Response to the comments of Reviewer #1

General remark: This paper is based on the surface (ship-borne) measurements of ozone, CO and methane over the Bay of Bengal (BoB) during summer/monsoon months of year 2009. The main objective of this study is to investigate the spatiotemporal variations of trace gases. The WRF-chem simulations have been compared. Case studies mostly reduced levels of ozone during rainfall events have been investigated in details. Inferences from surface measurements over the land (India subcontinent) have been used to study the en route transformation (net O3). The data and analysis is good but the discussion needs somewhat better and concrete interpretation. The paper may be accepted but following aspects need improved or more considerations.

We thank the reviewer for careful evaluation of the paper. The paper is suitably revised by incorporating the reviewer's suggestions and comments. Please note that the line number mentioned in the reply is corresponding to the revised version ("CTCZ-BOB-R1_Track_Changed").

(1) Dividing BoB in regions southern, central and north, etc. is not impressive as transport of air mass is highly variable. I agree with categorization based on the trajectories. Characterizing the air masses measured over coastal and open oceans are valuable.

Following reviewer's suggestion, we removed the discussion based on BoB region's division (Page: 1, Lines: 21-26; Page 7, Line: 244-248) and primarily used categorization based on trajectories. The computations of region wise mean values are only used for comparison with other seasons (subsection 5.4) obtained from previous papers. Such consistency of region is required for comparisons considering strong spatial variability over the BoB, more pronounced during winter (David et al., 2011, Nair et al., 2011)

(2) Significant data measured during the stationary phase (of ship) has been used. Typically, researchers reject such data. I think data measured during this period should not be used.

A contamination of ship-based observation could occur when ship is stationary and winds are calm. Samples taken upwind are not contaminated when winds are strong carrying the exhaust downwind. Here, when ship is stationary and wind speed is above 5 ms^{-1} in a direction opposite to the instruments as if ship is in motion relative to the air getting sampled, observations are not affected.

To further discard any effects, we used continuous NOx measurements as tracers of ship exhaust. Though NOx observations are not reported in the paper as the NOx levels were mostly below or around the detection limit of the instrument (1 ppbv), except during the effects of ship exhausts. A case of data filtering is shown below:

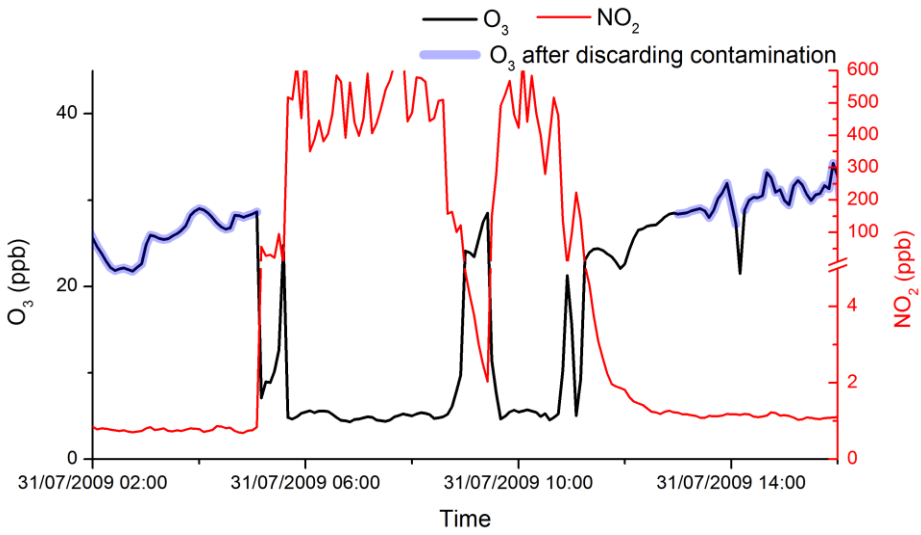

Figure shows variation of O_3 and NO_x on July 31, 2009. It is clearly seen that NO_x level was ~1 ppbv up to 05:00 hours and abruptly increases to 100 ppb reaching up to 600 ppb under the influence of ship-exhaust, this clearly discriminate the contamination. We have discarded the observations for such contaminations. This exhaust episode occurred when ship was rotated for oceanographic measurements.

(3) En route transformation of ozone has been assessed with reference to several station based measurements over the land. Instead of relying on observations, model data should be used / compared to estimate the en-route transformation of ozone.

We conduct simulations on a larger domain to estimate the en route ozone formation. Model simulations indicate that ozone production rate is about 4.6 nmol mol^{-1} day^{-1}. (Fig. 7; page-11, line: 381-387)

(4) Methane data have been overlooked, or else, can be removed from the draft.

As replied also to referee #2, we have extended the Methane discussion. Despite of longer chemical lifetime, observed spatial heterogeneity in CH_4 highlights the influence of transport from different source regions located in India to the BoB during the summer monsoon. Further analysis using CH_4 retrievals from SCIAMACHY have been conducted, showing that stronger methane sources are located in central/northern Indian region as compared to southern India (new Figure 6). This is, in agreement with the result based on trajectory assisted analysis, showing that air masses from the central India have significantly higher CH_4 over the BoB than other air masses. The discussion is suitably revised in the manuscript (Figure 6; Page 10, Lines 328-343). It is further inferred from sector-wise analysis of emissions over the hotspot region (i.e. eastern IGP) that these higher CH_4 emissions are due to rice cultivation, waste treatment and livestock (Page: 10, Lines 338-339).

Modeling CH_4 from the WRF-Chem setup used here was not possible as this tracer is updated only through chemical boundary conditions from a global model (MOZART). Nevertheless the observational values are presented here as reference for future studies. The correlation between

the presented in situ CH_4 measurements with retrievals from AIRS satellite instrument was found to be statistically insignificant (not shown) which further highlights the need of reporting in situ measurements from this region. This is also discussed in the revised manuscript (Page 10, Lines: 339-343).

(5) There are too many Figures (16), some data plots are repeated.
Therefore, improved representations of figs and tables are also required.
Following reviewer's suggestion, representation of figures and tables has been improved and total number of figures has been reduced in the revised manuscript (from 16 to 12).

(6) There is scope of improving English. Excessive use of "WE" "OUR" "FIRST", etc. is not desirable.
Use of "we", "our" and "first" has been minimized. English improvement has been made.

Following detailed and specific comments should be considered.
Abstract:
Page 1, Line 32-33 "simulations for a low-O3 event on August 10, 2009.....transporting ozone-rich airmasses" This sentence seems contradictory as low -ozone is explained by transport of ozone rich air?
Sentence is suitably modified.

Introduction:
Page 2, Line 64 "The marine regions adjoining South Asia, despite being far from direct anthropogenic activities," It is not really true, marine regions of AS and BoB are surrounded by polluted land of SEA and SA. If authors like to convey that there are no significant emissions (except ships) then it is well understood and do not require to mention "despite being far from...."
As suggested, the segment "despite being far from direct anthropogenic activities," is removed.

Page 3 line 69: "The airmasses exposed to ....." better to rewrite, "exposed" is not an appropriate choice.
Thanks for the suggestion. "Exposed" replaced by "influenced".

Page 3 Line 70-72: "In situ measurements over the.....transformation." This is not well written, please re-write.
Sentence is re-written as "In situ measurements over the marine regions are required to understand the effects of direct outflow, en route chemical transformation, and the chemistry in the transported airmasses".

Page 3, Line 81-83: "Both the export of .....transport of ...synoptic scale dynamics and monsoonal circulations" This is not well written what is the difference (scientifically) between export and transport.
The sentence is re-written as "Transport of airmasses between Indian subcontinent and adjacent marine regions has strong seasonal dependence associated with the monsoonal circulation"

Page 3, Line 85: "highly conducive for the accumulation of trace species". It is not clear, do the outflows from continents stop over BoB? I mean ACCUMULATION is not an appropriate choice?
"accumulation" word is removed and sentence is suitably modified.

Page 3, Line 96: "and an unnamed campaign" this does not sound good, write under "other campaigns "etc.
Suggestion is incorporated.

Pag3, Line: 103 ", which influences the oxidation capacity of the atmosphere" This is not required, in other seasons oxidation capacity can also be influenced due to higher levels of VOCs. So it is not unique for this season.
Yes, modified accordingly

Pag4, Line: 106 "remote sensing of.." here, there is no need to give examples of TES or AURA, it is in general true for any remote sensing technique.
Suggestion is incorporated.

Page4, Line: 114-115 and elsewhere "spatial and temporal variations in ozone" should be "spatial and temporal variations of ozone"
Suggestion is incorporated.

Page4, Line: 117 "We investigate ...we have ..greater detail." Please re-write "we" has come twice?
Suggestion is incorporated.

Page4, Line: 118-120: This may be deleted
Suggestion is incorporated.

2. The cruise track and background conditions.
Page 4, Line 123-124: Revise as "Figure 1 shows the cruise track of the Oceanic Research Vessel (ORV) Sagar Kanya during the CTCZ campaign (cruise number SKC3 261).
Sentence is revised as suggested, except "SKC3 261". The cruise number "SK 261" given by NCAOR (National Centre for Antarctic and Ocean Research), Goa, India is used.

This section is partly explained in a rather lengthy caption (Fig1). Try to adjust most explanations in this text sections but not in captions.
The caption of Figure 1 is shortened significantly and explanation is given now in the section 2.

"To take time series measurements, the ship was kept stationary at 89_ E, 19_ N for fifteen days (July 22 to August 06, 2009)." Usually researchers reject the measurements when ship is stationary, as it is proved that exhaust from ship influences the measurements of trace constituents. An explanation is required how it is ensured that ship's exhaust did not influence measurements around the measurement location?
Please see the response corresponding major comment #2.

Line 129-131: "The average prevailing wind patterns at 925 hPa during the cruise period are obtained from NCEP/NCAR reanalysis (http://www.esrl.noaa.gov/psd; Fig. 1). The prevailing westerly or south-westerly winds are conducive for the transport of ozone and its precursors from the Indian landmass to the BoB during the summer monsoon season." revise as here: "The average wind pattern at 925 hPa (NCEP/NCAR reanalysis; http://www.esrl.noaa.gov/psd) during the cruise period is shown in Figure 1. The prevailing westerly and southwesterly winds transport ozone and its precursors from the Indian landmass to the BoB during study period."
Suggestion is incorporated.

Line 133: Phase B (INTEX-B), Does this correspond to Indian summer season?
INTEX-B inventory provides annual mean emissions. References using INTEX-B inventory are provided in the manuscript (Page 6-7, Lines 214-216).

Line 134: Relatively high NOx emissions are located over parts of eastern and southern India.? This is arguable if compared with emissions over IGP and western India (which is not shown). So change the sentence accordingly.
Sentence modified accordingly, NOx emissions are higher over eastern and southern India as compared to that of central Indian.

3. Experimental details and data
Page 5, Line 146 "This instrument was based on the principle.." something like this is better "This instrument works on the principle.."
Corrected accordingly.

Page 5, Line 135-154 "Trace gas measurements affected by the ship exhaust ....." This is the issue when ship is stationary, irrespective of wind direction.
In strong winds, measurements made upwind direction are not affected by ship exhaust, Nevertheless additional filtering have been implemented as mentioned in the response to major comment #2.

Page 5, Line 164-165 following should be better "at 5-minute of integration time using an automatic weather..."
Sentence modified accordingly.

So far, it is not clear why measurements at Thumba, Thiruvananthapuram have been discussed in this draft. Objective of using the Thumba data is missing.
The suggested discussion is added in revised manuscript (Page-6, Lines 183-184).

5. Results and Discussion
Page 6 Line 205: "period of the summer monsoon season" can be deleted. "summer season" is being repeated again and again.
It is deleted.

Page 6 Line 206-207: revise as here "The mixing ratios of trace gases show large spatio-temporal variations over the BoB."
Revised accordingly.

Page 6 Line 207-209: can be summarized as "Levels of O3 and CO varied in the ranges of 8-54 nmol mol-1 (with average of 29.7±6.8 nmol mol-1) and 50-198 nmolmol-1 (average of 96±25 nmol mol-1), respectively. "
Changed accordingly.

Page 7, line 220-221: " In addition to sailing across the BoB....." use of stationary ship data is questionable, strong justification is required or remove this data.
Please see the response corresponding major comment #2.

Page 7, line 238-242: " Similar variations in O3 mixing ratios and residence time over continental India indicate the influences of transport from ..... and en route photochemistry." This does not go well. In this paper, it is explained that there is en route formation of ozone, so ozone is formed also over the oceanic region. Therefore this relationship study between residence time (over land) and a secondary species is meaningless. However, to some extent it is meaningful for primary pollutants such as CO and also for CH4. These processes: source strengths, vertical mixing or dilution, and en route photochemistry (or their variability) are not occasional but are continuous. Therefore, and overall, consistent discussion using the residence time calculation is required. It is expected to see best relation between CO and residence time, at least better than O3.
We have revised the calculation of residence time by taking only those hours when air parcels is typically within an altitude of 1.5 km over land, as residence of air masses over continent (but aloft) might not get direct influences of surface emissions. The revised analysis shows better correlation for primary species (R = 0.4 for both CO and CH_4) and slightly lower for secondary species (R=0.3 for O_3). The manuscript has been suitably revised to incorporate these changes (new Fig. 4 and Page 8, line 272-277).

Why CH4 is not influenced by the change in residence time?
Please also see the response to your previous comment. CH_4 is also found to be influenced by residence time as shown in the revised analysis (Fig-4, page-8, line 276-277).

Page 7-8, line 250-287: This discussion should be shortened, this lacks completeness. All inferences have been derived using two point measurements at Thumba and Anantpur. I feel, if model is doing good job then rely on model data for such discussion. Otherwise question may arise ; 1. Os the distribution of ozone and southern India homogeneous over central/southern India? 2. Is en-route transport from India the only factor controlling ozone over BOB? downdraft of O3-rich/poor air, or transport from other regions such as SEA are not relevant? Therefore, detailed discussion insightful analysis is required, otherwise, just shorten this part.
We agree that estimation here has been based on very limited observations. Following reviewer's suggestion, now we investigated the en route ozone production by analyzing model simulated O_3 along the air mass trajectories at several representative locations in the BoB.
Based on modeled O_3 along airmass trajectories ending over BoB, we find that ozone production rate is about 4.6 nmol mol^{-1}. The new analyses and discussions are incorporated in revised manuscript (new Fig. 7; Page:11, Lines: 380-387)

5.2 WRF-Chem simulations:

Throughout the draft: It is not nice to see frequent use of "we", "our" and "first time". please try to minimize the use of such words.
Thanks for the suggestion. The use of “we”, “our” and “first” is minimized.

Page 9, line 292-293: "variations in the meteorological parameters simulated by the model are briefly evaluated" This analysis is beyond the scope of this paper, I suggest to remove Figure 8.
Figure is removed as suggested. Comparison between simulated and observed meteorological parameters is mentioned in one sentence.

Page 9, line 312: " that is the mean value subtracted from the mean diurnal pattern,.."
This is not clear?
Following reviewer#2’s suggestion, this section (Diurnal variation) is removed in the revised manuscript.

Page 9, line 315: "Ship exhaust contaminated the observations for a period of time between 5 to 14 hours long;.." Here is the entire issue of using stationary data. Questions:
How it is ensured that rest of hours were not impacted by ship exhaust? Why this period (5-14 hr) is fixed on each day? Second, how about residual air mass (aging of ship exhaust), which can definitely change photochemistry of O3 during rest of the hours. Suggestion: Do not use stationary phase data, you have got great deal of other results to focus on. How it is ensured that rest of hours were not impacted by ship exhaust?
The ship was rotated for oceanographic measurements, typically conducted between 5–14 hours making sampling inlet downwind ship exhaust. Please note that the section being referred to showing the average diurnal variation has been removed following reviewer#2’s suggestion

Second, how about residual air mass (aging of ship exhaust), which can definitely change photochemistry of O3 during rest of the hours.
The direct influences of exhaust are swept away by strong southwesterly winds. As wind direction remained same, there would be minimal effects of the aging downwind on the upwind measurements, residual effects being similar to what would be caused by mixing of the background with emissions caused by ship transportation in the BoB.

Page 10, line345-346: "We suggest that, in the presence of ozone-poor airmass aloft, a downdraft would result in reductions in surface ozone mixing ratios." This is reasonable but if mid-troposheric air (typically O3 higher than at surface) is down-drafted then one may have opposite scenario. This needs to be mentioned.
This is mentioned now. (Page-12, lines 424-425)

Page 12-13, line 432-442: The cause of seasonality is explained in very general terms, and these are well known. Or at least new insights have not been presented about the seasonality. I suggest removing this part.
Here, we wanted to show how the measurements during CTCZ experiment fills the gap in seasonal variation of trace gases over BoB. This is especially important considering that it is minima of CO seasonal cycle during the monsoon so the CTCZ measurements complete the

information to get overall seasonal variability. Also following the comment of Reviewer#2, we are retaining this section.

Table:

Table1: Only references are not enough. Please prove unique/salient features of the option for different atmospheric processes in second column and third can be used for the references. Little more elaborated table is required which will justify the options used for atmospheric processes.
Table is revised and an extra column about the features of the schemes is added.

Table 2: Revise as here: "Table 2. A comparison of average surface O3 mixing ratios measured at various sites during summer monsoon period.*boundary layer ozone over the Arabian Sea. " Also Arrange the Table properly, for example, Ahmedabad data are coming at 3 different rows. Better to show "mean +/-1-sigma format" rather showing mean and 1-sigma in two different columns.
Table 2 and its caption are revised accordingly.

Table 4: Last row "No name" , better leave it blank
Suggestion is incorporated.

Figure

Figure 1. Caption is very lengthy. What is the unit of wind speed (m/s)? Is NOx data corresponding to period of observations? Revise the map so that NOx emission over entire (southern/central) continents is covered. This is required as back trajectories pass through the beyond the domain shown in present map.
Figure 1 is revised accordingly. Caption is shortened and detail is mentioned in the text. NOx emissions are shown over India covering southern and central regions.

Figure 2. Revise the map as suggested for Figure1 (to show the distributions of trace gases over entire southern and central India).
Figure 2 is revised accordingly. Distributions of trace gases over entire southern and central India are shown.

Figure 4. The color scale should be further resolved (1km is not good enough, at least 500m would be better), as I only see the red (mostly). Also show symbol along the trajectories for each back day. This will help to understand "residence hour" calculation.
Figure is revised accordingly. Colour scale is resolved to 500m. The cross symbol along the trajectories for each day is also shown. (Please see Fig. 3)

Figure 5. Why UT is used in time series plot, while LT is used in a diurnal plot? Better to use LT in all plots (other figures also). Again captions are too long, legends and colors are good enough, no need to mention or repeat same in text (caption).
LT is used now in all the plots as suggested. Caption has been shortened.

Figure 6. This figure is not impressive and not required (already you have 16 figs). Instead, A few lines in text should be okay.
This figure is removed.

Figure 8. This is redundant figure, already results have been summarized in Table 3. I suggest to remove this Figure.
This figure is removed.

Figures 5 and 9 can be combined: I do not understand why same data (residual hours) have been plotted in 3 different panels. Keep just one (may be in bottom panel). Instead of residual hours (right y-axis), plot WRF chem results.
Figure 5 and 9 are combined now. While residence time is shown in bottom panel, WRF chem results are shown corresponding right y-axis. (See Figure 4)

Figure 10. Again, there is need to shorten the caption, do not explain the legends in details.
This caption is also shortened.

Figures 11 and 12 can be merged: Instead of Figure 12, plot temp data in left-y axis in Fig11. No need for Figure 12.
Figure 11 and 12 are merged. (See Figure 8)

Response to the comments of Reviewer #2

This manuscript presents measurements of O_3, CO and CH_4 made from a ship sailing in the Bay of Bengal (BoB) during 2009. The work investigates the spatio-temporal variation of these trace gases, looking at the relationship between their observed mixing ratios and air mass origin and also investigates how well WRF-chem simulations can reproduce the observations. The paper is suitable for ACP and should be accepted subject to the following minor revisions:

We thank the reviewer for careful evaluation of the manuscript and valuable comments. All the comments and suggestions are incorporated as discussed below. Please note that the line number mentioned in the reply is corresponding to the revised manuscript "CTCZ-BOB-R1_Track_Changed".

General comments:
Section 5.1: The first part of the analysis looks at the variation in concentrations of the trace gases along the cruise track and attempts to explain them by looking at air mass origin (the % of residence time over land). This is shown nicely in figure 5, however I feel figures 2 and 3 could be merged (in general the paper has too many figures).
Figure 2 and 3 are merged in revised manuscript (please see Figure 2). Number of figures is now reduced to 12 (from 16).

The data seems to be divided into two regions (central and northern BoB) and I am not sure this is necessary. The difference in data taken in different areas in more likely to be driven by air mass origin rather than the area that the ship was in so I would stick to this analysis.

We have removed the discussion based on BoB region's division (Page: 1, Lines: 21-26; Page 7, Line: 244-248) and primarily used categorization based on trajectories (see also comments to reviewer#1). However, the computations of region wise mean values are only used for comparison with other seasons (subsection 5.4) to be consistent with previous papers. Such consistency is required for comparisons considering strong spatial variability over the BoB more pronounced during winter (David et al., 2011, Nair et al., 2011)

Section 5.2: In general I feel this section could be expanded. Why has CH_4 data not been investigated with the model here? From figure 5 it seems that there is reasonable agreement between the observed CH_4 and residence time over land of the air so it would have been interesting to see how well the model reproduced the CH_4. In general CH_4 data is often overlooked in the paper, even though the dataset seems reasonably complete and CH_4 is mentioned in the title. If the authors are not confident in the CH_4 measurements then they should be removed.

The section is expanded by adding the analysis of the influence of India's anthropogenic emissions to O_3 over BoB using model sensitivity simulation (See revised Fig 4, Page-11, Lines: 375-379). CH_4 from WRF-Chem was not analyzed as the existing model setup did not include explicit treatment of regional emissions of CH_4, being included through chemical boundary conditions from global model MOZART. Nevertheless the observational values are presented here for their use in future studies. To investigate the spatial variability in observed CH_4, retrievals of SCIAMACHY are now analyzed (Fig. 6) which reveals higher methane concentration over central Indian region compared to southern Indian region during the study period, complementing the trajectory assisted analysis.

We are confident that our measurements and CH_4 data are reasonably complete to derive the spatial variation. Interestingly, despite of longer chemical lifetime, the observed spatial heterogeneity in CH_4 highlights the importance of transport from different source regions located in India to the BoB during the summer monsoon. It is further inferred from sector-wise analysis of emissions over the hotspot region (i.e. eastern IGP) that high CH_4 emissions are due to rice cultivation, waste treatment and livestock (Page: 10, Lines 338-339). The relevant discussion is also suitably revised in the manuscript (Page 10, Lines 328-343).

