Author response to referee comments on "Atmospheric gas-phase composition over the Indian Ocean" by Tegtmeier et al.

We thank all three referees for their valuable comments. We have changed the manuscript according to the comments listed below. Most important, we have shortened Sections 1, 2 and 3 and merged substantial fractions of Section 6 and 7 thus providing now a clear overall summary of the current state of knowledge, highlighting new findings as well as current knowledge gaps. Following the referee suggestions, we have added paragraphs on VOCs, a discussions of trace gas chemistry based on modelling results and have rewritten the section on CH_4. As a result of these and other changes, the manuscript has clearly improved being now more comprehensive and at the same time more focused.
Comments are reproduced below, followed by our responses in italics.

Anonymous Referee #1

This paper provides a detailed review of atmospheric gas phase composition over the Indian Ocean. The species includes ozone and pollutants, greenhouse gases and short-lived biogenic gases. It is a good update after Lawrence and Lelieveld (2010) and suitable for publication in ACP. However, I have the following suggestions, which may be considered for final version.

It is too big to read and goes beyond 'atmospheric gas phase composition'. These extra details such as too much discussion on various meteorological and oceanic processes in Sections 2.1 and 2.2 may be reduced significantly. Similarly, since there is no discussion on the distribution of trace species in oceanic water (it not required also), discussion on salinity, SSTs and productivity may be reduced.

We agree that this material goes somewhat beyond the scope of the manuscript and have shortened sections 2.1 and 2.2 accordingly. Large parts have been moved to supplement, while a short and essential discussion of the atmospheric transport regimes remained in Section 2.1 and a short discussion of Salinity, SSTs and productivity remained in Section 2.2.

Similarly, Sections 6 and 7 may be reduced and merged.

We have merged a substantial fraction of Section 6 with section 7. In particular, we have combined Section 6.1 'Synthesis of scientific progress after 2010' with the former Section 7 'Summary and current knowledge gaps' (now Section 8) and thus reduced the overall length of the two sections.

Definition of various monsoons and transition periods with months is given in Section 1.2. However, these are not followed in various diagrams starting from Fig.1.

The sentence 'Seasonal mean plots here and in the rest of the paper are shown for core monsoon and transition periods, i.e., June to August (JJA) for the summer monsoon, December

to February (DJF) for the winter monsoon, April to May (Apr-May) for the boreal spring transition period and October (Oct) for the boreal autumn transition period.' can be found at the beginning of section 1.2. In all following plots that show seasonal variations of emissions or trace gas distributions, we use this classification.

Emissions and discussions related to various countries surrounding the Indian Ocean are given at many places in the text. Also, names of various regions are used such as East Asia, South-east Asia, South Asia etc. However, these is no explanation which are the countries in these regions nor marked in the diagrams. This should be taken care.

We have now defined our region of interest upfront in the manuscript (in Section 1.4 Scope and organization of this study) based on Figure 1 including a list of the continental regions considered here. However, we have decided to not include lists of countries in all these regions in order to not make the manuscript longer.

Percentage changes in various species from the year 2000 to 2015 are given in diagrams and well as in Tables for various regions. The unit for change from 2000 to 2015 in the diagrams is %/yr and the range for all such diagrams is -20 to +20. Whereas in the tables this change is given as total from 2000 to 2015 and the % changes are very high. Please check and clarify if the numbers are ok in the diagrams and suggest plotting as a total change from 2000 to 2015 as in the corresponding tables.

We have double checked the values in the tables and in the figures, everything is correct. For the figures, the emission maps are high resolution (0.1°x0.1°) and can show very pronounced relative increases found for some grid cells. For the tables, the increases are calculated based on the regional average of emissions in 2000 and in 2015. These regional relative increases are not identical with just simply averaging all relative increases over all grid cells. Therefore, the tables and figures contain complementary information and we have decided to leave them in their current form.

Like the INDOEX, there was a JGOFS-India campaign for the Arabian Sea almost in the same period or maybe a few years prior to the INDOEX. Detailed measurements of physical, chemical and biological processes were done including the flux measurements of CH4 and N2O in different seasons. These studies, at least the flux part, may also be referred in this work.

We have added a reference to the JGOFS-India campaign to the manuscript.

With developments and related emissions increasing in the African region, a detailed discussion on the transport of pollution over the Indian Ocean from this region and its effect will be desirable. Impact of air pollution transport from the Indian continent over the Indian Ocean has been studied in detail by now.

Indeed, emissions from the African regions and their transport across the Indian Ocean is a very interesting and relevant topic. Wherever we have found material (e.g., seasonal variations of CO measurements at two stations on Reunion Island, contribution of long-range transport from southern Africa to the GEM and PBM budgets, impact of African emissions on O_3 vertical profiles at Ahmedabad, CH_4 emissions observed from east African wetlands, etc.)

we have included these discussions in the manuscript. Overall, however, the majority of the work published over the last years has focused on pollution transport from India and Asia, which therefore presents the major part of this review article.

