

Interactive comment on “Interhemispheric differences in the chemical characteristics of the Indian Ocean aerosol during INDOEX” by M. Norman et al.

Anonymous Referee #1

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GENERAL REMARKS

The paper presents data on the chemical composition of submicron aerosol in the marine boundary layer of the Indian ocean obtained from several ship cruises. The authors present interesting data which highlight the role of the ITCZ as an efficient barrier for transport. However, the manuscript in its present status I do not consider to be of sufficient quality to be published in ACP.

The paper contains new data and some relevant analysis and therefore, in general, merits publication. But quite a number of issues need to be addressed as explained in the following. The authors should be encouraged to submit a carefully revised version of this manuscript to ACP.

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In the introduction, the authors state that two of the main objectives of the INDOEX study were to learn about the role of the ITCZ in the transport of aerosol (and gases) and to assess regional radiative forcing. Section 3 of their manuscript addresses the first of these two issues. As for the discussion presented in Sections 4-6 (chemical composition variations in relation to air mass origin, and interannual variability) only weak motivation is presented in the Introduction. The relevance of the findings in Sections 4-6 of the paper is not made very clear. Sections 4-6 could be written in a more concise style. The question of aerosol radiative forcing is not further addressed in the paper.

As for the general outline of the paper, one might find it more straightforward to discuss first absolute concentrations observed, their variability and possible explanations for this variability, and then proceed to discuss the gradients across the ITCZ. I suggest therefore to essentially swap Sections 4-6 and Section 3.

Analytical errors including blank variability should be addressed by the authors in more detail. This is important because they discuss in the manuscript how variations in concentration levels observed during different ship cruises relate to air mass origin. It is not clear from the manuscript in its present status if the observed variability cannot not be (partly) related to observational uncertainty. See specific comments below.

The concept of comparing the measured chemical composition with air mass origin and travel time since last being in contact with surface sources is an important element in this paper. However, the method chosen by the authors to relate statistics of chemical data with transport times (highlighted in Table 2) is either wrong or needs much clearer explanation. Again, see specific comments below.

SPECIFIC REMARKS

Title: The title is not specific enough. The discussion in the paper is confined to the aerosol of the marine boundary layer (MBL). This should be added to the title. Or, alternatively, state that measurements were ship based. Furthermore, the term "inter-

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hemispheric differences" should not be used here, because most readers would expect a more global coverage of observations than presented in this paper. The authors report on measurements north and south of the ITCZ in the Indian ocean, but mainly only within tropical latitudes. I would suggest something like: "Differences in the chemical characteristics ... across (or north/south of) the ITCZ ..."

Introduction, page 2375, line 10. Why is it a "unique" region? What is this referring to?

Introduction, page 2375, lines 21/21. Reference for expected (climatological) location of ITCZ should be added. Furthermore, I suggest to add one or two sentences on the role of the ITCZ for "interhemispheric" exchange in the lower troposphere along with references to earlier work on this.

Section 2.2.1, page 2377, lines 8/9. "The size ... was determined ..." I would rather suggest to refer to a size range. Define briefly the meaning of "D50".

Section 2.2.1, lines 14-19. How frequent were contamination events? In other words: For how much time during sampling (percentage) were pumps switched off on average? Explain the switch-off as a consequence of a "rapid increase" of particles >15 nm. Was there a threshold for number concentration or the rate of change of number concentration?

Section 2.2.2, page 2378, lines 3-5. Could there be significant interference of the SO₂ signal from reactions of gases with particles collected on the filter?

Section 2.2.2, page 2378, line 8. I do not understand "calculated reproducibility"? Please explain.

Section 2.3, page 2378, lines 23-26. I am surprised that there is a significant blank contribution to MSA. But more importantly, why is the blank contribution for sodium so high (on average 30%?). This is important, because sodium is used later as a reference element to derive nss fractions of various compounds. Was the blank subtracted? What is the variability of the blank? What are the resulting uncertainties for each compound?

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Isn't the agreement of duplicate samples of 25 % (on average) quite poor?

I think the data presented in this paper need a more thorough discussion on errors, blanks, blank variability and detection limits. Of particular importance are the errors for derived quantities like nss-fractions (could be differences of two large numbers) and north/south ratios. The authors might want to consider adding a table summarizing the blanks and estimated errors for each quantity discussed.

Section 2.4, page 2379. There is no information given in the paper on MBL heights. If these were not measured, what is to be expected? How well do trajectories for arrival heights of 500 m or 950 hPa represent air mass transport to the sampling system on board the ship (at 18 m height)? Furthermore, some information should be given on the amount and relevance of precipitation in the area investigated.

Section 3, page 2380, first paragraph (and Figures 2, 3 etc.). Please explain how the location of the ITCZ was actually defined in this paper. As already stated, in the discussion of Figure 3 (and in Figure 3 itself) some information on the uncertainty of data points needs to be added.