The correlation between presented in situ CH4 measurements with retrievals from AIRS satellite instrument was found to be statistically insignificant (not shown) which further highlights a need of reporting in situ measurements from this region (Page 10, Lines: 339-343).

Can the authors comment on the main in source of the increased ozone (e.g. anthropogenic / biogenic emissions).

We performed additional model simulation by switching off anthropogenic emissions in the model domain. As shown in revised Fig. 4, the spatio-temporal variations in O_3 over the BoB are mainly controlled by the regional anthropogenic emissions over the South Asia. On average, O_3 mixing ratios over the BoB are predicted to be reduced of about 14 nmol mol^{-1} in lack of anthropogenic emissions in South Asia. The manuscript is suitably revised to include the new analyses and related discussion (Fig. 4, Page-11, Lines 375-379).

What levels of NOx are seen in the model?
Mean NOx mixing ratios along the ship are calculated to be 135 ± 90 pmol mol^{-1}.

The comparison of meteorological parameters from the model does not add much to the analysis and the authors should consider removing it (which helps reduce the number of figures).
As suggested by the reviewer, we removed the comparison in the revised version of the manuscript.

Could the authors also compare model data to the measurements at the surface sites? This would help assess how well the model predicts the air coming into the region and whether this contributes to any discrepancies in the data after emissions and processing.
Detailed evaluation of WRF-Chem simulated ozone over surface sites in India has been conducted by Kumar et al., (GMD, 2012b). A comparison of O_3 measurements at Thumba with model showed a good agreement (R^2=0.6) and mean values compared typically within 1-standard deviation. We have shown surface O_3 is simulated within the 1-sigma variation at another stations (Gadanki) in southern India during monsoon (Ojha et al., 2016, Fig-8). This information is now provided in the revised manuscript (Page: 11; lines: 369-374).

Section 5.3: It seems that much of the data here has had to be removed due to contamination from the ship exhaust. This causes a large gap in the diurnal average where there is no data between 0600 and 1300, a time of particular interest for photochemistry. Because of this the authors should consider removing this analysis.
Following reviewer's suggestion, this section is removed in the revised manuscript.

Section 5.4: Figures 11 and 12 seem to essentially show the same thing – could the authors combine them somehow.
Suggestion is incorporated (Please see Fig. 8).

Section 5.5: The seasonal variation is investigated by examining data from a series of previous publications of measurements in the region, presented in table 3. The analysis here is good, however I find table 3 hard to interpret. Could the data presented as a figure?
The data of table 4 (seasonal variation) is already shown in Fig 16 (which is now Fig. 12 in the revised manuscript)

Minor comments:
Both 'O_3' and 'ozone' are used throughout the text. The authors should pick one and stick to it.
"O_3" is now used throughout the revised manuscript.

Line 151: How were the analysers calibrated? A few lines of detail and references should be given here.
Suggestion is incorporated (Page-5, Lines 168-171 and 181).

The authors should try to avoid excessive use of the terms 'we' and 'our' when describing the results.

Excess use of “we” and “our” is avoided in revised manuscript.

Figure 4 is very hard to interpret – could the authors find a clearer way of showing air mass origin for the different positions on the cruise track?

To make the figure clearer now only representative trajectories (instead of all) are shown. Following suggestion of Reviewer #1, a symbol is added along to trajectories representing a time difference of one day. (Please see Fig. 3)

Variations in O_3, CO, and CH_4 over the Bay of Bengal during the summer monsoon season: Ship-borne measurements and model simulations

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Abstract

We present ship-borne measurements of surface ozone (O_3), carbon monoxide (CO) and methane (CH_4) over the Bay of Bengal (BoB), the first time such measurements have been performed~~taken~~ during the summer monsoon season, as a part of the Continental Tropical Convergence Zone (CTCZ) experiment during 2009. O_3, CO, and CH_4 mixing ratios exhibited significant spatial and temporal variability in the ranges of 8–54 nmol mol^{-1}, 50–200 nmol mol^{-1}, and 1.57–2.15 µmol mol^{-1}, with means of 29.7±6.8 nmol mol^{-1}, 96±25 nmol mol^{-1}, and 1.83±0.14 µmol mol^{-1}, respectively. ~~While the airmasses were mainly from northern or central India over northern BoB, they were from southern India over central region of BoB.~~ The average mixing ratios of trace gases over ~~northern~~ BoB in airmasses from ~~northern or~~ central/northern India (O_3: 30±7 nmol mol^{-1}, CO: 95±25 nmol mol^{-1}, CH_4: 1.86±0.12 µmol mol^{-1}), ~~in airmasses from northern or central India, did not differ much~~ were not statistically different from those in airmasses from southern India~~over central BoB~~ (O_3: 27±5 nmol mol^{-1}, CO: 101±27 nmol mol^{-1}, CH_4: 1.72±0.14 µmol mol^{-1})~~, in airmasses from southern India~~. Spatial variability is observed to be most significant for CH_4 with higher mixing ratios in the airmasses from central/northern India, where higher CH_4 levels are seen in the SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY) data. ~~n mean O_3rate of about 4.6 nmol mol^{-1} day^{-1}. The ship-based observations, in conjunction with backward air trajectories and ground-based measurements over the Indian region, are analyzed to estimate a net ozoneO_3 production of 1.5–4 nmol mol^{-1} day^{-1} in the outflow.~~ ~~Ozone~~O_3 mixing ratios over the BoB showed large reductions (by ~20 nmol mol^{-1}) during four rainfall events. Temporal changes in the meteorological parameters, in conjunction with ~~ozone~~O_3 vertical profiles, indicate that these low ~~ozone~~O_3 events are associated with downdrafts of free-tropospheric ~~ozone~~O_3-poor airmasses.

While the observed variations ~~in~~of O_3 and CO are successfully reproduced using the Weather Research and Forecasting model with Chemistry (WRF-Chem), this model overestimates mean concentrations by about 6 and 16~~20~~% for O_3 and CO respectively, generally overestimating O_3 mixing ratios during the rainfall events. An analysis of modeled O_3 along airmass trajectories show mean en route O_3 production rate of about 4.6 nmol mol^{-1} day^{-1} in the outflow towards the BoB. Analysis of the ~~chemical~~ various tendencies from model simulations during ~~for a low-~~ ~~O_3~~an event on August 10, 2009, ~~captured successfully~~ reproduced by the model, shows ~~the key role of~~ horizontal advection ~~in~~ rapidly transporting ~~ozone~~O_3-rich airmasses from near the coast across the BoB. ~~Our~~ This study fills a gap in the availability of trace gas measurements over the BoB, and when combined with data from previous campaigns, reveals large seasonal amplitude (~39 and ~207 nmol mol^{-1} for O_3 and CO, respectively) over the northern BoB.

1. Introduction

Tropospheric ozone (O_3) is the third most important greenhouse gas, contributing to global warming and climate change with a~~its~~ radiative forcing of 0.40±0.20 Wm^{-2} (IPCC 2013). O_3 is also a pivotal trace gas in tropospheric chemistry, as it is a major source of hydroxyl radical (OH), which removes most of the organic compounds and pollutants from the atmosphere and controls the oxidation capacity of the troposphere (e.g. Brasseur et al., 1999; Finlayson-Pitts and Pitts, 2000; Seinfeld and Pandis, 2006). Further, enhanced concentrations of surface O_3 have detrimental effects on human health and vegetation (Heagle, 1989; Seinfeld and Pandis, 2006). Approximately 80% of tropospheric O_3 is produced by in situ photochemical reactions in the presence of nitrogen oxides (NO_x = NO + NO_2) involving the precursor gases of methane, non-methane hydrocarbons (NMHCs), and CO (Fishman et al., 1979; Crutzen et al., 1999; Seinfeld and Pandis, 2006). The remaining 20% of tropospheric ~~ozone~~O_3 is attributed to intrusions of stratospheric air during frontal activities or to tropopause folding events (Lelieveld and Dentener, 2000; Sprenger et al., 2007). Depending upon meteorological conditions and the availability of the aforementioned precursors, a net production or destruction of O_3 prevails. The average lifetime of ~~ozone~~O_3 is about one week in the lower troposphere, which leads to large variability in its spatial and temporal distributions, as compared to the long-lived greenhouse gases. The budget of tropospheric ~~ozone~~O_3 and its implications for human health, crop yields, and climate are, however, not yet well quantified, especially over regions in Asia. This is mainly due to a lack of~~insufficient~~ in situ measurements (e.g. Cooper et al., 2014; Monks et al., 2015).

Carbon monoxide (CO) is an indirect greenhouse gas which also has adverse effects on the health of humans and animals (WHO 1999). Although it does not have a direct greenhouse effect like methane and~~or~~ carbon dioxide, its role in atmospheric chemistry is estimated to cause an indirect radiative forcing of 0.23 (0.18–0.29) Wm^{-2} (IPCC 2013). The major sources of CO are fossil fuel combustion, biomass burning, and oxidation of hydrocarbons such as CH_4 and isoprene (e.g. Jacob, 1999; Bergamaschi et al., 2000; Seinfeld and Pandis, 2006).

Methane (CH_4) is one of the major greenhouse gases, with a direct radiative forcing of 0.48±0.05 Wm^{-2} (IPCC 2013). This gas plays a major role in the climate and in atmospheric chemistry. CH_4 is emitted from variety of

natural and anthropogenic sources (Jacob, 1999) and is removed primarily through its reaction with OH radicals (Fung et al., 1991, Seinfeld and Pandis, 2006).

The marine regions adjoining South Asia~~, despite being far from direct anthropogenic activities,~~ have been observed to have elevated levels of surface O_3 due to the outflow of continental pollution (Lawrence and Lelieveld, 2010) and minimal chemical loss by titration (e.g. Lal and Lawrence, 2001; Ojha et al., 2012). Suggested sources for this elevated ~~ozone~~O_3 and other trace gases observed over the marine regions surrounding India are anthropogenic, biomass burning, and biogenic emissions over continental India (Naja et al., 2004; Lawrence and Lelieveld, 2010; Nair et al., 2011; David et al., 2011). The airmasses influenced by ~~exposed to~~ continental emissions undergo chemical transformation, including ~~ozone~~O_3 production, during their transport to the cleaner marine regions. In situ measurements over the marine regions are required to understand the effects of direct outflow, en route chemical transformation, and the chemistry in the transported airmasses.~~In situ measurements over the marine regions adjacent to the South Asia are therefore required to understand the chemistry in the airmasses transported to the marine regions, the effects of direct outflow, and en route chemical transformation.~~ (Lawrence and Lelieveld, 2010, and references therein).

The experiments that have been conducted to date over the marine environment adjacent to the Indian region have revealed considerable spatial heterogeneity in the distribution of trace gases and aerosols, influences from source regions such as the Indo-Gangetic Plains (IGP), and radiative impacts (Nair et al., 2011; David et al., 2011; Mallik et al., 2013; Moorthy et al., 2009; Nair et al., 2010). Observations made during the Indian Ocean Experiment (INDOEX; Lal and Lawrence, 2001) and model simulations (Ojha et al., 2012) both found the ~~ozone~~O_3 mixing ratios over these remote marine regions to be even higher than those over the upwind continental regions, due to complex ~~ozone~~O_3 chemistry. Lawrence and Lelieveld (2010) provided a detailed review of the outflow of trace gases and aerosols from South Asia to the surrounding marine regions. ~~Both the export of continental airmasses and the transport of marine air to the continental regions have~~ Transport of airmasses between Indian subcontinent and adjacent marine regions~~of continental airmasses to marine regions and marine airmasses to continental regions~~ has strong seasonal dependence ~~which is~~ associated with the ~~changes in synoptic scale dynamics and~~ monsoonal circulation~~s~~ (e.g. Kumar et al., 2015).

The marine environment of the Bay of Bengal (BoB), the largest bay in the world, is surrounded by landmasses on three sides, making it highly ~~conducive~~ suitable to observe ~~for the accumulation~~elevated enhanced concentrations of trace species. Further, seasonal changes in synoptic winds make this a unique region to study variations in trace species due to transport and en route photochemistry. Considering the aforementioned special characterisitics of the BoB, as well as the considerable heterogeneity of trace gas and aerosol distribution, in situ measurements covering large areas are essential for investigating the distribution of pollutants and the controlling processes. Extensive in situ measurements of various trace gases over the BoB have been conducted in the following field campaigns: INDOEX during the winter months of 1998 and 1999 (Lelieveld et al., 2001; Muhle et al., 2002); the Integrated Campaign for Aerosols, gases, and Radiation Budget (ICARB) during the March–May (pre-monsoon season) of 2006 (Nair et al., 2011; Srivastava et al., 2011; Srivastava et al., 2012); the winter-ICARB (W-ICARB) during December–January 2009 (Girach and Nair, 2010; 2014; David et al., 2011); the Bay of Bengal Experiment

(BOBEX)-I during February–March 2001 (Lal et al., 2006); the Bay of Bengal Process Studies (BOBPS) during September–October 2002 (Sahu et al., 2006); BOBEX–II during February 2003 (Lal et al., 2007); and ~~an unnamed~~ the other campaign conducted during October–November 2010 (Mallik et al., 2013).

Although earlier studies have covered the spatio-temporal distribution of trace gases during most seasons over the BoB, there is still a lack of observations over the BoB during the summer monsoon season (June–August). The Asian summer monsoon circulation provides a pathway for pollution transport into the stratosphere (Randel et al., 2010), and observations taken during monsoon season capture a time of high water-vapour loading over the BoB~~, which influences the oxidation capacity of the atmosphere~~. Deep convection during the summer monsoon can uplift boundary layer pollution to higher altitudes, which is then distributed over a larger region, thereby influencing air quality and climate over much larger regions (Lawrence and Lelieveld, 2010), extending as far as, for example, over the Mediterranean (e.g. Lelieveld et al., 2002; Scheeren et al., 2003). Such in situ measurements are also essential given the fact that satellite remote sensing of boundary layer O_3~~ozone using, for example, the Tropospheric Emission Spectrometer (www.tes.jpl.nasa.gov) on board the Aura satellite,~~ has relatively higher uncertainty ~~(Verstraeten et al., 2013)~~. The uncertainties in satellite retrievals of trace species are particularly high during the summer monsoon season, as the view of satellite instruments is frequently obscured by thick clouds.

In the present paper, ~~we present~~the ship-based measurements of surface O_3, CO, and CH_4 over the BoB are presented for the summer monsoon season of~~year~~ 2009~~, the first time such measurements have been taken during the summer monsoon season in this region~~. These observations were carried out as a part of the Continental Tropical Convergence Zone (CTCZ) experiment (http://odis.incois.gov.in/index.php/project-datasets/ctcz-programme) under the Indian Climate Research Programme (ICRP) of the Government of India. In this study, ~~we analyse~~ the spatial and temporal variations of O_3~~in ozone~~ over the BoB and the effects of transport are analysed. These observations are compared with simulations from a regional model, Weather Research and Forecasting coupled with Chemistry (WRF-Chem). ~~We investigate~~The sharp reductions ~~that we have~~ observed in O_3 during rainfall events are investigated in greater detail.

~~The manuscript begins with a description of the ship cruise in Section 2, followed by experimental setup and observations in Section 3, description of model simulations in Section 4, and results of the study in Section 5. A summary and main conclusions are presented in Section 6.~~

2. The cruise track and background conditions

Figure 1 shows the cruise track of the Oceanic Research Vessel (ORV) *Sagar Kanya* during the CTCZ campaign ~~experiment~~ (cruise number SK 261). The arrows marked on the track show the direction of the ship, which sailed from Chennai (80.3° E, 13.1° N, marked by a circle) on July 16, 2009. The cruise offered greater coverage in the northern BoB than the southern or central BoB areas. To take time series measurements, the ship was kept stationary ~~at 89° E, 19° N~~ for fifteen days (July 22 to August 06, 2009) at 89° E, 19° N as marked by a triangle in the figure. After several tracks, covering latitude sector 11.0 to 21.1° N and longitude sector 80.3 to 90.1° E, the cruise ended on ~~-~~August 17, 2009 at Chennai, for a total of 32 days of voyage. ~~The average prevailing wind patterns at 925 hPa~~

~~during the cruise period are obtained from NCEP/NCAR reanalysis (http://www.esrl.noaa.gov/psd; Fig. 1). The prevailing westerly or south-westerly winds are conducive for the transport of ozone and its precursors from the Indian landmass to the BoB during the summer monsoon season.~~ The average wind pattern at 925 hPa (NCEP/NCAR reanalysis; http://www.esrl.noaa.gov/psd) during the cruise period is shown in Figure 1. The prevailing westerly and southwesterly winds transport O_3 and its precursors from the Indian landmass to the BoB during study period. The spatial distribution of emissions of NO_x, an ~~ozone~~O_3 precursor gas, is also shown as colour map in Fig. 1. NO_x emissions are obtained from the Intercontinental Chemical Transport Experiment – Phase B (INTEX-B) inventory (Zhang et al., 2009), which is representative of the year 2006. ~~Relatively high~~ NO_x emissions are relatively higher~~located~~ over parts of eastern and southern India as compared to central India. The square tagged with Thiruvananthapuram shows the location corresponding to the measurements shown in Fig. 9.

3. Experimental details and data

Surface O_3 measurements were carried out using an online ultraviolet (UV) photometric ozone analyzer (Model O3 42), manufactured by Environnement S.A, France. The analyser utilises the absorption of UV radiation by ~~ozone~~O_3 molecules at 253.7 nm and derives ~~ozone~~O_3 mixing ratios using the Beer–Lambert law. This UV absorption-based analyser has an uncertainty of about 5% (Tanimoto, 2007), corresponding to ~1.5 nmol mol^{-1} for the observed range of ~~ozone~~O_3. Zero noise of the instrument is 0.5 nmol mol^{-1}. The instrument has a lower detection limit of 1 nmol mol^{-1} and a linearity of ±1%. An individual measurement is performed at a minimum response time of 10 seconds. The analyzer was operated on auto-response mode, whereby responses could~~–~~ 10–90 seconds depending upon changes in ~~ozone~~O_3 mixing ratios. However, data were recorded continuously at 5-minute intervals.

CO measurements were made using an online CO analyzer (Model CO12 Module) manufactured by Environnement S.A, France. This instrument work~~as based~~ on the principle of Non-Dispersive Infrared (NDIR) absorption by CO molecules at the wavelength of 4.67 µm. The instrument has a lower detection limit of 50 nmol mol^{-1}, a linearity of 1%, and a response time of 40 seconds. The overall uncertainty in hourly CO measurements is estimated to be ~ 10% at a CO value of 150 nmol mol^{-1} (Sawa et al., 2007; Tanimoto et al., 2007).

Air was drawn from a height of approximately 15 meter above the sea surface through a Teflon tube. Before and after the cruise, both analyzers were calibrated, with calibration factors not found to be significantly changed. The calibrations of both analysers were carried out using appropriate calibration standards traceable to NIST and a multi-channel calibrator, following the procedure mentioned in the manuals of analysers. While O_3 analyser was calibrated for mixing ratios of 30 nmol mol^{-1}, the CO analyser was calibrated for mixing ratios of 1.1 µmol mol^{-1}. Meteorological parameters such as pressure, temperature, and relative humidity were measured continuously onboard the ship. Trace gas measurements affected by the ship exhaust were identified and discarded using onboard wind direction and NO_x measurements.

In addition, a total of 29 air samples were collected in 1-liter glass flasks during the cruise and were analyzed for ~~methane~~CH_4 using a Gas Chromatograph (GC) coupled with a Flame Ionization Detector (FID), as described in Tiwari and Ravi Kumar (2011). These CH_4~~methane~~ measurements are traceable to the WMO standard scale. CH_4~~Methane~~ standards were obtained from the WMO Central Calibration Laboratory (CCL) at the National Oceanic

and Atmospheric Administration (NOAA)/Earth System Research Laboratory (ESRL)/Global Monitoring Division (GMD), located in Boulder, Colorado, USA. The precision for CH_4 ~~methane~~ measurements was approximately ±0.1 µmol mol^{-1}. A detailed description of the analytical procedure for CH_4~~methane~~ measurement and calibration of GC is given in Ravikumar et al. (2014).

To further study the observed low-O_3 events over BoB, measurements made at Thumba, Thiruvananthapuram are used as a case study. Using the same ~~ozone~~O_3 analyzer as the one used for surface O_3 measurements over BoB, continuous measurements of surface O_3 were taken at Thumba, Thiruvananthapuram (David and Nair, 2011; Girach et al., 2012) in July 2011. Along with various meteorological parameters, rainfall measurements were also taken at Thumba at 5-minute ~~accumulation intervals~~ of integration time using an automatic weather station manufactured by Dynalab Weathertech Pvt. Ltd, India. The site, Thumba, is situated just ~500 m away from the west coast, with sandy terrain, and is a less populated area in the city of Thiruvananthapuram (8.5° N, 76.9° E) at southern tip of India. For more details about the Thumba site and measurements please see, for example, Nair et al. (2002) and David and Nair (2011).

A vertical profile of O_3 was measured on July 28, 2011 at Thumba using an electrochemical concentration cell ozonesonde (EN-SCI 2ZV7 ECC; Komhyr, 1969, 1995). The accuracy of such ozonesondes is reported to be about ±5-10 % up to ~30 km (Smit et al., 2007). More details of this measurement technique can be found in Ojha et al. (2014).

The accumulated rainfall for every 3-hour interval from the Tropical Rainfall Measuring Mission (TRMM; with a horizontal grid size of 0.25° × 0.25°) is also utilized in this study to complement the onboard rainfall measurements. The 3B42 algorithm -is used to calculate precipitation and root-mean-square precipitation-error estimates; these two estimates were then used to compute hourly and daily rainfall estimates (Huffman et al., 1995).

The gridded (2° × 2°) monthly column averaged CH_4 (level-3, version 6) retrievals from SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY) instrument on board Envisat spacecraft were used to infer concentrations over Indian land regions. IMAP-DOAS (Iterative maximum a posteriori - differential optical absorption spectroscopy) algorithm retrievs CH_4 utilising the spectra (i.e., 1000–1750 nm) at near infrared channel#6 (Frankenberg et al., 2005).

