

RESPONSE TO REVIEWERS

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Point-by-point responses to reviewers are included below. Reviewer comments are in blue. Responses are in black, and text added or altered is quoted in orange. Line numbers are those in the updated tracked changes PDF document.

Response to Reviewer RC2:

Wei et al. use different aircraft measurements combined with a global model simulation to characterise upper tropospheric NO_y and assess our understanding of the processes governing it. The manuscript is generally well written and easy to follow. The topic is important and timely, affecting for example tropospheric oxidation capacity and ozone formation.

Below, I have listed certain areas where I would still like to see more detail, followed by a list of minor comments.

1. The comparison between the IAGOS and DC-8 flights. There is a rather large difference in NO_y levels between the two, which is explained to result from differences in flight altitudes (lines 241-242). As the comparison of these two flight measurements forms a core part of the manuscript, I would like to see more detailed comparisons here, like altitude profiles of NO_y from the two types of measurements. Do they match up?

We now include a supplementary figure (Figure S1; pasted below) showing the seasonal mean vertical profiles of collocated DC-8 and IAGOS NO_y and we discuss the features in this figure in the text to support the distinct altitude ranges sampled by DC-8 and IAGOS and consistency in NO_y for the few instances that sampling is vertically collocated.

“The two campaigns sample distinct altitude ranges of the upper portion of the upper troposphere centred at ~240 hPa (~10 km) for IAGOS and a wider vertical extent of the lower portion of the upper troposphere centred at ~360 hPa (~1.5 km below IAGOS) for DC-8 (Figure S1). There is a general pattern of a steep increase in NO_y with altitude, with the exception of IAGOS layers located near 300 hPa in March-May and September-November (Figure S1). Average NO_y is similar between the two campaigns for the rare instances that DC-8 and IAGOS sample the same pressure layers (Figure S1).” (lines 316-322)

