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Comment on acp-2021-707

Anonymous Referee #2

Referee comment on "Redistribution of total reactive nitrogen in the lowermost Arctic stratosphere during the cold winter 2015/2016" by Helmut Ziereis et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-707-RC2>, 2021

The manuscript titled "Redistribution of total reactive nitrogen in the lowermost Arctic stratosphere during the cold winter 2015/2016" by Ziereis and co-authors describes the results from measurements of NO_y carried out from mid-December 2015 until mid of March 2016 aboard the German research aircraft HALO. The measurements consisted of more than twenty flights that probed the lowermost stratosphere in the Arctic region. The analysis uses measurements of N₂O, Nitric acid, ozone, and water to interpret the NO_y measurements. With their measurements, Ziereis and Co-authors observed 3 distinct phases of the evolution of the nitrification of the polar stratospheric clouds (PSC) during the strong 2015/2016 polar vortex. First in early winter (17-21 December) they observed a tight correlation between NO_y and N₂O indicating that the NO_y distribution is controlled by the gas-phase production of NO_y from N₂O (undisturbed conditions). In mid-winter (January, beginning of February) the observed NO_y exceeds the expected value calculated based on N₂O concentrations suggesting evaporation of sedimenting particles from PSC at higher altitudes. In late winter/early spring (end of February, mid-March) they observed NO_y below the expected values, concurrently with high potential temperatures, indicating that the distribution of NO_y is controlled by the downward transport of air masses that have undergone removal of nitric acid by heterogeneous processes. The observations were compared with CLaMS model simulations which confirmed, that the ensemble of all observations is representative for the vortex-wide vertical NO_y-redistribution.

The paper presents results that are very interesting for the scientific community and well within the scope of the journal. The data presented are very valuable and the analysis is sound. However, the presentation of the results needs to be improved in both text and figures before publication.

General comments

The manuscript is generally well written in a sentence-by-sentence sense, however, the text is sometimes too vague and leaves the reader guessing what the authors mean. With a few tweaks, especially in the abstract, I think the paper could be easily improved in a form that will be also appreciated by a larger group of atmospheric scientists that are not necessarily experts in reactive nitrogen in polar regions. The sentences are short and clear, however, sometimes it's hard to understand how they are connected to each other.

Specific comments

In the abstract, the authors talk about redistribution of NO_y without specifying that they are talking about the vertical redistribution of NO_y within the polar vortex. When tracer-tracer correlation is mentioned the author can make it clear that they are talking about N₂O-NO_y and N₂O-O₃ correlations. They talk about nitrification and de-nitrification or excess NO_y and missing NO_y without clearly defining with respect to what.

The findings are quite clear and well presented in lines 453-456. They could be briefly summarized in the abstract as well.

Line 20 "During winter 2015/2016 the Arctic stratosphere was characterized by extraordinarily low temperatures in connection with the occurrence of extensive polar stratospheric clouds" mention that this is connected with a very strong polar vortex

Line 26 "The redistribution of total reactive nitrogen was evaluated by using tracer-tracer correlations." Add how the correlation between N₂O and NO_y allows establishing if the air mass is in equilibrium – denitrified or nitrified.

Line 31: "These observations support the assumption of sedimentation and subsequent evaporation of nitric acid containing particles leading to redistribution of total reactive nitrogen" add "at lower altitudes" here

Line 32: "Between end of February and mid of March also de-nitrified air masses have been observed in Using tracer-tracer correlations, missing total reactive nitrogen was estimated to amount up to 6 ppb. Using tracer-tracer correlations, missing total reactive nitrogen was estimated to amount up to 6 ppb. This indicates the downward transport of air masses that have been denitrified during the earlier winter phase." Move "Using tracer-tracer correlations, missing total reactive nitrogen was estimated to amount up to 6 ppb" at the end of the sentence as this refers to denitrification+ high potential temperatures

Line 49: the sentence "Depending on temperature, 50 composition and physical state,

different types of polar stratospheric clouds can be distinguished: liquid supercooled droplets, binary or ternary solutions (SBS, STS), nitric acid hydrates (NAD, NAT) and water ice particles (e.g. Fahey et al., 2001; Hoyle et al., 2013; Khosrawi et al., 2017; Tritscher et al., 2021)." This sentence seems unnecessary/not relevant.

Line 53: "It does not only prepare the surface for heterogeneous reactions, it also removes ..." Unclear maybe use "supply" instead of "prepare"?

Line 55: "Heterogeneous reactions also enable the de-noxification of the stratosphere, the conversion of NO_x to nitric acid" confusing. Maybe replace the comma with "by"?

Line 58: "The removal of nitrogen compounds from the stratosphere allows continuing ozone destruction that increases with increasing illumination of the polar vortex" the use of "increasing illumination" is not very clear maybe add "at the end of the polar winter"

Line 59: "PSCs" acronym not defined

Line 67: "UTLS" acronym not defined

Line 10: "So, the questions could be addressed:" change into "So, the following questions could be addressed:"

Lines 110-114: add a table in to help the reader following the timeline of the campaign

Lines 179-181: add which reagent ion is used

Line 184 remove extra parentheses before "Friedl"

Line 185 the parenthesis should be moved from before "Johansson" to after "et al." i.e., "discussed by Johansson et al. (2018)"

Line 214: "because their lifetime is long compared to transport time" vague sentence. Please add ranges for lifetime and transport time.

Line 221: a schematic figure of N₂O vs NO_y could be added to explain this.

Figure 1 is used to support the sentence at line 253 "As expected for undisturbed conditions, NO_y and N₂O are anticorrelated". For this reason, NO_y and N₂O should be in the same panel. Or plotted elsewhere as a scatterplot. Or at least add a vertical grid.

Line 235 it looks like NO_y* was determined from the least-square fit in figure 6a but in the text, it's not clear that this is the case. Add in the text (either here or at line 269) how this is used for the analysis.

Line 268: why a value of 320 ppb was chosen? Please add to the text.

Line 269: remind the reader that this slope is the same as the "f" in equation (3) and more in general how each term of eq3 is treated to get NO_y* from the slope in Fig 6s

Line 270-276 add ranges/uncertainties to the slopes

Line 278: is the value 0.067 (mid-latitude) chosen as a reference from PGS-5? Please clarify.

Line 287: "The uncertainty in the estimation of NO_y* resulting from the uncertainty of the tropospheric NO_y contribution is highest directly at the tropopause and decreases with decreasing N₂O concentration and increasing stratospheric character of the air mass" not obvious why this is the case. Please add an explanation in the text

Line 319: "... more than 85 % of the total flight time in the lower stratosphere with PV values of more than 2 PVU" PV is not defined. Also please explain briefly in the text what it means to have a PV >2 PVU

Line 322 add the year of TACTS

Line 325 "Significantly higher NO_y concentrations" add a value here, e.g., "up to ..."

Line 325 "than during the flight in December" add max value here

Line 334 "Values changed from around 0.004 to values up to about 0.01." Unclear if its' referring to dNOy or to the ratios from the sentence before

Line 381 "As an example, the flight on 26 February (Figure 4 and 6d) may serve" change into "the flight on 26 February (Figure 4 and 6d) may serve as an example"

Line 398: "Down to about 260 ppb N₂O, observed NO_y and calculated NO_y* agreed within a reasonable uncertainty range." Add uncertainty range in parenthesis

Figure 7, lower left panel: add NO_y line; left panels: add a horizontal line at zero

Line 549: the equation should be numbered (5) not (3)