We thank the reviewer for these constructive and helpful comments. Our replies (in blue/red) to each comment (in black) are listed below. Red text indicates changes to the manuscript.

Referee 2
General Comment
This well written manuscript by Sobanski et al. outlines a comprehensive study of NO3 mixing ratios and lifetimes in the context of a field campaign (referred to by the acronym PARADE) in the German mountain region, Taunus, during Summer 2011. The campaign was equipped with a broad range of instrumentation to detect most atmospheric species (as well as meteorological data) that are relevant for NO3 generation and destruction in order to interpret the key findings, which included unusually large and also highly variable NO3 mixing ratios, and long lifetimes (up to 1 hr and more). The general discussion uses a steady state model which is based on the most relevant NO3 production reaction at nighttime (NO2 + O3). The manuscript also considered none-steady state conditions and contains novel aspects such as the influence of Criegee intermediates on the NO3 production rates through reactions with NO2; also the reaction of OH and HNO3 was considered. Loss mechanisms of NO3 are discussed based on reaction with NO and volatile organic compounds (VOC, biogenic and anthropogenic). Data are additionally interpreted on basis of meteorological conditions and in this context it is argued that the high NO3 concentration on some occasions are likely to be due to a "low lying residual layer" over the boundary layer with a significant positive NO3 concentration altitude gradient. Key instruments are (i) a cavity ring down spectrometer (CRDS) for the detection of NO3 and N2O5 (in a heated channel) as well as (ii) a 4-beam long-path differential optical absorption spectrometer (LP-DOAS). A comparison of the corresponding data is also included in the paper mostly to argue for the residual layer hypothesis. Generally, the manuscript is well structured and there is a good flow of the text. The discussion of data dwells on the most significant events and discussion of data dwells on the most significant events and atmospheric scenarios during the 3 week campaign and the items that require discussion and explanation have been carefully selected. Based on the methodology the data inspire confidence and the majority of conclusions are supported by arguments that are anchored in the data, however some discrepancies remain. This manuscript clearly merits publication in ACP, however, it is recommended that the comments and minor corrections listed below are considered prior to acceptance.
We than the reviewer for this positive assessment of our manuscript.
Specific Comments
Section 1: Page 1&2: The introduction is giving a good overview of the most relevant atmospheric reactions that influence the NO3 concentration. This part of the manuscript would however benefit from more quantitative data on the reaction rates as they are known in the literature (IUPAC and Atkinson et al.). Reaction rates could be incorporated in the R-equations or presented in form of a table and would provide the reader with kinetic information on the relevance of each reaction for the NO3 chemistry.
The discussion at this point is largely qualitative. Later in the manuscript we discuss rates of production and loss of NO_3 in quantitative terms and also list rate coefficients in a Table. We prefer not to reduce the readability of this section by introduction of reaction rate constants.
Section 2: The information on the instrumentation specification is brief, as most of the experimental setup utilized during PARADE have been published before. However, the

reader would benefit from some more information on the key instruments. Page 5: What zeroing/calibration procedures for the CRDS instruments were applied? Page 5: The description of the NO_2 instrument (Page 5) is particularly short (Thieser et al. 2015).

We have added text to describe how the CRDS measuring NO_3 and N_2O_5 was zeroed: Zeroing was performed by addition of NO (R6).

We have extended the information on the NO_2 instrument:

The instrument was zeroed using dry, synthetic air and corrections were made for humidity differences between the zero air and ambient. Typical ring-down times were ~ 30 μs and the instrument has a detection limit for NO_2 of ≈ 20 pptv in 1 min and an accuracy of 6 % where the dominant contribution is uncertainty in the NO_2 cross sections (Voigt et al., 2002).

Page 5 & 6: The literature used for the cross-sections for NO3 and NO2 does not seem to be mentioned (neither for the CRDS nor the LP-DOAS setups).

We now write (for NO_3) : The total uncertainty was ~15 % for both NO_3 and N_2O_5, with the largest contributions arising from uncertainty in the NO_3 cross section (Orphal et al., 2003; Osthoff et al., 2007) and NO_3 losses.
The source of the NO_2 cross sections is now also listed (see reply to comment above).