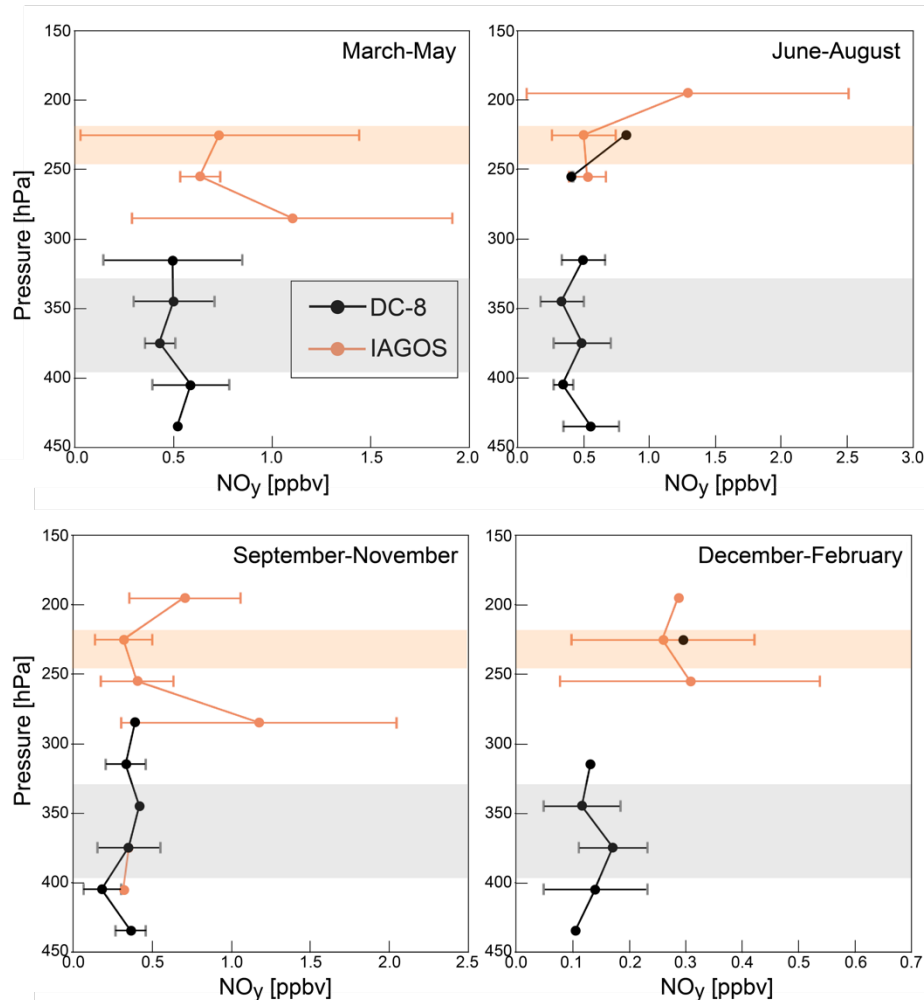


Figure S1: Comparison of seasonal mean vertical profiles of total reactive nitrogen (NO_y) from spatially collocated DC-8 and IAGOS aircraft observations. Symbols are means from averaging upper troposphere (450-180 hPa) observations into 30 hPa bins. Lines are standard deviations. Shading indicates the typical vertical sampling range (pressure standard deviation) of DC8 (grey) and IAGOS (orange). Pressure range selection and screening for stratospheric influence and plumes are detailed in the main manuscript.

2. SEAC⁴RS stands out from the other campaigns in Figs. 4 and 5. These differences are expected to arise from the high contribution of MPN to NO_y in that campaign. Especially the poor correlation seen in Fig. 4 leads me to suspect there may be something wrong with these measurements, or then that MPN would not be properly reflected in the NO_y measurements. Could you analyse further, whether these MPN measurements are indeed high and correct, or may there be some interference in them?

Further analysis assessing a potential high bias in MPN is detailed in the first part of the response to reviewer # 1, Comment (3). Even with this smaller MPN concentration, the conclusion that the model substantially underestimates MPN still holds.

3. Altitude definitions: upper troposphere is here defined as 8-12 km in altitude. However, tropopause may be kilometers higher in the tropics: can you justify the choice of altitude range further?

This is addressed in response to Reviewer #1, Comment (1).

Minor comments:

Abstract, lines 23-24: fractional/percentage values of the over/underestimation would be useful here

Added as 10-90% for PPN and 31-65% for NO₂. (lines 23-24)

Line 54, Fig. 1: can you provide references justifying these are the main species & reactions?

To better represent the key studies that have informed dominance of NO_y components and the reactions in Figure 1, we have edited the text and cited these studies as below:

“Chemical cycling of dominant daytime NO_y components, informed by past review and measurement compilation studies of the free troposphere (Emmons et al., 1997; Bradshaw et al., 2000), is illustrated in Figure 1.” (lines 73-74).

Lines 61-63: add ref for the photolysis vs thermal decomposition

Huey (2007) added. (Line 82)

Lines 65-68: add refs

Schultz et al. (1999) added for NO_x reservoir compound transport and subsidence. (Lines 88-87)

Text edited to clarify that existing citations substantiate slow loss processes in the upper troposphere:

“Loss processes in the dry upper troposphere are slow and dominated by subsidence, resulting in long NO_y lifetimes of 10-20 days (Logan, 1983; Prather and Jacob, 1997). Similarly, NO_x

has a lifetime of about a week compared to less than a day in the boundary layer (<2 km) (Jaeglé et al., 1998).” (Lines 87-89)

Lines 76-78: add ref

We have edited the text, so that the citations appear before the sentence starting “These studies have either focused on ...”. For completeness, we have also added other relevant citations. The updated text reads as:

“Modelling studies evaluating best understanding of NO_y in the upper troposphere routinely identify stark discrepancies between observed and modelled total NO_y, NO_x, and the ratio of NO-to-NO₂ in the upper layers of the troposphere (Jaeglé et al., 1998; Talbot et al., 1999; Bertram et al., 2007; Hudman et al., 2007; Liang et al., 2011; Nault et al., 2015; Huntrieser et al., 2016; Travis et al., 2016; Fisher et al., 2018; Silvern et al., 2018; Lee et al., 2022; Cohen et al., 2023). These studies have either focused on a few NO_y components, or a single aircraft campaign.” (Lines 107-112)