4. Model Simulations

The Weather Research and Forecasting model with Chemistry (WRF-Chem; Grell et al., 2005) version-3.5.1 was used to simulate meteorological and chemical fields during the campaign period. The model domain (Fig. 2d-e) is defined on the Mercator projection, centred at 80~~6~~° E, 15.5~~6~~° N, at a spatial resolution of 15 km x 15 km. The model has 51 vertical levels from surface to 10 hPa. The simulations were conducted for the period of June 29 to August 31, 2009, covering the complete measurement period. The meteorological inputs have been adopted from ERA-interim reanalyses by the ECMWF. Horizontal winds, temperature, and water vapour are nudged above the planetary boundary layer using a nudging coefficient of 0.0003 s^{-1} (Kumar et al., 2015), employing the Four Dimensional Data Assimilation (FDDA) technique. Anthropogenic emissions of CO, NO_x, SO_2, and NMVOCs are provided by a regional emission inventory that was developed to support the Intercontinental Chemical Transport

Experiment – Phase B (INTEX-B; Zhang et al., 2009; Kumar et al., 2012b; Ojha et al., 2016). This inventory is representative of the year 2006. Aerosol emissions are provided by the Hemispheric Transport of Air Pollution (HTAP v2) inventory (Janssens-Maenhout et al., 2015). Biomass burning emissions from NCAR Fire Inventory (FINN; Wiedinmyer et al., 2011), and biogenic emissions calculated online using MEGAN (Guenther et al., 2006) were used in the simulations.

Gas-phase chemistry in the model is represented by the second-generation Regional Acid Deposition Model (RADM2; Stockwell et al., 1990), and the aerosol module is based on MADE SORGAM (Binkowski and Shankar, 1995; Ackermann et al., 1998; Schell et al., 2001). Initial and boundary conditions for chemical fields are provided by the MOZART-4/GEOS5 data. The options used to parameterize different atmospheric processes are given in Table 1. For more information about meteorological nudging, chemical mechanisms, emissions, boundary conditions, and evaluation of WRF-Chem against in situ measurements and satellite data over the Indian region, please see, for example, Kumar et al. (2012a; 2012b; 2015), Ansari et al. (2016), and Ojha et al. (2016). Model-simulated mean spatial distributions of O_3 and CO over the model domain during the study period are shown in Figure 2d-e.

5. Results and Discussion

5.1 Variations in O_3, CO, and CH_4 over the BoB

Figure 2a-c~~3~~ shows the observed variations in O_3, CO, and CH_4 along the ship track during the July 16 to August 17, 2009~~. period of the summer monsoon season. In Figure 3, the solid black lines define two regions, central BoB (80-91° E, 11-16° N) and northern BoB (81-91° E, 16-21.5° N). All of the measured~~ The mixing ratios of trace gases show large~~significant~~ spatio-temporal variations~~heterogeneity~~ over the BoB~~ region during the summer monsoon season. O_3 levels are found to vary from as low as 8 nmol mol^{-1} to as high as 54 nmol mol^{-1}, with average mixing ratio derived from the complete data of 29.7±6.8 nmol mol^{-1}. CO mixing ratios are observed to be in the range of 50 nmol mol^{-1} falling below the detection limit of the instrument to 198 nmol mol^{-1}, with an average value of 96±25 nmol mol^{-1} from all observations.~~ Levels of O_3 and CO varied in the ranges of 8–54 nmol mol^{-1} (with average of 29.7±6.8 nmol mol^{-1}) and 50–198 nmolmol^{-1} (average of 96±25 nmol mol^{-1}), respectively. As CO mixing ratios below the detection limit of the instrument are discarded from the analysis, the reported minimum and average values of CO mixing ratios are therefore slightly higher than their actual values. CH_4 mixing ratios are observed to range from 1.57–2.15 µmol mol^{-1}, with average of 1.83±0.14 nmol mol^{-1}. ~~We further separated the observations into two defined geographical regions: northern BoB and central BoB (Figure 3). The average mixing ratios for O_3, CO, and CH_4 are observed to be ~ 30±7 nmol mol^{-1}, 95±25, nmol mol^{-1}, and 1.86±0.12 µmol mol^{-1}, respectively, over northern BoB. These ratios are comparable or only slightly higher than those over central BoB: O_3: 27±5 nmol mol^{-1}, CO: 101±27nmol mol^{-1}, and CH_4: 1.72±0.14µmol mol^{-1}.~~ Average CH_4 mixing ratios, ~~however,~~ showed a significant difference of ~0.14 µmol mol^{-1} between northern (81-91° E, 16-21.5° N) and central (80-91° E, 11-16° N) BoB during the ~~summer monsoon season~~study period. ~~The average mixing ratios of O_3 and CO are comparable or only slightly higher than those over central BoB.~~

In addition to sailing across the BoB, the ship was also kept stationary for fifteen days, from July 22 to August 06, 2009 at 89° E, 19° N. During this time period, surface O_3, CO, and CH_4 mixing ratios are observed to fall into the range of 9–46 nmol mol^{-1}, 58–144 nmol mol^{-1}, and 1.71–1.89 µmol mol^{-1}, respectively, with temporally averaged mixing ratios of 28±7 nmol mol^{-1}, 91±19 nmol mol^{-1}, and 1.81±0.06 µmol mol^{-1}, respectively.

The HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model was used to simulate five-day backward airmass trajectories arriving at 500 m (a height that falls within the marine atmospheric boundary layer) above the measurement locations (Draxler and Rolph, 2003; Rolph, 2003; http://www.arl.noaa.gov/ready.html), as shown in the Fig. 34. Trajectories are colour-coded to show the altitude variations of the air -parcels along ~~its~~their path. The influences of two different airmasses are observed over the BoB during the CTCZ experiment. Over the central BoB, the backward air trajectories ~~are seen to~~ cross southern India (i.e. <13° N), where a belt of elevated anthropogenic emissions (5–20 mol km^{-2} hr^{-1} of NO_x; see Fig. 1) is located. In contrast, most of the air trajectories over northern BoB come ~~through~~across the central Indian region, where anthropogenic emissions are ~~significantly~~ relatively lower. For example, with the exception of a few hotspots, NO_x emissions ~~above~~north of 13° N are in the range of 1–10 mol km^{-2} hr^{-1} (Fig. 1). The O_3 and CO mixing ratios over BoB in airmasses from central/northern India (Fig. 3a) are slightly higher or comparable (O_3: 30±7 nmol mol^{-1}, CO: 95±25 nmol mol^{-1}) to those (O_3: 27±5 nmol mol^{-1}, CO: 101±27 nmol mol^{-1}) in airmasses from southern India (Fig. 3b).

The observed spatio-temporal variations ~~in the~~of trace gases are investigated by calculating the ~~percentage~~fractional residence time of airmasses over land, using HYSPLIT simulated 5-day backward air trajectories. Figure 45a–c shows the temporal variations ~~in~~ of O_3, CO, and CH_4 during the CTCZ experiment along the cruise track. The percentage ~~residence~~ of time of airmasses over continental India is also shown (Fig. 4d~~blue line~~), as estimated by the ratio of residence time over land to the total trajectory time of 5 days. The hours of residence have only been included in the analysis if the altitude along trajectory is less than 1.5 km, as the surface emissions might not been directly influence the airmasses aloft. Red vertical bars depict the sharp reductions in O_3 ~~as well as CO~~ mixing ratios associated with rainfall events (see Section 5.34). O_3, CO and CH_4 show correlated variability with the estimated residence times over the Indian subcontinent with slightly higher correlation in the case of primary species (R = 0.4 in case of CO and CH_4), as compared to O_3 (R = 0.3). Similar variations in ~~O_3~~ mixing ratios of these trace gases and residence time over continental India indicate the influences of transport from the Indian subcontinent on the observed spatio-temporal variations over the BoB during the summer monsoon season. The occasions on which such a one-to-one correspondence are not observed can be attributed to varying source strengths, vertical mixing or dilution, and en route photochemistry. ~~As seen in Fig. 5b, CO is also associated with residence time, although not as strongly as in the case of O_3. CH_4 does not show a considerable correlation with residence time over the Indian subcontinent (Fig. 5a).~~

Generally, during the summer monsoon season, relatively cleaner marine airmasses from the Arabian Sea are transported to the Indian region. These airmasses are then exposed to regional emissions and subjected to photochemistry depending upon the availability of solar insolation under the cloudy conditions of monsoon. The airmasses in which precursors have accumulated, and to some extent photo-chemically processed, outflows into the BoB. As a result, the airmasses out-flowing at the eastern coast of India could have higher ~~ozone~~O_3 mixing ratios

than the background air coming from the Arabian Sea into the western coast of India. The difference between these two values is a representative of the ~~ozone~~O_3 build-up that can be attributed to regional pollution; this difference would also reflect the extent of photochemical processing that would have taken place.

As the observational site Thumba, Thiruvananthapuram, is situated just at the Arabian Sea coast, the monsoon-time observations here could be approximated to represent the background ~~ozone~~O_3 mixing ratios entering from the Arabian Sea. In August 2009, using only daytime monthly average O_3, the ~~ozone~~O_3 at Thumba during the monsoon season was observed to be 23±7 nmol mol^{-1}. Since the objective of investigation is the additional O_3 over the BoB produced by en route photochemistry, daytime O_3 values at Thiruvananthapuram are therefore compared with all the observations over the BoB. The average mixing ratio observed over the BoB during monsoon season for July 16–August 17, 2009 was 30±7 nmol mol^{-1}, which was ~7 nmol mol^{-1} higher than the Arabian Sea airmass. This additional amount of ~7 nmol mol^{-1} could be attributed to the effects of regional and en route photochemical ~~ozone~~O_3 production. Net ~~ozone~~O_3 production rate in the outflow is estimated to be in the range of 1.5–4 nmol mol^{-1} day^{-1} ~~(Fig. 6)~~. Note that the ~~ozone~~O_3 mixing ratio is reported to be ~30±2 nmol mol^{-1} during July 2009 over Ananthapur, a rural site in central India, indicating the enhancement due to regional ~~ozone~~O_3 production ~~(Fig. 6)~~. As shown in Table 2, while average O_3 mixing ratios over the west coast of India and the Arabian Sea are in the range of 9–25 nmol mol^{-1} during the monsoon season, the average O_3 mixing ratio is ~ 30 nmol mol^{-1} over the central Indian station and the BoB.

~~As shown in Fig. 6, during the cruise observations,~~ O_3 mixing ratios were 27±3 and 28±5 nmol mol^{-1} for July 21, 2009 and August 15, 2009, for which back-trajectories (not shown here) crossed Thiruvananthapuram on July 20, 2009 and August 13, 2009, with daytime O_3 values of 23±6 and 25±6 nmol mol^{-1}, respectively. The difference of 3–4 nmol mol^{-1} between ~~ozone~~O_3 mixing ratios over the BoB and Thurivannthapuram represents the en route photochemical production of ~~ozone~~O_3 in the airmasses toward the observation points over the BoB. It is further found that the airmasses were typically below 700 meters, and generally within the marine boundary layer (e.g. mean boundary layer height ~897 m during winter over the BoB; Subrahamanyam et al., 2012). The enhancements in O_3 are attributed here to in situ photochemical build-up while moving towards the BoB, which has been noted in previous experiments and model simulations (e.g. Lal and Lawrence, 2001; Ojha et al., 2012).

CO showed a sharp enhancement (denoted with red arrows in Fig. ~~5~~4b) on August 7 and 11, 2009, coinciding with a longer residence time over the Indian region. Figure ~~5~~7 shows backward airmass trajectories above the measurement locations, along with the distribution of anthropogenic CO emissions from the INTEX-B inventory, representative of the year 2006. The airmasses over the BoB are found to be influenced by emission hotspots (corresponding emission of 250–350 mol km^{-2} hr^{-1}). The airmasses took about half a day to be transported from the emission hotspot to the observation location over the BoB. CO mixing ratios measured at Bhubaneswar (20.30° N; 85.83° E), a station within the hotspot region, is ~251±58 nmol mol^{-1} during the monsoon season (June–August 2011–2012; Mahapatra et al., 2014), with the elevated CO emissions in the Bhubaneswar region being attributed to industrial activities. The higher CO mixing ratios ~200 nmol mol^{-1} is inline with the monsoonal values observaed at Bhubaneswar. The CO mixing ratios around 150 nmol mol^{-1} were sampled on August 11, 2009 near the coastal source regions. Additionally, CO mixing ratios over central BoB (101 nmol mol^{-1}) were only slightly higher than those over

northern BoB (95 nmol mol^{-1}). ~~We~~It is suggest that this is partially due to higher emissions over southern India, against the shorter residence of airmasses over land and the relatively longer lifetime of CO.

The mixing ratios of surface CH_4 were higher in the airmasses from central/northen Indian over northern BoB (1.86±0.12 µmol mol^{-1}) as compared to those in the airmasses from southern India (1.72±0.14 µmol mol^{-1}). As CH_4 is a relatively well mixed trace gas, the average values over column approximates uniform mixing ratio within troposphere (Seinfeld and Pandis, 2006). The monthly column averaged tropospheric CH_4, retrievals from SCIAMACHY, for August 2009 shows (Fig. 6) higher values around 1.85 µmol mol^{-1} over central/northern India as compared to that of southern India (~1.80 µmol mol^{-1}). The higher tropospheric CH_4 over the central/northern Indian landmass during the summer monsoon season has been also reported by Kavitha and Nair, 2016. The observed higher CH_4 over the northern BoB are attributed to the influences of emissions from central/northern Indian regions as also suggested by backward trajectories. Owing to the longer lifetime of CH_4, diffusion of CH_4 from a hotspot region over the eastern IGP to northern BoB might be the other source of higher CH_4 levels over northern BoB during summer monsoon season. An emission inventory analysis by sector over the hotspot region (i.e. eastern IGP) indicates that these higher CH_4 emissions are associated with rice cultivation, waste treatment and livestock. The correlation between presented in situ CH_4 measurements with retrievals from satellite instrument (AIRS–Atmospheric Infrared Sounder) was found to be statistically insignificant (not shown) which highlights a need of more such in situ measurements in this region to validate the satellite products especially during the summer monsoon.

5.2 WRF-Chem simulations

WRF-Chem simulations, as described in Section 4, are used to evaluate the performance of the model in reproducing the~~our~~ measurements, and to investigate the underlying processes that caused the observed variabilities in O_3 and CO. A comparison between model simulated and measured meteorological parameters shows only small mean biases, such as -1.9 hPa in pressure, -0.6° C in temperature, and -1.1% in relative humidity (Table 3). ~~Before evaluating the specific chemical species, variations in the meteorological parameters simulated by the model are briefly evaluated. Figure 8 compares WRF-Chem simulations with the in situ measurements of meteorological parameters along the cruise track. Overall, WRF-Chem reproduces these meteorological parameters with only small mean biases, such as -2.3 hPa in pressure, -0.5° C in temperature, and -1.4% in relative humidity (Table 3). However, model shows limitations in capturing some of the sharp reductions in air temperature and associated enhancements in relative humidity.~~

Figure 4b-c~~9~~ compares WRF-Chem-simulated O_3 and CO with in situ measurements taken along the cruise track. WRF-Chem is found to reproduce the observed variations ~~in~~ of O_3 and CO over the BoB during the summer monsoon season with an overestimation of absolute O_3 levels by ~~6.2~~1.9 nmol mol^{-1} (i.e. ~~21~~6% of averaged O_3 value, 29.7 nmol mol^{-1}) and absolute CO levels by ~~22~~18 nmol mol^{-1} (i.e. ~~23~~16% of averaged CO value, 96 nmol mol^{-1}). It should be noted ~~here~~ that the average CO mixing ratio of 96 nmol mol^{-1} is slightly higher than its actual value, as data points below the detection limit of the instrument are discarded. Biases in the model simulations can

be attributed to the uncertainties in the simulated meteorology and in the ~~input~~ emission~~s~~ datasets; however, in the present study, ~~we use~~ the model fields are used mainly to investigate temporal variations rather absolute mixing ratios. The squared correlation coefficients between daily averaged in situ measured and simulated O_3 and CO are 0.~~67~~58 and 0.19, respectively. The higher value of the squared correlation coefficient for O_3 demonstrates WRF-Chem's ability to reproduce the observed broad features in surface O_3 over the BoB. Note that the sharp reductions that caused very low ~~ozone~~O_3 during rainfall episodes are not captured by WRF-Chem, except during the event of August 10–11, 2009. This will be discussed in detail in Section 5.3~~4~~. WRF-Chem simulated O_3 has also been evaluated against several surface observations in India previously (Kumar et al., 2012b). Modeled O_3 was found to be within 1-σ standard deviation of mean from observations at Gadanki in the southern India during summer monsoon (Ojha et al., 2016). At Thumba also, model simulated O_3 variations correlated reasonably with measurements (R^2=0.6). More information on evaluation of WRF-Chem simulations of O_3 and CO over India can be found elsewhere (e. g. Kumar et al., 2012b; Ojha et al., 2016).

An additional simulation was conducted by switching off the anthropogenic emissions over the model domain ~~(~~Fig. 4b-c; dotted blue curves). Mean O_3 (17.7 nmol mol^{-1}) is found to be lower by 14 nmol mol^{-1} with smaller variability of 2.4 nmol mol^{-1} compared to standard WRF-Chem simulation. Similarly, also the mean CO level is lower by 36 nmol mol^{-1} with smaller variability of 4.9 nmol mol^{-1}. This shows that enhanced levels and observed variability ~~of~~in O_3 and CO mixing ratios over BoB are attributable to the regional anthropogenic emissions.

The limited collocated measurements showed en route O_3 production rate of 1.5–4 nmol mol^{-1} day^{-1}, as discussed in section 5.2. Here, the O_3 production rate is estimated by analyzing the model simulated O_3 mixing ratios along the airmass trajectories ending at few representative locations over the BoB (Fig. 7a). Model simulated chemical evolution of air parcels clearly shows an increase in the O_3 mixing ratios towards the marine region of BoB. The temporal variations of O_3 mixing ratio averaged corresponding all the trajectories shown in Fig. 7a is shown as a boxplot in Fig. 7b. A linear regression analysis (blue curve in Fig. 7b), is used to estimate the mean en route O_3 production rate of ~4.6 nmol mol^{-1} day^{-1} in the outflow. The enhancement in average O_3 over the BoB as compared to the continental Indian region is also shown in the Fig. 2d, averaged for the study period.

~~**5.3 Diurnal variation**~~

~~Figure 10 shows the mean delta diurnal variation, that is the mean value subtracted from the mean diurnal pattern, in surface O_3 from observations and model simulations for the location where the ship was kept stationary (89° E, 19° N) for a period of about 15 days. The delta diurnal variation is calculated by subtracting daily mean value from values at individual hours. Ozone at each hour shown here is an average of 10 to 15 observations. Ship exhaust contaminated the observations for a period of time between 5 to 14 hours long; data corresponding to this period is therefore discarded from analysis, leading to a gap. Both the WRF-Chem model and observations showed only small variability from mean values (delta ozone = -2 to +2 nmol mol^{-1}) during the summer monsoon season. Neither our limited measurements nor the simulations exhibited any tendency towards net photochemical build up in ozone after sunrise during the monsoon. The observations available during 5 to 14 hours on July 23 and 24, 2009 also do not show any daytime build up. A net daytime photochemical build up has been reported over the BoB during both pre-~~

~~monsoon (Nair et al., 2011) and post-monsoon season (Mallik et al., 2013), as shown for comparison in Figure 10. The absence of net day-time photochemical build-up and the highly correlated variability of ozone with residence time over the Indian region (Section 5.1) suggest that spatio-temporal variations in surface ozone over the BoB during monsoon season are associated with the direct transport, supplemented with en route photochemistry. Note that, due to the insufficient number of observations, diurnal variations in CO and CH_4 are not discussed.~~

~~5.4~~5.3 Ozone variations during rainfall events

An interesting phenomenon observed during the CTCZ experiment is the abrupt reduction in ~~ozone~~O_3 mixing ratios that accompanied the onset of heavy rainfall, despite the low solubility of ~~ozone~~O_3 in water. In this section ~~we investigate~~ the possible causes of these low-~~ozone~~O_3 events during rainfall ~~over the BoB~~are investigated.

Figure ~~8~~11 shows variations in O_3 (black circles) mixing ratios, surface temperature (orange curve), TRMM-retrieved rainfall (thick grey vertical bars), and WRF-Chem-simulated vertical winds at pressure levels ranging from 950–750 hPa (coloured bars) during four such events on July 21, 26, and 28–29 and on August 10–11, 2009. As high time-resolution in situ measurements of rainfall were not available aboard ship, Figure ~~8~~11 therefore uses 3-hourly rainfall retrievals from the TRMM, co-located with ~~ozone~~O_3 measurements. During these events, CO mixing ratios also show a reduction of about ~56 nmol mol^{-1}, with observed values falling below the detection limit of the instrument during the first event of July 21, 2009 (not shown). Although CO measurements are not available for the second and third event, during the fourth event (August 10–11, 2009) CO mixing ratios showed an enhancement due to transport from strong source regions (see Section 5.1). While the first three low-O_3 events are not captured by WRF-Chem (Fig. ~~9a~~4c), the fourth event is reproduced.

Wet scavenging does not directly reduce ~~ozone~~O_3, as its water solubility is low; as a result, some dynamic process could be responsible for the observed reductions in ~~ozone~~O_3 during rainfall. Airmasses could undergo downdrafts during heavy rainfall (Kumar et al., 2005) through air drag by the falling rain drops and in mesoscale subsidence that compensates convective updrafts. We suggest that, in the presence of ~~ozone~~O_3-poor airmass aloft, a downdraft would result in reductions in surface ~~ozone~~O_3 mixing ratios. An opposite scenario leading to O_3 enhancement could take place if downdrafts bring mid-troposheric air, where typically O_3 is higher than at surface. The model-simulated meteorology shows occurrences of downdrafts at different pressure levels during the first three events on July 21, 26, and 28–29 (Fig. ~~8~~11a–c), which ~~we~~ is further corroborated with measurements of air temperature aboard. Downdrafts of free tropospheric air could lead to a reduction in near-surface temperature by as much as 10 °C within a few minutes (Ahrens, 2009). Air temperature measured aboard ship showed a sharp decrease of 2–4 °C, coinciding with the first three low-~~ozone~~O_3 events (Fig. 8a-c~~12~~). The reductions in temperature caused by downdrafts are generally short-lived (Ahrens, 2009), it is confirmed in the case of these events (Fig. 8a-c~~12~~).

Model-simulated vertical winds and variations in air temperature suggest that downdrafts did occur during the first three rainfall events. As in situ measurements of ~~ozone~~O_3 vertical profiles are not available over the BoB during the summer monsoon season, ~~we instead used~~ the observations taken at Thumba, Thiruvananthapuram, in the southern Indian region are used as a case study to investigate this hypothesis. For general details of the typical diurnal and seasonal variations ~~in~~of O_3 at Thumba, please see Nair et al. (2002), David and Nair (2011), and Girach et al.

