

Interactive comment on “The detection of post-monsoon tropospheric ozone variability over south Asia using IASI data” by B. Barret et al.

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We would like to thank the referee's for their thorough evaluation of our paper. Answering their comments and concerns allowed us to improve the content and the clarity of the paper. Because some important comments raised by both referee's are identicals, we wrote a single document to answer to both of them.

The **first point of concern of both reviewers** is the ability of IASI to capture the O₃ variability around Hyderabad highlighted by Fig. 7. They insist upon the fact that IASI fails to reproduce the lowest O₃ tropospheric columns over Hyderabad:

Referee 1: *however, the authors may also point out that IASI failed to capture low*

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ozone abundance during the study period (also shown in Figure 7) in the abstract and the conclusions.

Referee 2: *IASI significantly overestimates the low ozone events*

At the same time, **Referee 1** agrees with the authors that IASI can capture fast variability of tropospheric ozone on daily scale as illustrated in Figure 7 and admit that the validation part of the paper is solid and convincing. We acknowledge these statements because we have made the validation study prior to the case study in order to prove that IASI tropospheric O₃ data were able to capture variations both at high and low latitudes. Based on this thorough validation we are sure that the fast drops in O₃ observed by IASI consistently with MOZAIC are not artefacts even though they are of lower amplitude than detected with MOZAIC.

According to **Referee 1**, *The reason for such failure requires more investigation. The a priori biased towards mid-latitudes may only be one of the reasons.* **Referee 2** thinks that the reason for IASI to overestimate low ozone events is that MOZAIC data were not smoothed and then it is difficult to meaningfully compare the variability in the MOZAIC and IASI data - i.e. Figure 7 is not very useful.

Referee 2 is right, in Figure 7, the MOZAIC data were not smoothed with IASI averaging kernels. He is also partly right when he states: *My guess is that transforming the MOZAIC data is a challenge because the profiles do not extend into the UTLS.* To overcome this limitation we use MOZAIC profiles complemented above 220 hPa with profiles from MLS assimilated data that are part of the database used to build the a priori data. A better description of the MLS assimilated data with the appropriate references and numbers concerning their validation is now given in section 2.2. We can therefore apply the averaging kernel smoothing to the complete MOZAIC-MLS profiles

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and check whether the agreement, especially concerning the low ozone events is better.

In order to make things really clear about the validity of IASI TOC measurements over India and to give strength to the case study of the second part of the paper, we have completed the validation part with a comparison of IASI and MOZAIC Hyderabad data (with MLS data above) for the same period (July-December 2008). The results (see new Figure 5 and updated Table 2) show the ability of IASI to detect the O₃ variability without ambiguity with a correlation coefficient of 0.86 when the comparison is made with smoothed data. The smoothed data are consistently biased low by 12% relative to IASI which is in good agreement with the ozonesonde validation even though it is higher than the results from low latitudes (5%).

Referee 2 makes a further statement about the IASI bias for low TOC: *On the other hand, if the MOZAIC data were smoothed, the authors must explain the overestimate by the IASI data, since this represents a significant failure of IASI to capture the observed ozone variability. Is this because of contamination from the stratosphere in the upper tropospheric retrievals?*

The lower variability detected by IASI comes from the fact that we are dealing with satellite data that are less sensitive than in-situ data and correspond to a limitation rather than a failure. The variations of TOC detected by IASI are clearly reduced as can be seen in Figure 4 and 5, with the slopes of the correlation plots close to 0.5 (Table 2). The contamination with the stratosphere is probably rather low at the tropical latitude of Hyderabad, according to the TOC averaging kernel of Figure 2 which is close to 0 at the altitude of the tropopause (around 100 hPa). Furthermore, such a contamination would not play a significantly different role for high and low TOC and would not impact the ability of the sensor to capture variability. Anyway, we now clearly mention the fact that the TOC variability detected by IASI is lower than the variability measured by in situ data as a result of the smoothing effect, in both the abstract and the conclusions.