Anonymous Referee #2

The paper is essentially a review of atmospheric composition over the Indian ocean which follows an earlier review by Lawrence and Lelieveld in 2010. The authors attempt to summarise current knowledge, with particular emphasis on post-2010 research, identifying trends and gaps in our current understanding. There is some new work presented, but this is a relatively small contribution to the overall document.

In many ways the paper is a major achievement and would be a valuable resource for researchers interested in this region. However, although the paper is very thorough and generally well written, it is probably over ambitious, and I found it to be overly long and somewhat repetitive. There is a lot of detailed information regarding such things as ocean parameters (currents, salinity, etc.) and climatology (MJO, IOD, etc.) and some of this material, although clearly important for the region generally, is probably not of specific interest to readers of ACP. There is a lot of "background" material which is available elsewhere. Indeed, it is not until page 14 that atmospheric composition begins to be discussed. Some of the introductory atmospheric chemistry is rather basic for ACP readers and could be cut down.

We agree that the material in Section 2 goes somewhat beyond the scope of the manuscript and have shortened sections 1.2 and 2.2 accordingly. Large parts have been moved to supplement, while a short and essential discussion of the atmospheric transport regimes remained in Section 2.1 and a short discussion of Salinity, SSTs and productivity remained in Section 2.2. We decided to leave the introductory atmospheric chemistry as it is, to provide some background to motivate the overall interest in the gases discussed for a wider audience.

The region clearly suffers from a paucity of data which is a common theme throughout the discussion, and it makes the drawing of clear conclusions very difficult. Because of this, the document tends to drift into speculation. This was particularly apparent in the latter sections (section 6) where there are a number of cases where it was suggested that a specific process "could" have an impact whilst presenting little actual evidence. Section 6 could be reduced in length considerably (as could several other sections) with the main points made more succinctly, and with less speculation.

We have merged a substantial fraction of Section 6 with section 7. In particular we have combined Section 6.1 'Synthesis of scientific progress after 2010' with the former Section 7 'Summary and current knowledge gaps' (now Section 8) and thus reduced the overall length of the two sections. As part of this process, we have also changed sections that were too speculative by adding more references and rephrasing or shortening the text.

The lack of clear conclusions and future directions was somewhat disappointing.

By rearranging Section 6 and former section 7, we now have the summary in one place of the manuscript. This provides a clear overall summary of the current state of knowledge highlighting the new findings from 2010 onwards and indirectly motivating future directions by pointing out current knowledge and data gaps.

The paper appears to be part of a special issue but doesn't seem to refer to any other papers in the issue. As an overview, or review paper, this is perhaps something that could be considered.

We have added references to other papers in the special issue to sections 2.2, 2.3, and 7.2.

The paper is somewhat difficult to review as, being largely a summary of previously published work, there are hopefully few mistakes or errors. The authors appear confident that they have included all relevant literature, although I note that they have not included any results from the recent StratoClim campaign.

Thanks for pointing this out. The StratoClim campaign focused on air composition inside the Asian monsoon anticyclone with nearly all flights over the Indian continent and South Asian land masses. Thus, the StratoClim results do not offer much information on the composition over the Indian Ocean. Indeed, transport simulations analysing air masses observed during StratoClim demonstrate that the Indian Ocean contribution to the Asian monsoon anticyclone air masses was very small (Bucci et al., 2020, ACP).

I was also surprised to see that there are so few longer-term ozone measurements in the region. Did the authors refer to the recent TOAR assessment?

We have included all the references that are used in the TOAR assessment and several others, which were not included in the TOAR assessment. The TOAR database for example did not include any observations from moving platforms. We did however overlook one more citation, which has now been included in the manuscript (Lal and Lawrence, 2001).

It is not clear whether the South China Sea region is included or not. If so, there are various datasets (including WMO GAW stations) from this region which have not been considered.

We do not include the South China Sea in this review and removed the one mention of it in Section 2.

The paper would benefit from having a simple regional map (in section 1.1) which clearly defines the region of interest.

We have now defined our region of interest upfront in the manuscript (in Section 1.4 Scope and organization of this study) based on Figure 1 including a list of the continental regions considered here. As the same region of interest is shown in nearly all Figures, we have decided to not add another map.

I don't feel the title does the document justice. It is far more than a review of gas-phase composition. Perhaps need to refer to the marine aspect in the title as well.

True, the manuscript also includes discussions of oceanic and air-sea-exchange processes as well as anthropogenic emissions. Since the atmospheric composition is the major part though and in order to avoid different titles for the ACPD and ACP versions of this manuscript, we have decided to leave the title as it is.

Please explain how the various figures are derived. Which model is being used to show the emission regions and the surface mixing ratio contour plots?

