# Springtime variability of lower tropospheric ozone over Eastern Asia: contributions of cyclonic activity and pollution as observed from space with IASI. Dufour et al.

The authors thank the referees for their interest in the article and their suggestions for improvements.
The comments are addressed below. The referee's comments are indicated in italics and the reply to each comment is given just below. Quoted text from the revised manuscript is given in blue. Following the recommendations of referee #2, major changes have been made in the manuscript, it is then difficult to report all the changes in details here without providing the full text. We then edit the changes done in the revised manuscript only when they remain sufficiently short to be inserted here. A full revised manuscript will be provided soon. An English native speaker should edit the revised version.

**Reply to Referee #1:**

*Specific comments 1:*
*"1. Validation of IASI against ozonesonde Ozonesonde at Tateno (Tsukuba) are launched in the early afternoon (specifically 14:30LT). Tsukuba site is located in sub-urban area (appox. 50 km away from Tokyo), and there are typically diurnal variations observed, with a maximum in the early afternoon and a minimum during the night due to photochemical build-up and titration by NO, respectively. This means that ozone data by sondes is recorded when lower trop. ozone (say, boundary layer ozone) is at the maximum during the day, resulting in overestimates by sondes versus other methods like UV absorption at the surface sites, when we simply compare daily or monthly means. I think this would be the main cause for the difference between sonde and IASI at Tateno/Tsukuba. I am not familiar with other sites including Beijing, Hong Kong, Naha, and Sapporo, but if the sondes are launched in sub-urban area, we can expect the same bias. In fact, as the authors noted at the footnote of in Table 1, most sonde observations are made in the early afternoon, hence they had to relax time-matching criteria from 6 hr to 24 hr. This can allow the authors to increase the number of matched (concomitant) data, but this does not necessarily contribute to statistical robustness nor improve the validation of IASI against sondes, due to differences in local-time sampling between satellite and sondes, along with substantial diurnal variations at Tsukuba. I would suggest the authors to stick to narrow time band, say 6 hr (by the way, is this +/- 6 hours, correct?) for the sites where diurnal cycles are presumably negligible (say, Naha, Sapporo). This might result in a number that is different from currently estimated (-2DU, -9%) for lower trop. ozone in East Asia.*

*Also, please specify if "correction factor" is used for ozonesonde in this comparison, and mention the database used here (WOUDC?).*

*In spite of higher sensitivity of IASI to ozone in the upper atmosphere, the biases for UTLS and STRATO are larger than for LT and TROPO, while correlation coefficients are excellent (0.95). What is the explanation of this?"*

Reply:

We follow the recommendation of the referee and keep the 6 hr criterion as coincidence criterion for all the stations including Asian stations. We redo the validation analysis with this criterion for the Asian stations. The number of coincidence is less but similar results are obtained with a negative bias of 2.2 DU and a correlation coefficient of 0.70. Considering each station individually shows a similar large negative bias (2.6 DU) for Beijing, Hong Kong and Tateno whereas the bias is weak for Sapporo (+0.8 DU). We thank the referee to stress the implication of afternoon observation in suburban area like Tateno. The mean time difference between IASI and ozonesondes is ~ +5 hours (IASI measuring in the morning). The difference for Beijing, Hong Kong and Tateno are then explained by this time difference and the build-up of ozone in suburban areas. We include this discussion in the paper as follow: "A significant bias of 2.2 DU (9.5%) with IASI underestimating ozone partial columns is determined. The bias is similar for Beijing, Hong Kong, and Tateno (-2.6 DU) and different for Sapporo (+0.8 DU). Most of the ozonesonde measurements are performed in the early afternoon. The ozone build-up is then maximal in polluted urban or sub-urban sites like Beijing, Hong Kong, and Tateno. IASI observations are performed in the morning, about 5 hours earlier on average. The time difference between IASI and ozonesonde osbervations in polluted suburban sites may partly explain the larger bias in this case. Indeed, the bias for the Sapporo region where the diurnal cycle of ozone is limited is reduced. However, the small number of coincidences does not allow one to firmly conclude on the origin of the observed bias over East Asia."