(2012). Figure ~~13~~9a shows the temporal variation in surface O_3 on July 15, 2011 at Thumba, along with 5-minute accumulated rainfall. Here, surface ~~ozone~~O_3 is observed to decline from 25 to 13 nmol mol^{-1} within 15–20 minutes, coinciding with the occurrence of intense rainfall (3.5–0.5 mm rain over a period of 5 minutes). Measurements of the O_3 vertical profile are not available for this day due to the rainy conditions; a profile measured on July 28, 2011 is therefore shown in Fig. ~~13~~9b. This profile has ~~significantly~~ lower ~~ozone~~O_3 mixing ratios aloft (~22 nmol mol^{-1} at ~1 km) than near surface (~42 nmol mol^{-1}). ~~A typical~~The observed mixed layer height is about 0.15–0.6~~±0.2~~ km over Thumba, Thiruvananthapuram (Anurose~~Nair~~ et al., 2016~~1~~) during ~~the summer monsoon season~~July 2011; above this height, O_3 mixing ratios sharply decrease with altitude. The present case study suggests the presence of ~~ozone~~O_3-poor airmasses aloft than those near the surface over the south Indian region during summer monsoon. With an ~~ozone~~O_3 distribution as observed in the present case study at Thumba, the downdraft during intense rainfall could lead to the mixing of free-tropospheric air with near-surface air, or to the replacement of surface air with free-tropospheric ~~ozone~~ O_3-poor air.

Although air temperature measurements could not be made during the fourth event (10–11 August 2009) due to a technical problem, model meteorology does not indicate a downdraft during this event (Fig. ~~11~~8d), indicating the dominance of a different process. As WRF-Chem-simulated ~~ozone~~O_3 variability is in good agreement with observations during this event, ~~we used~~ various tendency terms (Barth et al., 2012) from WRF-Chem are used to investigate the relative influences of different processes. The variations in instantaneous values for horizontal advection tendency, vertical advection tendency, and net tendency (i.e. the sum of chemical, vertical mixing, convective, vertical advection, and horizontal advection), along with modelled O_3 over the two locations during the event are shown in Figure 10~~4~~. The tendency values shown here are derived by subtracting the accumulated tendencies at (n-1)th hour from the accumulated tendency at nth hour. The vertical dotted lines show the time of a low-~~ozone~~O_3 event.

Both the horizontal and net tendencies (Fig. 10~~4~~b-c,e-f) show negative values, indicating that they are contributing towards a reduction in ~~ozone~~O_3 mixing ratios (Fig. 10~~4~~a,d). However, as the time of the event approaches, it is the horizontal advection tendency term that is significantly negative (Fig. 10~~4~~c, f), while other terms are small and close to zero. Horizontal advection is therefore suggested to dominate during the low-~~ozone~~O_3 event of August 10–11, 2009. The influence of horizontal advection on O_3 during this event is shown more clearly in Fig. 11~~5~~, which shows the spatial distribution of O_3 and CO from WRF-Chem before the event (16:00 and 19:00 UT) and during the event (22:00 UT) on August 10, 2009. The white triangles show the two locations where the event was observed. During 16:00 and 19:00 UT, a patch of high ~~ozone~~O_3 mixing ratios (~~4~~35 nmol mol^{-1} and higher) is seen to be distributed over a large region surrounding the measurement location. This large patch of elevated ~~ozone~~O_3 mixing ratios is horizontally advected eastward from 16:00 to 19:00 and then towards 22:00 UT (event time). As a result of this rapid advection, the high-~~ozone~~O_3 airmasses are transported from the coastal regions to deeper into the BoB; by the time they reached the location of observation, ~~ozone~~O_3 mixing ratios are observed to be lower (25–35 nmol mol^{-1}) during the event time (22:00 UT). A patch of higher levels of CO (~300 nmol mol^{-1}) was also found to be distributed across the east coast of the Indian region. Transport and dilution of this CO patch is, however, less pronounced than the high-~~ozone~~O_3 airmasses, possibly due to the relatively longer lifetime of CO. Thus, ~~we suggest that~~ in nutshell,

the horizontal advection played a key role in transporting O_3-rich airmasses deeper into the BoB region, while it diluted O_3 levels near the coastal regions in southern India during the fourth event.

~~5.5~~5.4 Seasonal variation in trace gases over the BoB

In this section, ~~we combine~~ the ~~first~~ monsoon-time measurements of ~~ozone~~O_3 taken in the present study~~,~~ are combined with data from previous campaigns (see Table 4~~3~~) to investigate the seasonal variation in ~~ozone~~O_3 over the BoB (Fig. 1~~2~~6). O_3, CO, and CH_4 mixing ratios are averaged over northern (81-91° E, 16-21.5° N) and central (80-91° E, 11-16° N) BoB regions~~, as defined in Fig. 3~~.

Overall, higher O_3 mixing ratios are present over both northern and central BoB during the winter, while ~~significantly~~ lower O_3 levels are observed during the spring–summer (with more scatter in the data over central BoB). The O_3 seasonal amplitude (i.e., the range from maxima to minima) is estimated to be ~ 39 nmol mol^{-1} over northern BoB and ~ 27 nmol mol^{-1} over central BoB. The monsoonal surface ~~ozone~~O_3 mixing ratios (~30±7 nmol mol^{-1}) are nearly half those observed during winter (63±5 nmol mol^{-1}) over northern BoB. During December 2008–January 2009, February 2003, March 2006, and November 2010, the ~~ozone~~O_3 mixing ratios were ~~significantly~~ higher (by ~3–22 nmol mol^{-1}) over northern BoB than those over central BoB. However, over the course of February 2001, ~~ozone~~O_3 mixing ratios were higher over central BoB (~38 nmol mol^{-1}) than that over northern BoB (~14 nmol mol^{-1}). In contrast, during summer monsoon season, average ~~ozone~~O_3 mixing ratios are comparable or only slightly higher over northern BoB (30±7 nmol mol^{-1}) as compared to that over central BoB (27±5 nmol mol^{-1}).

As compared with the summer monsoon season, when CO mixing ratios were lower, over northern BoB, CO mixing ratios were higher during the winter, while over central BoB, CO mixing ratios were higher during the pre-monsoon season. For O_3, spring–summer had the lower mixing ratios in both regions. The seasonal amplitude in CO mixing ratios is estimated to be ~205 nmol mol^{-1} over northern BoB and ~124 nmol mol^{-1} over central BoB. The monsoonal CO mixing ratio (~95 nmol mol^{-1}) is about one third that of the winter season (302 nmol mol^{-1}) over northern BoB. During the present study, average CO mixing ratios were comparable over northern (95±25 nmol mol^{-1}) and central BoB (101±27 nmol mol^{-1}).

A clear inference about seasonal patterns is difficult in the case of CH_4, however a tendency of lower levels towards winter can be seen. Higher mixing ratios ~1.95 (~1.91) µmol mol^{-1} were observed during November 2010 over northern BoB, and during February–March 2001 over central BoB, as compared to those from other studies. ~~In the present study, average mixing ratios of methane are significantly higher over northern BoB (1.86±0.12 µmol mol^{-1}) as compared to over central BoB (1.72±0.14 µmol mol^{-1}) during the summer monsoon season. The higher tropospheric CH_4 that has been observed over the central and northern Indian landmass during the summer monsoon season (Kavitha and Nair, 2016) could be responsible for the higher CH_4 that is observed over northern BoB in the present study. Owing to the longer lifetime of CH_4, diffusion of CH_4 from a hotspot region over the eastern IGP to northern BoB might be the other source of higher CH_4 levels over northern BoB during summer monsoon season. An analysis of an emission inventory by sector over the hotspot region (i.e. eastern IGP) indicates that these higher methane emissions are due to rice cultivation, waste treatment and livestock.~~ The surface CH_4 observations obtained during the present study show the highest variability (i.e. the difference between maxima and minima) when

compared to earlier studies: 0.53 µmol mol^{-1} over northern BoB and 0.39 µmol mol^{-1} over central BoB. ~~We attribute t~~This high variability is attributed to the relative source strengths over central and northern India as compared to southern India, highlighting the regional differences in CH_4 variability across India (Kavitha and Nair, 2016).

Seasonal variations in trace gases over the BoB are attributed to seasonal changes in the meteorological conditions, emissions, and photochemistry over the South Asian region, as well as to synoptic scale transport patterns. Wintertime stronger westerly winds transport the pollution from South Asia including that of the Indo-Gangetic basin to the BoB region. Monsoonal circulation, in contrast, carries cleaner marine airmasses to the BoB from the oceanic regions. However, as observed during the CTCZ, polluted continental or coastal airmasses can also occasionally be transported deeper over the BoB. Intense monsoonal rainfall generally leads to wet removal of ~~ozone~~O_3 precursors, while cloudy and rainy meteorological conditions suppress ~~ozone~~O_3 formation. Along with the importance of monsoonal convection in cloud formation, rainfall, and uplifting the boundary layer pollution, rapid horizontal advection is also an important process during the summer monsoon, especially affecting the near-surface variability of trace gases over the oceanic regions adjacent to India.

6. Conclusions

In this paper, we presented ~~the~~ ship-borne in situ measurements of O_3, CO, and CH_4 that were carried out as a part of the CTCZ experiment over the BoB during the summer monsoon season, July–August 2009~~, the first time that such measurements had been taken over this region during the summer monsoon season.~~ We analyzed the spatial and temporal variations in ~~our~~ the observations and compared them with results from simulations performed ~~from~~ with a regional chemistry transport model (WRF-Chem), as well as with observations from previous campaigns over the BoB. The main conclusions ~~from the study~~ are:

1. These first monsoonal observations of O_3, CO, and CH_4 show ~~significant~~ large spatio-temporal variability over the BoB, with mixing ratios varying in the range of 8–54 (mean: 29.7±6.8) nmol mol^{-1}, 50–200 (mean: 96±25) nmol mol^{-1}, and 1.57–2.15 (mean: 1.83±0.14) µmol mol^{-1}, respectively. The O_3~~,~~ and CO~~, and CH_4~~ mixing ratios in airmasses from central/northern India are slightly higher or comparable (O_3: 30±7 nmol mol^{-1}, CO: 95±25 nmol mol^{-1}~~, CH_4: 1.86±0.12 µmol mol^{-1}~~) ~~over northern BoB as compared~~ to those in airmasses from southern India ~~over central BoB~~ (O_3: 27±5 nmol mol^{-1}, CO: 101±27 nmol mol^{-1}~~, CH_4: 1.72±0.14 µmol mol^{-1}~~). The ~~difference (~0.14 µmol mol^{-1}) between~~ CH_4 mixing ratios in airmasses from central/northern (1.86±0.12 µmol mol^{-1}) are higher (~0.14 µmol mol^{-1}) compared to those in airmasses from southern India (1.72±0.14 µmol mol^{-1})~~over northern and central BoB is most significant.~~ This could be due to higher CH_4 levels over central/northern India, also found in SCIAMACHY data.
2. Back-trajectory analysis shows effects of long-range transport from northern or central India to northern BoB, and from southern India to central BoB. The correlated variations of these trace gases~~O_3 mixing ratio~~ and percentage residence time of air parcels over the Indian regions suggest that the enrichment of ~~ozone~~O_3 and precursors in air parcels over the BoB is associated with both emissions and photochemistry over the Indian region. The trajectory analysis ~~and mean diurnal variations~~ show that the observed variation in surface O_3 is primarily due to transport and en route photochemistry~~, rather than to local photochemical production~~ over the

BoB during monsoon season. An analysis of modeled O_3 along airmass trajectories show mean en route O_3 production rate of about 4.6 nmol mol^{-1} day^{-1} in the outflow towards the BoB.

3. The observed spatio-temporal variations ~~in~~of surface O_3 and CO during summer monsoon season are generally reproduced by the WRF-Chem model, although the absolute mixing ratios of O_3 and CO are typically overestimated by about 6 and 16 % respectively~~20%~~.
4. ~~We observed~~The four low-~~ozone~~O_3 events coinciding with intense rainfall were observed over the BoB. After analysing the observed variability in air temperature, model simulations of vertical winds, and an ~~ozone~~O_3-profile case study from southern India, we suggest that first three low-~~ozone~~O_3 events were due to strong downdrafts of ~~ozone~~O_3-poor airmasses. Analysis of the fourth ~~low-ozone~~ event, which is successfully reproduced by the model, shows the pivotal role of horizontal advection in transporting ~~ozone~~O_3-rich airmasses deeper over the BoB.
5. Finally, ~~we combined our monsoon time~~ the measurements during the monsoon are combined with previous campaigns over the BoB during other seasons to investigate the seasonal variability in trace gases over the BoB. O_3 and CO are shown to have pronounced seasonality, O_3 having amplitudes of about 39 and 27 nmol mol^{-1}, and CO having amplitudes of about 207 and 124 nmol mol^{-1} over northern and central BoB, respectively.

Our study data fill~~s~~ a gap of observations~~experimental data~~ during the summer monsoon over the BoB, providing information on the extent of seasonal variability. We recommend supplementing these findings with ship-borne experiments featuring collocated vertical profile observations from balloon-borne and aircraft-based platforms over the oceanic regions surrounding India to better understand the role of both large-scale dynamics (e.g. Ojha et al., 2016) and of regional influences due to South Asian outflow (see Lawrence and Lelieveld, 2010, and references therein). Such a future study would also improve our understanding of the changes that take place in the atmospheric oxidation capacity during the summer monsoon season.

Acknowledgements

We thank ~~all of~~ the CTCZ and ICRP organizers for ~~providing~~ the opportunity to participate in the 2009 CTCZ experiment. We are thankful to the Director of the National Centre for Antarctic and Ocean Research (NCAOR), Goa, for providing ship-board facilities. We gratefully acknowledge Prof. G. S. Bhatt (Indian Institute of Science, Bengaluru, India) and his team for providing the measurements of meteorological parameters ~~onboard ship~~. We also thank the chief scientist on board *SagarKanya* for providing necessary support during the cruise. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the providing the HYSPLIT transport and dispersion model and READY website (http://www.arl.noaa.gov/ready.php) used in this publication. The rainfall estimations (3B42) from the TRMM satellite were obtained from the NASA/GSFC via their website http://mirador.gsfc.nasa.gov/. The monthly CH_4 retrievals (IMAP-DOAS) of SCIAMACHY were obtained from their website, http://www.temis.nl/climate/methane.html. Use of INTEX-B and HTAP (http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123) anthropogenic emissions is gratefully acknowledged. Initial and boundary-conditions data for meteorological fields were used from the ERA interim of ECMWF. Use of MOZART-4/GEOS5 initial and boundary conditions data for chemical fields is acknowledged.

Data/processors for anthropogenic emissions, biogenic emissions, and biomass burning obtained from NCAR ACD website are gratefully acknowledged. The authors acknowledge the use of MPG supercomputer HYDRA (http://www.mpcdf.mpg.de/services/computing/hydra) for model simulations. Constructive comments and suggestions from anonymous reviewers are gratefully acknowledged.

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Tables

Table 1. The WRF-Chem options used for parameterization of atmospheric processes.

Atmospheric Process	~~**Option**~~ **Scheme used**	**Features of the scheme**
Cloud microphysics	Lin et al. scheme (Lin et al., 1983)	Sophisticated parameterization including ice, snow and graupel processes, suitable for high-resolution simulations.
Longwave radiation	Rapid Radiative Transfer Model (RRTM; Mlawer et al., 1997)	Accurate scheme utilizes look-up tables for efficiency, accounts for multiple bands and microphysical properties.
Shortwave radiation	Goddard shortwave scheme (Chou and Suarez, 1994)	Two-stream multi-band scheme using O_3 from climatology and includes cloud effects
Surface Layer	Monin–Obukhov scheme (Janjic, 1996)	Based on Monin-Obukhov with Zilitinkevich thermal roughness length and standard similarity functions from look-up tables
Land surface option	Noah Land Surface Model (Chen and Dudhia, 2001)	Unified NCEP/NCAR/AFWA scheme with soil temperature and moisture in four layers, fractional snow cover and frozen soil physics. This includes the modifications for better representation of processes over ice sheets and snow covered areas.
Urban surface physics	Urban Canopy Model	3-category urban canopy model with surface effects for roofs, walls and streets.
Planetary boundary layer	Mellor–Yamada–Janjic scheme (Janjic, 2002)	One-dimensional prognostic turbulent kinetic energy scheme, local vertical mixing is included.
Cumulus parameterization	~~New~~ Grell 3D Ensemble scheme ~~(G3)~~(Grell, 1993; Grell and Devenyi, 2002)	Improved version of the GD scheme suitable for coarse as well as high resolution simulations

Table 2. A comparison of averaged surface O_3 mixing ratios ~~over~~measured at various sites during summer monsoon period. *boundary layer ~~ozone~~O_3 over the Arabian Sea.

Observation site	Longitude (° E)	Latitude (° N)	Observation period ~~during monsoon season~~	~~Mean~~ Surface Daytime ~~Ozone~~O_3 (Mean ± Standard Deviation)	Reference
Arabian Sea					
Arabian Sea	69 –76	9 –19	July–August 2002	9	Ali et al., 2009
~~Ahmedabad~~	~~72.6~~	~~23~~	~~July–August 2003–2007~~	~~25*~~	~~Srivastava et al., 2012~~
Western coast of India					
Thiruvananthapuram	76.9	8.5	August 2009	23±7	Present Study
Thiruvananthapuram	76.9	8.5	June–August 2008	19±6	David and Nair, 2011
Kannur	75.4	11.9	July 2010–2011	11±4	Nishanth et al., 2014
MtAbu (1.6km amsl)	72.7	24.6	August 1993–2000	25±9	Naja et al., 2003
Ahmedabad	72.6	23	July 1991–1995, August 1991–1995, July–August 2003–2007	22±8, 17±4, 25*	Lal et al., 2002; Srivastava et al., 2012
~~Ahmedabad~~	~~72.6~~	~~23~~	~~August 1991–1995~~	~~17~~	~~Lal et al., 2002~~
Central India					
Anantpur	77.65	14.62	July 2009	30±2	Reddy et al., 2011

Eastern coast of India					
Bhubaneswar	86.4	20.5	June–August 2011–2012	29±6	Mahapatra et al., 2014
Bay of Bengal					
Bay of Bengal	80.3–90.1	11–21.1	July–August 2009	30±7	Present Study

Table 3. A comparison of mean values from observations with model-simulated parameters along with the mean bias. The squared correlation coefficients correspond to the linear regression analysis between daily averaged in situ and simulated parameters.

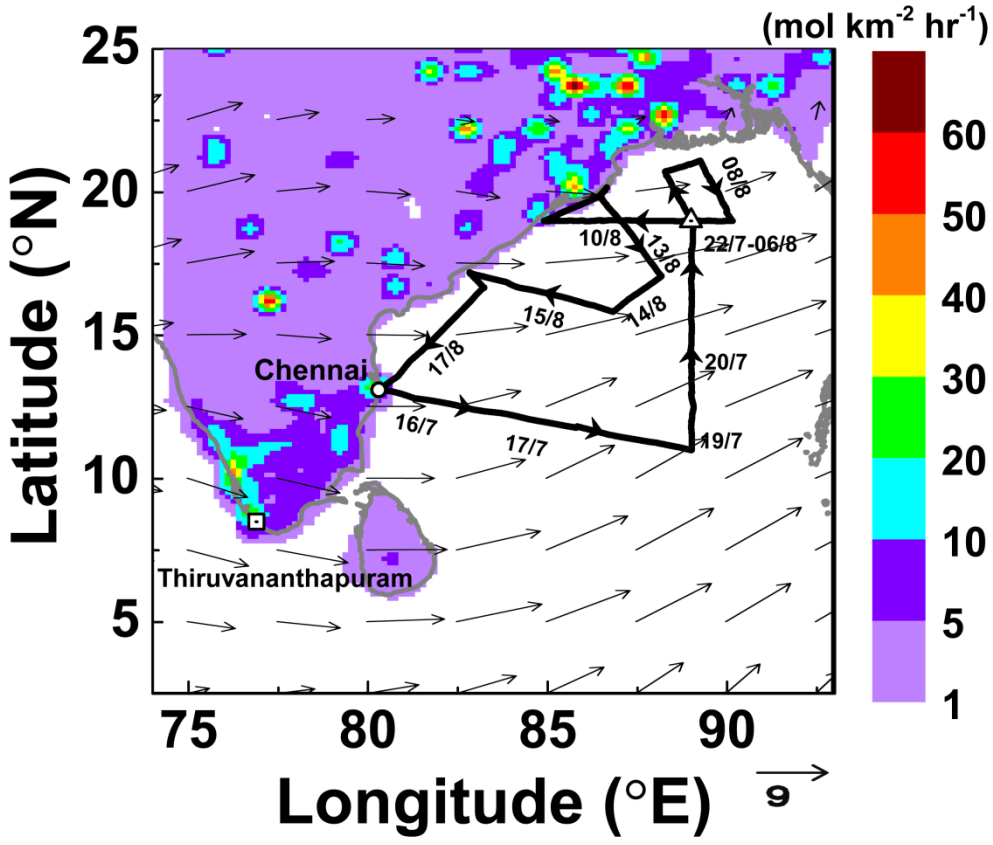
Parameter	**Observation**	**Model (WRF-Chem)**	**Mean bias**	**R^2**
Pressure (hPa)	1001.3±2.1	999.04±2.42	-2.31.9	0.93
Temperature (°C)	29.3±0.9	28.87±0.6	-0.56	0.132
Relative Humidity (%)	87.9±4.2	86.58±2.8	-1.14	0.3654
O_3 (nmol mol^{-1})	29.7±6.8	35.91.6±86.36	6.21.9	0.5867
CO (nmol mol^{-1})	96±25	1148±307	2218	0.19

Table 4. A comparison of average mixing ratios of surface trace gases measured over northern BoB (81-91° E, 16-21.5° N) and central BoB (80-91° E, 11-16° N) in different seasons as measured during different experiments. The range of mixing ratios (i.e. minima–maxima) is given in the brackets. *CO mixing ratios below the detection limit (i.e. 50 nmol mol^{-1}) are not considered in the analysis.