The second paragraph in section 4 (starting with 'We use the latest versions of the Emissions Database for Global Atmospheric Research (EDGAR), in order to present continental pollution and greenhouse gas emissions over the last two decades. …') describes how the CO, NO_x, SO_2, CH_4, N_2O, and CO_2 emission maps were derived. Atmospheric trace gas distributions were derived from satellite data sets as given in the caption of each figure, with the satellite measurements being described in Section 3.2.

I found the wind arrows in Figure 1 a little hard to see (print version). Could these be made a little clearer?

We think that this might be a problem of the current pdf version of the discussion paper. In case the paper will be accepted for publication, we will provide all figures as high-resolution vector graphics, so that the arrows will be clearly visible.

Do you really need to include all the campaign details on page 14? Much of this is superfluous or could have been restricted to Table 1.

We agree that the information in the text and in table overlapped and have removed large parts of the text.

In section 6 there are several paragraphs which have no associated references (e.g. p47-48) although refs are included later in the section (p49). Was this deliberate?

These sections have now been merged with the former Section 7 'Summary and current knowledge gaps' (now Section 8). We have removed all references in order to keep the summary short and concise.

It is not always clear where some of the calculations come from. For example, the isoprene fluxes on p29, line 4. Were these derived by the authors are they taken from the Booge papers?

We use the Booge papers (on which C. Marandino and B. Quack are both authors) to compute the isoprene fluxes. We modified the following statement in the original manuscript (end of p28):

'Here we use data from the OASIS campaign and a modelling study with input variables from 2014 to assess seasonal isoprene fluxes to the atmosphere from the Indian Ocean (Booge et al., 2016, 2018).' to

'Here we provide data from a published modelling study from the OASIS campaign with input variables from 2014 to assess seasonal isoprene fluxes to the atmosphere from the Indian Ocean.'

P36, line 20: do you mean the Straits of Malacca?

Thank you for pointing out our mistake. The name was changed accordingly.

Check spelling of Lelieveld throughout the document (e.g., p48, line 29, 32)

Thank you – done!

Anonymous Referee #3

Major comments

1. The article need to be concise as content in some of the sections (especially, section 1 to 3: making 17 pages) do not link well with the central theme of the article, i.e., "atmospheric gas-phase composition". Additionally, there are overlapping information in introduction and section 2.1.

We agree that the material in Section 2 goes somewhat beyond the scope of the manuscript and have shortened sections 1.2 and 2.2 accordingly. Large parts have been moved to supplement, while a short and essential discussion of the atmospheric transport regimes remained in Section 2.1 and a short discussion of Salinity, SSTs and productivity remained in Section 2.2. We have also shortened Section 1.2 in the Introduction in order to avoid overlap with Section 2.1. Finally, we have shortened Section 3.1 and removed parts of the text that overlapped with information provided in Table 1.

2. The article mainly represents emission or concentration levels of trace gases and associated transport patterns. However, discussions on the findings related to the chemistry of trace gases also considering modelling results (for example, Mallik et al., 2013; Nair et al., 2011; Girach et al., 2017; Ojha et al., 2012) need to be added / strengthened. Recently a few studies have used chemical reanalysis (e.g., CAMS) for better insight into chemical and dynamical processes over the IO (e.g., Girach et al., 2020). Add discussion on chemical reanalysis fields and regional/global chemistry transport modelling.

Thanks for pointing this out. We have added discussions of trace gas chemistry based on modelling results in various sections of the manuscript. Most important is the addition of a discussion of ozone variations and diurnal cycle based on modelling and chemical reanalysis results.

3. What additional inferences do the authors draw while compiling all the literature related to emissions, in situ and satellite observations?

We have compiled a summary of the combined outcome of all new studies including additional information in Chapter 6 of our previous manuscript version. Following the suggestions of the reviewers, we have shortened and combined this section with our overall summary and knowledge gap section, now to be found in Section 8 of the new manuscript version. This shortened and more concise version of new findings, overall summary and knowledge gaps highlights our conclusions based on all related literature.

4. CH_4

Authors used AIRS version 6 (v6) data of CH4 to discuss the surface distribution and seasonality. AIRS is mainly sensitive to CH4 in the upper troposphere (300 hPa) and has no skill in retrieving the surface amounts of CH4. Due to this fact, surface CH4 data has been removed from the latest version 7 of AIRS (refer user guide of v7). It was also reported that AIRS v6 does not capture the variability in surface methane (also at 925 hPa) over the Bay of Bengal (BoB; http://dx.doi.org/10.1007/s12040-017-0915-y). Thus, instead of AIRS version 6 data, please discuss CH4 variabilities using in situ measurements available.

