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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Total column ozone variations over oceanic region around Indiansub-continent during pre-monsoon of 2006" *by* M. C. R. Kalapureddy et al.

M. C. R. Kalapureddy et al.

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Dear ACPD Editor,

Here, we made sincere efforts to respond to the reviewer’s critical comments.

In this paper authors describe the diurnal and day-to-day variation of Total Column Ozone (TCO) over oceanic regions around India during pre-monsoon period under ICARB06 campaign. Interesting diurnal feature of TCO has been noticed. Atmosphere over Bay of Bengal (BoB) was noticed to be more polluted than Arabian Sea (AS) and hence TCO found be less over the BoB than over AS. It has also been observed that

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Arabian Sea shows relative coarse mode particle domination where as Bay of Bengal shows more anthropogenic fine mode particle domination (Kalapureddy and Devara, 2008, Atmos. Env.).

We look forward for your kind cooperation in reconsidering the article for publishing it in ACP. Thankyou very much.

With best regards Sincerely yours P E Raj (email: ernest@tropmet.res.in)

Replies on 1st Referee’s comments

GENERAL COMMENT: I strongly doubt the suitability of this type of instrument to measure systematic diurnal ozone variation. Microtops-II instruments have been used to trace day to day variability of total ozone with reasonable accuracy. Some similarities of day to day total ozone variability between satellite and the Microtops-II data (with a large unexplained offset of 23 Dobson units) have been found in the submitted study (see Fig. 2 in the paper). However, the systematic diurnal variation is basically a different feature than day to day variation and therefore the similarity in day to day variability between Microtops-II and satellite column ozone measurements hardly can be used to support the systematic diurnal variation. A systematic diurnal variation would change dramatically our present textbook knowledge, because no systematic diurnal variation is known from stratospheric ozone and 10% in total ozone variability would make a tremendous diurnal change in tropospheric ozone. Ozone in the troposphere systematically varies on a diurnal scale in the strongly polluted planetary boundary layer (namely as consequence of titration of ozone by NO during night and diurnal changes in inversion layer structure), but such a change is too low to explain a systematic variation of the magnitude claimed in the paper. At some distance away from strong emission sources (more appropriate for the measurements on the ship cruise) I don’t see any possibility for explanation of a large systematic diurnal variability and particularly the decrease in ozone concentration in the late afternoon seems very suspicious to me. Indeed, ozone in the troposphere can change substantially as

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consequence of changes in transport of polluted air masses but this would not appear in any systematic diurnal way. The authors speculate about the effect of water vapor, but I don’t see any plausible explanation for a dominant water vapor influence diminishing ozone in the afternoon when solar radiation becomes weaker. Because of my strong doubts concerning the suitability of the used instrument I don’t support the publication of the paper.

Reply: Authors understand the reviewer’s concern about the Instrument used for the systemic diurnal variation of ozone. The used instrument is Microtops-II sun-photometer and Ozonometer. This instrument is capable of making measurements on total atmospheric column aerosols and trace gases using the optical attenuation of solar energy in the specified wave lengths and field of view (FOV). This instrument is now well calibrated and accepted due to its compact size and reliable measurements on both daily and diurnal observations on the total atmospheric columnar aerosols and trace gas measurement using radiometric/sun-photometric technique (Beer-Lambert law). Hence, we strongly believe the capability of the instrument used for this work. The second part, we don’t agree fully that the statement “because no systematic diurnal variation is known from stratospheric ozone and 10% in total ozone variability would make a tremendous diurnal change in tropospheric ozone”. Besides to the ground based observational evidence of diurnal variation of surface ozone, there are some observational evidences (e.g., using a ground-based 110 GHz radiometer; Ahonen et al., 1999, IEEE@ 10.1109/ IGARSS.1999.773598) observations on short-term and long-term variations of stratospheric (in between 30 and 60 km) Ozone, which shows that there exists clear diurnal variations in stratospheric ozone as well. Furthermore, the recent, report in Science (Dominik Brunner, et al., 2008) shows there is evidence of large-scale nitrogen oxide plumes in the tropopause region and its implications on Ozone. This paper also points out about unusual high concentrations of ozone over ocean and land in contrast to seasonal change due to large scale convective plumes as also supporting our results. So, convective updraft and its interaction with stratosphere through tropopause by the vertical velocities play a vital role