Study period	**Name of Experiment**	**Reference**	**O_3 (nmol mol^{-1}) over northern BoB**	**O_3 (nmol mol^{-1}) over central BoB**	**CO (nmol mol^{-1}) over northern BoB**	**CO (nmol mol^{-1}) over central BoB**	**CH_4 (µmol mol^{-1}) over northern BoB**	**CH_4 (µmol mol^{-1}) over central BoB**
December 2008–January 2009	W_ICARB	David et al., 2011	63.0±4.7 (50.8–73.8)	40.9±6.7 (27.7–63.5)	302±68 (140–450)	188±53 (50–320)	No data	No data
February 2003	BOBEX-II	Lal et al., 2007	~34.1 (15.8–50.4)	~26.8 (13.9–35.0)	~238 (187–292)	~192 (159–224)	~1.77 (1.70–1.85)	~1.73 (1.68–1.77)
February–March 2001	BOBEX-I	Lal et al., 2006	~23.8 (16.1–38.3)	~38.0 (19.4–62.9)	~194 (165–235)	~227 (97–339)	~1.94 (1.89–2.02)	~1.91 (1.74–2.06)
March–April 2006	ICARB	Nair et al., 2011; Srivastava et al., 2012	27.4±2.9 (21.4–32.6)	13.4±4.2 (3.1–24.6)	~189 (157–235)	~132 (96–167)	~1.84 (1.80–1.88)	~1.80 (1.75–1.84)
July–August 2009	CTCZ	Present Study	30.0±6.9 (8.50–54.1)	27.5±5.0 (8.8–40.5)	95±25 * (50-198) *	101±27 * (50-157) *	1.86 ±0.12 (1.62–2.15)	1.72±0.14 (1.57–1.96)
September–October 2002	BOBPS	Sahu et al., 2006	~27.3 (17.8–33.8)	~30.6 (22.5–35.2)	~152 (109–179)	~141 (108–211)	~1.79 (1.72–1.86)	~1.73 (1.68–1.80)
November 2010	~~No name~~	Mallik et al., 2013	~46.0 (26.7–59.6)	~38.7 (17.8–60.8)	~223 (131–280)	~188 (42–266)	~1.95 (1.85–2.06)	~1.79 (1.67–1.93)

835

Figures


(mol km-2 hr-1)
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Longitude (°E)
Chennai
Thiruvananthapuram
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19/7

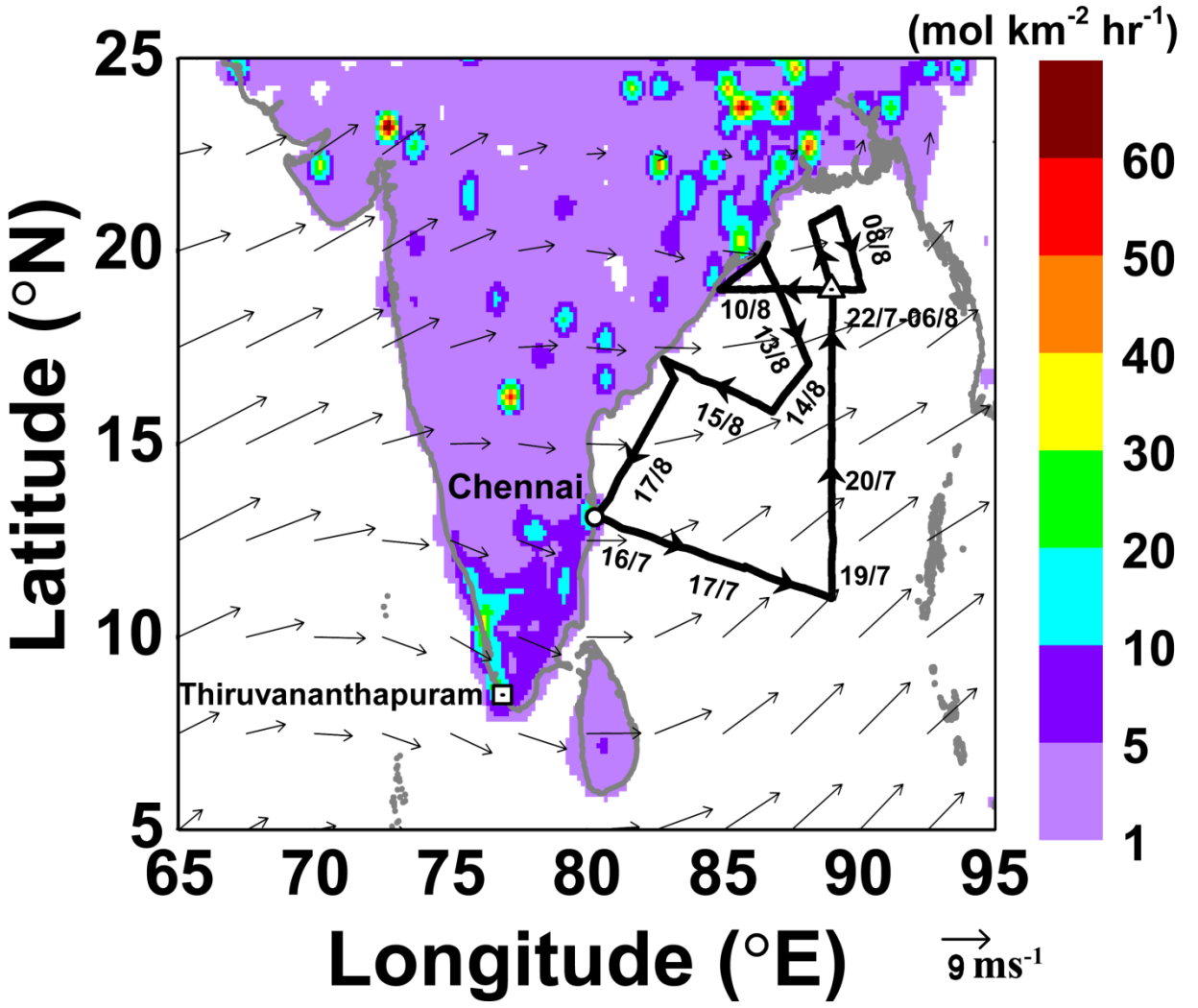
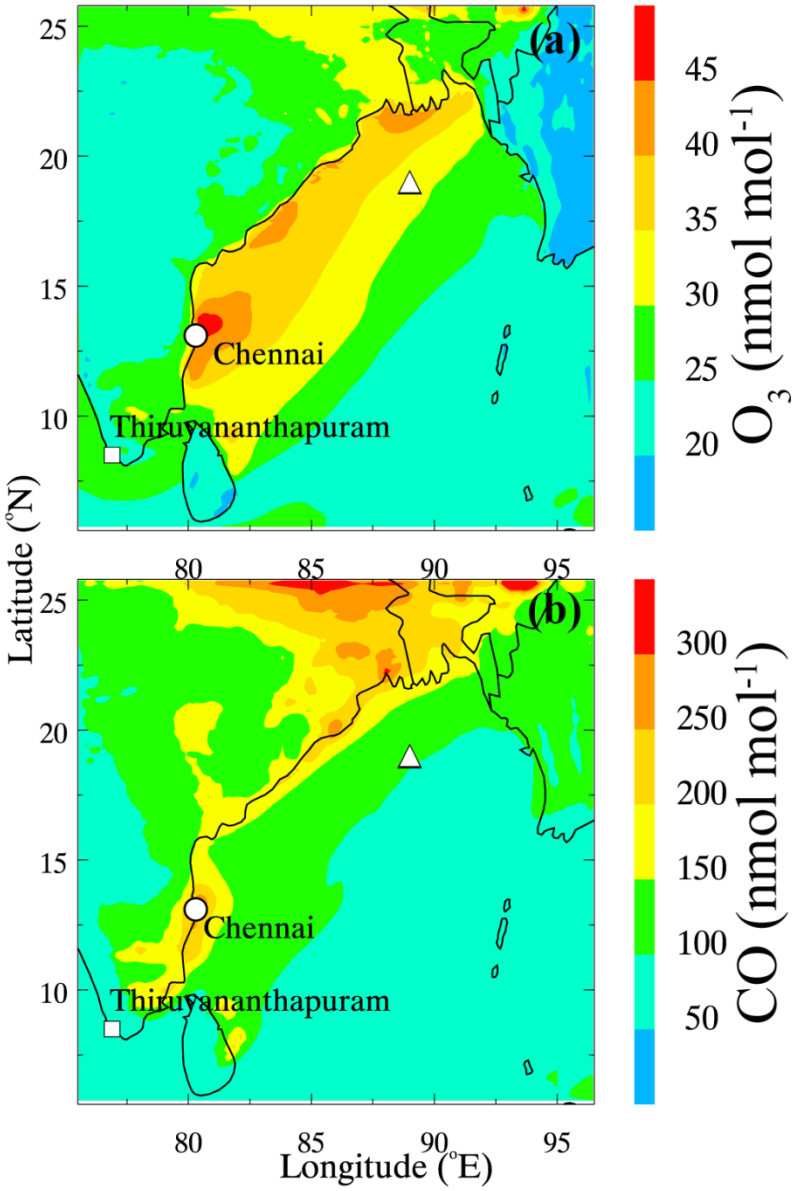


Figure 1. Cruise track (continuous black line) of the Research Vessel *Sagar Kanya* along with synoptic winds at 925 hPa (black thin arrows) and NO_x emissions in background colour map~~averaged over the cruise period~~. ~~Arrows marked on the track shows the ship direction.~~ The dates corresponding to approximate ship positions are marked along the track. The start and end position of the cruise, stationary position of the ship, and Thiruvananthapuram are shown by the circle, triangle and square respectively. ~~The circle shows the start and end point of the cruise. The square tagged with Thiruvananthapuram shows the location corresponding to the measurements shown in Fig. 15. The location at which the ship was kept stationary (July 22–August 06, 2009) is denoted with a triangle. The background colour map shows the NO_x emissions over the Indian landmass for year 2006 as obtained from the INTEX-B inventory.~~


(a)
(b)
Chennai
Thiruvananthapuram
Latitude (°N)
Longitude (°E)
25
20
15
10
80
85
90
95
45
40
35
30
25
20
O3 (nmol mol-1)
300
250
200
150
100
50
CO (nmol mol-1)

850

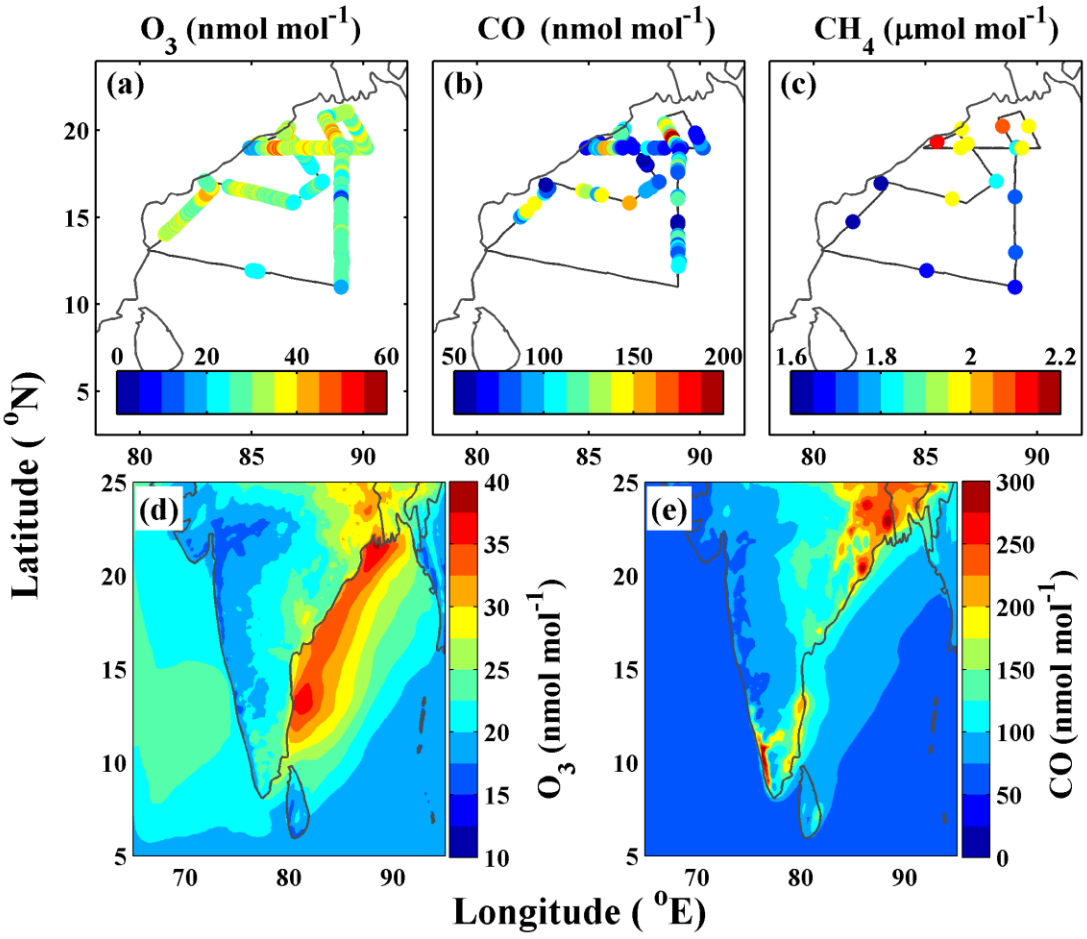


Figure 2. Spatial variation of surface O_3 (a), CO (b), and CH_4 (c) mixing ratios along the cruise track during the CTCZ campaign. WRF-Chem-simulated spatial distribution of surface O_3 (~~a~~d) and CO (~~b~~e) averaged during the July 16–August 17, 2009 period. ~~The location of the ship cruise start and end (Chennai), the ground-based measurement site at Thiruvananthapuram, and the location where the ship was kept stationary are shown by the white circle, square, and triangle, respectively.~~

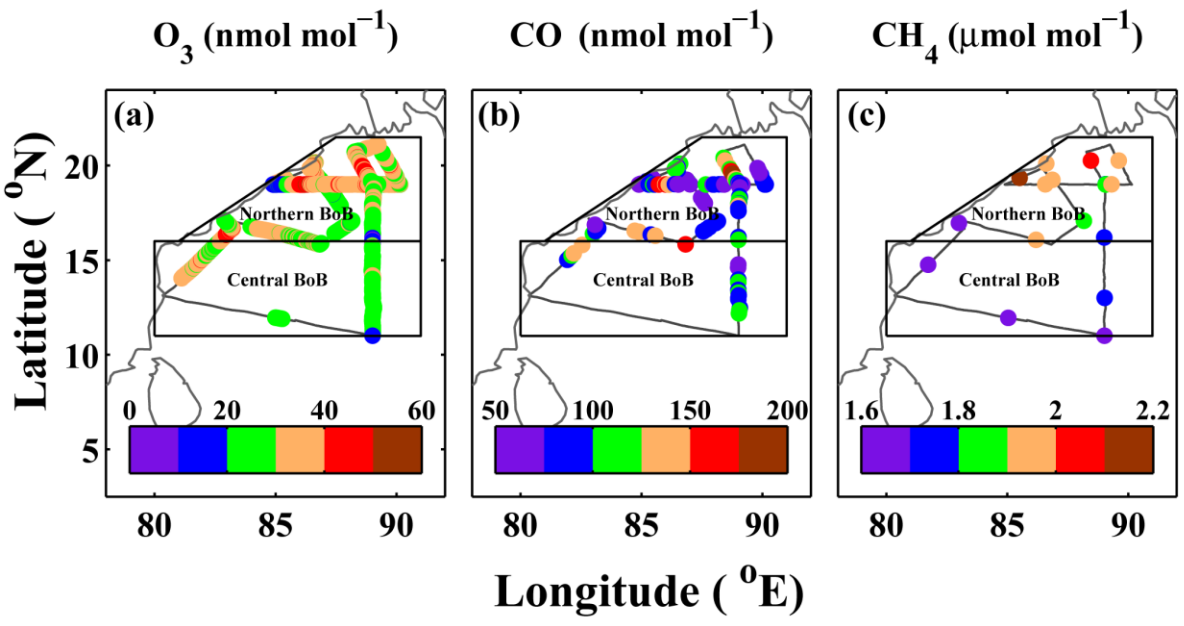
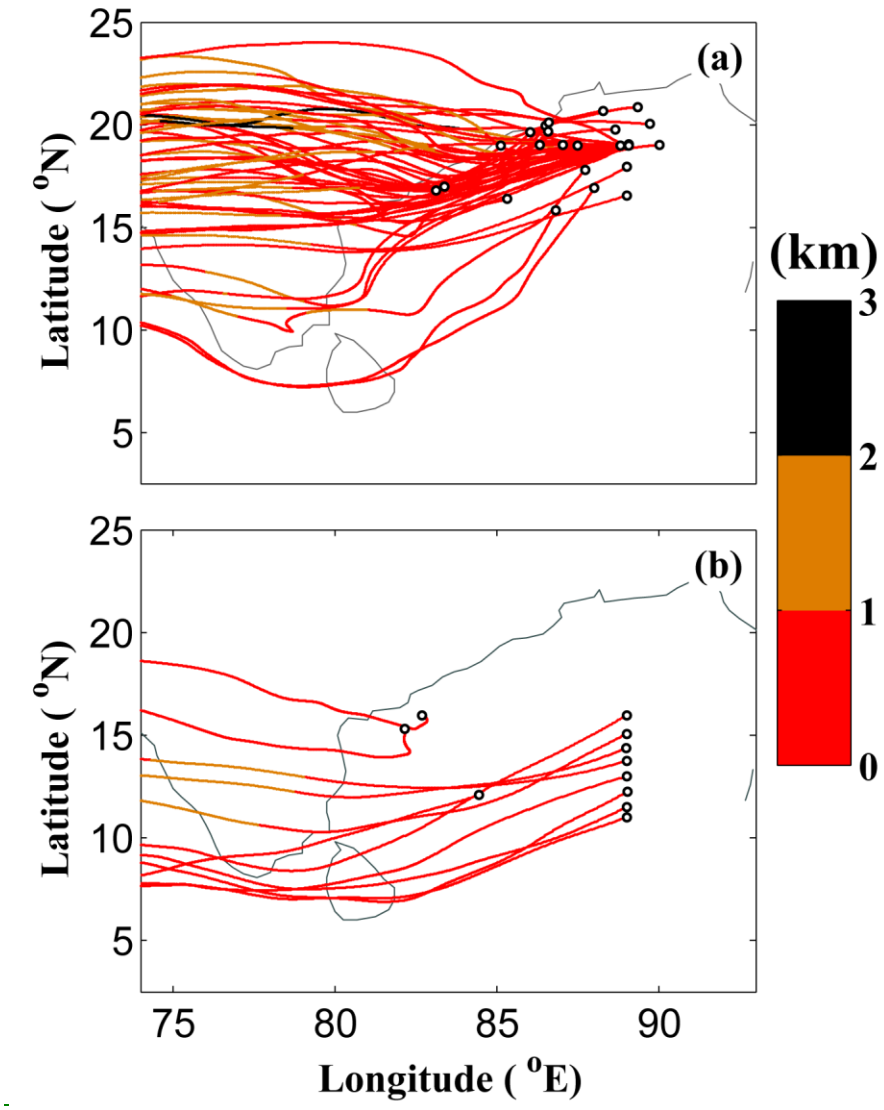
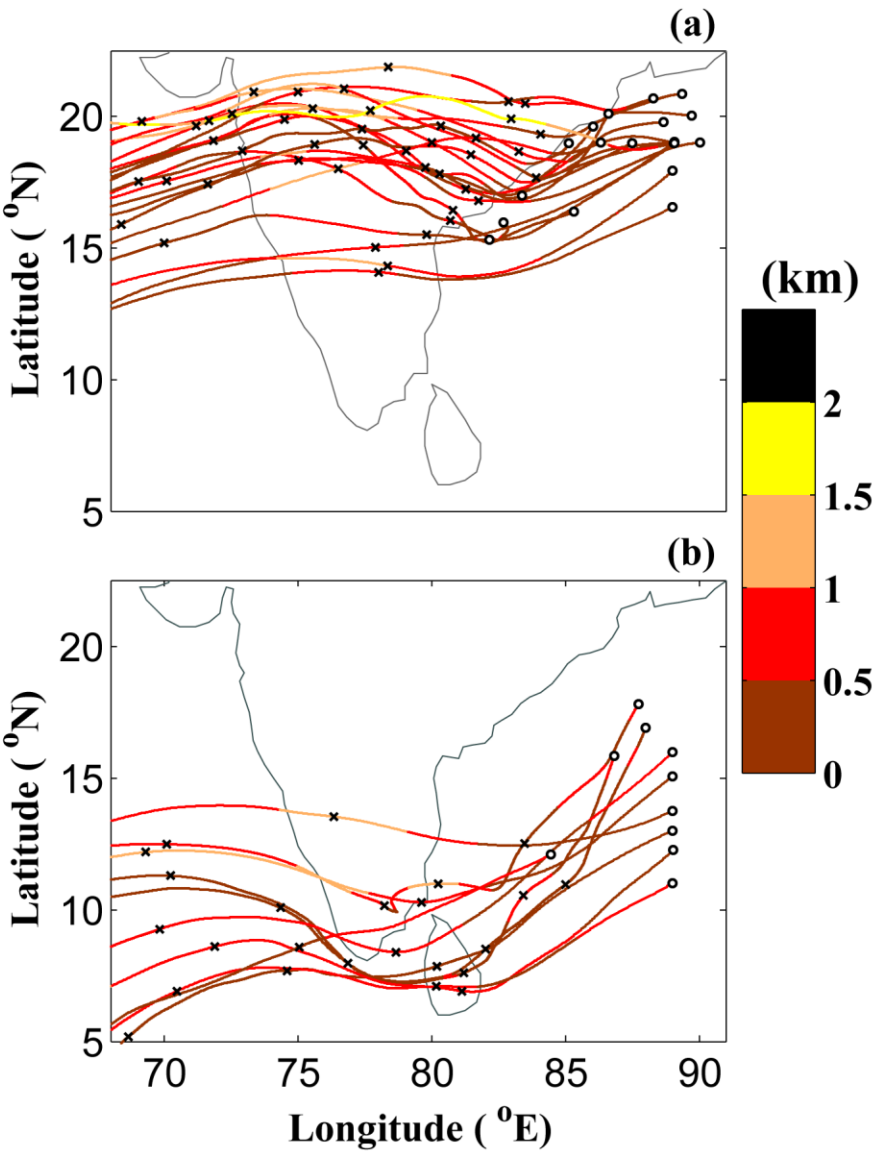
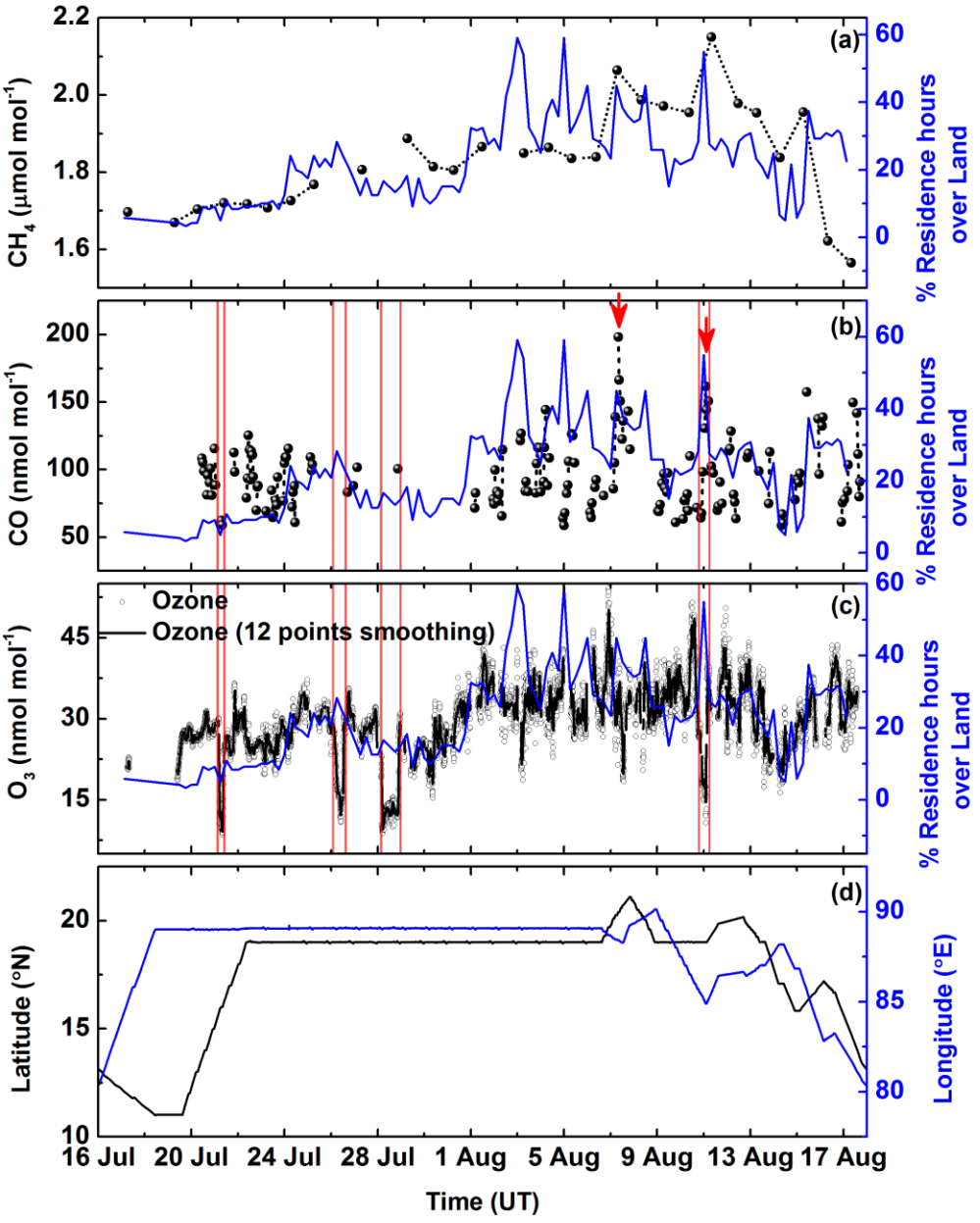