Line 104: add ref

Singh et al. (2006) added. (Line 155)

Lines 105-107: mention that the exact definitions for screening will follow

Thank you for the suggestion. We rather decided to delete “... and that have limited influence from stratospheric air”, as this data screening aspect was not a factor in eliminating INTEx-NA and DC3. (Line 156)

Line 126: would times relative to sunrise and sunset be more appropriate? You screen for jNO₂ as well, but this leads to different representation of high- and low latitudes (as mentioned on lines 139-140). I'm not requesting you to redo all the analyses, but preferably comment on if this has an effect on the results

We do already use the reported solar zenith angle (SZA) values to confirm that are time range and jNO₂ filter removes data with large SZA (sunrise/sunset). (Line 201). Given this, we do not expect any effect on our results if we instead used SZA.

Line 127: full vertical extent. But this does not include full vertical extent, esp. in tropics

This text has been updated to address the similar Reviewer #1, Comment (1).

Screening criteria: how much data do these criteria exclude? In other words, how typical are the sought-for background conditions?

We now state provide these statistics and the main cause for data loss:

“The proportion of observations at 450-180 hPa is 42-50% for ATom and 16-37% for the other campaigns. After applying all other data screening, 20% of all data are retained for ATom and 7-11% for the other campaigns.” (Lines 202-203).

Lines 135-136: refs for these screening criteria

These are already provided as Hudman et al. (2007) and as Shah et al. (2023). (Lines 196-198)

Line 137: what does approximately zero mean?

Zero within the range of uncertainty of the instrument.

Line 149: ref talks about TD-CIMS specifically

Thank you for pointing this out. We have replaced the Slusher et al. (2004) reference with Huey (2007). (Line 212)

Lines 151-153: ref

This information is obtained directly from the dataset.

Line 154: ref for TD-LIF

Day et al. (2002) added. (Line 217)

Eq. 1, also in the text: NO and NO₂ should be in square brackets

These are intentionally not in square brackets, as the units are distinct (pptv) from the compounds in squares brackets (molecules/cm³).

Line 181: how is HO₂ measured?

HO₂ is measured using a laser induced fluorescence.

Line 184, RO₂ relatively insignificant: I could not easily find this in the reference. Is it so?

Shah et al. (2023) state that it makes a small contribution in the free troposphere, so we have reworded our text to more closely match theirs:

“... but we ignore this reaction as it is relatively insignificant throughout the free troposphere (Shah et al., 2023).” (Lines 248-249).

Line 202: are O₃, CO and jNO₂ also measured on the commercial aircraft?

O₃ and CO are, as is now made clear in the text:

“... and daytime filtering as is applied to DC-8 data (Sect. 2.1) using IAGOS O₃ and CO measurements.” (Line 273).

There are no jNO₂ measurements, but there is no need to include this filtering step, as none of the coincident flights extend to the high latitudes. Now also stated in the text:

“There are no NO₂ photolysis frequency measurements, but the requirement for spatial coincidence with ATom excludes polar twilight and night measurements at high latitudes.” (Lines 274-275)

Line 222: do you mean below-cloud?

Yes. Corrected (Line 293).

Lines 242-243: ref or show data

Data are now shown in response to your Comment 1.

Fig. 3 b: is panel b needed?

Yes. The panel helps illustrate the size of seasonal variability in total NO_y. Despite large differences in absolute concentrations of DC8 and IAGOS NO_y, both exhibit relatively similar seasonal changes, whereas the model (state of knowledge) seasonal shifts in NO_y are too modest.

Lines 261-266: would it be easier to read if common measurements were listed, and then campaign-wise which compounds were included?

We now summarise this information in a new Table 1 (pasted below) for greater clarity and refer to the table in the Figure 1 caption and in the text (Line 350).

