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Atmospheric Chemistry and Physics Discussions

Interactive comment on "IASI measurements of tropospheric ozone over Chinese megacities: Beijing, Shanghai, and Hong Kong" *by* G. Dufour et al.

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The authors thank the referee for his/her interest in the article and his/her suggestions for improvements. The comments are addressed below.

General Comment

"However, concerning the explanation and scientific interpretation of the ozone observations the paper remains speculative and just scratches the surface. Particularly the separation and discussion of upper tropospheric ozone values seems to be critical without a discussion of the tropopause altitude given the averaging kernels of the instrument in the upper troposphere. Lower tropopauses lead to a stronger influence of

stratospheric ozone considering the strong ozone gradient at the tropopause and the width of the kernel. Even if the main intention of the authors is to present the potential of a relatively new observational data set, they should support their hypotheses with meteorological informations. Particularly for the discussion of stratospheric influence this is important. Since the degrees of freedom in the troposphere are typically smaller than 2, observations of upper tropospheric ozone can be artificially enhanced due to a broad averaging kernel. As presented here, I doubt that the evaluation and discussion of upper tropospheric cycles makes sense without additional data. Therefore I recommend to the paper for publication after the following points have been adressed."

Reply to the general comment

The authors agree that the ACPD version of the paper was somewhat speculative. Indeed, the first aim was to present the potential of a new satellite instrument (IASI) for probing ozone in the lower part of the troposphere at the regional scale of megacities as well as to show that with the corresponding data set, even not perfect, realistic hypothesis can be elaborate. Nevertheless, the authors agree with the referee that the quality of the paper gains adding external information to support the different hypotheses. This has been done in the revised version of the manuscript and some details are given in the responses. The authors thank the referee for his/her suggestions.

Comment #1

"p.8: Vertical cross sections and seasonal cycles (Fig.5 and 6,8,10): The authors show vertical cross sections of ozone (Fig.5) and conclude that phase shift of the cycles at different altitudes in the troposphere can be resolved by the observations. Of course the cycles around 3 km and between 7-10 km are different, but I doubt that these differences are of tropospheric origin. A close inspection of Figure 5 and the upper cycles (Figs 6,8,10: black curves) reveal, that the cycles seem to be related to the location of the tropopause (taking the strongest change of color gradient from orange to yellow in Figure 5 as proxy for the ozonopause). Due to the kernels as shown in

Figure 2 the upper tropospheric values can be affected by stratospheric ozone even if no transport, mixing etc. takes place. Therefore the discussion of sources of ozone for the upper cycles is at least ambiguous. I'm not convinced that the upper cycles can be undoubtfully used to conclude on ozone sources between 6-10 km without additional informations (e.g. other tracers like CO, meteorological data, long-range transport and trajectories etc.). This could be easily tested, when including the respective tropopause altitudes (e.g. PV, thermal tropopause from analysis data) into Figure 5. Further: Why are the 'upper' ozone cycles in Figs. 6,8,10 are taken at different altitudes (8,10, and 12 km, respectively)? Why are occurrences of maxima of the the Beijing ozone cycles (Fig. 6) different from Ding et al., 2008 (i.e. what is special in 2008 compared to the 1995-2005 period in Ding et al.?)"

Reply #1

Concerning Figure 5, the authors estimated the monthly tropopause heights using the NCEP meteorological analysed fields for each city. The tropopause height is usually larger than the upper limit of the graph for each city. For Beijing, the tropopause height is slightly lower than 11 km (between 10 and 11 km) for the months of January, February, March and December and reaches values up to 15 km in July. For Shanghai, the tropopause height exceeds 12 km (about 15 km), except in February and March (slightly lower than 12 km). For Hong Kong, the tropopause height is almost constant all over the year and is about 16 km. The authors agree that the broad averaging kernels can lead to a "stratospheric contamination" of the upper tropospheric retrieved levels, especially during winter when the tropopause is low. The difference noticed in the paper between the lower and upper/free tropospheric levels over Beijing and attributed to transport from the lowest layers arise during late spring, early summer, when the tropopause height is high. The contribution of the stratosphere is less important in these conditions. In the cases of Shanghai and Hong Kong, the tropopause height is much higher and the contribution of the stratosphere is small. However, the aim of the paper was to focus mainly on the ability of IASI to provide insights on interesting