Figure 3. Spatial variation of surface O_3 (a), CO (b), and CH_4 (c) mixing ratios along the cruise track during the CTCZ experiment, which took place during the summer monsoon season. The solid lines demarcate the regions of central and northern BoB.


(a)
(b)
Latitude (°N)
Longitude (°E)
25
20
15
10
5
75
80
85
90
(km)
3
2
1
0



865 **Figure 34.** Five-day airmass back-trajectories during the study period ending at the measurement locations (small black circles) grouped for corresponding airmasses from (a) central/northern India and (b) southern India. ~~over (a) northern BoB and (b) central BoB.~~ The cross symbols along the trajectories represent each back-day. The colour scale shows the height (in km) of the trajectories.


(a)
(b)
(c)
(d)
CH4 (µmol mol-1)
CO (nmol mol-1)
O3 (nmol mol-1)
Ozone
Ozone (12 points smoothing)
% Residence hours over Land
Latitude (°N)
Longitude (°E)
16 Jul
20 Jul
24 Jul
28 Jul
1 Aug
5 Aug
9 Aug
13 Aug
17 Aug
Time (UT)

870

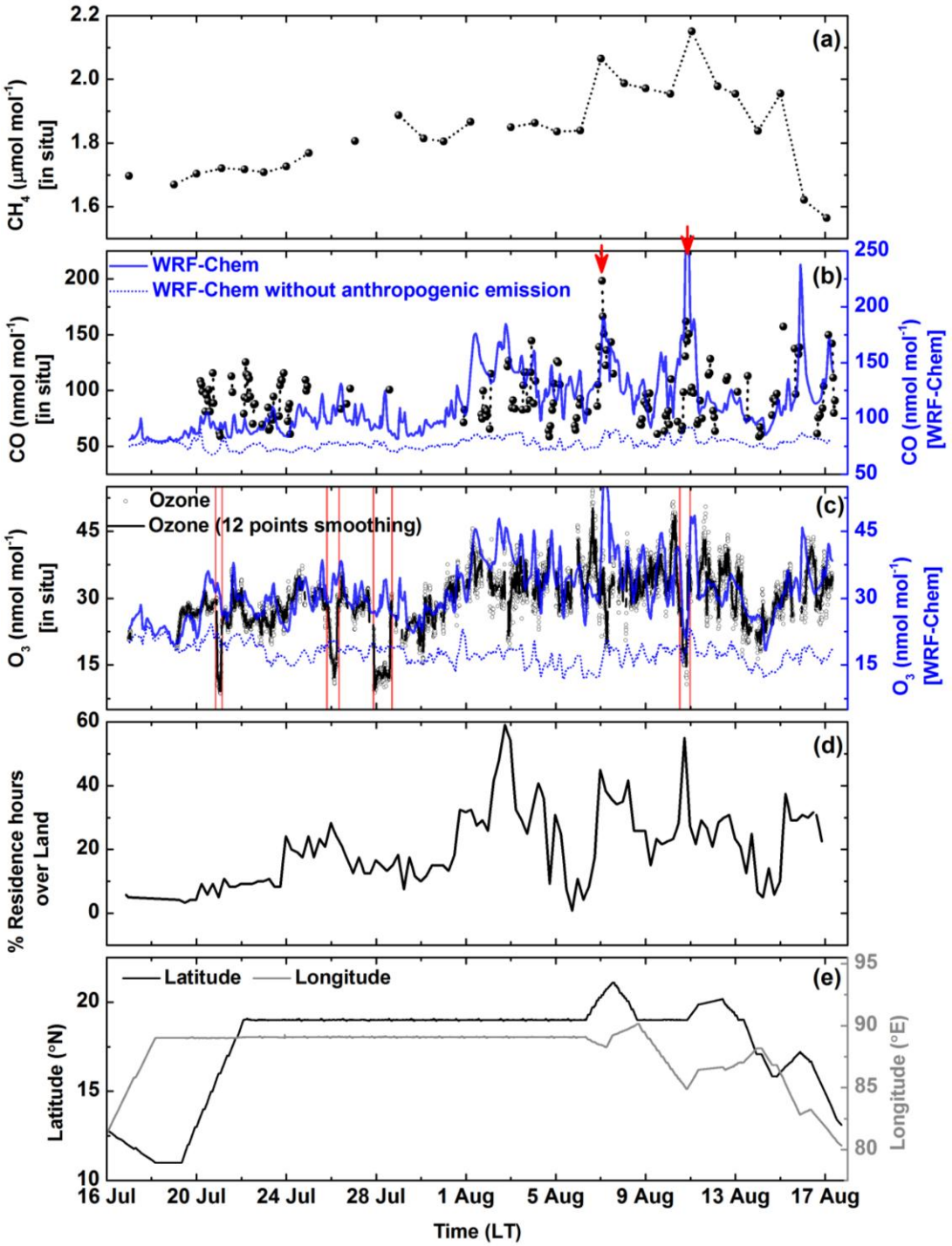
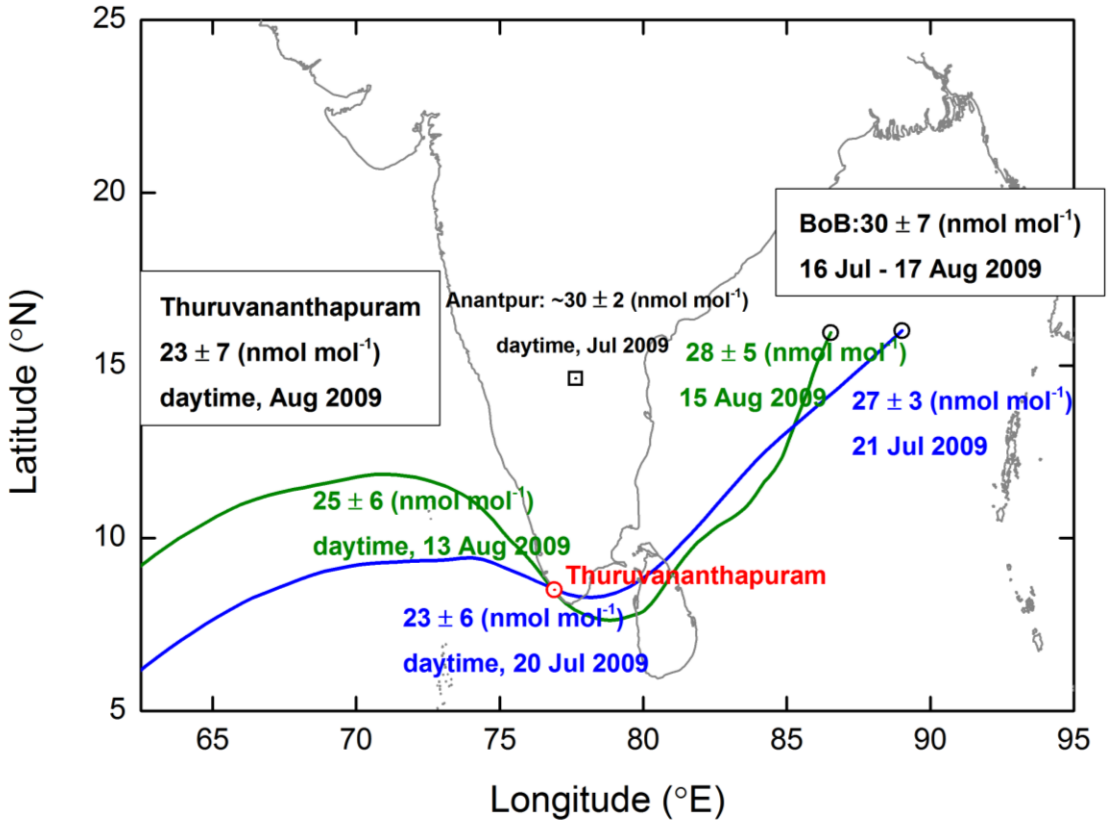


Figure 45. ~~Spatio-temporal v~~Variations ~~of~~ in observed ~~surface~~ CH_4 (a), CO (b), O_3 (c), ~~CO (b), and CH_4 (a)~~ and percentage residence time over land (d) along with WRF-chem simulated O_3 and CO ~~percentage residence time~~ (blue line) ~~over land~~ during the campaign~~summer monsoon season~~. ~~The red vertical lines show the four e~~Events of sharp decrease in ~~surface~~ O_3 ~~and CO~~ during rainfall are marked by vertical red lines (Fig. 8~~11~~). (~~d~~e) Variations in measurement locations~~., latitude (black line), and longitude (blue line) corresponding to trace gas measurements shown in (a–c). Red arrows in 5~~4b highlight elevated CO mixing ratios.



880 **~~Figure 6.~~** ~~Airmass back-trajectories (blue and green curves) reaching 500 m altitude over the two locations (black circles) of observations over the BoB for July 21 and August 15, 2009. The trajectories crossed an observational site, Thiruvananthapuram (red circle), on July 20 and August 13, 2009. Monsoon-time average mixing ratios over Thiruvananthapuram (which is representative of the Arabian Sea airmasses), Ananthapur (which is representative of airmasses over the central part of southern India), and the BoB are also shown.~~

885

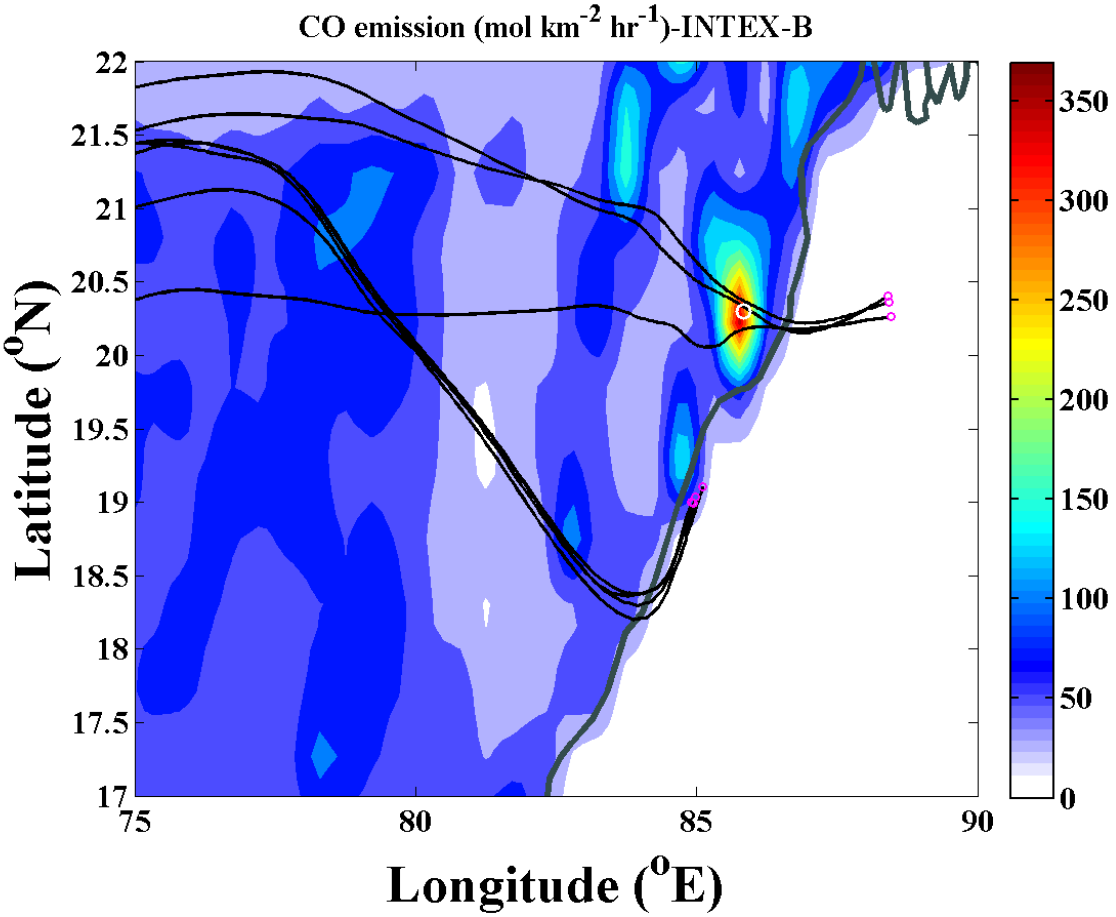


Figure 5~~7~~. Backward ~~A~~airmass trajectories (black curves) 500 m above the location of higher CO observations as marked by red arrows in Fig. 5b during August 7 and 11, 2009. The background colour map shows the spatial distribution of anthropogenic CO emissions over the Indian region for the year 2006 from INTEX-B inventory. The small circles in magenta represent the points where observations were made, as well as the end-point of trajectories. The white circle over the hot-spot region denotes an observational site, Bhubaneswar.

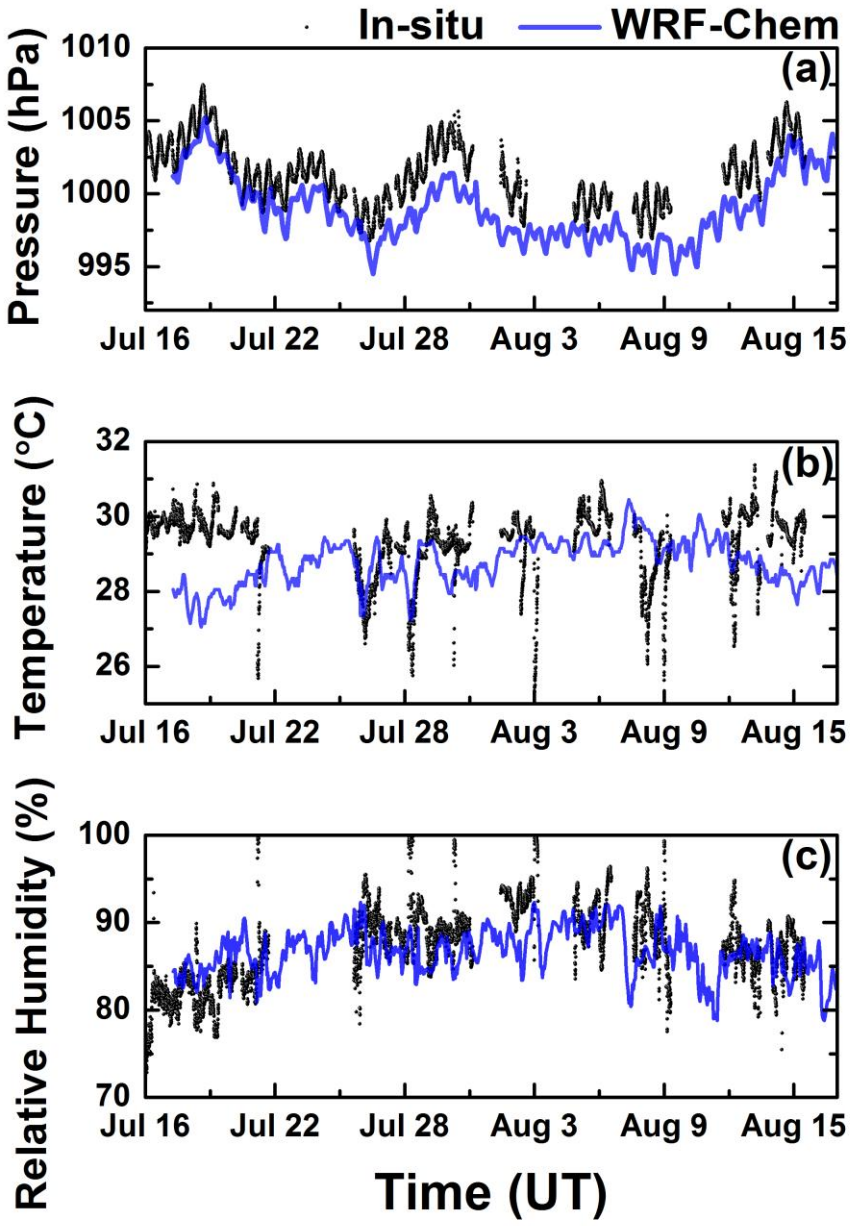
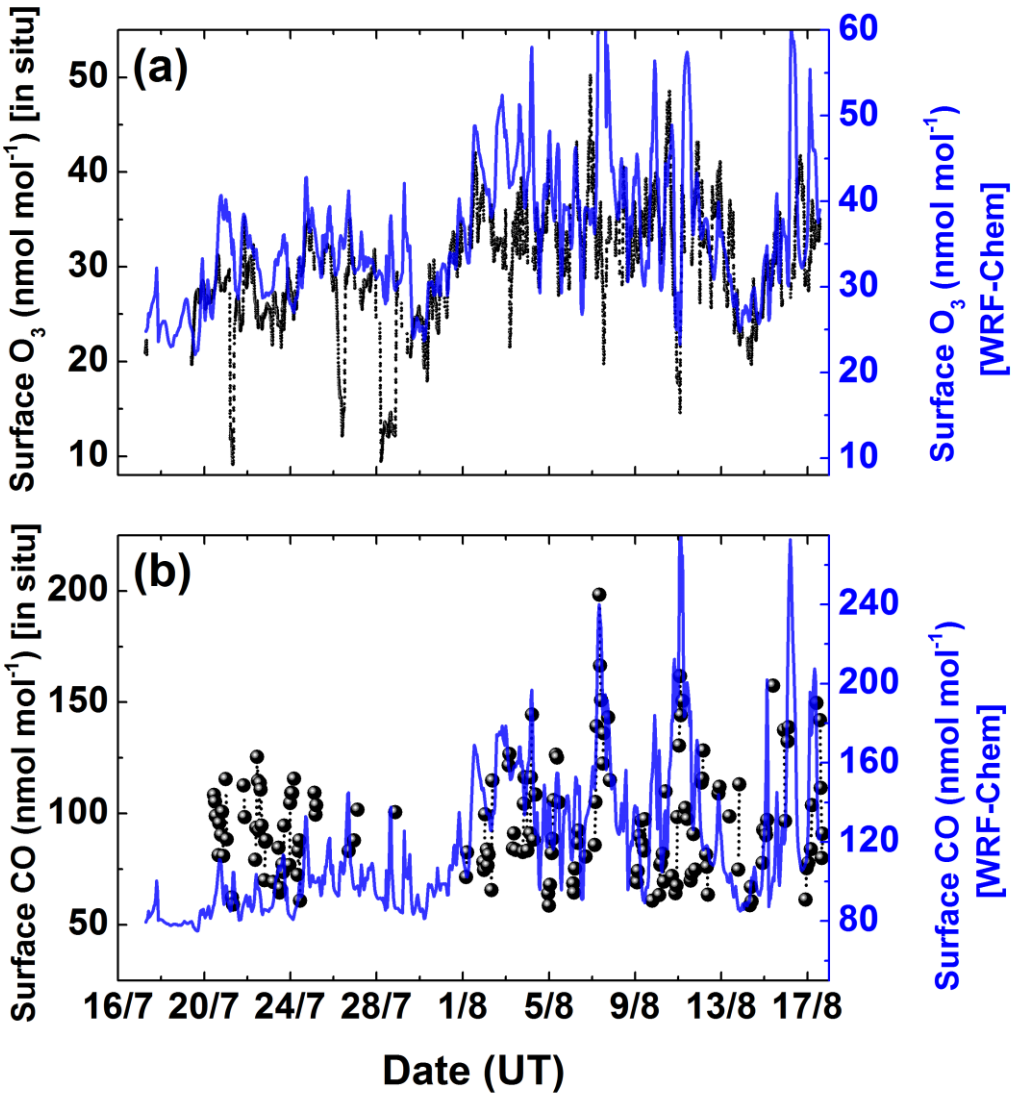


Figure 8. Comparison of the WRF-Chem-simulated meteorological parameters (a) pressure, (b) temperature, and (c) relative humidity with in situ measurements aboard ship during the CTCZ experiment.


