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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric Ozone Variability during the East Asian Summer Monsoon as Observed by Satellite (IASI), Aircraft (MOZAIC) and Ground Stations" *by* S. Safieddine et al.

S. Safieddine et al.

sarahsaf@mit.edu

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We would like to thank Reviewer 4 for her/his comments. Below is a point-by-point response for the comments and suggestions raised in the review. The review is copied here (in italic), and then we dissect it and give our answers.

The authors showcase a number of different datasets that capture atmospheric variations associated with the East Asian Monsoon. The subject is interesting and appropriate for the journal, the paper is clearly written and the presentation is good, but ulti-

mately lacking in substantive analysis or new information. The authors show six years of IASI ozone, and then show the IASI ozone for the 2011 season alongside output from the ECMWF model. It is good to see that IASI captures the EASM phenomenon, but this has been shown before. This work shows maps of IASI data for multiple years, which has not been shown previously, but the existence of the EASM and the fact that it shows up in IASI data has already been documented in previous studies. The authors compare IASI 0-6 km columns with MOZAIC aircraft measurements for three different airports. Why do this for only one year? Presumably MOZAIC data are available for multiple years? It could be interesting to see how well IASI captures the seasonal cycle that is observed in the MOZAIC profiles at the different airports, but this is not clear from any of the figures. If IASI/MOZAIC comparisons were performed for multiple years, then it would be possible to quantify how well IASI captures the inter-annual variability of the EASM as observed at those sites.

As suggested by the reviewer, we updated the plots and the discussion to include the analysis of [2008-2013] MOZAIC/IAGOS aircraft data. We initially chose to present 2011 only as the aircraft data was most complete for the monsoon period of 2011 (as new Figure 5 shows), but analyzing more years indeed brings useful information.

Figures 3 and 4 are updated; we added new Figures 5 and 6. In this response we will start by showing the updated validation of IASI by the MOZAIC/IAGOS profiles during the monsoon of the study period, then we will study how well IASI captures the inter-annual variability of the EASM as observed at those sites.

The new discussion of Figure 4 is now as follows (changes are in bold):

"Figure 4 shows the correlation of [0–6] km O3 column retrieved from spatio-temporal coincidence of 363 IASI and MOZAIC/IAGOS smoothed profiles during **May–August of [2008-2013] (except for 2010 where no aircraft profiles were available), and over the airports located in the study domain (see Fig. 3 for the location). Over the five years, a good agreement between the two datasets is found with corre-**

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lation of 0.73 and absolute relative bias of 12%±9%. Analysis of each year data leads to correlations ranging between 0.7 and 0.8 and bias ranging between 11 and 19%. Our results suggest a good ability of IASI to reproduce O3 variability in the troposphere over this region. Discrepancies arise from the spatial resolution of the IASI footprint resulting in an observation averaged over tens of kilometers around the airport and therefore may include other surface O3 contributions. Moreover, the aircraft observation takes place at different times during the day whereas IASI observation is at around 9:30 a.m. and 9:30 p.m. local time. With its limited sampling time during the day and its lower sensitivity towards the surface, IASI observation is not able to capture the diurnal variation of O3 like an aircraft profile. **Our results show a declined correlation between IASI and aircraft products as compared to Barret et al. (2011) where they reported a correlation coefficient of 0.87 (12% ± 6%). This could be due to the different retrieval algorithm used: SOFRID (Barret et al., 2011) vs FORLI (Hurtmans et al., 2012). A discussion of the differences between the two algorithms can be found in Dufour et al. (2012). Another source of difference may arise from the different season and time period studied (Barret et al. (2011) uses a 6-month profiles over the period July–December 2008). Over the monsoon period, and in particular in May and June, the diurnal variability of tropospheric ozone is much more pronounced and highly dependent on the local meteorology. Therefore discrepancies between IASI and the aircraft profile will carry larger discrepancies given the ± 10h coincidence criteria we used. Moreover, our study takes only the column from [0-6] km O3 column from IASI whereas the lower tropospheric column used by Barret et al. 2011 is based on the column from the surface up to 250 hPa (10 km) and IASI is known to have a better sensitivity in the upper middle troposphere (Boynard et al., 2009; Safieddine et al., 2013).”**

To address how well IASI captures the inter-annual variability of the EASM as observed by the aircraft data we created a new Figure 5. The discussion of this figure comes directly after Figure 4, as follows:

