

The manuscript entitled “Atmospheric gas-phase composition over the Indian Ocean” by Tegtmeier et al. provide a comprehensive literature review on gas-phase atmospheric composition over the Indian Ocean (IO) utilizing emission inventories, satellite observations and in situ measurements conducted during field campaigns. However, the manuscript need a major revision before publication in ACP.

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.
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 modeling.
3. What additional inferences do the authors draw while compiling all the literature related to emissions, in situ and satellite observations?
4. CH₄:
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.
P42L6-8: 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.
5. CO₂:
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.
P25L20: CO₂ emission from the BoB is associated with cyclones during postmonsoon. But whether the BoB is “net” source of CO₂? Provide suitable references else a general statement may be avoided.
6. CO:

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

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?

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

Is there signature of CO enhancement over shipping lane over the BoB? Black carbon aerosols shows 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.

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).

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.

7. NO₂:

Authors referred NO₂ as NO_x. I don't find any reason to refer as NO_x unless it is NO₂ + NO. NO₂ should be referred as NO₂, not NO_x.

P33L8: NO₂ pattern is NOT "very similar" to CO. Due to short lifetime of NO₂, 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.

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

P49L45-46: Trend in NO₂ over India is referred here. NO₂ being a short lived species, what is the impact of long-term change of NO₂ over the IO? What's the long-term trend over the IO? Is it significant?

8. O₃:

As shown for other species, include also the distribution for the seasons including monsoon and post-monsoon (Figure 14). 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 O₃ over the IO? What would decide presence of specific diurnal pattern?

What are possible causes for low-O₃ 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/NO_x-limited) is active where (coastal versus open ocean)?

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.

Also, discuss the plumes of CO, NO₂ and O₃ from coastal megacities over the IO.

9. SO₂:

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

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 SO₂ has not been reported over the study region. Hence, title “Sulfur dioxide” is a bit misleading. Authors should mention explicitly that using sulfate as proxy for SO₂ and be concise for the section.

10. Ammonia

Can NH₄ be used for the proxy of NH₃? See Aryasree et al., 2015 and Aswini et al., 2020.

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.

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

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.

Minor comments

1. Abstract and P54L5-6: What is “unusual type of wind patterns”?
2. P47L33: subscript 2 in SO₂. Similar mistakes are seen over numerous instances (e.g., P48L16, L17, L25). Correct here and in entire manuscript.
3. Indicate latitude-longitude range for referring different regions of the IO (for example, P35L16; P34L8-20; check for the entire manuscript).
4. P31L6: Use an em-dash to represent the range of values here and other places in the manuscript.
5. P2L3: Doesn't “dynamics” include “transport”?
6. P2L19: “unusual” what are you referring to?
7. P2L41: “the atmospheric composition...other regimes.” What do you mean? Please rewrite and be specific.
8. P2L44-45: “Here we review...” this is repetition of L10-11. Rewrite slightly differently.
9. P3L10-11: provide reference.

10. P3L9-10, 15: "race, religions, political competition" these may be dropped to make revised version more concise.
11. P4L8: Lelieveld et al. (2018) should also be mentioned; this reference is already in the list of references and should be mentioned here.
12. P4L3-4: "Atmospheric pollutant levels are low and typical open ocean background conditions can be observed" where?
13. P4L42: NH₃ is discussed at later stage (P18L43-45) but not mentioned in section 1.4
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.
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.
16. P6L17: add: Air mass undergoes purification (lightning driven OH) during monsoonal convection and significant amount of pollutants get removed before air mass gets into stratosphere as well as getting circulated globally (Lelieveld et al., 2018)
17. P7L9: "ITCZ typical within" -> "ITCZ, typically within"
18. P7L22-24: Let the reader know which altitude range you are referring for the oscillation.
19. P8L8-10: mention a few trace gases which get influenced by oceanic transport patterns.
20. P8L15: "...Trades..." Why is T capital here?
21. Figure2: makes (a) and (b) of same size.
22. P10: Mention OMZ region of IO (Arabian Sea) and some studies linking OMZ with VOCs.
23. P12L6-7: Many "and"s here. Use proper punctuations and reframe the sentence.
24. P12L31-32: provide reference.
25. P13: What is the expected trend in VOCs and CO₂ due to long-term changes in phytoplankton?
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/>)
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.
28. P16L42: Pollutants? Why plural?
29. P17L31: It's NO₂, not NO_x.
30. P19L11-12: Is it true for recent time? Provide reference.
31. P20L7: "Manufacturing industries"..? Be more specific. "... (electricity production and heat production)..." -> "... (electricity and heat production)..."
32. P20L11: Use more recent reference, Kurokawa et al., 2014
33. P22L13: which site does 407 ppm correspond to?
34. P22L19: Use more recent number for lifetime of CH₄ (~ 12 years) and other species. See IPCC AR5 Ch-8
35. P24L10-12: what about per capita CO₂ emission from Asia as compared to developed countries? Mention briefly.
36. P24L24: Expand OMZ appearing for the first time in the manuscript.
37. P25L5: use "%" only once here and elsewhere. Same applies to other units.
38. P25L18: Is ENTIRE northern IO source of CO₂?
39. P27L15: S flux? Sulfer flux?

40. P29L8-9: Why is DMS flux higher over the northern IO during winter?
41. P29L19: Let the reader know the lifetime you are referring by “very short-lived”.
42. P32L2: “... short lifetime (<mention lifetime>)”
43. P33L16: “...tip...” -> “...southern tip...”. Refer this as “shipping lane at ~ 6N over the BoB”
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.
45. P38L8-12: it’s a broad statement. Is there study reporting the signature of stratospheric O3 to lower troposphere over the IO?
46. P40Table-4: “2003 BOBEX I” -> “2003 BOBEX II” (See Srivastava et al., 2012)
47. P46L10: How come mean value is represented as a range of values?
48. P47L1: trend in mixing ratio? Or emission trend?
49. P47L6: any quantification on RF?
50. P47L6-7: “air quality” What is the importance of air quality over oceanic region?
51. P48L8-9: Provide reference.
52. P49L6: “...trend...” Do you mean seasonal trend or long-term trend?
53. P49L4-15: reframe the sentence. The word “correlation” appearing twice.
54. P49L30-33: How does this statement connect to ‘long-term trend’-the title of sub-section?
55. P49L41: CO shows decreasing trend over the globe including the IO.
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.
57. P50L19: “...increased convective activity...” It’s a speculation and no evidence presented in the reported paper.
58. P50L29: Use O3 once “ozone” is defined/mentioned as O3. Follow the same for other instances.
59. P51L36: Impact of < ? >
60. P51L40: “...gaseous atmospheric pollution...”. Not all pollutants increasing? Whether CO is increasing? SO2? Be precise about the gas/species you are referring here.
61. P53L14-16: what’s the significance of O3 gradient? Provide reference.
62. P53L36-37: “... the Indian Ocean...” or “the northern Indian Ocean”?
63. P54L6: “Seasonal” variation of O3 is available over northern IO.
64. P54L33: “...physical...” which processes are you referring? Dynamics is already mentioned in previous statement.

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.