The WOUDC and the SHADOZ databases have been used to collect the validation dataset except for Aquila and Beijing. This is now mentioned in the corrected version of the paper.
As we focus the validation on the lower troposphere, we do not consider correction factor for the ozonesondes. Applying standard correction factor for tropospheric purposes is usually not recommended (see Chap. 11, p437 of the EUROTRAC report, Tropospheric Ozone Research, Oystein Hov (Editor), Springer, 1997).

Concerning the larger biases for UTLS and STRATO, they are similar to those derived from the previous version of the ozone product and reported in Dufour et al., 2012. The bias in the UTLS has been extensively discussed in this paper. Spectroscopic issues and the limited vertical resolution of IASI, which limits the capability of IASI to retrieve the strong vertical gradient of ozone, mainly explain the biases. Very recently, Boynard et al. (poster presented at the last ESA ATMOS2015 conference, June 2015) show that applying the 2012 version of HITRAN reduces the bias on the total columns (existing when UV and IR sounders are compared). This bias is mainly driven by the stratospheric part of the column. As we focus on the lower troposphere and as the validation results are not significantly different from those discussed in Dufour et al., 2012, we decided not to include the results for the other partial columns and to display the validation results for individual Asian stations instead of all the partial columns.

*Specific comments 2:*
*"2. O3-CO correlation as seen with IASI*
*In Abstract, the authors mention that they found significant correlation between lower tropospheric ozone and carbon monoxide, especially over North China Plain (NCP), and this O3-CO correlation indicates that the photochemical production of ozone from primary pollutants emitted over such large polluted regions. Later in Page 9217, they mention that the correlation (coefficient?) is 0.6 over NCP for one specific day, 5 May 2008 (Later in Page*

*9224, the correlation is said as 0.62). The fact that the authors found correlation is good, but more important factor is the slope of O3/CO – i.e., relative enhancement of O3 to CO, as this ratio can suggest the degree of photochemical O3 production per precursor emitted in a given season of the year. For example, in East Asia, Tanimoto et al. (2008) paper (Tanimoto et al., Diagnosing recent CO emissions and ozone evolution in East Asia using coordinated surface observations, adjoint inverse modeling, and MOPITT satellite data, Atmos. Chem. Phys., 8, 3867-3880, 2008) showed that the O3/CO ratios can vary from 0 to 0.3 as a result of photochemical evolution of the air masses transported from Asian continent. It would be interesting to describe how IASI can see the O3/CO ratios over NCP (and downwind area) and quantitatively discuss the O3/CO ratios in comparison to those in previous papers"*

Reply:
We agree with the referee that looking at the O3/CO ratio would be interesting. At the beginning, we felt reticent about deriving such ratio from IASI. Indeed, the ozone and CO used in the paper are integrated columns on different altitude ranges. The lower tropospheric (LT) columns of ozone are integrated from the surface up to 6 km whereas the CO columns correspond to the total columns. If we retrieve the ozone profile by ourselves, the CO columns used are those available from the Ether atmospheric database (www.pole-ether.fr). Only the columns are distributed (not the profiles). However, assuming that most of the CO enhancement observed from the total CO columns occurs in the lower troposphere, we then calculated the mean mixing ratio corresponding to the LT ozone columns and to the total CO columns to estimate the O3/CO ratios. We computed this ratio only for specific regions (NCP + regions where transport of ozone and CO is observed on 13 and 14 May 2008). The estimated ratios are in the range 0-0.3 given by Tanimoto et al. 2008. During the transported event mentioned in Section 5.4 of the paper, the ratio increases from 0.16 over NCP on 12 May to 0.28 over the Sea of Japan on 14 May, indicating the chemical processing of the transported air masses.
We have then included the following discussion at the end of Section 5.4 of the revised version of the paper: "To complete the study, we calculate the enhancement ratio of O3 to CO over NCP on 12 May, over Yellow Sea and Korea on 13 May, and over the Sea of Japan on 14 May. The ratios are respectively 0.16, 0.21 and 0.28. The increase of the ratio indicates possible photochemical processing during the transport. Part of the large lower tropospheric ozone is then due to the transport of ozone produced over NCP but also to ozone produced during the transport."
We also include the following discussion at the end of Section 4.2: "To evaluate the degree of photochemical production of ozone, we calculate the equivalent or mean mixing ratio corresponding to the CO and LT O3 columns. This allows us to estimate a relative enhancement ratio of O3 to CO of 0.14 and 0.08 on 5 and 6 May respectively. These values are in agreement with the typical values ranging between 0 and 0.3 reported over East Asia by Tanimoto et al. (2008). The estimated enhancement ratio remains quite low suggesting an early stage of ozone production."
The abstract and the conclusion have been updated accordingly.

