Point-by-point reply to referee comments

Paper: Spatial, temporal and source contribution assessments of BC over the northern interior of South Africa (acp-2016-934)

We thank Referee #1 and #2 for their detailed reviews of the manuscript. We have improved the manuscript by incorporating the comments and remarks of the referees and believe the manuscript has gained in clarity and scientific soundness. Below is a point-by-point reply (in blue font) to the comments of the two referees (in black font). Please note that each referee comment (and associated reply) is numbered, to enable cross referencing between comments of the two referees.

1. Anonymous Referee #1

"General comments"

1.1 *"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 thank Referee #1 for the positive remarks. We believe all the issues were adequately addressed in the revised version.

1.2 *"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."*
The authors agree that differences between EC and eBC must be indicated to the reader. The text was modified, as indicated by the screenshot below.

3.1 Spatial variation

In Figure 4, a box and whisker plot indicating the statistical eBC or EC mass concentrations for each of the sites is presented. The significant difference in number of samples (N) is due to the fact that at the DEBITS sites EC mass concentrations were only measured once per month over a 24-sampling period, whereas at the other sites, one-minute eBC data were collected that were converted to 15 min averages. Precaution should also be taken when directly comparing eBC and EC, since it was previously proven that eBC and EC concentrations can differ by up to a factor of 7 among different methods, with a factor of 2 differences being common (Watson et al., 2005). However, an unpublished 12 month intern-comparison of eBC and EC at the Welgegund measurement site, with the actual sampling and analysis equipment used to acquire data for this study, proved that EC and eBC were within the same order of magnitude (Sehloho, 2017). Therefore, notwithstanding the limitations in directly comparing EC and eBC data, Figure 4 gives the most realistic spatial perspective for the northern interior of South Africa, especially within the context of very little other data being available in the peer reviewed public domain.

Insert Figure 4

1.3 *"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."*
Thanks to Referee #1 for pointing out these issues, they were all addressed as indicated below.

"Detailed comments"

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

Referee #1 is correct in stating that it would be advantageous to indicate some assumptions. Therefore the text was changes. Below is a screenshot indicating these changes.

> Presently, the majority of aerosol radiative impact assessments are based on models (Bond et al., 2013; IPCC, 2013), both on local and global scales, which incorporate measured aerosol properties. However, this approach involves several assumptions (e.g. assuming aerosol properties and the use of global instead of regional emission inventories for under sampled/characterised regions).~~,~~ Considering the relatively short atmospheric lifetime of BC, such assumptions~~which~~ could lead to significant uncertainties, especially on regional scales (Andreae and Gelencser, 2006; Masiello, 2004; Bond et al., 2013; Kuik et al., 2015). For a better understanding of the transport, removal and climatic impacts of atmospheric BC, accurate and up-to-date measurements covering large spatial areas and long temporal periods are required.

1.5 *"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?"*

Firstly, the method described in Section 2.5 and Figure 2, was only applied to sites where active eBC was measured and not to sites were EC was measured. However, the questions asked by Referee #1 made the authors realise that the text should clarify this better to the reader. The text screenshot below indicates the changes made to the relevant section.

> **2.5 Linking ground-based measurements with point sources using back trajectories**
>
> This method was introduced by Maritz et al. (2015) who used it to link ambient organic carbon (OC) and EC concentrations to potential sources. The same method was applied here, to assess if large point sources and in- or semi-formal settlements contributed to ambient eBC concentrations at the sites where active eBC data was gathered (Elandsfontein, Welgegund and Marikana). The method was not applied to sites where 24-hour composite EC samples were taken (Louis Trichardt, Skukuza, Vaal Triangle, Amersfoort and Botsalano). The method relates eBC concentrations measured at a particular sampling site with the closest distance

Secondly, as correctly indicated by Referee #1, the baseline BC was not properly defined in the text, although it was correctly indicated in the figure. Therefore, the text associated with Figure 3 was augmented to clarify the matter.

> therefore associated with the source that emitted the H_2S. For each such plume the excess eBC (Δ eBC) was determined, with the baseline defined as the linear line between the start end eBC concentrations of the observed plume and~~with~~ Δ eBC defined as the eBC concentration above the baseline, as indicated in the top pane of Figure 3.

1.6 *"Page 11 line 9 remove of"*

Thanks for Referee #1 for pointing out this text error, it was corrected.

