

We thank the reviewers for their thoughtful reviews, which helped us to improve our manuscript. Point-by-point responses are provided as follows.

### Reviewer 2:

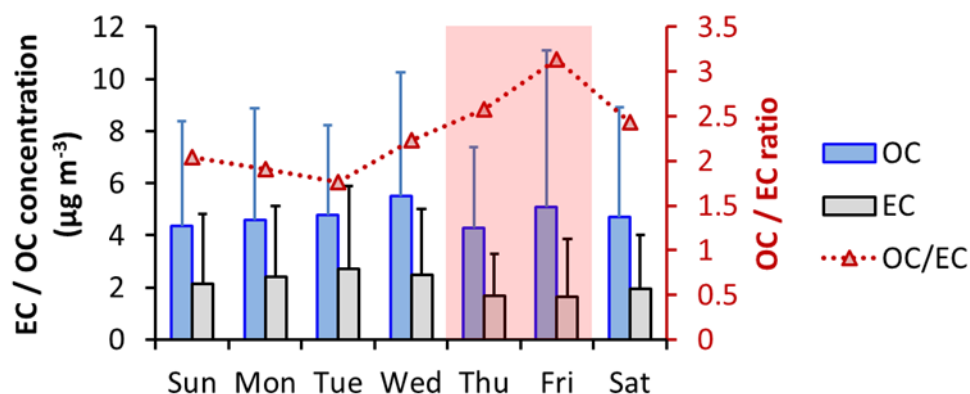
This is mostly an excellent submission, presenting results from a long-term measurement campaign in Saudi Arabia and source apportionment analysis. Questions I had were subsequently answered in the submission, which is usually a sign the authors have done a thorough job with the analysis. However, one glaring flaw appears to be that in 2012, the weekend in Saudi Arabia was Thu-Fri, not Fri-Sat. The authors should re-analyze their data accordingly. 2013 news article about the weekend switch: <http://english.ahram.org.eg/NewsContent/2/8/74730/World/Region/Saudi-Arabia-changes-working-week-to-SunThurs-Offi.aspx>

### Response:

Thank you to the reviewer for pointing out this flaw.

Upon re-analyzing the data for the correct weekend dates, we discovered that the analysis was correct (i.e., the weekend was defined as Thu-Fri) but the labels in the figure were incorrect. The axis label in Fig. 5 (now Fig. 6) was updated as shown below. The main text (lines 289-293) has also been corrected:

“To investigate whether a weekend effect could be discerned in the Riyadh dataset, two-sample t-tests assuming unequal variances were performed for hourly EC and OC samples, grouped according to whether they were obtained on weekdays (Saturday to Wednesday) or on weekends (Thursday and Friday).”



1. How often or when was the OC/EC filter changed? Was the OC/EC correction different at the beginning than at the end? Did the switch coincide with particular days of the week?

### Response:

OC/EC filters were changed after the laser intensity was reduced to 2000 or 3000 [a.u.] We have added a footnote to Table S1 to indicate this. The time to reach this threshold was mainly controlled by particulate concentrations, especially during dust storms. Some

filters lasted for only 24-48 hours, while some lasted for about 6-7 days. The OC/EC correction was the same over the entire filter lifetime, as long as there were refractive particles on the filters.

2. Lines 450-451 - the authors say the limited sample size means they can't quantify the local and regional contributions to OC and EC. However, this limitation only applies to the 24-hour metals analysis, which also appears to show that SOC, associated with Ca, may be regional. So couldn't the authors use the hourly-resolved OC/EC data to estimate local contributions to OC and EC?

*Response:*

We removed this sentence.

3. Table 1 could be rearranged to list the current study next to the 2007 middle-east study, as that is most relevant to the present analysis. I would have liked to see a more extensive comparison of the two sets of results.