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in it. Moreover, the observed large systematic diurnal variability could be due to the switching over of pre-monsoon to monsoon seasonal changes causing change in wind fields, convection and water vapor loading (which is early over ocean than over the Indian land). It is also noticed that the BoB region is observed to be relatively low pressure region compared to AS. This allows the BoB region to be convectively more active during the observational period and large scale subsidence during March causes the BoB region more polluted and causing destruction of Ozone and hence this might have been a reason for the observed difference. Initially, we took care on our observations not to be contaminated by possible cloud around the FOV. All those possible cloud contaminated data has been discarded during our quality check. Hence, we strongly believe in the instrument used and the observations taken. This kind of observations reported are believed to be first of its kind on diurnal variation of column ozone over oceans made from high resolution observations.

Other Critics:

1 COMMENT: Line 145: "If the three measured values of any parameter are not close in magnitude, the data set is rejected from further analysis": This sentence implies that the instrument produces sometimes numbers which obviously are erroneous.

The authors don’t make any statement, how they defined "parameters not close in magnitude" and how often such measurements occurred: did they exclude 5%, 10%, 20% or even more of the measured values?

Reply: Triplet observations are well known technique to take reliable observations on moving platforms. This technique has adopted to avoid any possible manual positioning errors in sun pointing during the observation period. Some of the incidents were observed when cloud traces are passing across FOV observations and may also be due to sudden track change of ship causing errors. Hence, there exists an obvious deviation from sun pointing due to the motion. So, this is not a problem with the instrument but it is only with its usage.

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2 COMMENT: Line 147: "An inter calibration/comparison of several similar ozonometers operated aboard the ship by different organizations was undertaken to ensure the reliability of measurements": How many instruments were on the ship? What means "similar" ? In what respect were the instruments different? What were the results of the comparisons? Please clarify.

Reply: About three more similar instruments used in the campaign and a sample comparison is shown in these replies for your quick check. Here our observations are shown in red closed circles (IITM) which were taken every 10 min compared to others which were made every 30 min or even hourly.

3 COMMENT: Line 149: "Data recorded around cloud passage on or near field-of-view have not been considered for the analysis": I don’t understand this sentence. How did you identify possible interferences by sub-visible cirrus clouds?

Reply: This point is made due to possible optical technique problem with the sunphotometers due to cloud in the FOV of instrument used. Yes, we do agree about interferences by sub-visible cirrus clouds on which we have no control or check. But, we took care about visible clouds.

4 COMMENT: Line 166-168: "Significant diurnal variation with well defined maximum during noon time": Particularly the coincidence of ozone maxima with local noon is suspicious to me (see general comments).

Reply: Please see the replies to the general comments.

5 COMMENT: Line 170-171: Comparison with Ernest Raj et al., 2004 (measurements at Pune): If I understood the paper of Ernest Raj et al. correctly, a Dobson spectrophotometer was operated at Pune, where also the Microtops II data showed a systematic diurnal variation: Were the systematical diurnal variations in total ozone measurements at Pune supported by Dobson data (the Dobson spectrophotometer is the standard instrument for total ozone measurements) ? Was the Dobson spectrophotometer at Pune

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well calibrated and maintained (how looked the results of the inter comparisons with standard Dobson instruments?)?

Reply: We made sincere efforts to make possible comparisons with those available at that time. We didn’t suspect the calibration because India Meteorological Dept carefully does it at systematic intervals. We may not be in a position to check that point now which is out of the scope of the paper. But, present instrument observations have been checked further with on board calibrated instruments and the sample figure comparison has shown its reliability.

6 COMMENT: Line 177-181: Significant differences between satellite and Microtops II measurements: I believe, that at least for the measurements coinciding in time with the overpass satellite measurements the reported bias between satellite and Microtops II measurements is much too large to be attributable to differences in slant path.