Page 5, Line 24: What does "quasi online" mean?

The term "quasi-online" was unnecessary and has been removed

Page 6: An overall uncertainty of the LP-DOAS setup of 10% for NO3 is stated. The cross-section uncertainty is of that order. The error evaluation seems to be too optimistic.

The NASA evaluation panel recommends a value of $2.25 \pm 0.15 \times 10^{-17}$ cm^2 $molecule^{-1}$ for the cross section at 662 nm, i.e. errors of 6.6 %.

Section 3: Page 9: Please check (Eq4). If L is a loss rate constant in units of [s-1] the power of "−1" seems out of place.

The comment is correct. An early version of this expression referred to lifetimes, hence the power term. This has been corrected.

Page 9, Line24: What does the index 'DER' stand for?

To clarify this we now write: The lifetime calculated via derivatives, τ_{DER} (NO_3) is given by.........

Page 9, Line30: Why is it meaningful to state an average value of the steady state lifetime, if the night to night variability is high? What is meant by values (plural)?

We give an average mixing ratio to highlight the fact that so that nights with lifetimes of e.g. >1000 s are to exceptional. The use of a plural was an error and has been corrected.

Page 10, Line 8: "Because the error made by assuming steady state is generally small . . .", can this be quantified?

Yes. We now write: The derivatives method does not provide data for the whole campaign. $\tau_{DER}(NO_3)$ are sometime very scattered and sometimes negative which arises from noise on the derivative terms. However, on average, the ratio $\tau_{ss}(NO_3)$ / $\tau_{DER}(NO_3)$ was 0.99 with ~66 % of the lifetimes agreeing within 30 %. The the analysis and discussion below is based on the lifetimes obtained by the steady-state analysis.

Page 11, Line 26: What is meant by "weaker day/night cycle"?

We now use the term "weaker diel variation"

Page 12, Line25: What is meant by "these time scales"?

The term "these time scales" was superfluous and has been removed.

Page 13: The arguments in Section 3.3.4 are well laid out and appear conclusive; nevertheless the section would benefit from explicit mentioning of reaction rates (e.g. for sCI +NO2; also see comment above).

Specific literature measurements of the rate constants for sCI + NO_2 are mentioned in the text and an "average" value listed in Table 3. We consider this sufficient.

Page 13 (bottom part): Even though sCI mixing ratios of 4 pptv seem clearly too high, the authors outline an interesting hypothesis that merits further research.

We agree. We have little information on the concentration of sCI in the atmosphere and their reactions.

Page 14: What is meant by "low-lying residual layer"? What constitutes this layer and how is it contrasted to the boundary layer, as the measurements were taken at some height of 825 m ASL.

See also reply to the similar comment of reviewer 1. The "low-lying residual layer" is essentially a very shallow boundary layer. We now write:

In summary, the data shown in Fig. 9 and the LP-DOAS measurements of NO_3 at different altitudes provide compelling evidence for a low-lying residual layer (or very shallow nocturnal boundary layer)

Page 15: Fig. S2 in the main text would be really helpful for the discussion of the observed NO3 vertical concentration gradient. The altitudes of the sender/receiver units and retro-reflectors of the LP-DOAS setups in comparison to the altitude of the CRDS instrument should be repeated here for the benefit of the reader.

Done. We now write:

In Fig. 12 (left panel) we plot the campaign NO_3 mixing ratios measured by the CRDS and DOAS,4 versus the DOAS,1 dataset. As mentioned previously and illustrated in Fig S2, the LP-DOAS light source and the CRDS inlet were co-located at a heigth of ~835 m whilst the LP-DOAS retro-reflectors were at ~959 and 872 m.

Page 15: Is there any evidence for mixing of the layers when comparing the four DOAS directions and different heights probed during the measurements?

We cannot derive information about mixing from the DOAS measurements alone. To clarify this we now write (page 17): The DOAS measurement of concentration at different altitudes does not contain information about the vertical exchange and mixing itself. Any estimation of vertical mixing for a reactive trace-gas like NO_3 would require a chemical model with transport, which would be difficult for such an environment with complex topography like the Kleiner Feldberg.