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"Figure 5 shows the time series of the columns plotted in Fig. 4 averaged over three latitude bands (the vertical bars are the standard deviation around the mean). The IASI-O3 product captures very well the inter-annual variability of the EASM as observed by the aircraft data over the different latitude bands. The decrease in the O3 columns is most important at [10-20]oN, and over the different years, a result that can also be seen in Fig. 1. Less data is available at [20-30]oN (no co-located observations are recorded for 2008, 2009 and 2013). For 2011 for example, a decrease -though smaller in magnitude than the one in the [10-20]oN- is recorded both by IASI and the aircraft observations between May and June. Between July and August, a decrease is recorded in 2011, and an increase is recorded in 2012, and both events are well captured by IASI. At [30-40]oN, the effect of the EASM on the tropospheric O3 column, as Figs. 1 and 2 showed, is weak or not clear. IASI captures very well the variability during both 2011 (the consistent slight decrease between May to August 2011), and 2013 (various increasing/decreasing behavior)."

As (new) Figure 5 shows, 2011 is the year with available co-located data for both IASI and aircraft at the different latitude bands. Therefore, we choose to also keep the old Figure 5 (now Fig. 6) to check the effect of the EASM on the vertical distribution of O3 at the different latitudes bands using 2011 as an example year. Instead of looking at specific airports, we perform a latitudinal average over [10-20]N, [20-30]N and [30-40]N, as we did in Fig. 5. The discussion of this plot was not modified much, as the previous Fig. 5 discussion holds for the current plot as well.

We brought the (little) following modifications to the text (in bold):

"We show in Fig. 6 the monthly averaged (with the horizontal bars as standard deviation) raw MOZAIC profiles during the EASM of 2011 taken as an example year. **At [10-20]N**, and from May to June, a clear decrease of around **20 ppbv in O3 volume mixing ratios (VMR) is recorded at 1 and 2 km.** At higher altitudes up to around 7 km, an important decrease is also detected from May to June and then from June

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to both July and August. With a small standard deviation around the O3 profiles, the observations **at [10-20]N**, suggest consistent averaged monthly behavior and shows a decrease in the O3 profile at different altitudes from 0 to 7 km, which was also seen over the whole [0–6] km O3 column from IASI in Figs. 1 and 2. **At [20-30]N**, the O3 VMR in the lower troposphere **(1 to 5 km, no monsoon signature at the surface)** decreases from May to June of 10-20 ppbv but then increases back in July and/or August. At [5–8] km, the different months averaged O3 VMR becomes comparable. Looking at this **latitude band** in Fig. 2, we can see how the decrease in the lower tropospheric O3 in June can be explained by the increase in the cloud cover and in particular an increase in the wind speed at 850 hPa coming from the west Pacific. **The profiles located at [30-40]N**, except at the surface, show a decrease in the O3 VMR is detected from June to July and August (till 6 km). **All three averaged profiles show no monsoon signature at the surface, probably due to enhanced O3 precursor emissions near the ascent and descent of the aircraft.**

The section on ground-based information does not seem well linked to the rest of the paper. Again, analysis of multiple years of data and how the variability relates to that observed in the MOZAIC and IASI measurements would make this study more interesting.

It wasn't very easy to get ground observation data from Chinese colleagues and unfortunately we only have those for 2011. We re-wrote this section in a way to improve the connection between the different sections of the manuscript as follows (changes are in bold):

"**[. . .] Hyderabad shows continuous decreasing O3 values from May to August, of total magnitude of 22 ppbv in accordance with Fig. 6 at the same latitude band. Jabalpur is located in a region where the monsoon effect is strong as seen in Fig. 1 and 2, and the O3 at the surface behaves similarly, with a decrease of 20 ppbv. Udaipur lies in a region where the monsoon is milder, leading to a small decrease in the summer-time O3 values that decrease of 5 ppbv between May and July and**

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then increase back in August to 32 ppbv. Panel b station data are the 24-h running average (and the associated standard deviation in shaded blue) of 12 stations in the PRD region and 7 stations in the NCP region. Since we are interested in the regional EASM effect on O3, we show the average of the stations here (the station are between 25 to 300 km away). For details and more timely resolved observations for each of the station, please check the supplementary material. **The ground observations of the PRD stations, represented also by the [20-30]oN MOZAIC profiles in Fig. 6, detect a decrease of > 35 ppbv from June to July coinciding with when the northwesterly winds from the Pacific become stronger (see Fig. 2).** The O3 VMR increase slightly afterwards during July and August due to the decrease in monsoon strength over this region (also seen in Fig. 2). Panel b for the NCP stations shows a weak monthly decrease in O3 concentrations from June to July and August of 5–10 ppbv. The IASI and meteorological data presented in Fig. 2, also suggest the same decreasing pattern in O3 concentrations driven by the slight increase in cloud cover.