*Specific comments 3:*
*3. Some other relevant work*
*P9297, LL27: Wang et al. 2009 paper does not really examine Asian monsoon effect on trop. ozone seasonality, but rather look at long-term trend. Please check. Other relevant references that are missing but dealing with monsoon effect on ozone seasonality (with a focus on spring) in East Asia is: Tanimoto et al. (2005), Significant latitudinal gradient in the surface*

*ozone spring maximum over East Asia, Geophys. Res. Lett., 32, L21805, doi:10.1029/2005GL023514.*
*There is also a paper looking at synoptic-scale transport of air pollutants in East Asia, that the authors might be interested in and add values when discussed in the paper. Miyazaki et al. (2003), Synoptic-scale transport of reactive nitrogen over the western Pacific in spring, J. Geophy. Res., 108(D20), 8788, doi:10.1029/2002JD003248.*

Reply:
Indeed, there was an error in the use of the reference Wang et al., 2009. It has been corrected and references to Miyazaki et al. 2003 and Tanimoto et al 2005 have been added in relevant sections of the introduction.

*Technical comments*

*"Title: "contributions" of cyclonic activity and pollution. I found it a bit unfitted to what is discussed in the paper, since "cyclonic activity" is a meteorological factor and "pollution" is a (sort of) source. Perhaps the "role" is better? Or do the authors mean "cyclonic activity on pollution transport"? Anyway it needs to be modified to be clearer."*
Reply: the title has been changed as follows: "Springtime daily variations of lower tropospheric ozone over East Asia: role of cyclonic activity and pollution as observed from space with IASI"

*"Figure 1 can be omitted since it is not very important and there are as many as 15 figures.*"
Reply: We removed this figure.

English corrections suggested by the referee have been done.

**Reply to Referee #2:**

*Major comment 1*
*"In Section 4 and 5, the authors tried to describe the evolution and change of vertical distribution from one synoptic process to the other. There are too many details but lack of key points, which make the paper a bad readability. So I suggest the authors restructure the two sections into two parts. The first part could demonstrate that the IASI satellite data could well-capture the vertical structure of O3 and CO based on correlation analysis with PV or other available data. The secondary part could explain the transport mechanisms for different processes, such as stratospheric intrusion and warm conveyor belt associated with cyclones. The authors should clearly demonstrate what are the unique advantages in using IASI satellite data in understand these processes, and whether there are anything new or anything disagreeing with the existing understanding?"*