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

The authors agree and have added the words "*…, which was expected.*" to the end of the relevant sentence.

1.8 *"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."*
Referee #1 is correct in stating that atmospheric stability has an influence. This was not left out, but discussed in the next section (3.2.2). To avoid repetition of text/ideas, a sentence was added at the end of the paragraph to indicate to the reader that this will be discussed in greater detail in the next section.

As is evident from these figures, there is a distinct and similar seasonal pattern observed at all three sites, with the highest eBC mass concentrations measured in June to October. These months coincide with the colder winter months of June to August, as well as the dry season on the South African Highveld occurring between May and middle October. Venter et al. (2012) previously indicated that household combustion for cooking and space heating in informal and semi-formal settlements during winter could be a significant eBC mass concentration source on a local scale. However, it has not yet been determined whether such household combustion could also make a significant regional contribution in South Africa. During the dry season, increased savannah and grassland wild fires occur, which contributed to increased atmospheric eBC concentrations (Bond et al., 2004, Saha and Despiau, 2009). The influence of both of these potential eBC sources, i.e. household combustion and wild fires, will be discussed later in Section 3.3. Obviously, increased atmospheric stability during the colder months (Garstang et al., 1996) will also lead to trapping of low level emissions, hence resulting in possible higher eBC concentrations. This is discussed in greater detail in the next section.¶

1.9 *"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."
The authors agree with Referee #1 that this text needed to be improved. Referee #2 also indicated this, but requested additional clarification on certain issues. Please refer to Correction 2.6, which indicates in detail the changes made to the relevant text.

1.10 *"Page 15 line 2 what about household combustion for cooking? Presumably that occurs all year round?"*
Referee #1 is correct in stating that household combustion for cooking will still take place in the hotter months. Therefore household combustion referred to here was specified, i.e. for space heating. An additional sentence was also added to clarify the use of household combustion for cooking during the summer months. The screenshot below indicates the text changes.

will not influence eBC levels in the northern interior significantly. In addition, minimal household combustion for space heating takes place in December to February, since it is the warmest months. During this time household combustion for cooking will still take place, but such daily emission periods are far shorter than the extended space heating period (typically early evening, throughout the night, until after sunrise the next day) occurring during the colder months. Considering the afore-mentioned, it is best to isolate industrial and traffic related eBC sources during December to February.¶

1.11 "*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.*"

The authors do not agree with Referee #1 that "power stations were ruled out as a source of eBC" in Section 3.2.2. The text in Section 3.2.2. of the revised version reads "*The Elandsfontein diurnal plots indicate that the **main source** of eBC is not high stack emissions.*" This clearly indicates that the power station is not the main source, but they might still contribute. However, to clarify the matter even further, the text on page 16, line 14 (in the original version) was modified as indicated below.

> Although indicated in Section 3.2.2 that it was unlikely that high stack emissions were the main source of eBC at Elandsfontein, the possible fractional contributions of industries still need to be assessed. In order to quantify th~~is~~e ~~relative contribution of large point sources at Elandsfontein~~, eBC peaks that coincided with peaks of other pollutants, which are characteristic of large point sources in that area, were considered for the December to February period. Two

1.12 "*Page 17 line 18 suggest replacing "thereof" with "of which".*"

The authors agree with this text change, although it was on page 18 and not on page 17.

1.13 "*Page 17 line 20 suggest replacing "thereof" with "of these pollutants"*""

The authors agree with this text change, although it was on page 18 and not on page 17.

1.14 "*Page 17 line 22 replace "have" with "has"*""

The authors thank Referee #1 for pointing out this grammar issue and have corrected it, although it was on page 18 and not on page 17.

1.15 "*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.*"

The authors agree with Referee #1 that the text should be clarified. The screenshots below indicate how the text sections associated with Figures 9a, 10a and 13 were improved.