*Response:*

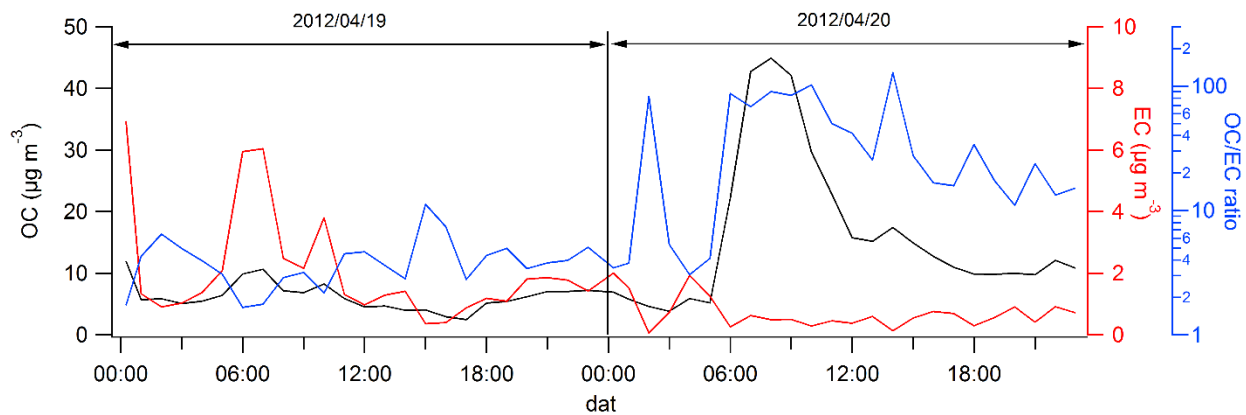
We have rearranged the table as suggested, revised the discussion of Table 1, lines 275-289.

4. Figures 2 and 3 just have EC/OC concentrations as the axis title, but the OC/EC ratios are also shown. Maybe put the ratio on the secondary axis with an appropriate title? Also, OC/EC ratios in the 100s - admittedly outliers - are interesting. Are those associated with low pollution levels?

*Response:*

Figs. 2 and 3 have been modified as suggested.

Most of the high OC/EC ratios (>100) were caused by rapid, large increases in OC and an accompanying decrease in EC during the measurement. An example is shown below, covering two consecutive days of data. On 2012/04/19, OC was  $6.24 \pm 2.27 \mu\text{g m}^{-3}$ , EC was  $2.08 \pm 1.79 \mu\text{g m}^{-3}$ , and OC/EC ratio was  $4.11 \pm 2.07$ ; on 2012/04/20, OC increased to  $16.09 \pm 12.20 \mu\text{g m}^{-3}$ , EC decreased to  $0.71 \pm 0.51 \mu\text{g m}^{-3}$ , and the OC/EC ratio increased to  $39.80 \pm 37.37$ . The highest OC/EC ratio on 2012/04/20 was 102. The total carbon on the second day was almost double that of the first day. This event may be a dust plume, as the methodology applied in the study cannot correct for carbonate interference in the OC/EC observations, although we have made inferences as to the presence of carbonate, as described in the text. While we do not have additional data to fully explore the causes of these excursions, we have no firm reason to remove these data, and thus we kept them in our dataset.



5. Figure 6(c) - Axis title is wrong. Also, the average ratios appear to be wrong, as almost all of them are higher than 75th percentile of the data. What do the caps represent - 90th or 95th or 99th percentile?

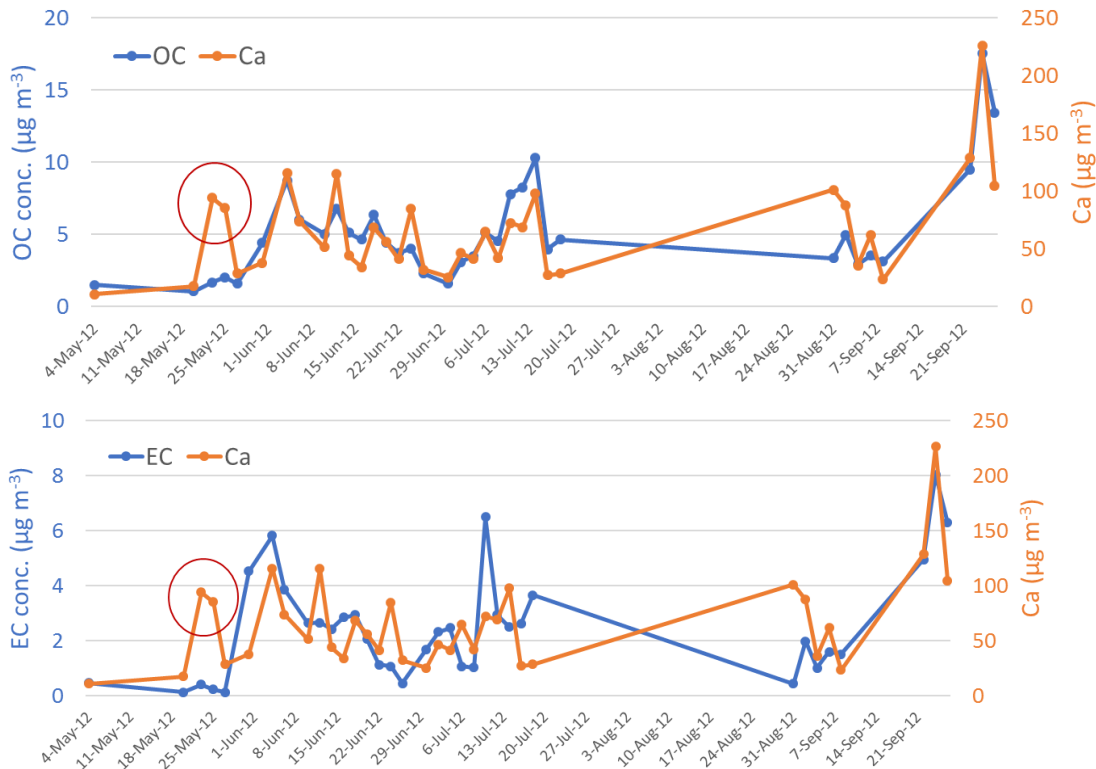
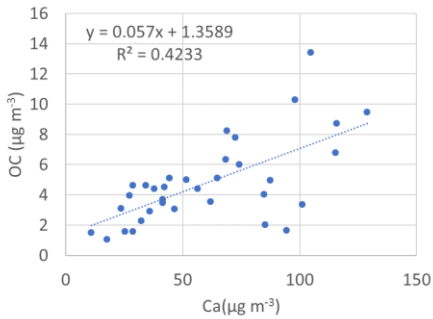
*Response:*