Alternatively, the authors might consider further analysis of the causes of the observed inter-annual variability in tropospheric ozone in this region. Is the observed variation in tropospheric ozone mainly due to meteorology? [. . .] Or in changes in cloudiness/photochemical activity?

Figure 1 now presents 6 years of IASI-O3 data, and we attempted to strengthen the discussion over this figure by looking at different sources to explain the year-to-year variability. We came to the conclusion (see below for more info) that the variability observed is due mainly to meteorology (including cloudiness as the Reviewer suggests), and that the other two possible sources of variability in O3 (transport from the stratosphere, and changes in emissions) are not conclusive enough to describe and depict the year-to-year variability.

To what extent do changes in stratosphere-tropopshere exchange come into play versus long-range transport within the troposphere?

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In our analysis, we chose to use the [0-6] km O3 column from IASI to get at least one degree of freedom of information on the vertical while minimizing stratospheric contribution. We agree that STE events could be a source of ozone; in particular in the north of the domain we are studying. To answer this we analyzed potential vorticity and water vapor mixing ratios as tracers of STE events during the monsoon seasons of [2008-2013], and we choose here 2011 as an example year. Data is extracted from the ERA-interim reanalysis at 3 pressure levels: 500 hPa (5.5 km), 350 hPa(8 km) and 250 hPa(10 km). Shown first here (Fig A1 here, see below) is the result at 500 hPa, which is the most relevant to check if in our [0-6] km O3 column, stratospheric intrusions is a source of ozone.

The results show that the potential vorticity, at latitudes <30 N is around 0.5 pvu and increase with latitude, but stays below 1 pvu (and far from the condition of possible stratospheric intrusions of 2 pvu). The specific humidity is high on average, and lowest around the North China Plain, where we have also the highest PV, but the PV/Qvap are not high/low enough to suggest stratospheric intrusions at this altitude.

On the other hand, the possible intrusions become more important when we go up in altitude as the Figures A2 and A3 suggest, in particular at 250 hPa (10 km).

The plots show that stratospheric intrusions become important at latitudes >30N at 10 km. The same analysis has been carried out for the 6 years of the study period [2008-2013] and similar seasonal behavior is recorded, with no particular events (in particular at 500 hPa) that suggest that the ozone regional distribution and year-to-year variation could be explained by stratospheric intrusions into the [0-6] km O3 column shown in figure 1.

Given the above analysis, we updated the discussion of Figure 1, with the following:

"**[. . .] To understand the year to year variability in tropospheric ozone, we looked at possible stratospheric intrusions into the [0-6] km O3 column. Potential vorticity (PV) and the water vapor mixing ratio (Qvap) measurements were used as**

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markers of transport from the upper troposphere–lower stratosphere (UTLS) to the troposphere, and extracted at different altitudes. We used data from the ERA-interim reanalysis (more info about this dataset is provided in section 2.3), and it was seen that downward transport from the UTLS can have an effect at altitude > 10 km and latitudes > 30°N (results not shown here). The analysis therefore suggests that the [0-6] km O3 monthly average columns studied here are not affected by transport from the stratosphere."

To what extent are the year-to-year variations associated with changes in regional emissions?

Year to year variations associated with changes in regional emissions and their effect on local production of ozone is an interesting point to be mentioned. Looking at the literature and different emissions databases we were not able (to our knowledge) to find recent and validated emission database for years 2011- present, the latest being to 2010. E.g. the EDGAR-HTAP project ($http://edgar.jrc.ec.europa.eu/htap_v2/index.php$) compiles a global emission dataset with emission estimates of different pollutants. In order to answer this question, we analyzed the emission estimates of NMVOCs and NOx as ozone precursors for the years available within the project, 2008 and 2010 individually. We calculated the total emissions by summing the individual sectors emissions (energy, industry, residential and transport). We regridded the data from 0.1x0.1 to a 1x1 grid. For the sake of simplicity we show in Fig S1 (that will be included in the supplementary materials), the difference (2010 emissions – 2008 emissions) for each of the months, reported in tons.

The plots show that between 2008 and 2010, there has been an increase on average over China, of 2 to 20 tons of NOx, the largest increase being over East China (between 2 and 20% increase). The North China Plain and the area around Beijing shows > 30% increase, that could be attributed to the unusual low emissions in 2008 because of the Beijing Olympic games. India shows regions with mixed trend but a uniform increase of around 5% is recorded over the whole Indian region. Japan is the only region that

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shows a clear decreasing signature.

