Author's Response to Referee #2

In this response, the referee comments (in black) are listed together with our replies (in blue) and the changes to the original manuscript (in red).

Friedrich et al. report on measurements of 'reactive nitrogen' as part of the shipborne 2017 AQABA campaign. Data were acquired using a custom-built thermal dissociation cavity ring-down spectrometer recently described by Friedrich et al. (2020) supplemented by numerous auxiliary measurements. In this paper, the authors dive into some of the analysis of this rich data set, focussing on the conversion of NO_x to NO_z in 3 regions, on HONO budgets and on ozone production efficiencies.

The paper is written well. It is a bit too long, and there were some organizational shortcomings (see below) that should be addressed before the paper is accepted.

We thank the referee for the constructive review of our paper and the detailed comments, which we will address in the following responses.

Major comments

1) Organizational / presentation issues

NO_z/NO_x ratios were investigated in 3 regions (3.1-3.3 - Mediterranean Sea, Red Sea, and Arabian Gulf). It is not sufficiently clear why the data set was divided in this way.

An extended explanation has been added in the introduction to Sect. 3.

Dividing the analysis into the three regions helps to highlight the chemically different environments encountered. An analysis of the Arabian Sea region was unfortunately not possible due to a gap in the NO_z measurements between 9 and 17 August 2017, caused by instruement failure during heavy seas and winds. The division into the regions was based on the prevalent NO_x mixing ratios displayed in Fig. S3c. In contrast to other AQABA publications (Eger et al., 2019; Pfannerstill et al., 2019; Tadic et al., 2020), the Gulf of Oman and the Suez Channel were included in the Arabian Gulf and the Red Sea regions, respectively, as a clear shift in NO_x to mixing ratios below ca. 1 ppbv occurred both upon leaving the Gulf of Oman into the Arabian Sea and upon exiting the Suez Channel to the north towards the Mediterranean Sea. The transitions between the Arabian Gulf and the Gulf of Oman, and between the Northern Red Sea and the Suez region are less obviously represented in the NO_x levels.

(b) A brief but incomplete analysis of HONO is presented in section 3.4. The introduction does not mention HONO (other than in R5) and this section appears "out of the blue". Consider removing this section if the plan is to write a separate paper on HONO anyways.

We would preferably keep Sect. 3.4 in the manuscript in its current form, introducing the potential role of HONO as a NO_x source on AQABA, as this is an important finding of this study. A more comprehensive analysis of the HONO data set is outside the scope of this paper, but will be subject of a future publication. We thank the reviewer for pointing out that Sect. 3.4 is not sufficiently included into the flow of the main text, which has been amended in several places:

a) HONO photolysis as a source of NO_x is now mentioned in the introduction:

NO_x can be reformed from HONO at daytime through photolysis, with a noontime lifetime of ca. 20-30 minutes (Stutz et al., 2000).

b) Sect. 3.4 is now referenced in the outline part of the introduction:

In this paper we present NO_x, NO_y and NO_z mixing ratios obtained by a thermal dissociation cavity-ringdown-spectrometer (TD-CRDS), together with a comprehensive set of ancillary measurements and an analysis of the results in terms of photochemical processing/aging of air masses, chemical sources of NO_x (e.g. from the photolysis of HONO), and the efficiency of ozone formation.

c) Sect. 3.4 is now referenced at the beginning of the results section (Sect. 3):

Chemical sources of NO_x e.g. from the photolysis of HONO or pNit are discussed in Sect. 3.4.

An additional literature source has been added to strengthen the discussion on HONO linked to ship emissions.

Ship-derived HONO has a substantial effect on the rates of photochemical O_3 formation in the remote marine boundary layer, largely as a result of higher RO_x production rates (Dai and Wang, 2021).

(d) Information already presented elsewhere should be removed and the previous paper(s) cited - e.g., Fig 1 of this paper is similar to Fig 1 / Fig 3 of Tadic et al. (2020), Fig 9 and 13 and Table 1 of this paper and Fig 7 of Eger et al. (2019). Likewise, rather than describing the TD-CRDS over 2 pages (section 2.1 - page 4 and 5) I would suggest simply citing the earlier Friedrich et al. (2020) manuscript.

Figure 1a has been moved to the supplement.

Section 2.1 has been substantially shortened to focus on information which is not given in Friedrich et al. (2020) and which is relevant to the presented AQABA data sets (see revised manuscript version with ‘track changes’).

