Review of Chiloane et al acp-2016-934

General comments

This paper presents the collation and analysis of equivalent black carbon (eBC) and elemental carbon (EC) data measured at several locations in the northern interior of South Africa. The paper includes an assessment of spatial variability across 8 locations and a detailed investigation of the contribution of several sources of eBC at one location. The analysis uses seasonal and diurnal climatologies and multiple regression analysis to indicate the contribution of industrial sources, traffic emissions, household combustion, Savannah and grassland fire plumes to eBC loadings.

This paper reports on eBC and EC data from an under-sampled region of the world and the approach used to analyse the dataset is sound and innovative given the paucity of support data. However before publication a few issues need to be addressed.

The authors should discuss and review the issue of the difference in EC and eBC and discrepancies that are found when the two methods (MAAP and thermal evolution) are compared. This is particularly important since the authors use both data sets to describe spatial variability in the data. It is important to ensure that the spatial differences observed are not simply due to bias introduced by the different measurement methodologies.

In a number of places the explanations and discussion is repetitive and circular and could be simplified. I have indicated these areas in the detailed comments below.

Detailed comments

Page3 line 30- list some of the assumptions in modelled aerosol radiative impact assessments, particularly the ones associated with BC.

Figure 2 and Section 2.5 (Page 9) How was the baseline BC determined? Was it a constant value at each site? What method did you use for the EC correlation analysis to identify sources at the EC sites?

Page 11 line 9 remove of

Page 12 line 13 This has been observed everywhere so it may be worth stating "as expected"

Page 12 Section 3.2.1 what is the influence of atmospheric stability? Is there greater stability and therefore less mixing during the winter months in South Africa as seen in other places (e.g. SE Australia)? Could this also be contributing to higher winter concentrations? Suggest an assessment of windspeed climatologies could provide information on this. I note that this atmospheric stability is discussed in section 3.2.2.

Page 13 line 7- this explanation can be simplified e.g. "The Elandsfontein diurnal plot indicates highest concentrations occur in the evening hours (18:00 to 24:00). The area in which Elandsfontein is situated, is a well-known international NO2 hotspot (Lourens et al., 2012) and it is widely accepted that NO2 in this hotspot mainly originates from coal-fired power stations. However the timing of the NO2 and eBC peak concentrations differ by several hours with the NO2 peak occurring at 11:00, so that eBC is most likely not due to emissions from the coal-fired power stations."
Also since this is discussed in a lot more detail in section 3.3.2 (where it appears the contribution of the power stations is considered) authors may consider rewriting this paragraph to show that the role of power stations as a source will be considered later in the analysis and are not completely ruled out.

Page 15 line 2 what about household combustion for cooking? Presumably that occurs all year round?
Page 16 Line 14 - This section needs to be clarified. For example, in section 3.2.2 because the NO2 and eBC diurnal patterns did not match, power stations were ruled out as source of eBC in this region. However on line 21 page 15 the authors suggest that "Although it is not shown here, eBC plumes that were associated with these species were confirmed to have originated from coal-fired power stations with back trajectory analyses" and that "From literature, it is known that plumes from coal-fired power plants on the South African Highveld are characterised by coincidental SO2, NO2 and NO increases (Collet et al., 2010; Lourens et al., 2011). Do these statements contradict the interpretation made in the Section 3.2.2? Perhaps show the evidence of the association between EBC, SO2 and NO2 and the trajectory analysis relating these to the power stations.
Page 17 line 18 suggest replacing "thereof" with "of which".
Page 17 line 20 suggest replacing "thereof" with "of these pollutants"
Page 17 line 22 replace "have" with "has"
Figure 9a, 10a, 13a, from the text in the manuscript it's not clear what is being plotted in these trajectories. The figure captions suggest that only trajectories were eBC and the other pollutant of interest are elevated are plotted. If this is correct the text in the manuscript associated with these plots needs to be clarified.
Page 18 line 3 Replace "Similar to what was done for large industrial point sources" with "similar to the analysis performed for the large industrial point sources".
Page 18 Line 12 suggest re-writing this sentence e.g. "Household combustion results in the emission of a number of different species (Venter et al., 2012). In this work tracers for household combustion were determined from species that simultaneously increased with eBC, including NO2, SO2 and H2S. Note that NO did not increase simultaneously with increased with eBC".
Page 18 Line 17 add used after i.e. commonly used
Page 18 Line 18 suggest replacing "thereof" with "of this coal";
Page 18 Line 22 replace "have" with "has"
Page 20 line 15 remove "However"
page 20 Line 23-27 and Figure 15 More discussion is required about what these ratios indicate. Why were particular species selected to ratio against? Suggest moving this figure and section to supplementary as currently it adds little to the papers conclusions.
Fig 1 specify in fig caption the site

Figure 5 is overall really annual?

Figure 9 what criteria were used to determine if H2S was elevated?

Figure 10 what criteria were used to determine if NO2 was elevated?

Figure 13 what criteria were used to determine if NO2, SO2 and H2S was elevated?