect and potential impact on enhanced absorption.***

Minor comments:

Figure 4 and 5: lines should not be connected since the data presented is not necessarily sequential.

***Modified as recommended***

Figure 7a and b: the difference in units between the two figures is not explained. The seasonal differences would be made clear if precipitation and temperature are also included in Figure 7.

***Figure 7 is not composed of only the seasonal UV-A bar chart, followed by the diurnal cycles of temperature and RH, by season, that show more clearly the meteorological differences by season. A time series of rain from May to November has been added as Fig. 4 that shows the start and end of the rainy season. We have added this figure in response to Reviewer #1 who suggested we emphasize more about wet removal processes. We discuss these processes in greater detail now in the revised manuscript.***

Was there a size-selective inlet on the PAX? Presumably the bulk of black carbon mass is less than PM2:5, but may be worth mentioning as they are directly compared with each other.

***In the section describing the PAX, it was discussed that both a PM2.5 cyclone was used, as well as a dehumidifier.***

p. 12561, line 9-11. strange wording: "The very long integral time scale is connected to the same secondary processes that led to the greater than 10h time scales for the eBC and PM2:5 (Fig. 11b)."

***We have revised the wording for better clarity.***

p. 12561, line 14-16. "The correlations are higher in the dry months than in the rainy season because of the additional contribution of light scattering particles formed under conditions of high humidity." This wording is also not very clear, and would benefit from explicit mention of the components contributing to light scattering. This is both due to chemical products of aqueous-phase uptake and reaction and also water itself?

***We have revised the wording for better clarity.***

Figure 8 y-axis labels are not formatted correctly.

***Corrected as noted.***

p. 12543, Johnson et al. 2007 should be Johnson et al. 2005
***Corrected as noted.***