Page 16, Line 16: How can be assumed that there are no significant NO3 gradients in the proposed "residual layer". Again, is there evidence for mixing owing to convective currents in the time series of the data on the relevant days?

The use of the term "no significant gradient" was misleading. We now write: The apparent dependence of the NO_3 mixing ratios and lifetime on the NO_3 concentrations reflects the fact that, when NO_3 is high, both instruments are sampling the residual layer in which NO_3 levels are expected to be higher and gradients in NO_3 are exected to be weaker than found close to the surface (Brown et al., 2007).

Page 16, Line7-9: Does Fig 12 (left panel) really allow this statement? Can the statement be underpinned by quantitative information on altitude dependence, and the term "closer" through R-values for example.

The Figure shows that at low NO_3 mixing ratios, the ratio of CRDS to DOAS,1 is nearly always below 1, where as that of DOAS,4 to DOAS,1 is nearly always greater than one. This is very strong evidence of a gradient. When NO_3 mixing ratios are high (above 100 pptv) the gradient is weaker. We have added text to illustrate this in a more quantitative manner:

To illustrate this, For NO_3 steady state lifetimes of < 1500 (i.e. all nights except for 31.08-03.09) s the average value of the $[NO_3]_{DOAS,4}$ / $[NO_3]_{DOAS,1}$ ratio is 1.23 ± 0.07, whereas for NO_3 steady state lifetimes of > 1500 s we derive a $[NO_3]_{DOAS,4}$ / $[NO_3]_{DOAS,1}$

ratio of 0.95 ± 0.05.
Page 16: The last paragraph in section 3.3.6 seems to be better placed in the previous section on the discussion of the residual layer and not in the comparison of CRDS and LP-DOAS.
This paragraph has been moved as suggested.
Generally, some material from the supplementary material would have been helpful in the main text; notably Fig S1, S2, S4 and S8. S1 and S2 are important to get an idea of the topography in the vicinity of the measurement site. Especially in the context of section 3.3.5 and 3.3.6 these figures are helpful.
Figures S2 and S8 have been moved to the main manuscript.
In many places mixing ratios are not expressed by volume. Examples are: P5,L23,L26,L27; P8, L15;P14,L19; P16,L4,L11.
Corrected
Page 3, Line 23: State the month(s) in 2011 explicitly here.
Done
Page 4, Line 13: "Fig. S1" should be highlighted.
Figure number highlighting will disappear in the final version
Page 6, Line 7: "broadbanding"? Is that the term to use? Moreover, the information in this line is redundant and was mentioned before on page 3.
Broadband is the correct term. This has been amended.
Page 9, L26: In Eq 5 the minus signs in the denominator are hardly discernible
We shall make sure that this is more legible in the type-set version.
Page 11, Line 6: IUPAC should get a reference.
IUPAC now referenced (Ammann et al., 2012)
Page 11, Line 29: "Fig. 7" should be highlighted.
Figure number highlighting will disappear in the final version
Fig. 1 & 2: There is a discrepancy on the third cold front arrival time (third blue vertical line). It may be better to merge Figs. 1 & 2 into one figure with six panels. If this could be done without loss of plot quality, this would help the reader.
The cold-front was on the 5th Sept. Figure corrected. We prefer to keep the Figures separate, but will make sure that they are located on the same page in final form.
Fig. 5 Units of seconds are missing on the vertical axis of the upper panel.
Corrected
Fig 6. The red and black trace refer to the left axis and radiation (in yellow) to the right axis. This is confusing, as the other panels are colour coded.
Figure modified
Fig 7. The lower panel does not seem to show LSUM, LNO. The stated time grid appears inconsistent with the dots in the figure (e.g. 30 min for LVOCS).
As stated in the text, NO mixing ratios were below the detection limit on the night of 30.08. Therefore LNO was not calculated and LSUM is not required. The sampling time of the VOC measurements was 20 mins (GC-MS) or 35 mins (GC-FID). The time resolution is however about 1 hour. This is now clearer in section 2.2.2.
The time resolution was ~ 1 hour (35 min sampling time), detection limits were around 1 pptv with an uncertainty of 10-15 %.
Page 22, Lines 2&3: ". . .of 4 the. . ." and "...at the 5 Großer Feldberg."?
Typos corrected