Reply: Agree about the large difference between ground based Microtops point observation and TOMS observations. This might have taken as constant bias between these two measurements platforms. Moreover, here in this work we are looking on relative difference in magnitude hence this wouldn’t show deviation much from the fact.

7 COMMENT: Line 178: When looking at Fig 2, top panel I would not say, that the day to day variations are "very much in phase with each other"

Reply: Yes, in some cases it is not in phase. Authors agree. But in almost all the cases the TOMS observations are well within the variance of the Microtops observations.

8 COMMENT: Fig. 4: Is this Figure obtained from the measurements interpolated along the ship track? I don’t believe, that the ship measurements reached 5o S in the Western part of the measurements

Reply: Right, it is an extrapolation down to 5 N from 8 N. It will be corrected in the revision.

9 COMMENT: The similarities of spatial correlations between OMI-TOMS and

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Microtops-II measurements are not obvious when looking at Fig. 4, 5a and 5b (at least the high values in the Western part of Fig. 4 are not visible in Fig. 5b).

Reply: Here our idea is to show the magnitude difference between the regions (1st leg for the BoB and 2nd leg for the AS) at two specified periods. One may not expect to see exact one-to-one similarity between ground-based and onboard satellite observations due their obvious special and temporal coverage differences besides the different techniques involved in the computations to derive the useful end products.

10 COMMENT: The discussion of the paper is not well structured: the paper of Lelieveld et al.(2004) discusses long-term changes of ozone measured from ships over the Atlantic, which is a different item than the systematic diurnal variation claimed in the paper. I don’t remember, that Lelieveld et al. reported about diurnal variability in surface ozone measurements of the Atlantic.

Reply: We will take more care about the discussions and over all outlook of the paper according to the reviewer’s critical suggestions/comments in our revision. Lelieveld et al. has not made any attempt on columnar ozone diurnal variation.

References: Ahonen, R. Hallikainen, M. Kyro, E. Observations of short-term and long-term variations of stratospheric ozone using a ground-based 110 GHz radiometer. Geoscience and Remote Sensing Symposium, 1999. IGARSS '99 Proceedings. IEEE 1999 InternationalPublication, Vol.1, 663 - 665 (1999).

Dominik Brunner, et al. Large-Scale Nitrogen Oxide Plumes in the Tropopause Region and Implications for Ozone, Science 282, 1305 (1998);

Lelieveld et al., The Indian Ocean Experiment: Widespread Air Pollution from South and Southeast Asia, Science, 291, 1031,(2001)

Replies on 2nd Referee’s comments

Scientific comments: The background total ozone value for the days studied seems to be about 260 DU which is a reasonable value for that time of the year. Also the polluted

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values obtained are consistent with those obtained by Fishman and his coauthors, and by Thompson and her co-authors. Stehr et. al, 2000 (JGR,107) made measurements on board a ship in the Indian Ocean of ozone concentrations during the INDOEX campaign. They report a diurnal variation of from 19 to 27%. They found that the minimum was in the afternoon and the maximum in the early morning (contrary to what one is used to). They attribute the phenomena to surface halogen chemistry. In more polluted atmospheres, over land, one also finds a strong diurnal variation, but with a maximum in late afternoon. So I cannot dismiss the results based on the science.

Reply: Suggestion received positively and necessary addition have been made in the revised version. All the earlier observations are either surface or radiosonde observations not on columner ozone. Hence, this could be the first of its kind on atmospheric columnar ozone diurnal observations.

2 COMMENT: The question does arise as to whether the instrument used has any diurnal bias. This could be tested for by looking at an area where the tropospheric pollution is small, that is where the column measurement is about 260 DU (i.e. the stratospheric value) Having said all that, I do not believe that the paper should be published as is. The statements I have made above could and should have been made by the authors. The authors need to do some further analysis to convince the reader of the veracity of their results.

Reply: Microtops II is now well accepted on its observations on both daily and diurnal measurements on the columnar aerosol and trace gas by the radiometer/sunphotometer community. Hence, we strongly believe in the technique and the capability of instrument used for this work for making column ozone measurements and hence reported our observations and results.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 3143, 2008.