features in the lower troposphere. The argument about the separation of the lower and upper troposphere was essentially made to support the fact that the features observed in the lower troposphere (during spring and summer) could be unambiguously attributed to tropospheric sources. The primary aim of the paper was not to put accent on upper tropospheric. In the revised version of the paper, efforts have been done to (i) better explain the performances of the retrieval in terms of averaging kernels and degrees of freedom and the ability of discriminating 2 pieces of information from the lower troposphere and the upper troposphere (lower stratosphere). Due to the lack of vertical resolution, the authors decided not to present the profiles (Fig. 5) and individual levels (Fig 6, 8, 10) but only partial columns. The remark of the referee has been accounted for in order to better assess when the different partial columns are or not contaminated by the stratosphere. Concerning the different altitudes of the 'upper' ozone cycles in Fig. 6, 8, 10, this choice arise from the averaging kernels (Fig. 2). Due to the difference in the tropopause between the midlatitudes and the tropics, the regularization used for the retrieval is different. The information content in the observation is not dramatically different, only the distribution of the information during the retrievals is slightly different. This leads to a different altitude for the separation of the lower and upper tropospheric part that can be retrieved (Fig 2).

Concerning the occurrences of the maxima of the ozone cycles over Beijing compared to Ding et al. 2008, the lower cycle from IASI has to be compared to the cycle corresponding to the altitude between 2 and 4 km from MOZAIC according to the averaging kernel. If one accounts for the errors in the IASI and MOZAIC measurements, the 2 cycles are in good agreement. For the upper cycle, the comparison with the 6-12 km partial columns (as done in the revised version of the paper) is in better agreement than for individual levels. A broader summer maximum is observed.

Comment #2

"p.13 and discussion of Figures 12,13,15: The authors try to explain the ozone changes at the lower layers over the three megacities with variations of circulation and winds.

It is speculated about wind directions at different levels at the different locations, but no further evidence for these hyptheses is given. It would be very easy to overlay the daily wind fields from e.g. ECMWF over the maps of regional variations of ozone at the three locations. This would give the speculations much more meteorological evidence and therefore confidence in the ozone observations. In particular the daily maps (Figs. 12,13, 15) would benefit from a discussion of the wind, since it is suggested that the observed variations in the PBL are partly related to different air masses (e.g. in April)."

Reply #2

The authors thank the referee for his/her suggestion. An analysis of the wind fields have been added in the revised version and this greatly helps for the discussion and support the hypotheses done in the ACPD version of the paper, especially when larger ozone amounts are observed during spring (April, May).

Comment #3

"Similarly, the variability of the daily ozone profiles, which is linked to downward transport from the stratosphere is performed without additional information. The authors suggest, that 'descent of stratospheric air' has influenced the tropospheric ozone signal. What is meant by this statement? Do the authors suggest downward mixing (in the sense of irreversible exchange)? A transient tropopause fold, associated with a low tropopause, could also cause high ozone columns, but not necessarily tracer exchange across the tropopause. Since the degrees of freedom are less than 2, how does this affect situations with a low tropopause? I would expect the stratospheric ozone signal affecting tropospheric column values stronger than during high tropopause conditions. To increase the credibility of the observations the authors could very easily support their hypothesis with maps or crossections of PV and ozone and the related temporal evolution of the synoptic situation. Further: What is the role of tropospheric long range transport from other sources (e.g. fires) or convection with regard to the daily vertical profiles?"

Reply #3

Backtrajectory calculations with a starting point at 6 km have been performed for this period. They show that for the first 2 days of the period selected in February the air masses come from the North West and from higher altitude (about 2 to 3 km higher), whereas the air masses come from the West and for lower altitudes for the other days. Moreover, the tropopause height (from NCEP) is more than 1 km lower for these 2 first days ($\sim$ 8 km against 9.5 km for the following days). This would support intrusion of stratospheric air that influences the 0-6 km columns retrieval. In addition it is worth noting that during winter the averaging kernels have their maximum higher (around 7 km) and are more sensitive to stratospheric influence (this is explained in the revised version of the manuscript).

Note that the backtrajectories over the whole year have been performed for the 3 cities and results of this calculation have been used to evaluate which latitudes were affecting mainly the domain considered for each city. This allows simplifying Fig. 4 for a better readability. Backtrajectories are added as supplementary material. Concerning the last point, this is an interesting question. Dedicated studies with model calculations would be necessary to clearly answer these questions. This is behind the scope of the current paper that is to provide an overview of the ability of IASI for probing lower tropospheric ozone at the regional scale of megacities.

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