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Finally, concerning the case study itself, the new Figure 7 (now Figure 8), including the MOZAIC TOCs computed from smoothed MOZAIC-MLS profiles clearly shows improvements and further answers to the referee's main concerns. The ozone drops observed by MOZAIC are reduced when smoothing is applied as mentioned by **Referee 2**. Nevertheless, these drops clearly remain important and in very good agreement to the drops measured by IASI highlighting the ability of IASI to detect these particular events. Therefore, Figure 8 now shows that IASI captures very well the fast variability of ozone detected by MOZAIC around Hyderabad in November 2008 even though both datasets are biased. The text of section 3.4 discussing the comparison of IASI and MOZAIC in November 2008 (Figure 8) has been updated accordingly. The coincidence criteria have been improved to compare IASI and MOZAIC leading to minor changes in Figure 8 relative to previous Figure 7. These changes concern in particular the location of the profile which is now the position of the aircraft mid-way between the ground and its flight altitude rather than Hyderabad (see answer to referee 2 minor comment 4).

Both referees suggest to use tropical sonde profiles.

Referee 2: *An alternative approach would be to use the sonde profiles from Thiruvananthapuram (8N, 77E) and Delhi (28N, 77E) (or even the Sepang airport in Kuala Lumpur (3N 102E)). These may be more limited temporally (i.e., just one or two per month), but they could be useful to show that IASI is capturing the ozone variability over the July to December period.*

Referee 1: *The authors may take more ozonesonde data from other stations in India in addition to Hyderabad so that a more complete picture can be drawn.*

This has already been done in part 1 of the paper. As is mentioned in the text, we

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have used all the data available from the WOUDC and SHADOZ networks at the time of preparation of the paper. Nevertheless, data for south and south-east Asia (part of the low latitudes in the paper) are really scarce: no data in 2008 at Dehli, 12 sondes from July to December 2008 at Sepang airport already taken into account in the validation, 9 sondes at Thiruvananthapuram available only since July 2010 and not taken into account in the paper (their addition would not make a big difference when we already have 146 profiles at low latitudes). It is clear from this inventory that these ozonesonde data are unable to validate the O_3 intra-seasonal variations detected by IASI over central India. With 40 profiles in 6 months measured at Hyderabad, MOZAIC (completed by MLS) provide the best available dataset for this purpose as is now demonstrated in the paper.

The **second important comment** made by both reviewers concerns the interpretation of FLEXPART simulations to identify the processes responsible for the mid-tropospheric O_3 variations in South Asia in November 2008.

Referee 1: *They attribute the rapid decrease in tropospheric ozone to two storm events. This is an interesting case. However, this part needs to be further elucidated. The proposed underlying mechanisms are not clearly presented. To improve this part, I suggest the following. Explain the Flexpart simulation and the output clearly (associated with Figures 9 and 10). The term, unit, and magnitude in Figures 9 and 10 are confusing (see Specific). Referee 1: Figures 9 and 10, unit is missing. If the unit is second, the magnitude seems wrong. The authors claimed a simulation of Flexpart for 10 days. Then the magnitude is way beyond 10 days. For 1011, the trajectory takes too long and it would not be reliable. Referee 1: What is the physical meaning of the retroplume residence times?*

The interpretation of the figures was difficult with the description of the model output
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(residence time) and the units as they were. We now have improved the description of FLEXPART dispersion simulations. It now appears clearly that residence time is not a travel time between emission and receptor (which is 10 days maximum in our simulations), but a sensitivity of source to receptor. A large number of particles is released from a receptor location (here around Hyderabad between 4 and 6 km) and the residence time is a four-dimensional variable that represents the response function to emission input. Because the relative values of residence time are more meaningful than the absolute values (that depends on the number of particles released), we have used mean residence time fraction (MRTF) to describe the transport pathways highlighted by FLEXPART simulations. This variable, given in %, is described in the text as follows: "the mean residence time fraction is computed for each cell of the grid, as the integration of the residence time over the selected vertical layer and over the 10 days backward simulation divided by the residence time integrated over the global domain and over the 10 days." The higher the MRTF is in a given cell of the grid, the more time the released particles have spent in that given cell and the higher the sensitivity of the receptor (Hyderabad) to sources coming from that location.