Non-methane hydrocarbons (NMVOCs) also show a similar behavior, with an increase over almost the whole domain, except for Japan. We choose to keep these plots in Supplementary materials, and talk about it in the text as follows:

"We also looked at the year to year variations associated with changes in regional emissions to study their effect on local production of ozone. We use emission estimates of NOx and non-methane hydrocarbons (NMVOCs), which are O3 precursors, from EDGAR-HTAP project ($http : //edgar.jrc.ec.europa.eu/htap_v2/index.php$**) available for only 2008 and 2010. An increase in both NOx and NMVOCS is observed between 2008 and 2010 over the studied domain and months, except for Japan (see supplementary materials Fig. S1). Note that strict controls on vehicles and industries emissions were implemented for the summer Olympic and Paralympic Games in Beijing between July and September 2008 (United Nations Environment Programme, 2009). Traffic was reduced by 22% during the Olympics (Wang and Xie., 2009) and restrictions were applied on polluting industries in Beijing and surrounding provinces (Li et al., 2009). While these standards did not include surface O3 limitations, O3 values were expected to be indirectly affected. When comparing 2008 to the rest of the years, O3 distributions do not show a significant decrease in Beijing and/or the surrounding regions (in fact August 2010 and 2012 have lower O3 values over Beijing than those recorded in 2008), a result also detected in previous studies looking at O3 surface measurements (Chou et al. 2011; Wang et al. 2010). Studies suggested O3 transport by winds from nearby polluted areas such as the North China Plain (Wang et al., 2009, 2010), or O3 formation in the boundary layer since the photostationary state of the nitrogen cycle was perturbed (Wang and Xie, 2009). Over the rest of the domain, the increase in emissions of O3 precursors, at least between 2008 and 2010, suggests that tropospheric ozone values also increased between these two year during the**

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different months, but the IASI-O3 columns do not show the same pattern. The analysis presented here suggests therefore that the meteorology associated with the EASM is the main driver of the O3 regional distribution and year-to-year variability. [. . .]"

The IASI ozone dataset is undoubtedly a great resource for investigating monsoon impact on the distribution of pollutants in the troposphere. The analysis presented here is an interesting start, but it would be good to see the authors take this study further in order to provide information that would add to the body of knowledge on this subject.

We hope that with the new version of the manuscript, addressing both Reviewers 3 and 4 comments in details, and updating the discussion and going more in depth into the analysis, we made the study more interesting for publication.

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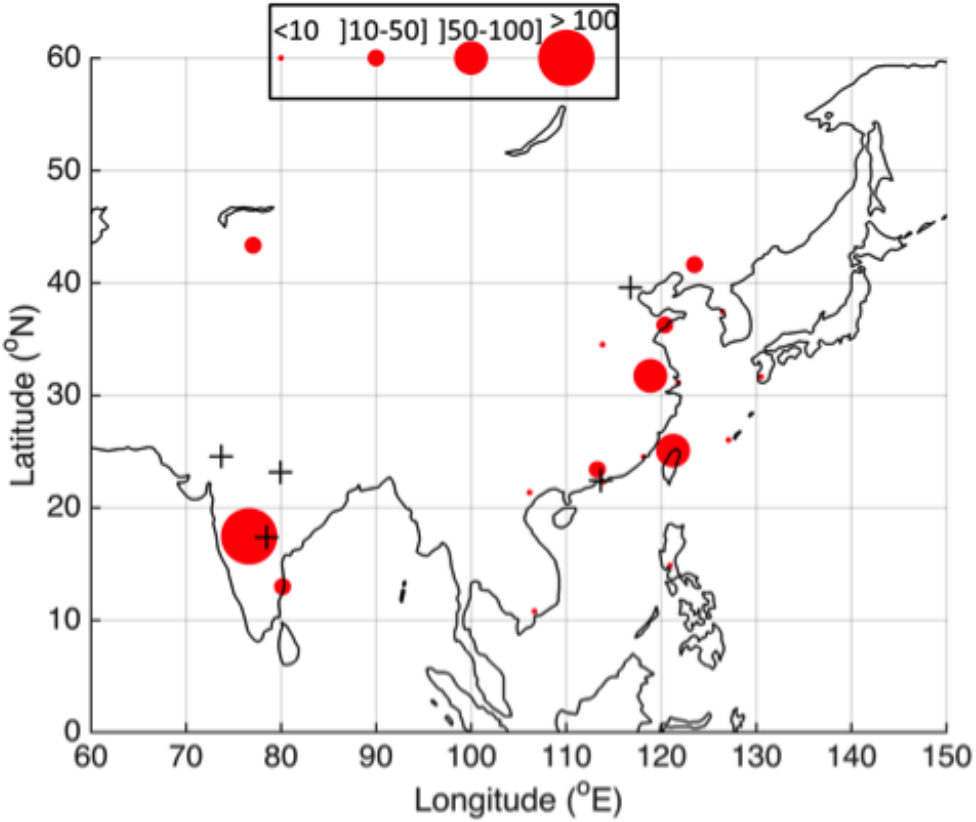