(a)
(b)
Surface O_3 (nmol mol^{-1}) [in situ]
Surface O_3 (nmol mol^{-1}) [WRF-Chem]
Surface CO (nmol mol^{-1}) [in situ]
Surface CO (nmol mol^{-1}) [WRF-Chem]
16/7 20/7 24/7 28/7 1/8 5/8 9/8 13/8 17/8
Date (UT)

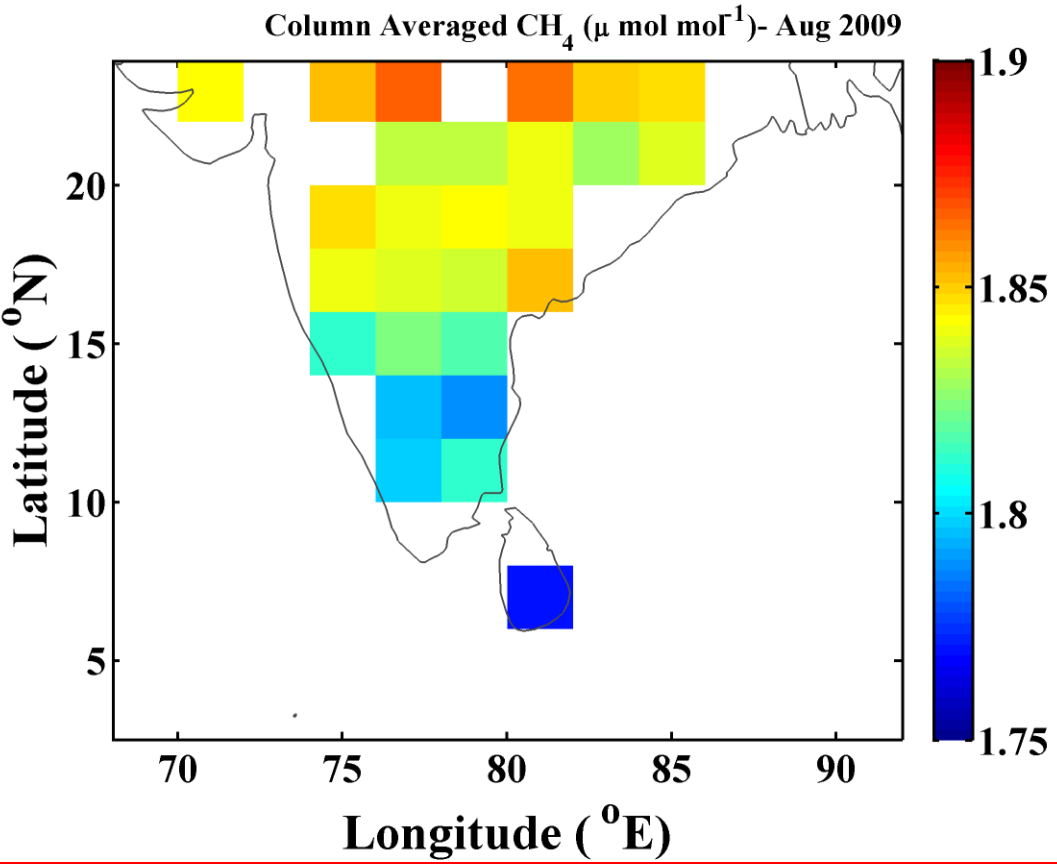
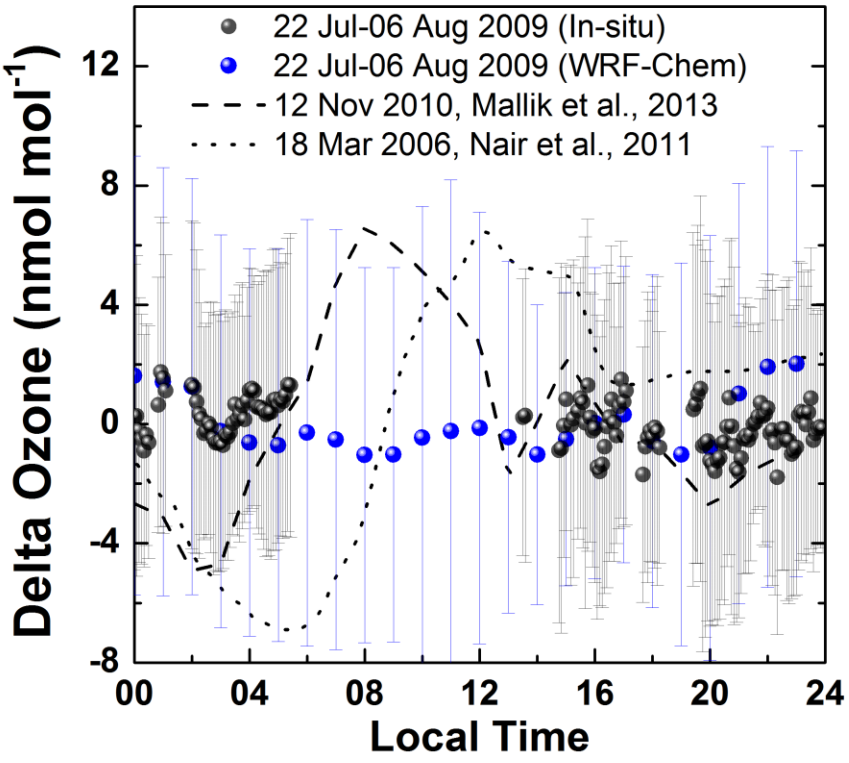


Figure 6~~9~~. Spatial distribution of column averaged CH_4 for the month of August 2009 as obtained from SCIAMACHY.~~A comparison of surface O_3 and CO from in situ measurements (black dots) with model results from WRF-Chem (blue line) along the cruise track over the BoB during the summer monsoon season. The scale of the right axis is adjusted for WRF-Chem according to the mean biases.~~

905


22 Jul-06 Aug 2009 (In-situ)
22 Jul-06 Aug 2009 (WRF-Chem)
12 Nov 2010, Mallik et al., 2013
18 Mar 2006, Nair et al., 2011
Delta Ozone (nmol mol-1)
12
8
4
0
-4
-8
00
04
08
12
16
20
24
Local Time

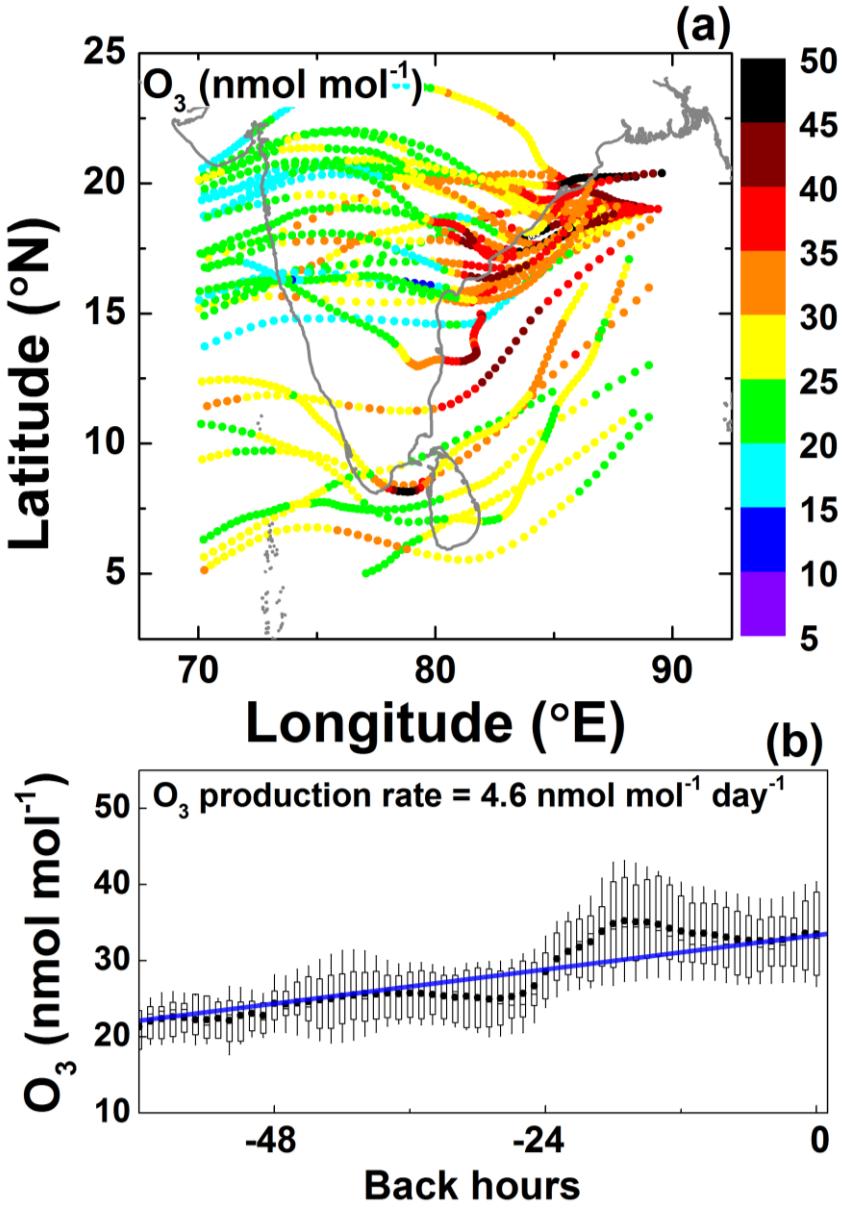
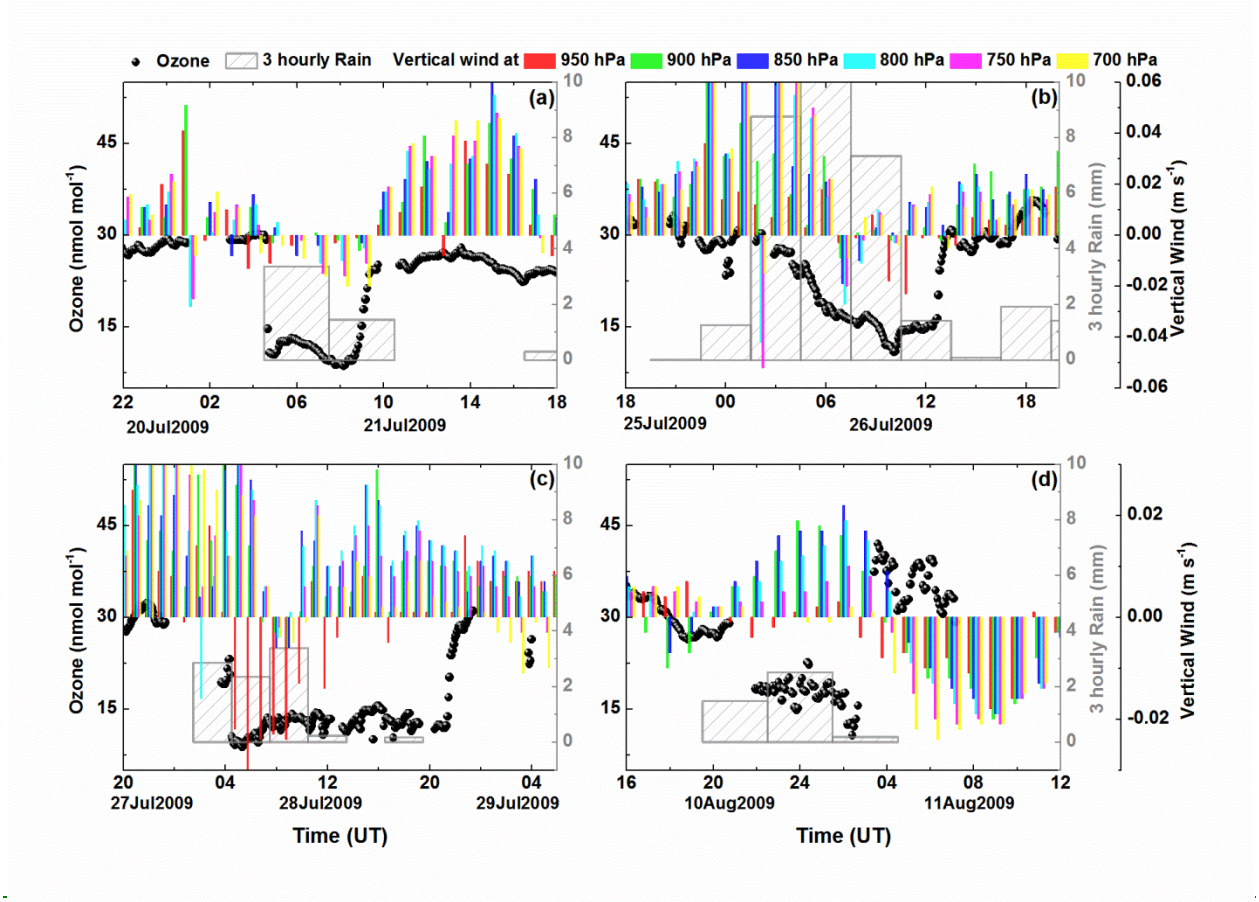
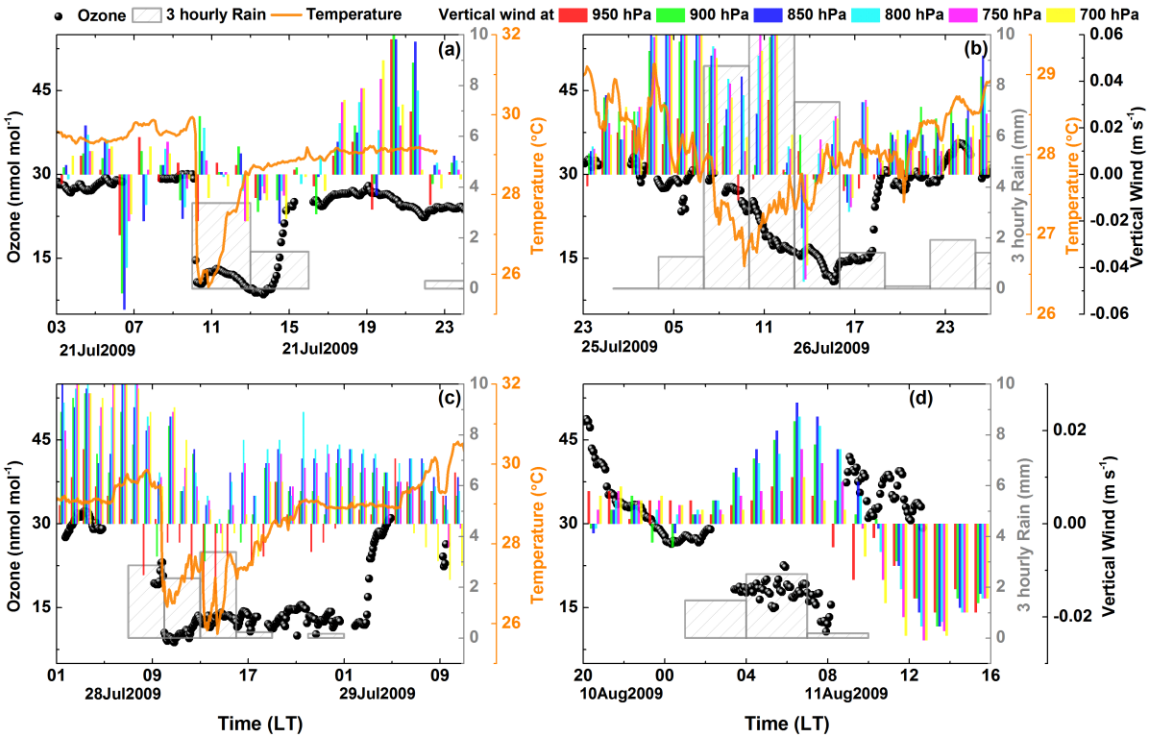


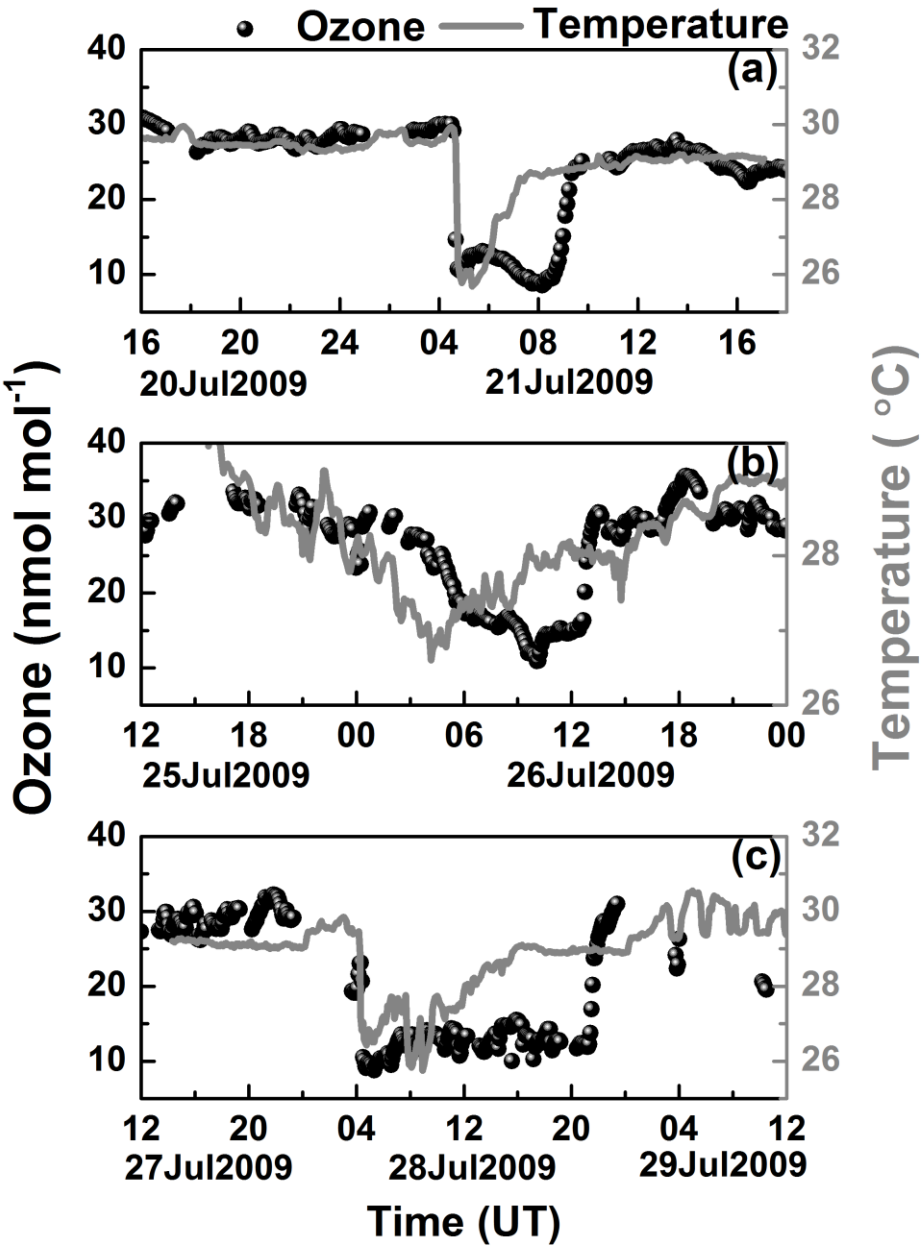
Figure 7~~10~~. ~~The mean delta diurnal variation of~~ insurface ozone ~~over the BoB at a stationary location (89° E, 19° N) in BoB from in situ measurements and along with that from WRF-Chem simulations. during the summer monsoon season. Error bars represent standard deviations. A comparison with The dotted and dashed curves show~~O_3 ~~diurnal variations in surface~~ O_3 ~~(as adopted from Nair et al., 2011, and Mallik et al., 2013) during the pre-monsoon (Nair et al., 2011) and post-monsoon (Mallik et al., 2013) are also shown.seasons, respectively.~~ (a) WRF-Chem simulated O_3 along the airmass trajectories ending over a few representative locations over the BoB. (b) Variation of O_3 mixing ratios with time along the trajectories as shown in 7a. In box plot, the black dots and lines inside the box represent the mean and median of the data respectively. While the lower and upper edges of boxes represent the 25th and 75th percentiles respectively, the whiskers represent standard deviations.