For Figure 9a

> dumps that burn as a result of spontaneous combustion. In order to identify the origin of the eBC peaks that were associated with H_2S only, a map on which all back trajectories that arrived at Elandsfontein during these eBC peaks (coincidental increases in eBC and H_2S) were plotted, is presented in Figure 9, together with a wind rose for such events. From these figures, it is

For Figure 10a

> approximately 38 km to the north and north-west. It therefore seems reasonable that the traffic-related eBC back trajectory map (Figure 10a, which was for coincidental increases in eBC and NO_2 time periods only) is somewhat biased toward the east and north, although limited contributions from other sectors are also evident. The wind rose showing the prevailing wind

For Figure 13a

> Figure 13a indicates back trajectories associated with household combustion contribution to eBC levels (for time periods with coincidental increases in eBC with NO_2, SO_2 and H_2S, but not NO). Most of these back trajectories passed over the Thubelihle and Kriel settlements,

1.16 *"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"."*

The authors thank Referee #1 for this text improvement suggestion, which was incorporated.

1.17 *"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"."*

The authors thank Referee #2 for this text improvement suggestion, which was incorporated.

1.18 *"Page 18 Line 17 add used after i.e. commonly used"*

Referee #1 is thanked for pointing out this omission, which was corrected.

1.19 *"Page 18 Line 18 suggest replacing "thereof" with "of this coal";"*

This is exactly the same correction, as already indicated in Correction 1.12.

1.20 *"Page 18 Line 22 replace "have" with "has""*

This is exactly the same correction, as already indicated in Correction 1.14.

1.21 *"Page 20 line 15 remove "However""*

The authors would prefer to retain the word "However" within the context, since they want to indicate the difference between the sources that seasonal, as opposed to the sources that contribute year round.

1.22 *"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."*

We agree with Referee #1 that the text associated with Figure 15 was not good enough. However, we do feel that the emission factors presented in this figure are a very valuable scientific contribution. Since so little BC measurements are conducted in South Africa, these emission factors will enable modellers to estimate BC levels better. Therefore, if allowed, we would prefer that this paragraph and associated Figure remain in the main text. The text was improved, as indicated below.

Vakkari et al (2014) used Δ eBC in relation to other species to characterise differences in plumes of savannah and grassland fires. In a similar manner, these ratios for Δ eBC divided by ~~other~~ species that were characteristic of the different plume types identified (i.e. representing industrial, traffic or house hold combustion)~~reported in this paper~~ were determined and are presented in Figure 15. Since so little BC data is available for South Africa, t~~T~~he median and/or mean values indicated in this figure could be used in subsequent modelling studies as emission factors to estimate~~quantify~~ eBC if only the concentration(s)~~emissions~~ of the~~other~~ species that were used in calculating these ratios are known.

Insert Figure 15

If Referee #2, or the editor, insist that we move this short paragraph and Figure 15 to a supplement, we will do so.

1.23 *"Fig 1 specify in fig caption the site"*

The authors thank Referee #1 for the suggestion, which was incorporated.

1.24 *"Figure 5 is overall really annual?"*

It is assumed that Referee #1 refer to Figure 6 where the word "overall" was used, and not Figure 5 as indicated in the comment. We agree with Referee #1 that the word "overall" on its own does not explain the context correctly. We therefore modified the caption as indicated below.

Figure·6. → Overall·(all·the·data)·and·seasonal·(each·season·separately)·average·eBC·diurnal· patterns·observed·for·Elandsfontein,·Welgegund·and·Marikana.··Summer:·DJF,· Autumn:·MAM,·Winter:·JJA·and·Spring:·SON. Page Break ¶

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

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

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

These three comments are answered as one, since all relate to the same method (as explained in Section 2.6). There was no specific limit or limiting value applied in determining when any species was elevated. What was considered is whether coincidental increases of a species (or more species) with eBC occurred, as indicated in Section 2.6. According to the authors it is important not to apply a limiting value above which a specific species was regarded as elevated, since doing so would bias the data. For instance, sometimes power plant plumes will have lower and sometimes higher concentrations of a specified species, but by including all values (low and high ones, as long as coincidental increases occurred with eBC) realistic "emission factors" (as presented in Figure 15 with the associated text) could be determined.

2. Anonymous Referee #2

"General comments"

2.1 *"This paper presents some very valuable measurements of Black Carbon made from South Africa, from a poorly sampled region of the globe. The data analysis in most part seems sound and the work is valuable, however in my opinion the paper needs some tightening up before it is suitable for publication in ACP."*

The authors thank Referee #2 for the positive remarks. We believe that all the issues indicated were addressed in the revised version.