Fig. 1. Figure 3. In red and scaled to number of observations: location of the MOZAIC/IAGOS flight data at the different airports in our study domain. The "+" sign locations corresponds to the ground stations

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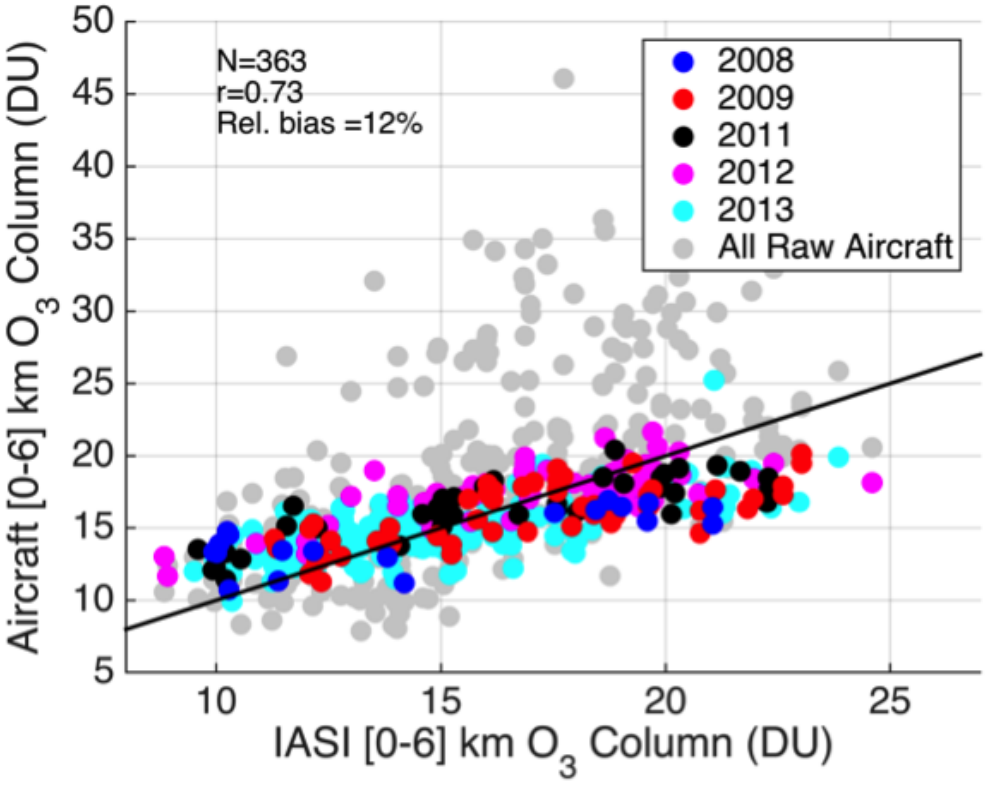

Fig. 2. Figure 4. The [0–6] km O3 columns retrieved from IASI correlation with 363 coincident MOZAIC/IAGOS profiles convolved with IASI averaging kernels for the period May–August of [2008-2013].

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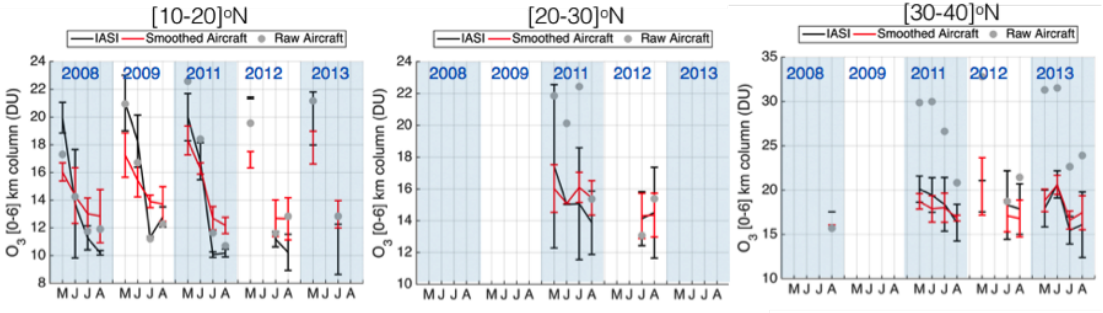