Thanks for pointing this out. Indeed, the AIRS CH_4 observations in the lowest layers should not be used and we have removed the figure and corresponding discussion from the manuscript. Instead, we have included GOSAT/TANSO-FTS TIR CH_4 observations at 800 hPa, which have been evaluated in the Indian subcontinent and Indian Ocean region (Belikov et al., 2021). We have restructured the CH_4 section so that it is mostly based on in-situ observations with some additional information on the large-scale CH_4 distribution above the boundary layer derived from GOSAT measurements.

Belikov, D.A.; Saitoh, N.; Patra, P.K.; Chandra, N. GOSAT CH_4 Vertical Profiles over the Indian Subcontinent: Effect of a Priori and Averaging Kernels for Climate Applications. Remote Sens. 2021, 13, 1677. https://doi.org/10.3390/rs13091677.

P42, L6-8: I do not agree to this. The higher CH4 levels (~2000 ppbv) measured over the northern Bay of Bengal (Srivastava et al., Mallik et al., 2013; Girach et al., 2017) are not seen in AIRS v6. Cape rama observations (Bhattacharya et al., 2009) would help to refer seasonality and CH4 levels, however, it might be older observations. Also see Bhattacharya et al., 2009 for other greenhouse gas variations. Thus, entire discussion of CH4 under section 5.2 (P42-43) needs to be revised.

We have restructured the CH_4 section so that it is mostly based on in-situ observations with some additional information on the large-scale CH_4 distribution above the boundary layer derived from GOSAT measurements.

5. CO_2

Why not use OCO-2 which has better sensitivity for lower troposphere than GOSAT? The distributions of CO2 for different seasons may be included, as for other species.

We have looked into the OCO-2 data and found that only column-averaged CO2 fraction (xco2) data is available but no profile data.

P25L20: CO2 emission from the BoB is associated with cyclones during post monsoon. But whether the BoB is "net" source of CO2? Provide suitable references else a general statement may be avoided.

We have revised the CO_2 subsection of 4.2 to include more specific information and references.

6. CO

Mention explicitly about data type (TIR or NIR or JIR) and retrieval time (day or night) used in the study.

We have added the following information to the manuscript: 'In this article, we use MOPITT V8 Level 3 monthly data (Near and Thermal Infrared Radiances, JIR) with day and night retrievals averaged to analyse the seasonal variation of surface CO distribution (Section 5.1).'

Discuss accuracy and uncertainties in presented CO distribution considering northern IO is covered by thick clouds during JJA? Since MOPITT's sensitivity for surface CO is limited, has there been a comparison/validation of MOPITT-surface-CO with in-situ measurements over the study region?

MOPITT CO data has been compared to in-situ measurements from the ICARB cruise taking place in the Bay of Bengal and Arabian Sea during the pre-monsoon season of 2006 (Srivastava et al., 2012a). The authors found good agreement between the satellite and in-situ datasets: 'These results are confirmed by CO distribution over the Indian region obtained from MOPITT'. We provide the reference to this paper and another study in the first sentence of the paragraph: 'The distribution of the major pollutant CO over the Indian Ocean is well known from MOPITT satellite measurements (e.g., Ghude et al., 2011; Srivastava et al., 2012a).'

Any report on change of mean level of CO (or trend) over the northern IO based on INODEX and recent measurements?

A discussion of CO trends was included in section 6.2 on trends, now section 6. We have expanded this section and now cite results from Srivastava et al. (2012a), Toihir et al. (2015), Girach and Nair (2014b) and Girach et al (2020a).

Is there signature of CO enhancement over shipping lane over the BoB? Black carbon aerosols show enhancement over this region (Ramana and Devi, 2016) and since like BC, CO is also a product of incomplete combustion, it is expected to see some enhancement in CO over the shipping lane.

We have added one reference (Girach et al., 2020b), which reported elevated levels of CO in close proximity to shipping lanes in the southern BoB for the summer 2018. To the best of our knowledge, there are no other publications discussing CO enhancement over the Indian Ocean shipping lanes.

Why is CO mixing ratio lower during monsoon over the Arabian Sea and BoB? In addition to ITCZ movement and cleaner airmass (as you mentioned already), convective uplifting/mixing also contributes for lowering CO over the BoB (Girach et al., 2014).

Thanks for pointing this out. We have added this information and the reference to the discussion of the CO distribution.

Figure 12: Since in-situ measurements of surface CO have been reported for all the seasons, it would be valuable to show spatial distributions alongside figure 12.

We agree that such a figure would be a valuable addition, but have decided to include only one figure per gas distribution in order to keep the number of figures at a reasonable level.

7. NO_2

Authors referred NO_2 as NO_x. I don't find any reason to refer as NO_x unless it is NO_2 + NO. NO_2 should be referred as NO_2, not NO_x.

In the detailed discussions, we distinguish between NO_2 and NO_x depending on if the cited literature reported NO_2 or NO_x abundances. For instance, David and Nair (2011) and Tadic et al. (2019) reported NO_x. Furthermore, EDGAR emission inventories provide NO_x emissions, but no NO_2 emissions. In the overall general sections of the manuscript, we decided to only use one term for simplicity and chosen NO_x as a reference to the discussion of NO_x and NO_2 emissions and distributions.