(e) Pages 2-3. The section on nitrogen oxide chemistry in the introduction is written well, but in my opinion is not needed - similar sections of text have been presented numerous times, including by the authors themselves in recent papers. It would have been more informative and interesting to tell the reader about what made the AQABA campaign interesting and worthwhile (e.g., effects of temperature/climate and mineral dust on nitrogen oxides, special/unique NOx sources in the regions etc.) and add more background on ozone production efficiencies (see g).

The chemistry part of the introduction has been shortened, focusing on reactions involving NO_z species which are later discussed in the results section (see revised manuscript version with 'track changes').

The scientific motivation for the AQABA campaign has been extended:

Emissions from oil exploration provide a complex atmospheric mixture of NO_x and anthropogenic VOCs. The presence of desert dust can have a significant impact on the budget of inorganic acids such as HNO_3. Finally, the overall elevated temperatures and actinic fluxes on AQABA promote rapid photochemical processing of NO_x. We therefore expect a more varied and complex chemistry than found in remote marine locations.

See g) for the novel paragraph on the OPE in the introduction.

(f) A table summarizing the various measurements and techniques would help.

Such a table is now provided (see Table 1).

(g) The OPE values calculated need to put more into context of existing literature (pg 19 lines 24-30). Consider stating in the introduction what values are typical or would be expected and expand the discussion.

The OPE explanation has been moved to the introduction and extended with more literature context:

The ozone production efficiency (OPE), a metric used in the analysis of the O_3 formation, quantifies the fractional transformation of primarily emitted NO_x to O_3 (Liu et al., 1987; Trainer et al., 1993) and thus reflects the relative importance of competing photochemical processes leading to O_3 and NO_z formation from NO_x. High values of OPE are favoured by low OH and VOC concentrations and values exceeding 80 have been reported for remote marine environments. Low, single digit values have been observed in polluted urban environments (Rickard et al., 2002; Wang et al., 2018). The location-dependence of the OPE can be further classified with previous observations from the literature. Minimal OPEs in urban environments between 1 and 2 have been reported from the Beijing area (Lin et al., 2011; Ge et al., 2013) and from the USA (Daum et al., 2000; Sillman, 2000; Nunnermacker et al., 2004). In rural and suburban environments, the OPE can increase to values between 10 and 15, as demonstrated in Northern America (Olszyna et al., 1994; Roussel et al., 1996; Fried et al., 1997; Ninneman et al., 2017) and in China (Sun et al., 2010). From oceanic samples, OPEs of 65 and 87 were observed on the south-eastern coast of the UK (Rickard et al., 2002) and on Sable Island, Canada (Wang et al., 1996). Flights over the Western Pacific Ocean found values of 102-246 in the tropical area (latitude 0-18 °N), and of 73-209 further north (18-42 °N) (Davis et al., 1996). For the AQABA campaign, we expect lower OPEs than those observed in remote oceanic locations, due to the variable influx from harbours, coastal pollution and surrounding ship traffic.

(2) (Perceived) lack of novelty.

There have already been at least 7 papers presenting results from the AQABA campaign, yet the introduction avoids telling the reader what was presented in the earlier papers. In the introduction, it should make clear to the reader what new information and/or analysis are presented in this paper and why this paper is worthwhile. In particular, it should be stated how this paper differentiates itself from Tadic et al. (2020), "Net ozone production and its relationship to nitrogen oxides" to avoid the perception of duplication (in particular of section 3.5).

We have now included a summary of previous AQABA papers into the introduction and explain the novelty of this paper:

Previous analyses from this campaign focussed on sources and sinks of non-methane hydrocarbons (Bourtsoukidis et al., 2019), the role of OH reactivity in ozone chemistry (Pfannerstill et al., 2019), formation of $ClNO_2$ (Eger et al., 2019), ethane and propane emissions from the Red Sea (Bourtsoukidis et al., 2020), emission factors in ship plumes (Celik et al., 2020), marine emissions of methane sulfonamide (Edtbauer et al., 2020), rates of net O_3 production (Tadic et al., 2020), and the abundance of carbonyl compounds .
In this paper we present NO_x, NO_y and NO_z mixing ratios obtained by a thermal dissociation cavity-ringdown-spectrometer (TD-CRDS), together with NO and NO_2 mixing ratios from a chemiluminescence detector ~~and~~, a comprehensive set of ancillary measurements and an analysis of the results in terms of photochemical processing/aging of air masses, chemical sources of NO_x (e.g. from the photolysis of HONO), and the efficiency of ozone formation..