Fig. 3. Figure 5. Time series of [0-6] km available O3 columns from co-located IASI (black) and aircraft (smoothed in red and raw in grey) data at the airports (Fig.3)averaged between different latitude bands

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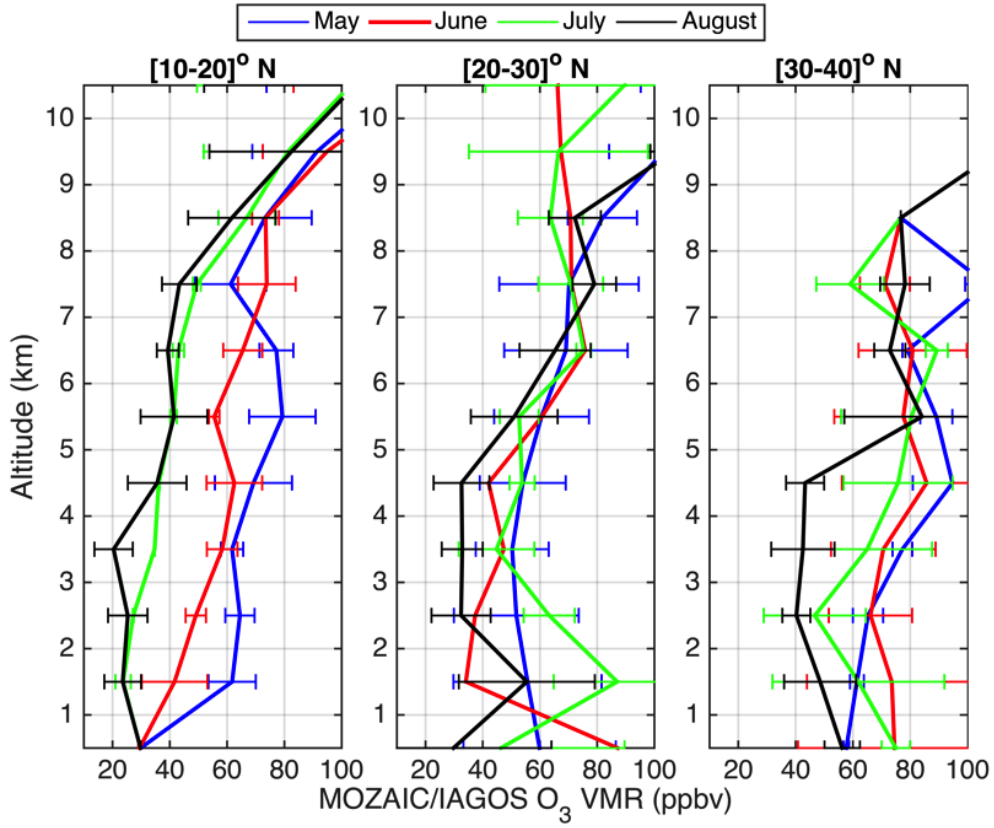

Fig. 4. Figure 6. Monthly averaged vertical profiles of tropospheric O3 from MOZAIC during the period May–August 2011 at [10-20]N, [20-30]N and [30-40]N. Error bars correspond to the standard deviation.

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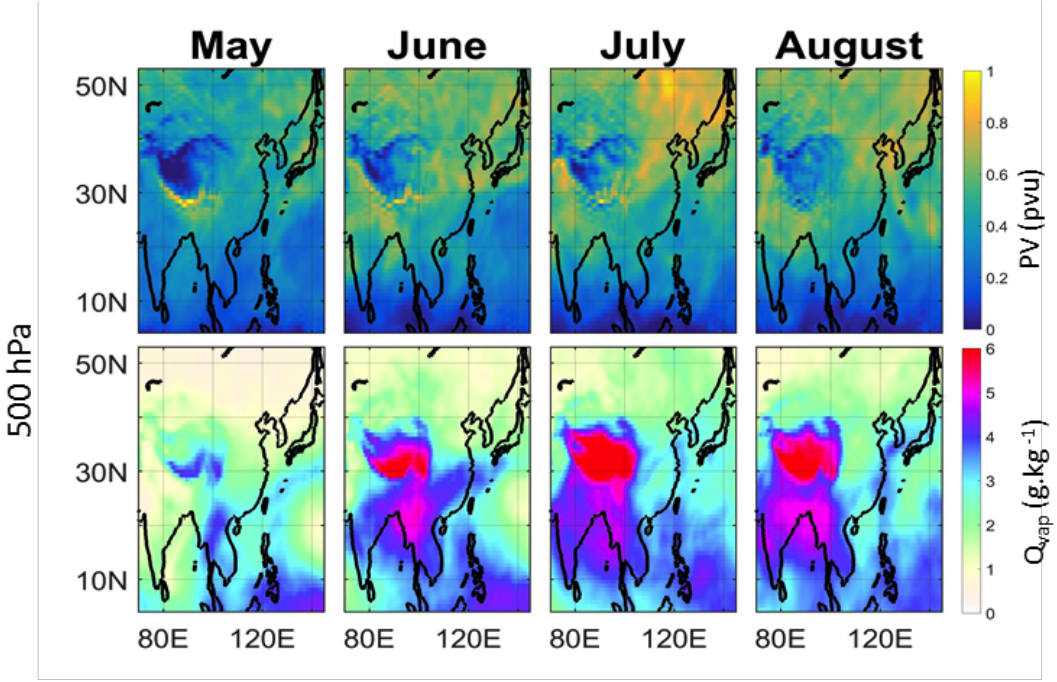