"Specific comments"

2.2 *"I suggest that the title uses the full term "black carbon" rather than shortening to "BC""*

The authors agree with this suggestion. The title now reads "Spatial, temporal and source contribution assessments of black carbon over the northern interior of South Africa"

2.3 *"The Abstract is long and introduces a lot of Acronyms that are later dispensed with. In particular the text is much easier to understand later in the document when the names of measurement sites are used in full, rather than shorten to initials. I suggest that the abstract is shortened perhaps by cutting down on the first paragraph of introductory text."*

We agree that the "Abstract" might be too long and that the first paragraph of introductory text is shortened. Below is a screenshot of the text changes made to paragraph 1 of the "Abstract". These changes resulted in a reduction of 84 words.

Abstract

After carbon dioxide (CO_2), aerosol black carbon (BC) is considered to be the second most important contributor to global warming. ~~Africa is one of the least studied continents, although it is regarded as the largest source region of atmospheric BC. Southern Africa is an important sub-source region, with savannah and grassland fires likely to contribute to elevated BC mass concentration levels. South Africa is the economic and industrial hub of southern Africa. To date, little BC mass concentration data have been presented for South Africa in the peer-reviewed public domain.~~ This paper presents equivalent black carbon (eBC) (derived from an optical absorption method) data collected from three sites in the interior of South Africa, where continuous measurements were~~have been~~ conducted, i.e. Elandsfontein ~~(EL)~~, Welgegund ~~(WG)~~ and Marikana ~~(MA)~~, as well elemental carbon (EC) (determined by evolved carbon method) at five sites where samples were collected once a month on a filter and analysed off-line, i.e. Louis Trichardt ~~(LT)~~, Skukuza ~~(SK)~~, Vaal Triangle ~~(VT)~~, Amersfoort ~~(AM)~~ and Botsalano ~~(BS)~~. ~~All these sites are located in the interior of South Africa.~~

The authors also agree that the use of measurement site name acronyms make the text more difficult to understand. Therefore, all site name acronyms were replaced with the full names in the "Abstract". This change does however make the "Abstract" longer, but it certainly does improve the clarity of the text.

After reconsidering the entire paper, the authors also realised that the site name acronyms were not consistently used in the rest of the paper. Therefore, to make the paper easier to understandable for the international reader (that might not know South Africa well), all site name acronyms were replaced by the full names, expect in the figures (e.g. Figure 4) that would become too crowded if full names were used. However, for such figures the acronyms were in the figure captions.

2.4 *"The introduction is clear as are the measurement site descriptions and methods."*

The authors thank referee #2 for the positive comment.

2.5 *“Page 12 – Figure 5 is confusing to me: did Welgegund measurements switch between PM10 to PM1 and back again during the time period shown?”*

Yes, measurements at Welgegund was switch between PM10 to PM1 and back again during the reported measurement period. Tiitta et al. (2014) reported on the chemical composition of “non-refractive submicron aerosols (NR-PM1)” at Welgegund, as measured with an Aerosol Chemical Specification Monitor (ACSM, Aerodyne Inc.). During the period when Tiitta et al. (2014) collected data with the afore-mentioned instrument, the BC inlet was changed to PM1 to correlate with the inlet used for the chemical measurements. However, the fact that the figure and associated text caused some confusion for Referee #2, implies that some text improvements are required to prevent the general scientific readers from being confused. The following changes were made:

- The text under Paragraph “*2.1.3 Welgegund” was modified and now reads “The Welgegund measurement station… A PM_{10} inlet was used from 1 June 2010 to 25 August 2010, as well as 1 September 2011 to 31 May 2012, while a PM_1 inlet was used from 26 August 2010 to 31 August 2011. The PM_1 inlet sampling period was undertaken to better quantify PM_1 aerosol chemical composition, which was reported in a previous paper (Tiitta et al., 2014).”*
- *The caption of Figure 5 was also changed to clarify the issue. Below is a screen shot of Figure 5 and the modified caption.*