P33L8: NO_2 pattern is NOT "very similar" to CO. Due to short lifetime of NO_2, its spread is in the vicinity of sources, making a spotty pattern. Whereas CO spreads deeper over the ocean from their coastal/continental sources. In addition, gradients of NO2 and CO across the coastal regions are different.

We agree that this was an oversimplification and have changed the text to 'The tropospheric distribution of NO_x is relatively similar to that of CO, however with a larger variability on small scales and steeper gradients between the continental source regions and the Indian Ocean'.

What is the impact of shipping lane emission on ozone formation/destruction or regional chemistry?

We have added 'In general, NO_x ship emissions can lead to substantial ozone enhancements and in turn to higher OH concentrations (Endresen et al., 2003).' to the text. Since there are, to the best of our knowledge, no detailed studies on the effect of shipping exhaust on ozone formation available for the Indian Ocean shipping lanes, we have not included any further discussion of the topic.

P49L45-46: Trend in NO_2 over India is referred here. NO_2 being a short-lived species, what is the impact of long-term change of NO_2 over the IO? What's the long-term trend over the IO? Is it significant?

To the best of our knowledge, there are no studies that address long-term NO_2 changes over the Indian Ocean, and therefore we cannot include this discussion in our review paper. In a follow-on study, we are currently investigating long-term NO_2 changes in the IO atmosphere and how they related to NO_2 changes over India.

8. O_3

As shown for other species, include also the distribution for the seasons including monsoon and post-monsoon (Figure 14).

Campaign based, in-situ ozone observations are mostly available for the winter and spring-transition period (see Table 4). To highlight the ozone distributions for these two seasons, we show exemplary ICARB campaign data (Figure 14). We have decided to not merge this Figure with panels showing data from other campaigns, but instead highlight the seasonal differences of the ozone distribution in Table 4, where ozone mean values, ranges, maxima and latitudinal gradients are provided.

While some studies reported the existence of specific diurnal patterns, some have not observed such. Putting all studies together, following questions can be answered. What's the general scenario of diurnal pattern of O_3 over the IO? What would decide presence of specific diurnal pattern?

Thanks for pointing this out. We have added a detailed discussion on diurnal ozone variations and the processes determining presence and absence of such variations. We have added this new part to the end of the ozone discussion in section 5.1.

What are possible causes for low O_3 over the Arabian Sea? [halides from sea salt aerosols (Ali et al., 2009); Heterogeneous chemistry indicating role of chloride ions (Nair et al., 2013)] Which O3 formation regime (VOC-limited/NOx-limited) is active where (coastal versus open ocean)?

We have added a discussion of the possible drivers of the low ozone levels over the Arabian Sea to the ozone section in 5.1.

P38L37-39: I do not agree to this statement. Over coastal oceanic region and in the downwind of shipping lane (6 N), NO2-driven photochemical production is evident. Far away from sources, in the open ocean OH-driven photochemical destruction (H2O act as a sink) is observed.

We have removed this sentence and added a comprehensive discussion of the ozone diurnal cycle to the end of the ozone section.

Also, discuss the plumes of CO, NO_2 and O_3 from coastal megacities over the IO.

We have added a discussion of ozone and pollution plumes in the downwind of megacities to section 5.1.

9. SO_2

Strengthened the discussions on SO_2 variations by including the distribution of boundary layer SO2 from OMI (or may be other satellite).

We downloaded and plotted the most up to date SO_2 satellite product from TROPOMI. However, the tropospheric SO_2 column data available does not allow to distinguish between anthropogenic boundary layer and volcanic UTLS SO_2 signals. In addition, in the boundary layer, the accuracy of the retrieved SO_2 abundance is directly affected by systematic errors in the SO_2 profile shape and surface albedo resulting in large retrieval uncertainties (Theys et al., 2015). While large anthropogenic signals can be detected, the SO_2 distribution over the ocean is strongly impacted by these uncertainties. We have therefore decided to not include a boundary layer or vertical column SO_2 product.

Theys, N., et al. (2015), Sulfur dioxide vertical column DOAS retrievals from the Ozone Monitoring Instrument: Global observations and comparison to ground-based and satellite data, J. Geophys. Res. Atmos., 120, 2470–2491, doi:10.1002/2014JD022657.

P36L21-44: Under the title of “Sulfur dioxide”, authors have mainly discussed sulfate aerosols which is a deviation from central theme of the article. Aswini has reported sulfate aerosols, but I think, in-situ measurements of SO_2 have not been reported over the study region. Hence, title “Sulfer dioxide” is a bit misleading. Authors should mention explicitly that using sulfate as proxy for SO2 and be concise for the section.

We added a statement in the SO_2 section that sulfate is used as a proxy, as the reviewer suggested.