Fig. 5. Fig A1. PV and Qvap at 500 hPa

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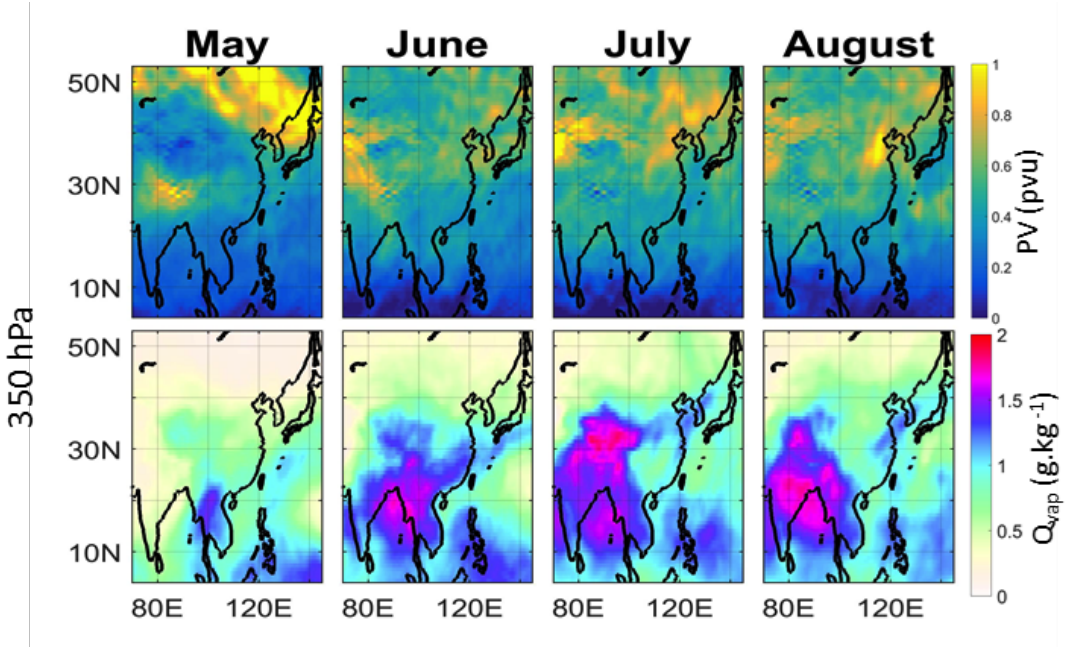