Referee 2: *Furthermore, the discussion about the FLEXPART analysis on page 10048 is very unclear. For example, on lines 4-5 the authors explain that "an important fraction of the particles reaching the middle troposphere over Hyderabad have spent some time in the BL", but Figure 9 shows residence time in the BL and upper troposphere. It and does not tell me what fraction of the air in the middle troposphere is from the BL.*

Figure 9 shows MRTF in the BL and in the UT because the particles reaching the free troposphere above Hyderabad on November 11 follow different transport pathways when they come from both altitude ranges. There is no contradiction between both plots which have even the same color scales. For instance, one can clearly see in

Figure 9 that particles reaching Hyderabad have spent similar time over Central Asia in the UTLS and over Bengale in the BL. Furthermore, the global integration of the MRTF for the two layers give the global sensitivity of the receptor to these layers. Following the comment of referee 2, this integrated values are now discussed in the text. On 11 November, the receptor is more sensitive to the UT/UTLS than to the BL while on 29 November the particles have spent more time in the BL.

Referee 1: *As the variation in tropospheric ozone in south Asia is influenced by chemistry, transport, and interaction between them, the role of chemistry needs to be addressed. The authors limited their discussion only on transport.* **Referee 2:** *Figure 9 needs to be better explained and the connection between the FLEXPART analysis and the ozone variations should be better established.*

The O₃ distribution is of course the result of a complex interplay between transport and chemistry. Fast variations (even diurnal) of O₃ in the BL are controlled by photochemistry and local emissions. Nevertheless, O₃ in the free troposphere is more related to transport processes than to local emissions and photochemistry. The FLEXPART runs demonstrate that the O₃ free tropospheric content changes are following radical modifications of the origin of the air masses. High O₃ is linked to both midlatitudes UTLS air masses (richer in O₃ than tropical tropospheric and UT air masses) and polluted air masses from northern India/Bengal while low O₃ is linked to the upward transport of O₃ poor MBL air masses from the Bay of Bengal by the westward propagating storms. Our analysis does not aim at quantifying the O₃ budget (chemistry, transport, deposition etc) during the post monsoon season over India, but at showing how severe weather conditions can dramatically modify the transport pathways and the tropospheric composition very fast as is observed by MOZAIC and IASI.

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Referee 1: *Is “m.a.g.l.” meters above the ground level?*

Yes m.a.g.l is for meter above ground level, this is clarified in the modified version.

Referee 2: *2) The ozone variability shown in Figure 7 was for the tropospheric column, averaged from the surface to 225 hPa. However, as shown in Figure 9, in their analysis of the impact of transport on the ozone abundances, the authors focused on the particles reaching the middle troposphere between 3500 – 4500 m. This is odd since Figure 2 shows that the IASI retrievals have low sensitivity to ozone at altitudes below about 400 hPa (e.g. Figure 2b shows that the error reduction in the retrievals is small at altitudes below about 400 hPa). It is not explained why the authors focused on the 3.5 – 4.5 km altitude range instead of looking at higher altitudes, where IASI is more sensitive to ozone.*

Before the FLEXPART transport analysis, we have shown that the O₃ variations, particularly the fast drops, were concerning the whole troposphere using the MOZAIC vertical profiles (see Figure 6). The choice of 3.5-4.5 km agl (4-5 km asl) was made to be above the boundary layer, impacted by local emissions/chemistry and not well observed by IASI, and at altitudes of higher sensitivity for IASI. Nevertheless, the highest IASI sensitivity is slightly above and we have made new FLEXPART simulations with the receptor region above Hyderabad between 4 and 6 km agl (4.5-6.5 asl), that is at the peak of sensitivity from the retrieved IASI TOC (see the corresponding averaging kernel on Figure 2). As could be expected, both figures are very similar because they concern close altitude ranges within the free troposphere which are impacted by similar transport processes. The interpretation of the FLEXPART runs is therefore unchanged.