Reply:
We agree that Sections 4 and 5 are somehow confusing and that the key points are not well underlined. Thanks to the recommendations of the referee, we decided to restructure the paper in order to better state the objectives of the paper and make a better use of the IASI observations.
One objective of the paper is to propose a way to use IASI O3 and CO observations to identify the different sources (UTLS reservoir and photochemical production) of the ozone enhancements in the lower troposphere. The second objective is to evaluate the stratospheric

and the photochemical sources of O3 on the daily variations of LT ozone distribution over East Asia using IASI. The main outline of the paper is then to use the first case study to elaborate the analysis method and to apply this method to analyze the second case study. Section 4 is now mainly dedicated to the description of the method based on the first case study and shows (we hope) the potentiality of IASI. We detailed how to use the different partial columns and profiles of ozone to determine (i) the regions under the influence of the tropopause perturbations associated to cyclones, (ii) the regions where STEs occur, (iii) the role of the photochemical production to explain ozone enhancement. We also follow the recommendations of the referee to exploit better the vertical capabilities of IASI (see reply to comment #2). In Section 5, we use this method to discuss the different processing leading to ozone enhancements in the LT with the same sub-sections than previously.
This restructuration of the paper allows us to avoid most of the repetitions, which were in the previous version of the manuscript and also allows us to simplify and reduce the number of figures leading to a better readability (we hope).
To illustrate the new guideline of the paper, we provide here the revised conclusion:
"Based on ozone and CO retrieval from IASI, we elaborate an analysis method to diagnose which processes contribute to ozone enhancement in the lower troposphere. We apply the method to evaluate the respective role of the stratospheric and the photochemical sources of ozone on the day-to-day variation of the lower tropospheric ozone distribution over East Asia. The study allows us to stress how satellite observations can help in monitoring and identifying these different sources. We focus on late springtime because the cyclonic activity – well known to drive the stratosphere-troposphere exchanges – is important and the photochemical production of ozone in polluted area can be significant at this time of the year.
We demonstrate that ozone profiles and semi-independent ozone columns between the surface and 12 km associated with simultaneous CO measurements from IASI provide a powerful observational dataset to identify the stratospheric and anthropogenic origin of lower tropospheric ozone. We show that UT ozone columns larger than 40 DU are a proxy to identify the region of subsiding ozone associated to the tropopause perturbation induced by low-pressure weather systems. Combined with LT ozone columns larger of ~30 DU, it identifies the areas in the lower troposphere affected by the UTLS reservoir of ozone. One of the advantages of IASI is to provide 3 dimensional observations of ozone distribution at synoptic scale when cloud free. The analysis of vertical section in longitude or latitude allows one to identify more precisely the areas where the lower troposphere is connected to the UTLS reservoir and the region of possible irreversible stratosphere-troposphere exchanges. On the contrary, we show that large LT ozone columns when not associated with large UT ozone columns but with enhanced CO total columns – used as pollution tracers – indicates the areas where the photochemical production of ozone takes part of the observed ozone enhancement in the lower troposphere. Once again, the 3D observational capability of IASI (vertical sections) allows one to evaluate if the ozone enhancement observed in the LT is disconnected from the UTLS reservoir and thus to assess the anthropogenic origin of the LT ozone enhancement or the mixing of the sources. We also show that enhancement ratio of O3 to CO, consistent with those from literature, can be derived from IASI.
As expected, the succession of low- and high-pressure systems strongly influences the day-to-day variations of lower tropospheric ozone over North East Asia during springtime, both leading to LT ozone enhancements. We show that the ozone subsiding transfer due to the tropopause perturbations associated with the low-pressure systems affect the free and lower tropospheric ozone over large regions. We determine the region of influence of such system, located mainly above 40°N but with some particular intense events (cut-off low from 11 to 13 May 2008) impacting southern regions such as NCP for few days. The vertical dimension provided by IASI allows the identification of the STE areas, which are located in the southern

part behind the cold front in case of frontal system and in the southern or southeastern flank of the low in the case of a cut-off low. Note that the STE are expected to occur preferentially in the western and southern flank of the trough.

Based on the case of a cut-off low travelling over NCP from 11 to 14 May 2008, we show that such systems, with potential convective capacity, when they travel over highly polluted regions, play a key role in the transboundary transport of pollutants. We identify from the O3/CO enhancement ratio estimated from IASI observations that significant ozone photochemical production occurs during the transport from NCP on 12 May to Sea of Japan on 14 May.