10. Ammonia

Can NH_4 be used for the proxy of NH_3? See Aryasree at al., 2015 and Aswini et al., 2020.

We added these references to the ammonia discussion, as the reviewer suggested.

11. VOCs

Discuss on the formaldehyde distribution and associated chemical processes (e.g., Chaliyakunnel et al., 2019; Chutia et al., 2019). Satellite-based mean distributions can be included. Also include studies on Glyoxal and VOCs.

VOCs (including formaldehyde) are now included in all respective sections.

12. Briefly state the uncertainties involved in in-situ measurements, emission inventory and satellite observations for each species represented here.

We agree that uncertainties provide valuable information on the reported measurements and inventories. In general, this information is available in the individual papers. Where possible, we have included the reported uncertainties and provide basic information on the quality of the measurements. However, we think that a detailed discussions of the uncertainties for different measurement techniques out of the scope of the here presented synthesis.

13. Table 4: Some of the campaigns (ARMEX-2002; Ali et al. 2009, CTCZ-2009; Girach et al., 2017; IIEO- 2 during 2018; Girach et al., 2020 and may be a few more) and their findings need to be discussed. Also add in section 3.1.

We have added the campaigns to table 4 and a discussion of their results to the respective sections.

Minor comments

1. Abstract and P54L5-6: What is "unusual type of wind patterns"?

Wind patterns that differ from the typical wind patterns observed during the specific season. We have changed the whole sentence to make this clearer.

2. P47L33: subscript 2 in SO2. Similar mistakes are seen over numerous instances (e.g., P48L16, L17, L25). Correct here and in entire manuscript.

We have corrected the subscripts throughout the whole manuscript.

3. Indicate latitude-longitude range for referring different regions of the IO (for example, P35 L16; P34, L8-20; check for the entire manuscript).

We have now defined our region of interest upfront in the manuscript (in Section 1.4 Scope and organization of this study) based on Figure 1 including a list of the continental regions considered here. The different regions of the Indian Ocean referred to in the manuscript such as Bay of Bengal, Arabian Sea and Southern Indian Ocean are common knowledge and we have decided to not provide their latitude-longitude ranges, consistent with many other publications cited here.

4. P31L6: Use an em-dash to represent the range of values here and other places in the manuscript.

The em dash can function like a comma, a colon, or parenthesis, while the en-dash is used between to signify a range of values. We have replaced the signs in all ranges with the en-dash.

5. P2L3: Doesn't "dynamics" include "transport"?
We have removed the word 'transport'.

6. P2L19: "unusual" what are you referring to?
Wind patterns that differ from the typical wind patterns observed during the specific season. We have added this information to the text.

7. P2L41: "the atmospheric composition...other regimes." What do you mean? Please rewrite and be specific.
This is explained in detail in section 1.2 on the next page. Since all reviewers asked for less repetition throughout the manuscript, we have decided to not repeat the information here.

8. P2L44-45: "Here we review..." this is repetition of L10-11. Rewrite slightly differently.
Thanks, done.

9. P3L10-11: Provide reference.
Reference has been added.

10. P3L9-10, 15: "race, religions, political competition" these may be dropped to make revised version more concise.
This part has been removed.

11. P4L8: Lelieveld et al. (2018) should also be mentioned; this reference is already in the list of references and should be mentioned here.
Reference has been added.

12. P4L3-4: "Atmospheric pollutant levels are low and typical open ocean background conditions can be observed" where?
Information has been added.

13. P4L42: NH3 is discussed at later stage (P18L43-45) but not mentioned in section 1.4
NH_3 has been added.

14. Winter monsoon period is mentioned as DJF on Page P5L26 but as Nov-March on P3L40. Please make it consistent. DJF may be more appropriate.
We have changed Nov-Mar to Dec-Feb.

15. Section 1.3: Prior to INDOEX-1999, pilot campaigns during 1996-1997 (Lal et al. 1998; Naja et al., 2004; Chand et al. 2001, 2003) were conducted. Add this information.
The information has been added.

16. P6L17: add: Airmass undergoes purification (lightning driven OH) during monsoonal convection and significant amount of pollutants get removed before airmass gets into stratosphere as well as getting circulated globally (Lelieveld et al., 2018).

The information has been added.

17. P7L9: “ITCZ typical within” -> “ITCZ, typically within”
Comma has been added.

18. P7L22-24: Let the reader know which altitude range you are referring for the oscillation.
This section has been removed.

19. P8L8-10: mention a few trace gases which get influenced by oceanic transport patterns.
We have added the information to the sentence.

20. P8L15: “...Trades...” Why is T capital here?
This has been changed to ‘southeast trades’.

21. Figure2: makes (a) and (b) of same size.
We have adjusted the size of panel b.

22. P10: Mention OMZ region of IO (Arabian Sea) and some studies linking OMZ with VOCs.
We now mention OMZs in the newly added sections on VOCs.