Table 1. Observations of individual NO_y components summed to assess budget closure in Figure

Component	NASA DC-8 aircraft campaign		
	ARCTAS, SEAC ⁴ RS, KORUS-AQ	ATom1-2	ATom3-4
NO ₂	PSS	PSS	PSS
NO	Chemiluminescence (CL)	CL	CL
HNO ₃	CIMS	CIMS	CIMS
HNO ₄	TD-LIF PN _s	CIMS	–
PAN	TD-LIF PN _s	PANTHER	PANTHER
PPN	TD-LIF PN _s	–	CIMS
other PAN _s	TD-LIF PN _s	–	CIMS
ALKN _s	TD-LIF ALKN _s	WAS C1-C5	WAS C1-C5
MPN	TD-LIF PN _s	–	–

Lines 294-296: does including the HCN observations improve the agreement?

At the risk of over-interpreting a rough estimate of HCN interference, we now state that the HCN interference estimates are consistent with the unaccounted NO_y in Figure 4:

“These lower-end interference estimates are similar in size to the percent missing NO_y (13% for ARCTAS, 3% for SEAC⁴RS, 8% for KORUS-AQ, 1-22% for ATom).” (Lines 390-391)

Section 3.2: includes long and complicated section on inferred concentrations, should this rather be in the methods section?

We prefer to keep it in this section, as the inference requires knowledge of the median values of the measured NO_y components presented in Figure 5.

We now state in the methods Section 2.1 that inference is needed for quantities not measured:

“The NO_y components not measured during specific campaigns are inferred. These include HNO_4 for KORUS-AQ, and ATom-3-4, PPN for ATom-1-2, and MPN for ARCTAS, ATom-1-4 and KORUS-AQ. The approaches used to infer these values differs, informed by the results, so a detailed description of this inference is in Section 3.2.” (Lines 251-254)

And in Section 3.2, we adjust the text of the 2 paragraphs to accommodate this pre-introduction to inference in the methods section and to focus paragraph 1 on HNO_4 and PPN and paragraph 2 on MPN:

Lines 425-442:

“Inferred DC-8 HNO_4 and PPN in Figure 5 use ATom-1 HNO_4 and ATom-4 PPN for combined ATom-1 and -4 components, and, similarly, ATom-2 HNO_4 and ATom-3 PPN for combined ATom-2 and -3. KORUS-AQ HNO_4 is estimated to be 37 pptv by multiplying the SEAC⁴RS median fraction of HNO_4 ($\text{HNO}_4/\text{NO}_y = 0.06$) by the KORUS-AQ median NO_y . SEAC⁴RS is used, as HNO_4 is thermally unstable (Ryerson et al., 2000) and so varies with temperature. Mean upper troposphere ambient temperatures for KORUS-AQ (252 K) are more consistent with SEAC⁴RS (246 K) than the other campaigns (238 K for ARCTAS, 238K-241 K for ATom).

The inferred ~10 pptv ARCTAS MPN is from the estimate by Browne et al. (2011). KORUS-AQ MPN is estimated by bounding a potential range from two approaches. The first is the median value of the difference between TD-LIF total PNs and the sum of all individual CIMS PANs and our inferred HNO₄ of 37 pptv, yielding MPN = 75 pptv. This likely overestimates MPN, as the CIMS instrument does not measure an exhaustive suite of PANs. Lee et al. (2022) estimated with a box model and KORUS-AQ measurements that unmeasured PANs account for ~20% of total PNs during KORUS-AQ, though this is for air masses impacted by petrochemical and other anthropogenic VOCs and NO_x emissions. Accounting for these unmeasured PANs yields a lower-bound KORUS-AQ MPN of 8 pptv. The MPN that we infer then for KORUS-AQ is 42 pptv, the midpoint of 8 and 75 pptv, accounting for 7% of KORUS-AQ NO_y. As the GEOS-Chem model MPN is consistent with DC-8 inferred MPN during ARCTAS, we multiply the GEOS-Chem A_{Tom} MPN fractions (MPN/NO_y ~0.01 for A_{Tom}-1 and -4 and ~0.02 for A_{Tom}-2 and -3) by A_{Tom} DC-8 NO_y to infer A_{Tom} MPN of < 6 pptv.”