In addition to the stratospheric influence on tropospheric ozone in the northern part of the domain, most of the enhanced lower tropospheric ozone columns are observed in regions mainly impacted by strong pollution level. Significant correlations between CO (used as a pollution tracer) and ozone in the lower troposphere have been found. Moreover, the analysis of vertical sections of ozone concentrations over NCP indicates that ozone concentrations are enhanced only in the lower troposphere in such regions, indicating the anthropogenic origin of the observed ozone enhancements. The maximal values of ozone are observed between 2 and 4 km in the cases where an anticyclonic situation is well settled over NCP (e.g. 5 and 15 May 2008). This is in agreement with in situ measurements (Huang et al., 2014), considering the limited vertical resolution of IASI and its limited sensitivity to surface ozone. Because of these limitations, it is not possible to determine more precisely the altitude of the ozone enhancements in the troposphere. This is all the more penalizing when stratospheric and photochemical events occur at the same time. The lack of vertical resolution does not allow one to separate the different contributions. Combined with modelling studies, advanced satellite products coupling UV and IR information such as the recent IASI+GOME-2 product (Cuesta et al., 2013) as well as the next generation of satellite instruments (Crevoisier et al., 2014, Veefkind et al., 2012) should help to address this issue."

*Major comment 2*

*"The vertical sections show some useful information about the structure, which is in fact one of the main unique advantages of satellite data. The authors showed some results in Figure 5 and Figure 11. However, the both figures were show along specific latitudes, and they should also show results along specific longitudes. In fact, most of works related to stratospheric instructions prefer to a figure along a specific longitude (e.g. Ding and Wang, 2006), which could demonstrate the tropopause folding more clearly."*

Reply:

Following the recommendation of the referee, we now provide vertical sections in latitude and in longitude to describe the different events and processes discussed in the paper. We use these vertical sections to evaluate when and where the lower troposphere is connected to the UTLS ozone reservoir and then to identify the areas of possible STE. The vertical distribution of ozone provided by IASI also permits us to identify when ozone enhancements is mainly due to photochemical production from emitted pollutant. We show for example that the ozone distribution retrieved from IASI over NCP during anticyclonic situations shows a maximum between 2 and 4 km in relatively good agreement with in situ observations.

*Minor comment 1*

*"1. The word of "variability" in the title: the paper was not really talk about "variability" but some specific processes. "Variability" is a term related to climatology. The current title*

*will mislead the readers that the cyclonic activity influenced the variability of springtime ozone from year to year."*

Reply: The term "variability" has been changed to "variation" in the title and elsewhere in the text.

*Minor comment 2*
*"2. P9205, L5-7, P9209, L5-8: the authors pointed out "May is typically the largest tropospheric ozone along the year". This is not true in East Asia. For example, ozone peaks in June in the NCP and western China and peaks in October in South China (Wang et al., 2009; Ding et al., 2008; Zhu et al., 2004). The reason of selecting May could be that late spring is one of the season having rather high ozone concentration and frequent cyclone/front activities."*

Reply: We agree that this statement is misleading. It reads now: "In a previous study, Dufour et al. (2010) show that IASI lower tropospheric ozone columns reach a maximum in late spring, early summer (May, June) in Beijing, Shanghai and Hong Kong. We then decided to focus our study on late spring (May), season for which high ozone concentrations and frequent frontal activities occur over East Asia."

*Minor comment 3*
*"3) Figure 2 and Figure 3: the figures are composed with many small figures. The figure captions are different from the label shows in the figures. For example, (a-c) vs. (a-i) in Figure 2. (a-d) vs. (a-l) in Figure 3. Please make corrections to these figures or capitations, and also check the text."*

Reply: The mistake in the figure captions has been corrected and the text checked.

*Minor comment 4*
*"4) A latest work by Ding K. et al. (2015) discussed similar processes using MOZAIC aircraft measurement and MOPITT CO data. Please make a comparison with that paper in the discussion part."*

Reply: A reference to Ding et al. 2015 has been added in the introduction and in section 5.4