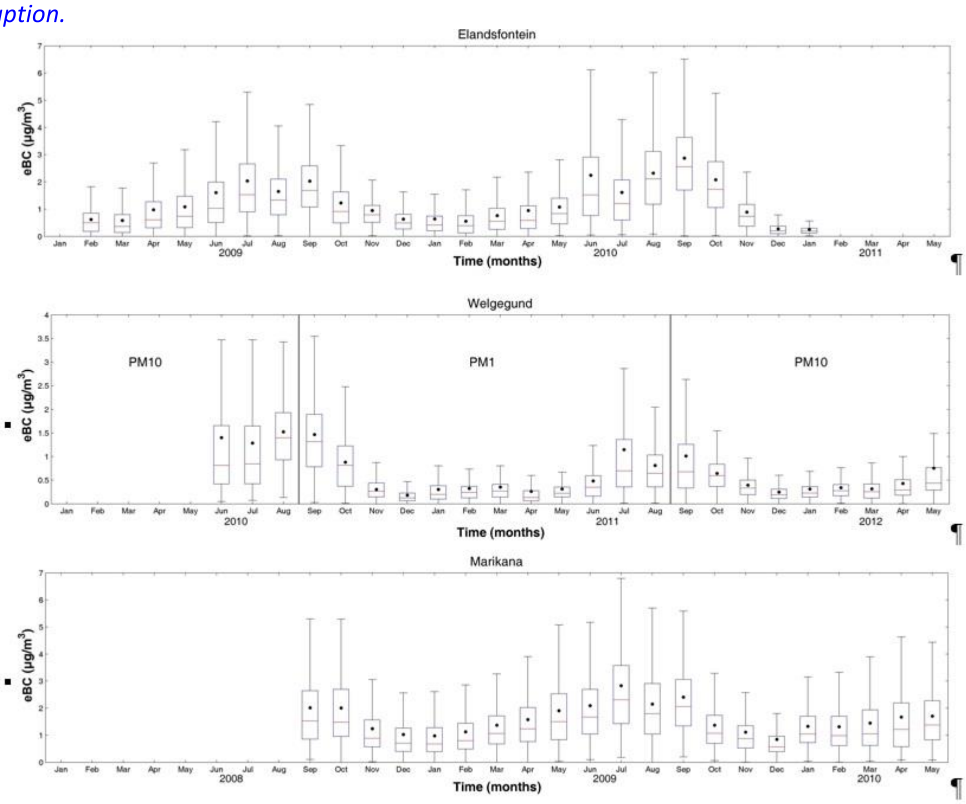

Figure 5. Monthly statistical distribution of eBC concentrations at the three sites where continuous measurement data were gathered, i.e. Elandsfontein, Welgegund and Marikana. PM_{10} inlets were used at Elandsfontein and Marikana, while measurements at Welgegund were conducted with either a PM_1 or PM_{10} inlet. The red line of each box is the median, the black dots indicate the mean, the top and bottom edges of the box are the 25th and 75th percentiles and the whiskers ±2.7σ (99.3% coverage if the data has a normal distribution).

2.6 *"Page 13. I am also confused by the arguments outlined here. They seem to say that eBC cannot be from the same source as the NO2 because they do not have the same diurnal cycles, however this is not obvious to me since NO2 may be photo-chemically produced from NO and does not have the same atmospheric lifetime as black carbon and so co-emitted species could have different diurnal patterns. Please clarify the reasoning here."*

The authors agree with Referee #2 that the explanation given here was not clear enough. To entire paragraphs was rewritten and now reads as indicated by the screenshot below.

