Response to Reviewer 2's Comments:

We thank the reviewer for their comments. The reviewer's comments are in black text and our responses are in red text. Any additions to the manuscript are in blue text. Reference to page and line numbers refer to the original manuscript.

Overview: The paper provides a comparison of PAN measurements from the MIPAS instrument on ENVISAT produced using two different retrieval methodologies. It also includes a comparison of the retrievals (and a suite of aircraft profiles) to a TOMCAT simulation of PAN. The paper is technically strong, and well written. Unfortunately, the paper does not really further our understanding of the sources of PAN in the atmosphere in any significant way. This makes it difficult to review, because there are no substantive conclusions drawn. That being said, this paper does represent the first presentation of a new dataset, which could have significant value. Thus, I recommend publication of the paper, but recommend that the authors consider the following idea for improvement.

We are happy to read that the reviewer feels the manuscript is suitable for publication in ACP. We would argue though that there are substantial scientific conclusions in our paper. Both the IMK and UoL groups have published papers on their MIPAS PAN retrievals and other studies have used this data (e.g. Fadnavas et al., 2014) or cited it (e.g. Fischer et al., (2014)). Therefore, our study highlights that there are inconsistences (page 20, lines 25-28) between these datasets and this needs to be acknowledged by future studies when discussing PAN concentrations in the UTLS retrieved from satellite. We also provide potential reasons for this in section 3.2. And finally, as the reviewer points out, we do include a new dataset (i.e. global investigation of PAN retrieved from ACE-FTS) in the manuscript.

Both datasets (Figure 2 and Figure 3) show a consistent PAN maximum in the tropical UT in SON. Can the authors use the data to attribute this feature? Casual mention is made throughout the text that the source is biomass burning, but no evidence is provided that this is the source. What does the model attribute this feature to? Is there a way to use any other simultaneously retrieved tracers (i.e. HCN or CO) to better understand whether the presence of this feature is driven by biomass burning or by lightning NOx forming PAN in the presence of isoprene oxidation products lifted in convection?

This is a good point as both mechanisms will result in PAN formation. Unfortunately, as shown in Figures 12c and 13c, the model struggles to capture the enhanced PAN signal over the tropical UT in SON with significant negative biases in both cases. Therefore, it is difficult to use the model to try and diagnose the potential source of the enhanced PAN in this region.