Ozone
3 hourly Rain
Vertical wind at
950 hPa
900 hPa
850 hPa
800 hPa
750 hPa
700 hPa
(a)
(b)
(c)
(d)
Ozone (nmol mol-1)
3 hourly Rain (mm)
Vertical Wind (m s-1)
20Jul2009
21Jul2009
25Jul2009
26Jul2009
27Jul2009
28Jul2009
29Jul2009
10Aug2009
11Aug2009
Time (UT)



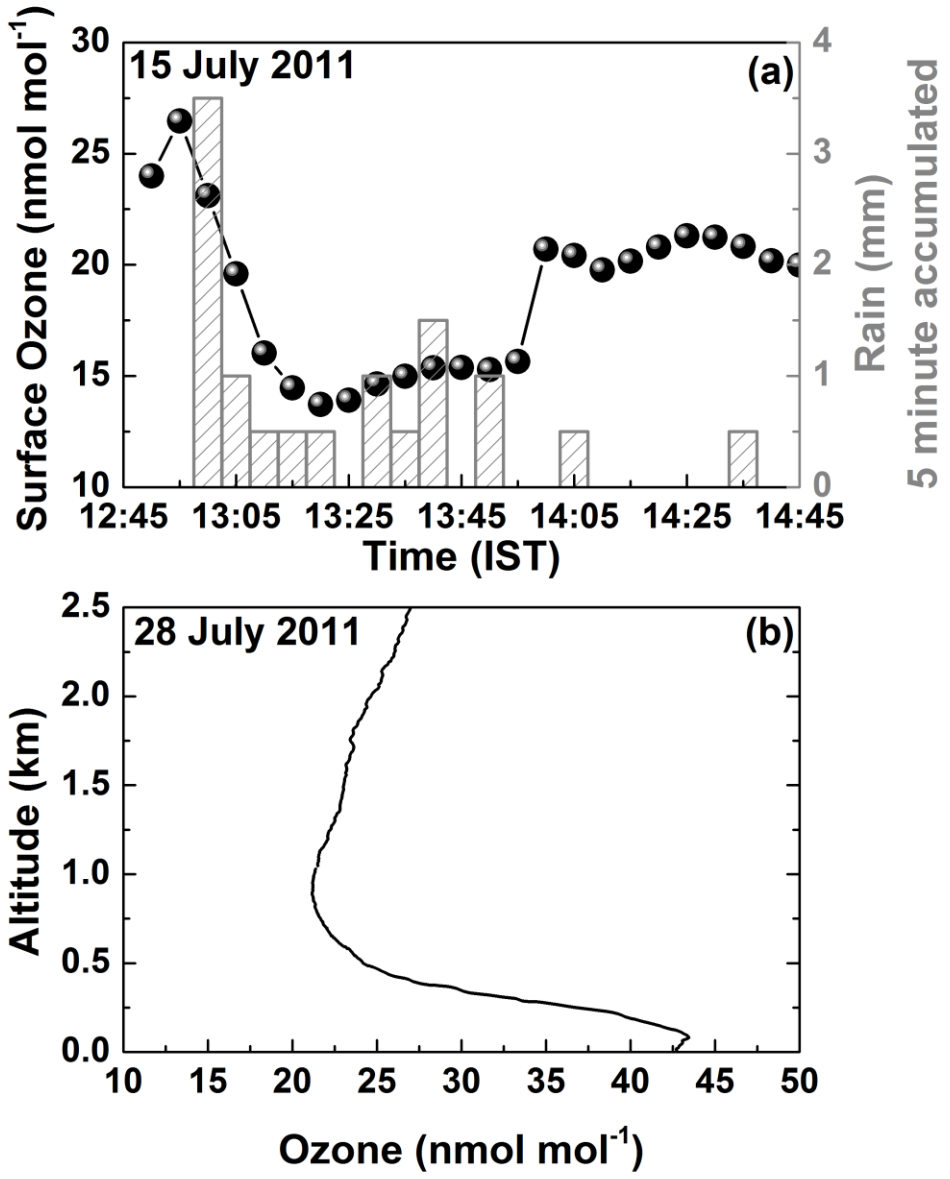
920

Figure 8~~11~~. Surface O_3 (black dots) along with temperature (orange curve) and 3-hourly rainfall (grey vertical bar) during the four events of sharp decline in ~~ozone~~O_3 (a–d) as marked in Fig. 4~~5~~c. Colours indicate the vertical wind as simulated by WRF-Chem.

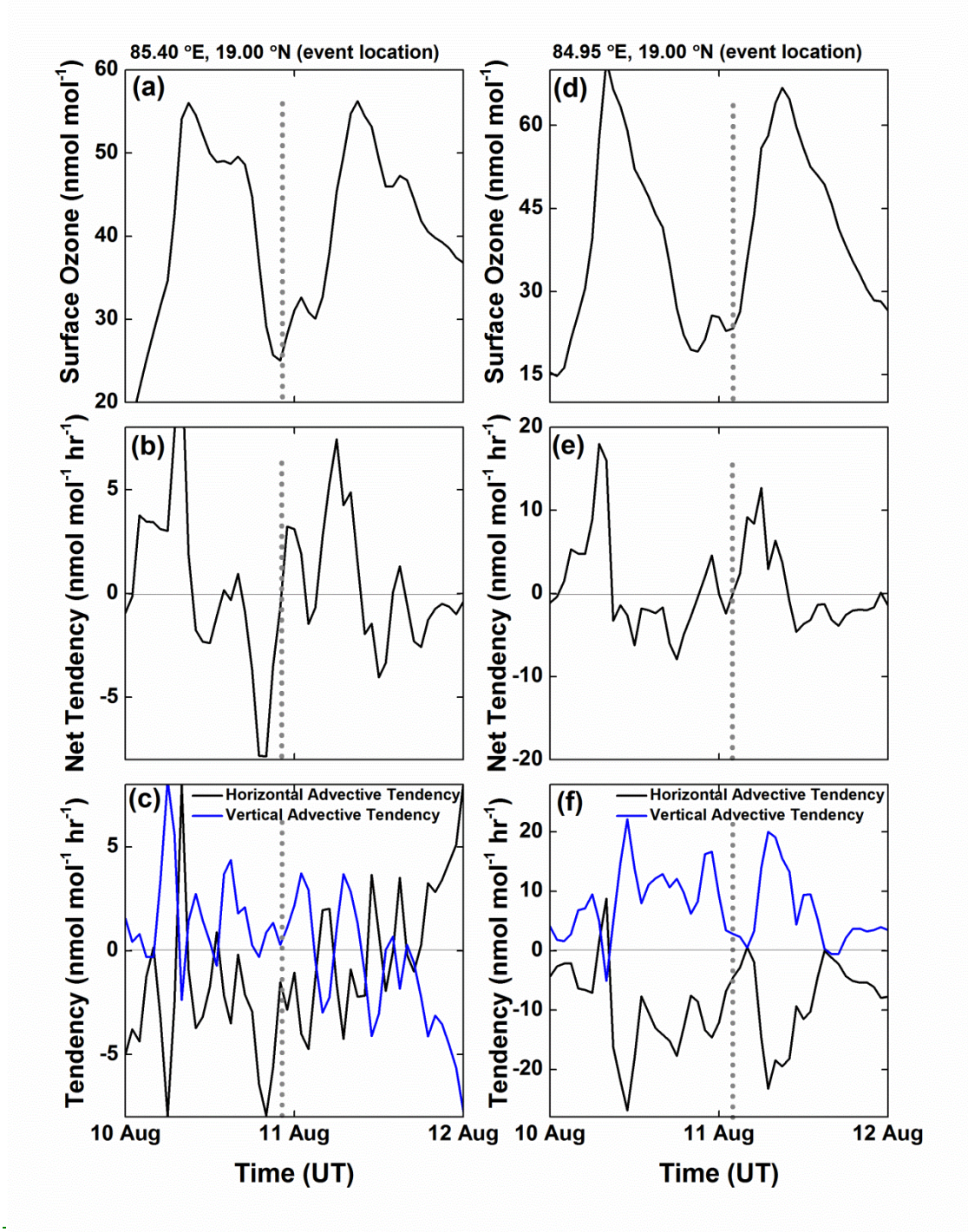


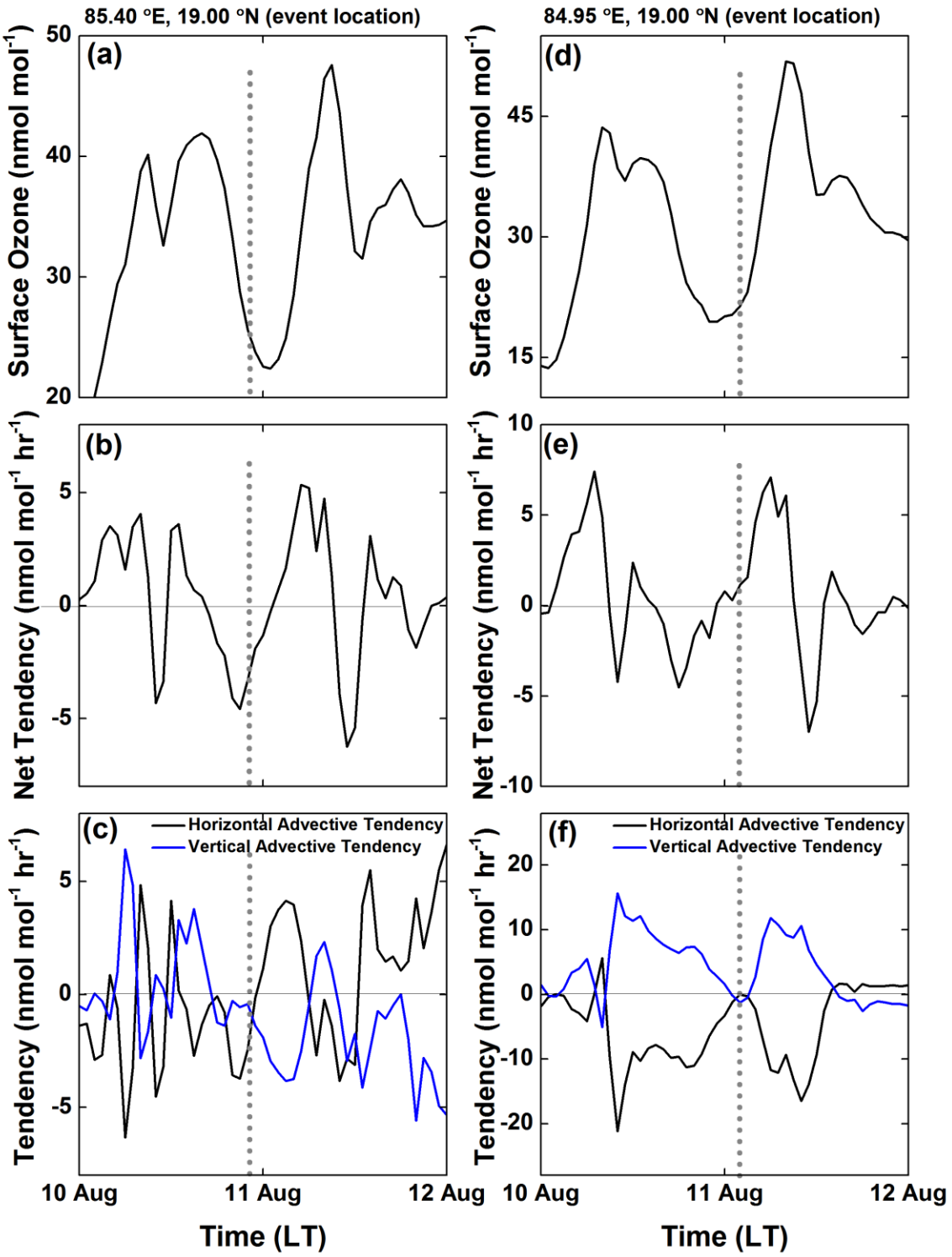
925

Figure 12. Surface O_3 (black dots) along with surface air temperature (grey line) during the three events of sharp decline in O_3 (a–c) as marked in Fig. 5.



930 **Figure 9~~13~~.** (a) Temporal variation in surface O_3 mixing ratio (black dots) along with 5-minute accumulated rainfall (grey vertical bars) over Thumba, Thiruvananthapuram (location of the site shown in Fig. 1 and 2) on 15 July 2011. (b) Vertical profile of O_3 mixing ratio over Thumba, Thiruvananthapuram as measured on 28 July 2011.

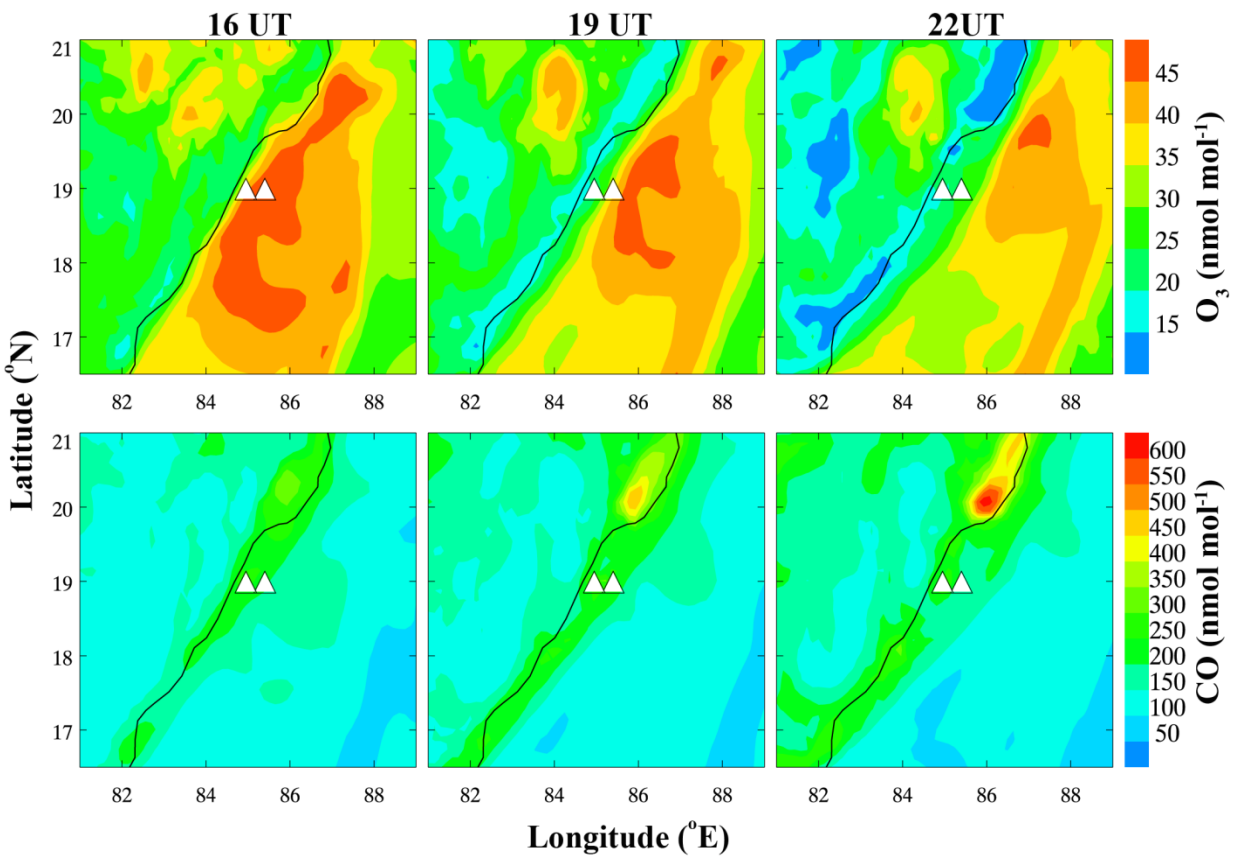

85.40 °E, 19.00 °N (event location)
84.95 °E, 19.00 °N (event location)
(a)
(b)
(c)
(d)
(e)
(f)
Surface Ozone (nmol mol-1)
Net Tendency (nmol mol-1 hr-1)
Tendency (nmol mol-1 hr-1)
Horizontal Advective Tendency
Vertical Advective Tendency
10 Aug
11 Aug
12 Aug
Time (UT)



935

Figure 104. Time series of surface O_3 (a) and various tendency terms (b and c) over the event location during the fourth low-~~ozone~~O_3 event, as obtained from WRF-Chem simulations. 14d–f are the same as 14a–c, but for another

location during the same event. These two event locations are also marked by triangles in Figure 15. Vertical dotted line shows the time of the event in the in situ observations of surface O_3 over the indicated locations.



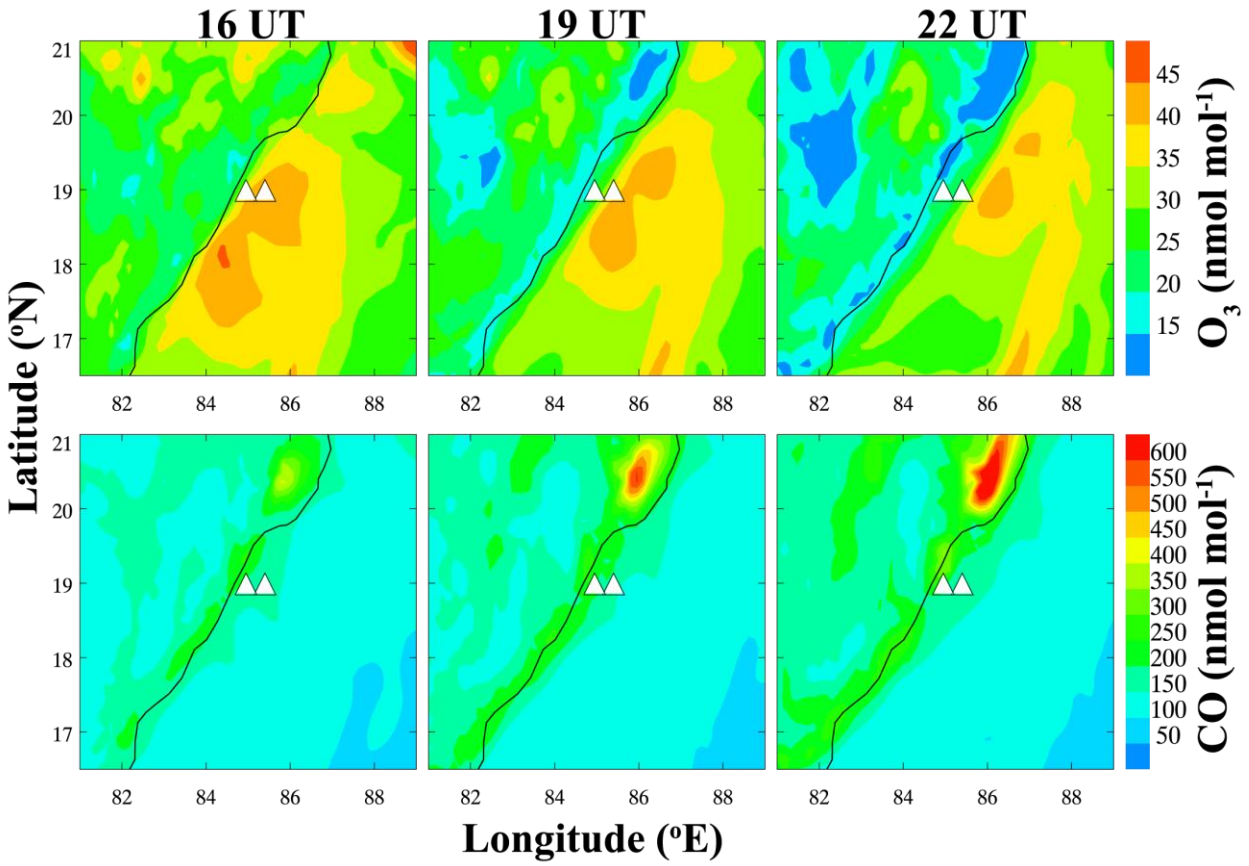


Figure 115. Spatial distribution of surface O_3 (top panel) and CO (bottom panel) at 16:00 UT and 19:00 UT on August 10, 2009, both prior to and during the fourth event, which took place 22:00 UT on August 10, 2009. White triangles show two locations (85.40° E, 19.00° N; 84.95° E, 19.00° N) corresponding to the event.

945

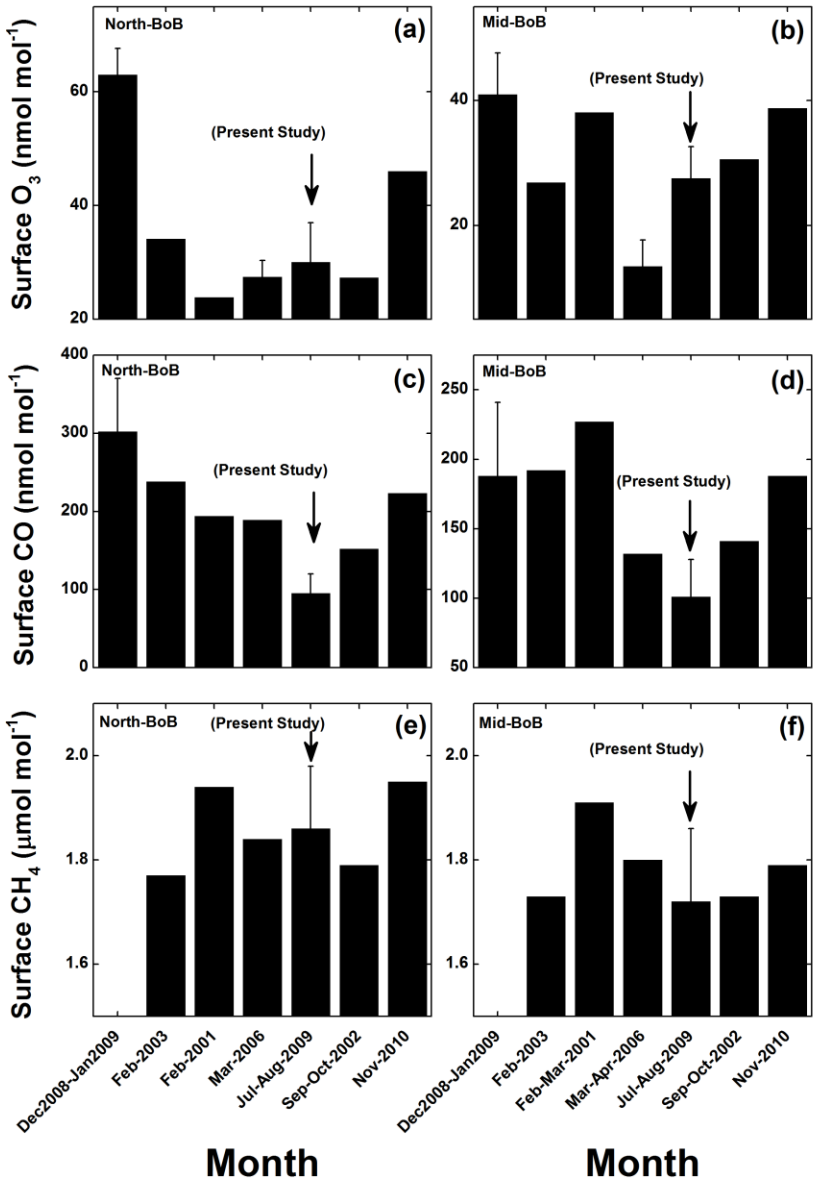


Figure 1~~2~~6. Seasonal variation in average O_3, CO, and CH_4 mixing ratios over (a, c, e) northern BoB and (b, d, f) central BoB ~~(see Fig. 3 for demarcation of these two regions)~~. Except for July–August 2009 (present study period), all average values are obtained from the literature (David et al., 2011; Lal et al., 2007; Lal et al., 2006; Nair et al., 2011; Srivastava et al., 2012; Sahu et al., 2006; and Mallik et al., 2013). Error bars show standard deviations for respective study periods. For any points for which high resolution measurements are not available, standard deviations are not shown.