The Elandsfontein diurnal plots indicate~~s~~ that the main source of eBC is not high stack emissions. ~~eBC would have peaked after 11:00, as has been indicated for NO_2 by Collet et al. (2010) if eBC originated mainly from industrial high stack emissions.~~ The area in which Elandsfontein is situated, is a well-known international NO_2 hotspot, with tropospheric column densities similar to what is observed over south-east Asia (Lourens et al., 2012; Lourens et al., 2016). It is widely accepted that NO_2 in this hotspot mainly originates from NO_x emission from coal-fired power stations. The troposphere over the Highveld is strongly layered, with several inversion layers occurring. These layers prevent vertical mixing to a large degree (Garstang et al., 1996). The afore-mentioned NO_x emission are released into the atmosphere via high stacks, which are typically taller than 300m. The effective stack heights (actual stack heights plus rise due to emissions being hot) were designed to ensure that the NO_x emissions are released above the lowest inversion layers, to prevent excessive local pollution and ensure distribution over a wider area. Collet et al. (2010) proved that NO_2 concentrations at Elandsfontein peak after 11:00 am, due to the breakdown of the lowest inversion layers, which allow downward mixing of the NO_x tall stack emissions. Therefore, if eBC mainly originated from these large point sources with tall stacks, eBC concentrations would also have peaked, ~~similar to NO_2,~~ after the breakdown of the night-time inversion layers that would allow downward mixing of tall stack emitted eBC. However, this is clearly not the case. Additionally, the winter diurnal plot for Elandsfontein indicates substantially higher values during night-time~~-~~ when the planetary boundary layer (PBL) is less well mixed (i.e. strong low level inversion layers that trap surface emissions), which re-enforces the notion that the major origin of eBC is from low-level sources, rather than industrial high stacks ~~that were designed to have effective stack heights above the low-level inversion layer heights~~. At Elandsfontein~~this site~~ the daily evolution of the PBL starts approximately three to four hours after sunrise (varies between 05:07 and 06:56 local time), which results in increasing atmospheric mixing down from the upper ~~atmosphere~~ troposphere, including high stack emissions (Korhonen et al., 2014). Considering all the afore-mentioned~~Therefore~~, the most likely eBC sources during winter (June to August) and the dry season (May to middle October) are~~can be attributed to~~ surface emissions from household combustion, and~~as well as~~ savannah and grassland fires~~, respectively, not industrial high stack emissions~~. The is an important finding, since industries on the Mpumalanga Highveld are often blamed for all forms of pollution, due to the NO_2 hotspot over this area ~~that is attributed to NOx emissions from industries and vehicle emissions from the Johannesburg Pretoria megacity (Lourens et al., 2012; Lourens et al., 2016)~~.¶

2.7 *"The use of different times of year to characterise the main sources is generally well explained, however by Page 16 the mention of the NO2 hotspot near Elandsfontein seemed repetitive. I think that some significant shortening of the text could be achieved with a re-write of this section and that this is likely to improve the clarity of the paper."*

Referee #2 is correct in stating that there is some repetition of ideas/text in this section, which was stated earlier. Several sentences were deleted and additional minor text changes made to remove the repetition. Below is a screenshot to indicate these changes.

3.3.2 Industrial contribution to eBC at Elandsfontein

Numerous large industrial point sources linked to coal utilisation occur in the South African interior, e.g. coal-fired power stations that produce most of South Africa's electricity, large petrochemical operations utilising coal gasification and numerous pyro-metallurgical smelters utilising coal and coal-related products as carbonaceous reductants for the production of various steels and alloys (Collet et al., 2010; Lourens et al., 2011; Beukes et al., 2012). ~~Previously, it has been indicated that some of these large point sources contribute significantly to certain pollutant concentrations, e.g. the NO_2 hotspot observed with satellite observations over the Highveld, mainly due to coal-fired power stations that do not de-SO_x or de-NO_x and traffic emissions (Lourens et al., 2012).~~ However, the possible contributions of these large point sources to atmospheric BC ~~concentrations~~ have not yet been investigated ~~for South Africa~~.

~~As previously indicated, Elandsfontein is situated within the well-known NO_2 hotspot, with various large points sources located in close proximity (Collet et al., 2010; Lourens et al., 2011). The diurnal eBC concentration plots of Elandsfontein (Figure 6) indicated that it is unlikely that industrial high stack emissions were the main source of eBC at this site. However, this postulation has to be proven.~~ In Figure 8, eBC concentrations measured at Elandsfontein were plotted against the shortest distances that back trajectories passed any large point source, during the summer months (December to February), when minimal household combustion, as well as savannah and grassland fires occur. Although there was no clear correlation (Figure 8), the results indicated that at least some trajectories passing closer to these large industrial point sources had higher eBC concentrations. This suggests that eBC contributions from large industrial point sources cannot be ignored, notwithstanding the diurnal patterns, indicating that high stack industrial emissions were not the main source (Figure 6).

2.8 *"Page 21, Figure 17 – it is not clear that the word "predict" is suitable here, because the text seems to imply that the whole dataset is used to generate the equation. Did I misunderstand and a subset is used to create the equation and then used to predict some later observations? Please clarify the text."*

Referee #2 is correct in stating that the word "predict" is not suitable for this context. The word "predict" was replaced with "calculate" at both places where it was used.

2.9 *"The conclusions section is clear, but maybe could be renamed Summary and Conclusions to better represent the contents."*

The authors agree with this suggestion. The header was renamed "Summary and Conclusions"

2.10 *"Typo: Pg11 line 5, "experience" should be "experiences""*

This typo was corrected.