Thank you for pointing this out; we have corrected the y-axis title. The upper and lower caps represent 90<sup>th</sup> and 10<sup>th</sup> percentiles, respectively. The high average OC/EC ratios were caused by several individual high OC/EC ratios, as explained in comment 4. We retained the median and removed the point indicating the average in Fig. 5 (now Fig 6) to avoid confusion, and made similar changes to the other box-and-whisker figures.

6. Figure 8 - the high correlation between OC and Ca appears driven by a single high-value sample. Is that really good enough to push the OC-Ca connection?

*Response:*

Removing the high-value data point indeed decreased  $R^2$ , however, the time series of OC and Ca matched each other well and better than that of EC and Ca (see the figures below). The relationship between OC and Ca looks to be real, but what caused the relationship was uncertain as we discussed in the main text: whether a methodology artifact or that these species actually shared the same source origins. We have included these time series in the supporting material to illustrate this relationship and modified the sentence 368-370 as follows “However, OC had a relatively strong correlation with Ca ( $R^2$  of 0.63) (Fig. 8 and Fig. S7) but, similar to EC, a poor correlation with other dust species (not shown).”



7. Figure 11 - what happened to the samples in August? Also, maybe the Aug 31 sample should be grouped with September?

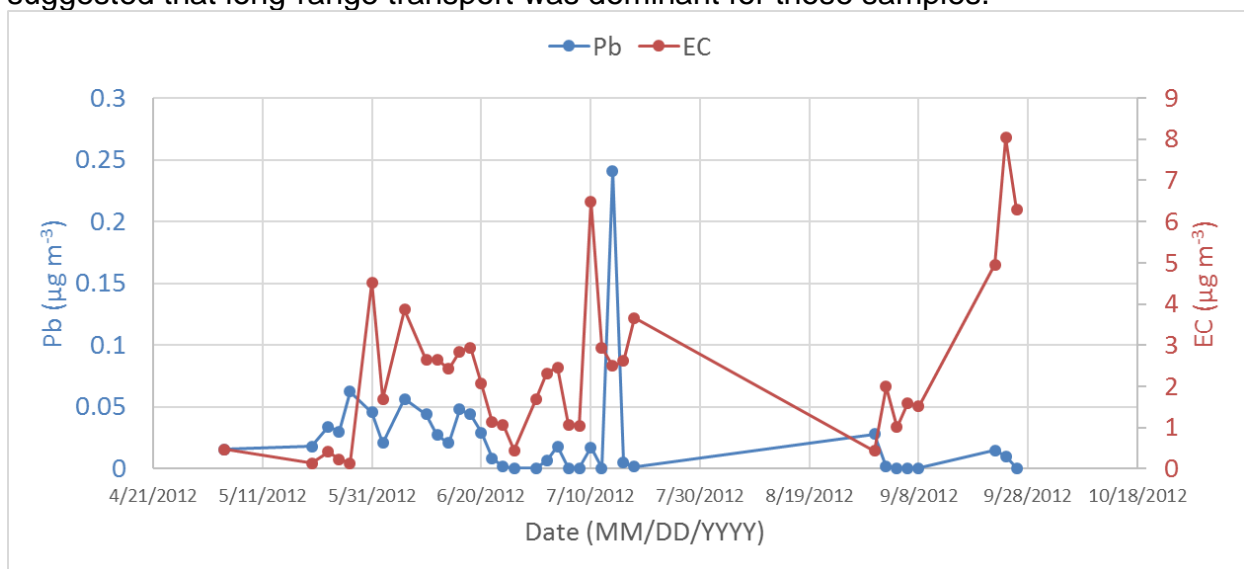
*Response:*

In 2012, Ramadan and Eid ran from July 20-August 24 and no measurements were made during this period. Thank you for the suggestion to regroup the Aug 31 sample with the September samples. We have done this and updated the numbers in lines 474 to 478. "The contribution of the mixed source ranged from 37% in May ( $0.7 \mu\text{g m}^{-3}$ ) to 95% in September ( $6.7 \mu\text{g m}^{-3}$ ). The EC concentration was also mainly attributed to the mixed source ( $1.9 \mu\text{g m}^{-3}$ , 92%)."

8. Figure 11 - was there no cement or gas flare or local vehicular contributions in May? That seems inconceivable. The authors should explain a bit more.

*Response:*

EC was indicated as a tracer for the mixed sources and Pb was the tracer for long-range transport, according to the PMF-resolved source profile (Figure 10). Daily-average EC concentrations were relatively low while Pb concentrations were high in May samples compared with those in the other periods, as shown in the following figure. This suggests that long range transport was dominant at that time. Due to the limited number of samples, PMF was unable to pick up the low contribution from mixed sources, with the result that minimal cement / gas flare / local vehicular contributions were found for most May samples. Lines 478 to 480 are revised as follows, “In some May samples, the mixed source contribution was negligible, as the source tracer, EC, was only 0.1-0.4  $\mu\text{g m}^{-3}$ , about one order of magnitude lower than that in other periods. The tracer analysis suggested that long-range transport was dominant for those samples.”