23. P12L6-7: Many “and”s here. Use proper punctuations and reframe the sentence.
We have rephrased the sentence.

24. P12L31-32: provide reference.
This comes also from the references (Singh et al., 2016; 2019b) given in the next sentence. We have moved the references to the first sentence to make this clear.

25. P13: What is the expected trend in VOCs and CO2 due to long-term changes in phytoplankton?
Given the uncertainty in the long-term changes of different phytoplankton functional types, any statement here would be highly speculative. Therefore, we have not included such statement in the revised manuscript version.

26. P14: section 3.1: Mention of aircraft measurements during ICARB-2009. There are limited measurements over the IO from IAOGS (https://www.iagos.org/)
The information has been added.

27. P16L7-12: In addition to Lal et al., 2014, what are additional findings reported from the observation of vertical profiles of ozone southernmost India (Ajayakumar et al. 2019)? Discuss.
This information has been added to Section 5.1.

28. P16L42: Pollutants? Why plural?
Since ‘Pollutant from MOPITT’ sounds strange.

29. P17L31: It's NO2, not NOx.
Has been changed NO_2.

30. P19L11-12: Is it true for recent time? Provide reference.
We are referring here to the year 2015 based on information from the EDGAR V5.0_AP data base. This becomes clear in the following two sentences.

31. P20L7: "Manufacturing industries"..? Be more specific. "...(electricity production and heat production)..." ->"...(electricity and heat production)..."
We have removed the first 'production'. We decided to not go into details of the manufacturing industries responsible for the continental emissions, in order to stay consistent with the overall level of detail for this section.

32. P20L11: Use more recent reference, Kurokawa et al., 2014
We assume this should be Kurokawa et al., 2013? We have replaced the reference.

33. P22L13: which site does 407 ppm correspond to?
This value gives a global annual mean average. We have included this information and updated the number and reference to give to the 2019 value.

34. P22L19: Use more recent number for lifetime of CH4 (~ 12 years) and other species. See IPCC AR5 Ch-8.
We have updated the lifetime and reference.

35. P24L10-12: what about per capita CO2 emission from Asia as compared to developed counties? Mention briefly.
We have decided to not include such detailed information as this number varies greatly among the Asian countries.

36. P24L24: Expand OMZ appearing for the first time in the manuscript.
OMZ had already been introduced on page 13.

37. P25L5: use "%" only once here and elsewhere. Same applies to other units.
We have corrected this throughout the manuscript.

38. P25L18: Is ENTIRE northern IO source of CO2?
We say that the entire northern IO is a net source (which is also demonstrated in Figure 8c.). We have added additional information to be more precise about the Bay of Bengal, specifically.

39. P27L15: S flux? Sulfer flux?
We have changed this to sulphur.

40. P29L8-9: Why is DMS flux higher over the northern IO during winter?

In fact, the highest fluxes in the northern IO occur during the summer according to Figure 9. The secondary peak in emissions in the winter is most likely due to high wind speeds. The Lana climatology flux calculation is based on the Nightingale et al. (2000) gas transfer parameterization, which contains a quadratic dependence on wind speed.

41. P29L19: Let the reader know the lifetime you are referring by "very short-lived".
The information 'with lifetimes shorter than 6 months' has been added to the sentence.

42. P32L2: "... short lifetime (<mention lifetime>)"
We have added the information to the sentence.

43. P33L16: "...tip..." -> "...southern tip...". Refer this as "shipping lane at ~ 6N over the BoB"
We have changed the sentence accordingly.

44. P38L2: frequently cloud-free conditions? I don't think so, it's a tropical oceanic region and expected to have frequent cloudy conditions, especially boundary layer clouds.
We have changed the sentence to '... which is particularly effective under cloud-free conditions …'.

45. P38L8-12: it's a broad statement. Is there study reporting the signature of stratospheric O3 to lower troposphere over the IO?
To the best of our knowledge no studies has reported this for the Indian Ocean.

46. P40Table-4: "2003 BOBEX I" -> "2003 BOBEX II" (See Srivastava et al., 2012)
We have corrected the table.

47. P46L10: How come mean value is represented as a range of values?
The range was based on mean value plus/minus standard deviation. We agree that this can be confusing and replace the range by the mean values and standard deviation information.

48. P47L1: trend in mixing ratio? Or emission trend?
Trends in atmospheric abundances, either reported in mixing ratios or tropospheric columnar amounts.

49. P47L6: any quantification on RF?
We feel that a quantification of the RF is beyond the scope of this manuscript.

50. P47L6-7: "air quality" What is the importance of air quality over oceanic region?
As discussed later in the section, gaseous atmospheric pollution over oceanic regions can impact the oceanic production and biogeochemical trace gas cycling, which in turn feedback on the overlying atmosphere.