Referee 2: minor comments

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1) Page 10037, line 12: Please change “i-th x j-th element” to i-th x j-th element”.

Done.

2) Page 10041, equation (4): Ozonesonde data were used in constructing the a priori (xa), but it is not clear if these same ozonesonde data are being used in the validation? If that is the case, it would be difficult to determine if a small bias between the sonde and the retrieval means a good retrieval or just low ozone sensitivity in the retrieval (which returns the a priori in the retrieval). I am concerned that this may be the case below 400 hPa in Figures 3a,b, and c.

The data used to build the a priori are described in section 2.2. They are based on ozonesonde and MOZAIC data for the whole year 2008. The data used for the validation are described in section 2.4 as a subset of this database, namely ozonesonde data and MOZAIC data for Hyderabad (in the revised version) for the period July-December 2008. Furthermore, the validation is made independently at low and high altitudes using two subsets of the validation subset. Therefore, the a priori and validation datasets are not identicals.

We show comparisons of profiles in order to discuss the impact of the a priori and of the smoothing and to validate the theoretical characterisation. First, we show from this comparisons that the disappearance of the large bias below 700 hPa when smoothing is applied (Figure 3e and f) demonstrates a lack of sensitivity below 700 hPa as evidenced from the averaging kernels. Second, the fact that the retrieved profiles diverge from the a priori and get close to the validation profile above 400 hPa (where the a priori and the true profile start to diverge) demonstrates that the retrieval is sensitive to O₃ above 400 hPa (Figure 3d) as also evidenced by the averaging kernels. We think that a good agreement between a priori, true and retrieved profiles below 400 hPa (this is also true at low latitudes above 700 hPa) cannot be interpreted as a lack of sensitivity of IASI to O₃ as stated by the reviewer. From the averaging kernels it is clear that the sensitivity of the retrievals to the O₃ profile decreases below 400 hPa.

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Nevertheless, it is important to notice that we show the averaging kernels (color lines in Figure 2) for the individual retrieval layers that are much denser at low altitudes, each of the averaging kernels therefore concerning a small altitude range. The addition of the information coming from all the lowermost layers is therefore responsible for an averaging kernel for the TOC which peaks at 500 hPa meaning that the TOC is mostly sensitive to O₃ variations around 500 hPa. Because our information content analysis demonstrates that IASI provides about 2 independent pieces of information from the ground to the UTLS that are identified as TOC and UTLS columns, we focused our validation discussion on this particular products rather than on the profiles that are not less meaningful. The case study presented in section 3 is only based on the analysis of TOC data.

3) Page 10041, line 10: Please change “combined to the low” with “combined with the low”.

Done.

4) Figure 3: It is not explained why the authors chose as their coincidence criteria 1 degree and 12 hours. A reference or explanation for this would be helpful.

There is a variety of choices of coincidence criteria that are valid for tropospheric O₃. Because IASI has a large spatio-temporal coverage we can use 1 degree and 12 hours which is the same criterium as the one used by Keim et al. (2009) to validate various preliminary O₃ products from IASI. The ref Keim et al. (2009) was already in the paper but we have added a citation to this paper as ref for our coincidence choice. For MOZAIC data we have slightly relaxed the spatal criterium to 1.5 degree in order to better take the distance traveled by the aircraft at landing and take-off into account. This is detailed in the text.

5) Figure 6 caption: Please put a comma before “(dashed line)” and before “and (dotted)”. Change “[vmr]” to “[ppb]”.

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Done.

6) Figures 9 and 10: What are the units for the residence time? The values peak at over $2.0E+11$ so it cannot be seconds since that would be more than 7 months.

See above the replies to comments about the FLEXPART simulations. The variable is now mean residence time fraction (MRTF) in %.

Referee 1: specific comments

It would be helpful to present the meteorological fields near the surface and at the midtroposphere separately. Ozone maps at low and middle troposphere would also be helpful, if IASI have some sensitivity to ozone in the low troposphere.