Fig. 6. Fig A2. PV and Qvap at 350 hPa

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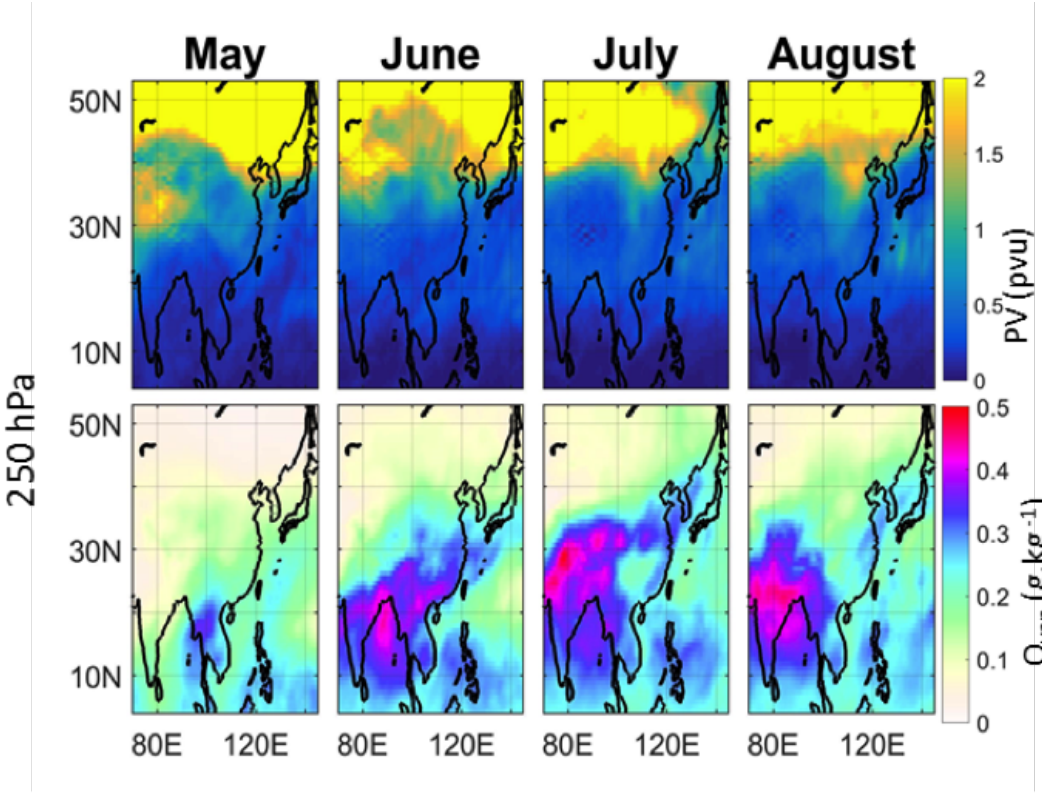

Fig. 7. Fig A3. PV and Qvap at 250 hPa

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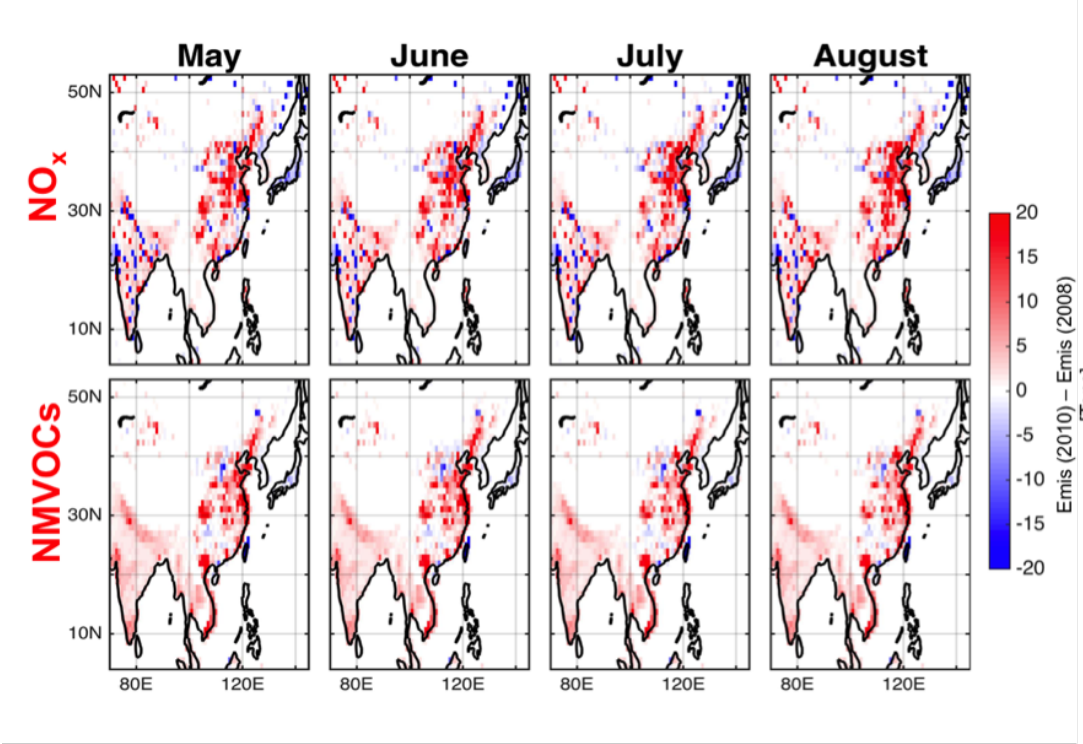

Fig. 8. Fig S1. Change (in tons specie) between 2010 and 2008 of NOx (upper panel) and NMVOCs (lower panel) emissions as derived from EDGAR-HTAP.

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