Following a comment from referee #1 we explained the differences between OPE and NOPR in Sect. 3.5:

In contrast to the OPE, NOPR accounts for the total amount of O_3 produced in one day, considering production (governed by the formation of NO_2 via reactions of NO with HO_2 and RO_2) and loss (via photolysis and reaction with OH or HO_2). The OPE, on the other hand, focusses on the product side and assesses the competition between O_3 formation and sequestering into NO_z from a given initial level of NO_x. By approximating the O_3 production rate via the NO_2 formation from NO reactions with HO_2 and RO_2, the NOPR thus neglects the alternative branch leading to NO_z.

(3) NOx lifetime - pg 9 line 30.

The equation given here is too simplistic in my opinion. Equation (1) should account for N_2O_5 formation, which can increase the $k_{13}[O_3]$ term by a factor of up to 2 (see Brown, S. S., et al. (2004), Nighttime removal of NO_x in the summer marine boundary layer, *Geophys. Res. Lett.*, *31*(7), L07108, doi:10.1029/2004GL019412.). There are also sinks such as the heterogeneous conversion of NO_2 to HONO/HNO_3 that may need to be considered (mentioned on page 18, lines 8-).

We have added explanations for the omission of these two processes:

By using Eq. 1 to approximate the NO_2 loss rate constant, we neglect two further processes which can, under some conditions, influence the lifetime of NO_2. Our approach assumes that the nighttime formation of NO_3 leads to the removal of one NO_2 molecule. This approach would be invalid, if a significant fraction of NO_3 would be lost via formation (and subsequent heterogeneous loss) of N_2O_5. Firstly, we note that formation of N_2O_5 was hindered during AQABA by the high gas-phase reactivity of NO_3 towards VOCs (Eger et al., 2019) and that the transfer of N_2O_5 to the particle phase was hindered by high temperatures. For example, taking an N_2O_5 uptake coefficient γ_{N2O5} of 0.03 (as found for polluted marine environments by Aldener et al. (2006)) and the median nighttime aerosol surface area (ASA) in the Mediterranean Sea of 1.78×10^{-6} cm^2 cm^{-3} (Eger et al., 2019), we estimated a loss rate constant for uptake of N_2O_5 of 3.5×10^{-4} s^{-1}, which is two orders of magnitude lower than the rate constant (4.9×10^{-2} s^{-1}) for thermal decomposition at 25.7 °C (the mean, minimum nighttime temperature in the Mediterranean Sea).
We also neglect the loss of NO_x via uptake of NO_2 onto black carbon (BC) particles. Using a literature uptake coefficient γ_{NO2} of ca. 1×10^{-4} (Longfellow et al., 1999) and the aforementioned ASA, the first order loss rate constant for the heterogeneous uptake would be 1.8×10^{-6} s^{-1}. Using an O_3 mixing ratio of 63.4 ppbv (= nighttime median mixing ratio in the Mediterranean Sea), we calculate a first-order loss rate constant for the reaction of NO_2 and O_3 of 5.5×10^{-5} s^{-1}, which implies that > 95 % of total NO_2 loss at nightime NO_2 is due to O_3. Uptake of NO_2 might therefore be relevant for HONO formation (see Sect. 3.4), but does not constitute a relevant loss process for NO_x.

Minor/Specific comments

In the future, please number continuously and do not restart numbering on each page. You are creating more work for the reviewer which is in nobody's interest.

Thank you for the advice, which we will be following in future publications. We had been using the .docx template provided by Copernicus without further considering the line numbering.

Abstract, line 20 - HONO. Consider stating how the role of HONO was assessed (were there measurements?)

Has been added:

The role of HONO was assessed by calculating the NO_x production rate from its photolysis.

Abstract line 24 - OPE. Consider stating how OPE were calculated (plots of ΔO_3 vs ΔNO_z ?)

We now mention the method in the abstract:

Regional ozone production efficiencies (OPE; calculated from the correlation between O_x and NO_z, where $O_x = O_3 + NO_2$) ranged from […]

pg 1 line 8 - Please define "M"

[…] where M is a collision partner.

pg 3 line 2 - "lifetimes of a few hours". The lifetime of PAN may be much longer aloft are in Arctic environments.

The sentence has been removed while shortening the introductionas requested.

pg 4 line 19 - please specify the make/model of the 3-way valve and state what the internal surfaces are made of.