Unfortunately, IASI is little sensitive to O_3 in the boundary layer as explained in the paper and shown in Figure 2. We therefore didn't focus on this atmospheric layer.

Throughout the paper, the authors used the term "radiosonde data" for ozone data. The ozone data used for the validation are from an ozonesonde. The authors may really mean "ozonesonde" throughout.

We did change from radiosonde to ozonesonde.

Table 2: add sampling number (N) for each case. Or add N in Figure 4.

Done.

Figure 2: for the Arabian Sea, please give domain in lat/lon.

The domain (15-20°N, 65-70°E) is now given in the figure caption.

Figure 3: for Figures 3b, 3c, 3e, and 3d, is each profile a mean of (IASA-RS)/RS, where (IASA-RS)/RS is calculated individually?

No, we compute the ratio of the mean of the difference to the mean sonde value.

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Figure 5, are white areas for missing data? Is the vertical velocity also a mean from 500-650 hPa?

White areas are indeed for missing data. This is now mentioned in the caption. Vertical velocities are also for 500-650 hPa. This is now clearly mentioned in the caption.

Figure 6, "3" should be in subscript. Unit of ozone is ppbv. Overlaying IASI vertical profiles for the same region and period would be helpful. Although the bars are understandable, please indicate how many standard deviations the length of the bars stands for.

We modified the Figure and the caption. This Figure is taking benefit of the vertical resolution of MOZAIC to demonstrate that the whole troposphere (at least below 250 hPa) is impacted by the drop of O_3 following the storms. IASI profiles provide less than 1 independent piece of information for the ground-250 hPa altitude range and they would only add confusion to the figure. Detailed comparison of IASI and in-situ profiles together with a full comparison of IASI and MOZAIC tropospheric columns at Hyderabad (in the updated version of the paper) are already provided in section 2.4. Furthermore, Figure 8 shows comparison of MOZAIC and IASI tropospheric columns, which are the most significant amount to be compared, for the studied period. We added that the bars stand for 1sigma standard deviation.

Figure 7, "3" should be in subscript in both x-axis and y-axis. Although the bars are understandable, please indicate how many standard deviations the length of the bars stands for.

This has been modified. We added that the bars stand for 1sigma standard deviation.

Figure 11, are white areas for missing data?

White areas are indeed for missing data. This is now mentioned in the caption.

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P10032, L6, "Level 1 IASI data". The authors may really mean IASI Level 2 ozone data. Level 1 data usually refer to radiance data.

The sentence is "IASI Level 1 data can be used to retrieve tropospheric O₃ columns (surface-225 hPa) and UTLS columns (225-70 hPa)." We do mean Level 1 or radiance data used to retrieve O₃.

P10035, where is the source of IASI ozone data? Are IASI data publicly available? A web link would be helpful.

The data are produced at Laboratoire d'Aerologie in Toulouse. They are not yet publicly available on a web or ftp site. Nevertheless, we have added a sentence stating that people interested should contact the corresponding author of the paper to get these SOFRID O₃ data.

P10038, are there any difference in ozone retrieval between this work and Boynard et al. (2009, ACP, 9, 6255-6271)? If so, please state them.

we state "...value used by Boynard et al. (2009) to retrieve O₃ profiles from IASI with the LBL radiative transfer code." this means that Boynard et al. (2009) use a different retrieval algorithm (LBL) than ours. In order to make things clearer, we now mention the name of this algorithm (Atmosphit) in the text. From the text, the reader knows that the algorithms are different (fast (RTTOV) versus LBL (Atmosphit) radiative transfer) but he has to read Boynard et al. (2009) if he is willing to have details about their retrieval settings.

P10046, L6, it is not clear if the anticyclone is near the surface.

In that particular case, we are discussing about the anticyclonic circulation near the surface which is ending with the northeasterly trades. We have made things clearer, adding "low-level..." about this point.

P10047, L16, missing a "?" after "DU".

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We corrected this mistake: there is a ? instead of DU.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 10031, 2011.

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