



Supplement of

Intercomparison and evaluation of satellite peroxyacetyl nitrate observations in the upper troposphere–lower stratosphere

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Tropical Southern Atlantic PAN Concentrations:

From Figure 1a there are clear source regions of NO₂ over both southern Africa and South America, with transport of NO₂ out into the tropical South Atlantic. Tropospheric O_3 (Figure 1c) has a similar and more distinct pattern over the Atlantic. As enhanced MIPAS PAN (Figure 1d) is also seen in this region, NO₂, O₃ and PAN are highly correlated. The gradient in retrieved tropospheric O₃ across the tropical Atlantic, suggests transport of PAN and NO₂ and subsequent O₃ formation, primarily from southern Africa (i.e. the ozone gradient is from East to West across the Atlantic; Figure 1c). Figure 1b 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 1d show locations of significant lightning events (> 5x10⁶ μ J/ster/m²/ μ m). 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 NO₂ sub-columns in regions of lightning activity, using a cloud-slicing technique, range between $0-0.5 \times 10^{15}$ molecules/cm², which can make up a significant proportion of the tropospheric column over the South Atlantic/West African coastline (e.g. 0.5-2.0 x10¹⁵ molecules/cm²). 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.



Figure 1: 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 NO₂ $(x10^{15} \text{ molecules/cm}^2)$, b) OMI-MLS (Microwave Limb Sounder; Ziemke et al. 2006) tropospheric O₃ (DU), c) Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) HCN (pptv) at 150 hPa and d) MIPAS PAN (pptv) at 150 hPa (Glatthor et al., 2007). White circles represent locations of significant lightning events detected by the Lightning Imaging Sensor (LIS; Cecil et al. 2014).

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