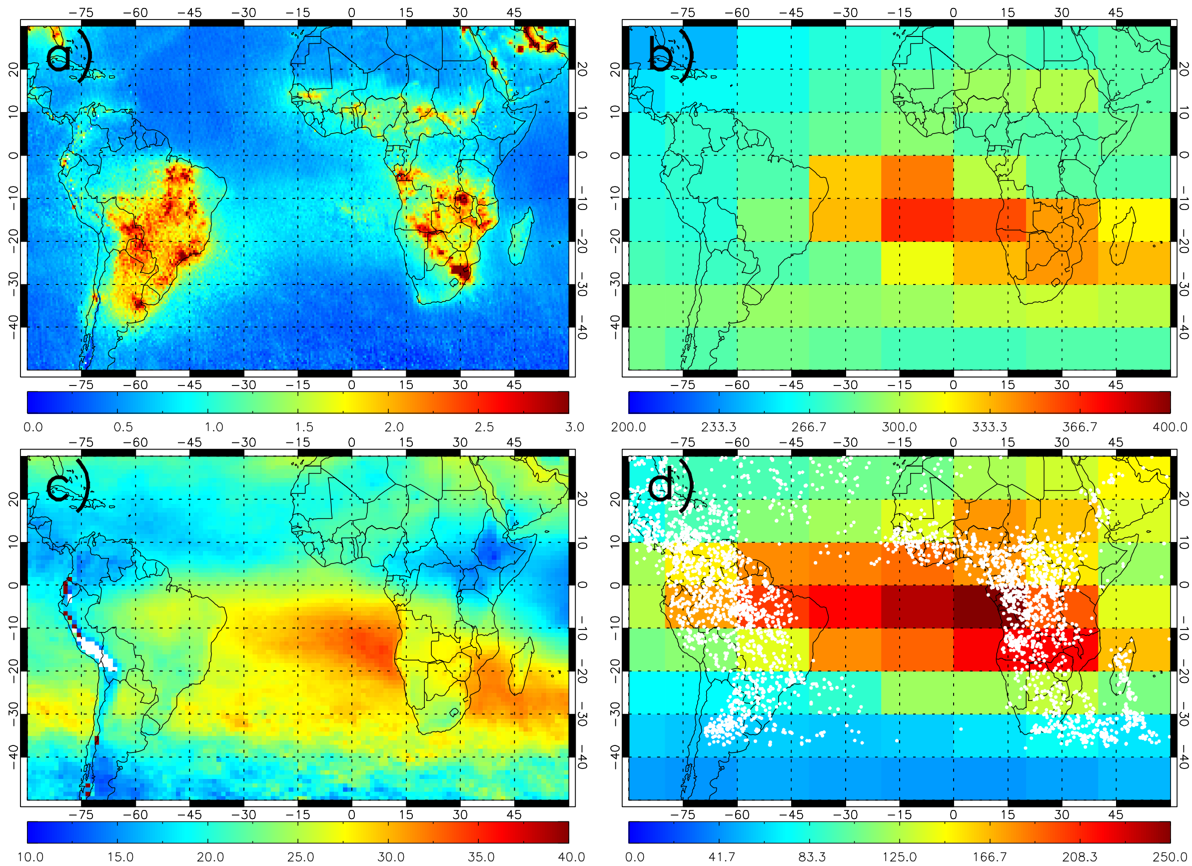

Figure SI: Atmospheric composition over the tropical South Atlantic observed by satellite in Sept-Oct-Nov 2008. a) Ozone Monitoring Instrument (OMI; Boersma et al., 2011) tropospheric column NO_2 (10^{15} molecules/cm^2), b) OMI-MLS (Microwave Limb Sounder; Ziemke et al., 2006) tropospheric O_3 (DU), c) Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) HCN (pptv) at 150 hPa and d) MIPAS PAN (pptv) at 150 hPa (Glatthor et., 2007). White circles represent locations of significant lightning events detected by the Lightning Imaging Sensor (LIS; Cecil et al., 2014).

In Figure SI, we have plotted SON 2008 averages of OMI tropospheric NO_2, tropospheric OMI/MLS O_3 and MIPAS HCN and PAN at 150 hPa. From Figure SIa), there are clear source regions of NO_2 over both southern Africa and South America, with transport of NO_2 out into the tropical South Atlantic. Tropospheric O_3 (Figure SIc) has a similar and more distinct pattern over the Atlantic. As enhanced MIPAS PAN (Figure SId) is also seen in this region, NO_2, O_3 and PAN are highly correlated. The gradient in retrieved tropospheric O_3 across the tropical Atlantic, suggests transport of PAN and NO_2 and subsequent O_3 formation, primarily from southern Africa (i.e. the ozone gradient is from East to West across the Atlantic; Figure SIc). Figure SIb shows MIPAS HCN, with peak concentrations over the southern Atlantic. HCN is produced by biomass burning with a tropospheric lifetime of approximately 5 months (Li et al., 2009). The HCN distribution suggests African biomass burning sources play a key role in the UTLS PAN budget in this region. White circles in Figure SId show locations of significant lightning events (> 5 x 10^6 W/ster/m^2/μm). These are clustered mainly over Southern African and South America. These are also potential sources of NO_2 in the mid-upper troposphere in this region. Belmonte Rivas et al., (2015) show that NO_2 sub-columns in regions of lightning activity, using a cloud-slicing technique, range between 0-0.5 x10^{15} molecules/cm^2, which can make up a significant proportion of the tropospheric column over the South Atlantic/West African coastline (e.g. 0.5-2.0 x10^{15} molecules/cm^2). Fischer et al., (2014), through a modelling study, suggest that lightning NO_x emissions can lead up to 50-60% of PAN formation in October total column. Therefore, we suggest that both pathways (biomass burning and lightning) lead to the enhanced PAN over the tropical Southern Atlantic.

In order to incorporate some of this discussion into our analysis, we have modified the following text in the paper:

Pages 9-10, Lines 20-22, 1 – "During SON, large PAN concentrations over the South Atlantic (>170 pptv) and along the SH dynamical tropopause (approximately 100 pptv) are linked to outflow from the African biomass burning plume" is replaced with "During SON, large PAN concentrations over the South Atlantic (>170 pptv) and along the SH dynamical tropopause (approximately 100 pptv) are linked to outflow from the African biomass burning plume and from lightning-generated NO_2 in the mid/upper troposphere. As shown by Belmonte Rivas et al., (2015), using a cloud slicing technique, there are significantly large sub-columns of NO_2 in the mid-upper troposphere co-located with lightning activity. In addition, deep convection transports African biomass burning emissions efficiently to the UT in this region. Fischer et al., (2014) indicate that up to 50-60% of PAN formation in the total column can be attributed to lightning NO_2 emissions in their modelling study. IMK MIPAS retrievals of HCN (see Supporting Information; SI), which is a long-lived tracer (5 months; Li et al., 2009) sourced from biomass burning, also shows a strong correlation with PAN in this region. Therefore, it appears that both lightning NO_x and biomass burning act as sources of PAN in this region. This is discussed further in the SI.".