We have also rewritten the first half of the ALKNs paragraph to focus it on the results in Figure 5, rather than the challenges of inferring >C5 ALKNs during A_{Tom}:

“Only the C1-C5 ALKNs are shown in Figure 5 for A_{Tom}. The remote measurements of total ALKNs available from ARCTAS that would be most suitable to assess the likely contribution of longer chain (>C5) ALKNs are on median 5 pptv less than the A_{Tom} C1-C5 ALKNs measurements. The total ARCTAS total ALKNs measurements are also very noisy, as indicated by a range of -113 pptv to ~333 pptv. The range in ARCTAS WAS C1-C5 measurements, by comparison, is 8-29 pptv. Contributions of >C5 ALKNs to total ALKNs for SEAC⁴RS (~50%) and KORUS-AQ (~60%), representative of the continental upper troposphere, suggest that >C5 ALKNs in remote regions are <50% of total ALKNs or <12 pptv (median of C1-C5 ALKNs for A_{Tom}1-4).” (p. 12 lines 443-447; p.13 lines 461-463)

Line 359: coincidence of the individual to total NO_y? What about disregarding ALKN as they make a relatively small fraction of NO_y to achieve higher southern hemisphere coverage?

Indeed, this approach could have been adopted instead, but as we do already include a paragraph dedicated to the southern hemisphere (Lines 488-493) that covers the relative

contribution of the dominant component PAN, and the seasonality in total NO_y that we find to be generally consistent with the northern hemisphere biased findings.

Lines 366-368: ref for negligible contribution

These are already provided following the initial opening sentence of the paragraph. Following that sentence, we elaborate that NO₃ and HONO have very short lifetimes, as both rapidly photolyze.

Line 370: check the reference. For 100 ppt NO the lifetime is 15 seconds, so for the max average NO campaign (SEAC4RS) it is close to a minute. Noontime clear sky photolysis lifetime is around 6 s, longer off-noon and at high latitudes. Not saying that NO₃ would be significant, but that people often underestimate NO₃ daytime lifetimes

Thank you for pointing out this issue. We have amended the text:

NO₃ has a lifetime of a few seconds during the day, due to efficient photolysis (Brown and Stutz, 2012). (Lines 497-498)

Lines 371-372: how much shorter?

We now state this in terms of the 50% faster photolysis rates expected for HONO from knowledge of the wavelength at which it photolyzes and the increase in actinic flux and hence photolysis frequencies with altitude at this wavelength:

Photolysis of HONO would be further enhanced (by ~50% at 390 nm) in the upper troposphere where photolysis frequencies are enhanced (Hofzumahaus et al., 2002; Reed et al., 2016). (p. 13, line 499; p. 14, lines 514-515)

Lines 391-392: ref, or is this your result?

Our result. This is clarified in updated text added to address Reviewer #1, Comment (5). To accommodate that update, we amend the text in these lines too:

“These are on median, $\sim 0.01 \mu\text{g m}^{-3}$ during ARCTAS, $\sim 0.07 \mu\text{g m}^{-3}$ during KORUS-AQ, $\sim 0.04 \mu\text{g m}^{-3}$ during SEAC⁴RS and $< 0.01 \mu\text{g m}^{-3}$ during ATom (Section 3.1).” (Lines 533-534)

Line 417: ref

Text slightly modified to clarify that this is our analysis of the data:

“The model high bias in HNO₃ could be because of a factor of 2 overestimate in our modelled H₂O₂ compared to observed H₂O₂ for SEAC⁴RS.” (Lines 569-580)

Lines 460-461: was this mentioned in the results section?

Yes. In Section 3.1, we discuss the role of lightning in affecting seasonality of total NO_y measured by IAGOS and DC8. (Lines 310-311)

Line 490: maybe cite all sources here again.

Done. (Lines 651-653)

References:

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