Section 3, page 2380, lines 27-28. Sea salt was calculated as a sum of the listed components. Is not Mg missing? How were "sea salt-SO₄", " sea salt -K" etc. determined? If there were calculated from the sodium concentration, why not simply say that the sea salt concentration was derived from sodium to total sea salt mass ratio in sea water (using the Wilson (1975) ratios).

Section 3, page 2381, lines 1-4. Is there any observational evidence that there is no north-south gradient in meteorological factors during the observations? Is there also no south-north gradient in the coarse mode sea salt aerosol concentration? (Some information on the coarse mode seems to be available, as discussed in the following section in context to MSA.)

Section 3, page 2381, lines 5-7. Figure 4 is discussed here. Why are there only 3 or

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4 data points of coarse mode MSA just in the vicinity north of the ITCZ. The argument would be more convincing if further information would be available also further north of the ITCZ. How do the concentrations (and gradients) of other compounds in the coarse mode fraction look like?

Section 3, page 2381, lines 18-25. While I do not doubt that the pre-existing surface size distribution plays an important role in determining the resulting MSA mass size distribution following DMS oxidation, the arguments put forward in the discussion are not entirely convincing. First of all, the total (fine plus coarse mode) of MSA is not roughly similar north and south of the ITCZ. It appears from Figure 4 it is rather a factor of two higher south of the ITCZ. To assess if higher oxidant concentrations north of the ITCZ would result in higher MSA production rates would require some chemical modeling. Furthermore, as the authors discuss, it appears that important information is missing if observations are concerned only with the sub-micron mass fraction only. Why were coarse mode measurements not done more extensively? The whole 2nd paragraph of Section 3 is quite long and appears to contain some redundant arguments. I suggest to be more careful with the conclusions and shorten the discussion.

Section 3, page 2382, lines 9-13. Parallel measurements of aerosol chemical composition, made during the same cruise but from different groups, are mentioned/discussed here (and also in the previous paragraph). References are given to Bates et al. (2002), Ball et al. (2002, Quinn et al. (2002) etc. It is stated, that a comparison of the measurements showed good agreement, but no details are given. What kind of comparisons have been made? Which parameters? In general, it appears to me a that publication of a joint paper using the joint chemical measurements of the different groups involved would have been more straightforward, in this case. (For example, in presenting merged data sets on fine and coarse mode chemical composition.)

Section 3, page 2382, lines 22-24. With respect to the "reversed" SO₂ concentration difference north and south of the ITCZ I do not find that the authors give substantial explanation in Sections 4.1 and 5.1. This interesting aspect should be addressed.

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Section 3, page 2382, lines 25-27. Is it correct that aircraft measurements were made in the MBL? Later on, page 2383, lines 1-3, reference is made to the gradient in the free troposphere. Are there MBL aircraft measurements to compare with the data presented in the manuscript?

Section 3, page 2382, lines 9-10. Does the displacement of the ITCZ (over 15 deg of latitude) not cause some exchange of air masses? What is time scale of that displacement? Is it large compared to aerosol life times?

Section 4, first paragraphs and Table 2. Table 2 contains an air mass type North- ITCZ, which appears to be not defined anywhere. With respect to the trajectory studies, some explanation should be given on the vertical travel history of air masses as air masses passing over certain areas at higher altitudes might not necessarily get in contact with surface emissions.

Table 2 and discussion of travel times (starting page 2385, lines 4-9). The authors compare observed concentrations with the time for the air mass to travel from the continent to the ship location. This comparison appears to yield some interesting insight into the causes of variability in aerosol chemical composition. However, the concept of how these travel times were determined is not adequately discussed. Table 2 presents for several types of air masses for several chemical compounds 25-, 50- and 75-percentiles of observed concentrations (or ratios). The authors also present 25-, 50- and 75-percentiles of transport times (since last contact with land). Are these really percentiles of the statistical set of all travel times (for each air mass)? If yes, these cannot be compared to percentiles of mass concentrations of chemical compounds because different statistical subsets of data would be addressed. So where is the link? How can a travel time be attributed to a median or percentile of a measured quantity? The aerosol samples showing, say, the 25 % of data with highest MSA concentrations cannot be expected to be identical with the 25 % of data with highest sea salt concentrations, for example.

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What should have been done is this: (1) divide data set into different classifications of air mass origin, (2) for each air mass category divide the data further into subsets of different travel times, (3) for these subsets the statistics (percentiles) of observed mass concentrations can be derived. Is this what the authors have done? It is not apparent from the text.

Section 4.1, page 2385, lines 10-11. What is meant by "lack of south to north gradient" in SO₂. The concentrations shown in Figure 5 are significantly different north and south of the ITCZ. This is a gradient.

Section 4.1. A conclusion is missing for this discussion (as well as partly for the following sections 4.2 and 4.3). What is the message for the reader here?