51. P48L8-9: Provide reference.
This is a summary statement based on results presented in Section 4.

52. P49L6: "...trend..." Do you mean seasonal trend or long-term trend?
We have added 'long-term' to the sentence.

53. P49L4-15: reframe the sentence. The word "correlation" appearing twice.
We have rephrased the sentence.

54. P49L30-33: How does this statement connect to 'long-term trend'-the title of sub-section?
True enough, there is no direct connection. However, such results can hint at long-term changes (not taken into account in the model simulations), therefore, we think this is interesting information in the section.

55. P49L41: CO shows decreasing trend over the globe including the IO.
We have rewritten the paragraphs on CO trends and are now providing more references for decreasing CO over the Indian Ocean.

56. P50L14-15: are you referring to trend in surface CH4? The trend reported based on version 6 of AIRS is questionable as AIRS has poor sensitivity for surface CH4.
This section reports CH4 trends across the troposphere.

57. P50L19: "...increased convective activity..." It's a speculation and no evidence presented in the reported paper.
We have changed this to 'The authors hypothesized ...'.

58. P50L29: Use O3 once "ozone" is defined/mentioned as O3. Follow the same for other instances.
For better readability, we have decided to keep the wording as it is.

59. P51L36: Impact of < ? >
We are referring to the impact of the atmospheric composition here. Now, the impact is presented in its section, so the context becomes clear from the introductory paragraph.

60. P51L40: "...gaseous atmospheric pollution...". Not all pollutants increasing? Whether CO is increasing? SO2? Be precise about the gas/species you are referring here.
Good point, we have rephrased the sentence.

61. P53L14-16: what's the significance of O3 gradient? Provide reference.
The deposition of tropospheric ozone and its reaction with marine iodide increases the input of inorganic iodine to the atmosphere (Carpenter et al., 2013). This was stated at the beginning of the paragraph.

62. P53L36-37: "... the Indian Ocean..." or "the northern Indian Ocean"?
We have changed this to ' … northern Indian Ocean'.

63. P54L6: “Seasonal” variation of O3 is available over northern IO.

This statement is referring to a ‘complete picture of the seasonal, latitudinal and vertical ozone variations’. While the available campaign data has been extremely useful in understanding such variations, we think that a complete picture is still missing.

64. P54L33: ‘’...physical...” which processes are you referring? Dynamics is already mentioned in previous statement.

The first sentence refers to atmospheric dynamics, while the second sentence refers to physical processes in the Indian Ocean.

A few additional references which would make this article more comprehensive:

Bourtsoukidis et al., (2020). The Red Sea Deep Water is a potent source of atmospheric ethane and propane. Nature communications, doi: 10.1038/s41467-020-14375-0.

Tripathi et al., (2020). Elevated levels of biogenic nonmethane hydrocarbons in the marine boundary layer of the Arabian Sea during the intermonsoon. Journal of Geophysical Research, doi: 10.1029/2020JD032869

Girach et al., (2020). O3 and CO in the South Asian outflow over the Bay of Bengal: Impact of monsoonal dynamics and chemistry. Atmospheric Environment, doi:10.1016/j.atmosenv.2020.117610

Chaliyakunnel et al., (2019). Constraining emissions of volatile organic compounds over the Indian subcontinent using space-based formaldehyde measurements. Journal of Geophysical Research, doi: 10.1029/2019JD031262

Girach et al., (2020). Tropospheric carbon monoxide over the northern Indian Ocean during winter: influence of inter-continental transport", Climate Dynamics, doi: 10.1007/s00382-020-05269-4

Ajayakumar et al. (2019), Dynamical nature of tropospheric ozone over a tropical location in Peninsular India: Role of transport and water vapour, Atmospheric Environment. Doi:10.1016/j.atmosenv.2019.117018

Chutia et al., (2019). Distribution of volatile organic compounds over Indian subcontinent during winter: WRF-chem simulation versus observations. Environmental Pollution, doi: 10.1016/j.envpol.2019.05.097

Girach et al., (2018). Variations of trace gases over the Bay of Bengal during the summer monsoon. Journal of Earth System Science, doi: 10.1007/s12040-017-0915-y

Lelieveld et al., (2018). The South Asian monsoon—pollution pump and purifier, Science, doi: 10.1126/science.aar2501

Girach et al., (2014). On the vertical distribution of Carbon monoxide over Bay of Bengal during winter: Role of water vapour and vertical updrafts. J. Atmos. Sol. Terr. Phys., doi:10.1016/j.jastp.2014.05.003.

Sahu et al., (2011). Seasonality in the latitudinal distributions of NMHCs over Bay of Bengal. Atmospheric Environment.

Sahu et al., (2010). Impact of monsoon circulations on oceanic emissions of light alkenes over Bay of Bengal. Global Biogeochemical Cycles, doi: 10.1029/2009GB003766.

Thanks for pointing out these additional references. We have added the missing ones to the document.