9. Figure A.2 shows that all the corrected laser values increase in transmittance at the beginning, which is rather strange - no EC should have left the filter in He1! What is going on - is the correction wrong?

*Response:*

Figure A.2a shows a thermogram for one of the “aged” blank samples. As expected, negligible carbon was detected in this sample, but the refractive particles that remained on the filter from previous samples still significantly influenced the laser variation. Fig. A.1a shows that the quadratic fit could not reproduce the signal at low temperature (<200 °C), so the increasing corrected laser signal during the He1 and He2 phases is likely an artifact of the correction methodology, and may not indicate EC evolution. However, we note that the studies of Wang et al. (2010) and Bladt et al. (2012) showed that refractory metal oxides / salts may cause premature EC evolution in an inert environment, thereby increasing the laser signal. A high loading of dust metals on the filters in our Riyadh samples may thus have caused premature EC evolution during the He stage. If this were the case, the original variable laser signal that we showed was

dependent on temperature may have masked that phenomenon, while the corrected one revealed it.

We revised the main text from lines 561 to 570 as follows:

“It is noted that although the quadratic equation correction produced a better laser signal for purposes of the carbon analyses, this correction did not work perfectly in the low temperature He phase, where the corrected laser signals exhibited unexpected increases. However, this shortcoming did not substantially influence the accuracy of the correction during subsequent carbon evolution. We note that premature evolution of EC, leading to an increasing laser signal in the inert environment due to the existence of refractory metal oxides, was observed in the studies of Wang et al. (2010) and Bladt et al. (2012). The increases in the corrected laser signal during the He stage in this study may be partially due to the same cause, as Riyadh samples contained abundant metal oxides.”

Reference:

Bladt, H., Schmid, J., Kireeva, E.D., Popovicheva, O. B., Perseantseva, N. M., Timofeev, M. A., Heister, K., Uihlein, J., Ivleva, N. P., Niessner, R: Impact of Fe Content in Laboratory-Produced Soot Aerosol on its Composition, Structure, and Thermo-Chemical Properties, *Aerosol Sci. Tech.*, 46, 1337-1348, DOI:10.1080/02786826.2012.711917, 2012.

Wang, Y., Chung A., Paulson, S.E.: The effect of metal salts on quantification of elemental and organic carbon in diesel exhaust particles using thermal-optical evolved gas analysis, *Atmos. Chem. and Phys.*, 10, 11447-11457, 2010.