The valve is made out of PTFE and was obtained from Neptune Research, Inc. (type 648T032, orifice diameter 4 mm). This information, however, was already given in Friedrich et al. (2020) and was removed from the revised manuscript in order to shorten the paper as requested by the referee.

pg 4 line 31 "adding 19 ppmv of O3" - Please clarify if this mixing ratio refers to the amount of O3 (in O2?) added (in which case also state the flow) or if "19 ppmv" refers to the amount of O3 after addition to the sampled air.

The O_3 mixing ratio of 19 ppmv was detected in the sampling flow of the TD-CRDS, i.e. after diluting in ca. 3 slm synthetic air. This information, however, was already given in Friedrich et al. (2020) and was removed from the revised manuscript in order to the paper as requested by the referee.

pg 5 line 4. Does the TD-CRDS respond to nitrate associated with mineral dust which may occur in the study area (e.g., https://acp.copernicus.org/articles/16/1491/2016/)?

We expect only a weak (if any) response to nitrates on mineral dust, as we write in Sect. 3.1.2:

"Detection of coarse mode pNit by the TD-CRDS (see Friedrich et al. (2020)) would lead to an overestimation of HNO_3. However, given that the thermal dissociation to NO_2 of $NaNO_3$ particles with 300 nm diameter is inefficient (~ 20 %) with this instrument, a significant bias by coarse mode nitrate (e.g. associated with sea salt or mineral dust) appears unlikely."

In Sect. 2.1 we also discuss that coarse mode nitrates were only encountered in short periods in high abundancies:

"The fractional contribution of coarse-mode particles to the overall mass concentration were derived using data from an Optical Particle Counter (OPC) and via the (PM_{10}-PM_1)/PM_{10} ratio (both PM_1 and PM_{10} were measured with the OPC). We see from Fig. S1 that the impact of coarse mode nitrate may have been largest on both legs in the transitional area between Southern Red Sea and Arabian Sea, where OPC PM_{10} mass concentrations exceeded 150 µg m^{-3} and the coarse mode fraction was consistently > ca. 90 %."

pg 5 line 20. Please state here how the detection limit was defined (move up from line 27) and also state how long data were averaged (longer averaging times => better detection limits).

The information has been moved and refined:

The total uncertainty (at 50 % relative humidity and one minute integration time) amounts to 11 % + 10 pptv for NO_x and to 16 % + 14 pptv for NO_z if we disregard the non-quantitative detection of coarse-mode, non-refractory nitrate (see below). Detection limits (5 s integration time) during the AQABA campaign were 98 pptv for NO_x, 51 pptv for NO_y, and 110 pptv for NO_z and are higher than those reported for laboratory operation owing to problems with optical alignment due to the motion of the ship. Detection limits are defined as the 2σ standard deviation between consecutive zeroing periods. Under laboratory conditions, NO_x detection limits of 40 pptv (1 min average) were obtained (Friedrich et al., 2020); 6 pptv (40 s) have been achieved with undegraded mirrors (Thieser et al., 2016).

pg 5 line 26. Why correct for humidity?

The discussion of systematic errors has been removed due to overlap with Thieser et al. (2016) and Friedrich et al. (2020). Correcting for ambient humidity is necessary due to the Rayleigh scattering of water and the zeroing of the TD-CRDS with dry synthetic air.

pg 6 line 14-16 How can the uncertainty of j data be 10% if upwelling radiation was not included?

We now write:

The overall uncertainty in *J* is ca. 15 %, which includes calibration accuracy (Bohn et al., 2008) and the negelect of upwelling radiation from the sea-surface

pg 6 line 25. Which meteorological field was used for the HYSPLIT trajectories?

[…], using the Global Data Assimilation System (GDAS1) meteorological model.

pg 17 section 3.4 "HONO formation"

The analysis appears to be only considering daytime processes in this section. How does the HONO budget during AQABA compared to the observations by Wojtal et al. (2011)?

We have included a remark about the daytime limitation of the analysis and give reference to the possibility of a nighttime pseudo stationary state between gas-phase HONO and HONO adsorbed to marine surfaces.

We emphasise that the analysis presented here focussed on the daytime chemistry of HONO. At nighttime, a pseudo stationary state, independent of fresh NO_x input, has been observed by Wojtal et al. (2011), and explained with a reversible deposition of HONO on marine surfaces. This will however be insignificant during the day.

pg 20 line 30. "HONO photolysis was as a significant source of NOx." It is not a net source if HONO is generated from NOx.

This comment has misunderstood the point we are making:

We argue that HONO is not generated photochemically from NO_x (i.e. from NO + OH) but from emissions of ships. If ships exhaust contains HONO (e.g on particles) then the NO_x that comes from HONO is not simply recycling NO_x.

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