Section 4.2, page 2386, lines 1-5. In the discussion of Ca/Na ratios the authors ignore that a substantial mass fraction of both, dust and sea salt, resides in the super-micron size range which they did not measure. Furthermore, soil dust and sea salt can be expected to have undergone different transport history. Sea salt could be of local origin whereas soil dust is not.

Section 4.2, page 2386, line 9. The effect of deposition should be explained in more detail. Are there any estimates on wet and dry deposition?

Section 4.3, page 2386, lines 11-22. Again, the discussion lacks consideration of the fact that the authors have only measured the fine mode fraction. That there is no covariation between local wind speed and sea salt concentration is a weak argument. The local wind speed may not be the right parameter to look at. Can the contribution of nss sodium be estimated quantitatively?

Section 4.2, page 2386, line 23. What kind of "distribution"? Spatial distribution?

Section 5.1, page 2388, lines 2-4. This conclusion appears to be somewhat simplified and is not convincing from the discussion made earlier in the paper. A discussion needs to consider sources and sinks, as well as atmospheric life time and transport.

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Section 5.1, page 2388, lines 5-12. Please be more specific. What ratio of MSA to nss sulfate south of the ITCZ was used to infer the biogenic fractions of nss sulfate north of the ITCZ. I would prefer if a range of this inferred fraction could be given.

Section 5.2, page 2388, line 26. Where does this equation come from? A fit to own data? Error of the fit? Where was the wind speed measured? Again, does this make sense if there is no information on the coarse mode sea salt fraction?

Section 5.2, page 2389, lines 6-9. I am missing a consideration of deposition effects, in particular wet deposition. Precipitation should be significant in the tropics near the ITCZ. The argument of "integrated wind speeds" needs to be explained in more detail (or skipped).

Section 6, page 2389, lines 14-16. "Relative chemical composition is remarkably constant ... north of the ITCZ" - is this discussed in Section 4? It would help, if the relative composition would be illustrated explicitly somewhere. Figure 9 does not support this. What is the role of Figure 9 in a Section on the topic of interannual variation?

Section 6, page 2389, lines 21-22. What is meant by "potentially larger deposition losses"? What is the difference between 1998 and 1999 in terms of deposition?

Section 6, in general. What is the role of this Section? I understand the authors want to address here mainly the question of temporal representativeness of their data. This is a valid question and should be addressed. But with one cruise in one year and two more in a second year there is no way to answer this conclusively. I suggest to shorten the very detailed discussion in Section 6, because the main conclusion ("variations in source strength, source area and transport time are important causes for variations in concentrations...") at the end of Section 6 is not a surprising new finding. It rather helps to set the ground for the discussion in Section 3 and should appear earlier in the paper.

Conclusions, page 2393, lines 2-5. The aerosol sea salt concentration should be derived from a suitable reference element. That chloride is depleted due to interaction

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of sea salt aerosol with acidic sulfate aerosol is a secondary effect and should not be seen as a variation of sea salt. Is there indeed a contribution of sea salt from continental sources?? Or is there rather a sodium contribution from continental sources?

Conclusions, page 2393, lines 6-11. There certainly exists some link between time of transport and deposition losses. But this has not been thoroughly discussed in the paper. There was no quantitative estimate given of wet and dry deposition for the various compounds discussed. This conclusion is very qualitative and not very original. The same holds true for the following paragraph (lines 12-15). It is no surprise that mineral dust is enriched in air masses coming from dry land masses.

Figure 2, caption, last sentence. ITCZ identification. This should be explained in the text and in some more detail. What kind of meteorological observations were used?

Figure 3. Error bars? See comments further above.

Table 2. Explain the North ITCZ category (not mentioned in the text). Explain in detail how data in last column were derived (time since last contact with land). See my comments further above.

TECHNICAL REMARKS

Abstract, page 2374, line 6: define ITCZ

Section 2.3, page 2378, line 16. I prefer "ion chromatography" instead of "IC".

Section 2.3, page 2378, lines 17-19. "columns" or "column" (three times). Use the term consistently.

Section 2.3, page 2379, line 3. For clarity add the temperature, STP is referring to.

Section 3, page 2380, first paragraph. In the discussion of Figure 3 it is referred to 3a, 3b etc. This numbering is missing in the figure.

Section 3, page 2381, line 22. "there" instead of "here".

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Section 4.2, page 2387, line 6. "is" is missing: "This is in line..."

Section 6, page 2389, line 12. I guess it is 1998 and "1999".

Section 6, page 2390, line 0. Add an "s": "This support-s- ..."

Figure 1. The line styles for SK 1998 and SK 1999 are not easy to distinguish. This could be changed.

Figure 2 and 6. The figures with trajectories are difficult to read. The figures should be enlarged. In ACP it is well possible to make use also of colored line styles.

Figure 4. Units missing in the axis titles.

Figure 7. Units missing for top panel.

Figure 9. Units and axis titles missing.

Almost all figures: "a", "b", "c" ... numbering of sub-panels is missing.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 2373, 2002.

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