We have added Figure SI as Supporting Information with the following text "From Figure SIa, there are clear source regions of NO_2 over both southern Africa and South America, with transport of NO_2 out into the tropical South Atlantic. Tropospheric O_3 (Figure SIc) has a similar and more distinct pattern over the Atlantic. As enhanced MIPAS PAN (Figure SId) is also seen in this region, NO_2, O_3 and PAN are highly correlated. The gradient in retrieved tropospheric O_3 across the tropical Atlantic, suggests transport of PAN and NO_2 and subsequent O_3 formation, primarily from southern Africa (i.e. the ozone gradient is from East to West across the Atlantic; Figure SIc). Figure SIb shows MIPAS HCN, with peak concentrations over the southern Atlantic. HCN is produced by biomass burning with a tropospheric lifetime of approximately 5 months (Li et al., 2009). The HCN distribution suggests African biomass burning sources play a key role in the UTLS PAN budget in this region. White circles in Figure SId show locations of significant lightning events (> 5×10^6 W/ster/m^2/µm). These are clustered mainly over southern African and South America. These are also potential sources of NO_2 in the mid-upper troposphere in this region. Belmonte Rivas et al., (2015) show that NO_2 sub-columns in regions of lightning activity, using a cloud-slicing technique, range between 0-0.5 $\times 10^{15}$ molecules/cm^2, which can make up a significant proportion of the tropospheric column over the South Atlantic/West African coastline (e.g. 0.5-2.0 $\times 10^{15}$ molecules/cm^2). Fischer et al., (2014), through a modelling study, suggest that lightning NO_x emissions can lead up to 50-60% of PAN formation in October in the total column. Therefore, we suggest that both pathways (biomass burning and lightning) probably lead to the enhanced PAN over the tropical Southern Atlantic.".

Page 10, Line 13 – "apart from the African biomass burning signals of approximately" with "apart from the African biomass burning/lightning NO_x signals of approximately".

Page 11, Line 19 – "linked to biomass burning over central and southern Africa" with "linked to biomass burning/lightning NO_x over central and southern Africa".

Page 11, Line 25 – "Similar biomass burning signals occur in MAM and SON" with "Similar biomass burning/lightning NO_x signals occur in MAM and SON".

Page 17, Lines 8-12 – "In SON, TOMCAT misses PAN associated with biomass burning plumes from southern Africa, which propagate out into the South Atlantic. In the IMK and UoL PAN products, PAN concentrations range from 150-200 pptv, while they are only 100-120 pptv in the TOMCAT PAN distribution." with "In SON, TOMCAT misses PAN over the South Atlantic, which is likely associated with biomass burning outflow and lightning NO_x from southern Africa. In the IMK and UoL PAN products, PAN concentrations range from 150-200 pptv, while they are only 100-120 pptv in the TOMCAT PAN distribution. This low model bias means that it is difficult to use the model to diagnose the relative contributions of biomass burning and lightning NO_x to the formation of PAN in this region and season.".

Page 18, Lines 6-7 – "produced from NO_x biomass burning emissions, which is seen the in IMK data " with "produced from NO_x biomass burning and lightning emissions, which is seen in the IMK data". The typo has also been corrected.

Page 18, Line 25 – "lower TOMCAT PAN concentrations in regions of biomass burning" with "lower TOMCAT PAN concentrations in regions of biomass burning and peak lightning activity".

I have only one minor comment. On Page 4, Line 21: PAN mixing ratios are on the order of several ppbv in heavily polluted air-masses. This is strange here.

Within the Russo et al (2003) paper, they look at polluted air masses from several regions. The reviewer is correct that in some regions, below 2 km in altitude, PAN concentrations, detected in aircraft campaigns and using backward trajectories, can reach several ppbv. However, in the majority of regions, the [PAN]s are less than 1.0 ppbv. In the mid-upper troposphere (>2 km in altitude), there are a few cases where max [PAN]s get to 900-1000 pptv. Therefore, we have altered the statement "rising to over 600 pptv in polluted air masses (Russo et al., 2003)." on page 4, line 21 to "rising up to 1000 pptv in some polluted air masses (Russo et al., 2003).".

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