Interactive comment on "Acetylene (C2H2) and hydrogen cyanide (HCN) from IASI satellite observations: global distributions, validation, and comparison with model" by V. Duflot et al.

Response to Anonymous Referee #1

The manuscript by Duflot et al. reports on global distributions of total column acetylene and hydrogen cyanide obtained from IASI for a three-year period. The dataset is ompared with ground-based FTIR spectrometer measurements at four sites and further with simulation results from the chemical transport model MOZART. The paper is well written and structured, the assumptions made are generally clearly stated and related work is adequately referenced. I consider that this paper constitutes a valuable addition to the literature on these atmospheric trace species, usefully complementing previous published studies. I therefore recommend publication in ACP.

The authors would like to thank the reviewer for reading the manuscript and suggesting changes which have helped us to improve it. The responses to individual comments are developed here below:

- The references given in section 3.3.1 (pages 14374–5) do not appear to explicitly describe how the anthropogenic source (fossil fuel and biofuel) of HCN that was used in the model simulation was estimated. Given that the HCN emissions are currently poorly constrained, it would be of particular interest, from the perspective of future modelling studies that could potentially try to build upon previous work in order to improve our understanding of the atmospheric behaviour of this species, to have the anthropogenic HCN source used in this specific case properly documented. The same would apply for the oceanic sink.

The emission dataset used in this study is now better described in Section 3.3.1 to address this comment:

"The surface anthropogenic (including fossil fuel and biofuel) emissions used here were taken from the inventory provided by D. Streets and University of Iowa and created for the ARCTAS campaign (see http://bio.cgrer.uiowa.edu/arctas/emission.html for more information). This inventory was developed in the frame of the POLARCAT Model Intercomparison Program (POLMIP) and is a composite dataset of regional emissions as representative of current emissions as possible: it is built upon the INTEX-B Asia inventory (Zhang et al.,2009) with the US NEI (National Emission Inventory) 2002 and CAC 2005 for North America and the EMEP (European Monitoring and Evaluation Programme) 2006 for Europe inventory to make up NH emissions (see Emmons et al. 2015 for an evaluation of POLMIP models). Emissions from EDGAR (Emissions Database for Global Atmospheric Research) were used for missing regions and species. Since only total volatile organic compounds (VOCs) were provided with this POLMIP inventory, the VOC speciation based on the RETRO emissions inventory as in Lamarque et al. (2010) was used. The anthropogenic emissions are constant in time with no monthly variations.

Daily biomass burning emissions were taken from the global Fire INventory from NCAR (FINN) version 1 (Wiedinmyer et al., 2010). The fire emissions for individual fires, based on daily MODIS fire counts, were calculated and then gridded to the simulation resolution (Wiedinmyer et al., 2006, 2010). The oceanic emissions are taken from the POET emissions dataset (Granier et al., 2005) and the biogenic emissions from MEGANv2 dataset inventory (Guenther et al., 2006)."

Note also that Viatte et al. (ACP, 2015) which analyses HCN and C_2H_2 in fire plumes over the Arctic using MOZART-4 with the same anthropogenic emissions as that used here is now referred in Section 3.3.1.

[C. Viatte, K. Strong, J. Hannigan, E. Nussbaumer, L. K. Emmons, S. Conway, C. Paton-Walsh, J. Hartley, J. Benmergui, and J. Lin: Identifying fire plumes in the Arctic with tropospheric FTIR measurements and transport models, Atmos. Chem. Phys., 15, 2227–2246, 2015, doi:10.5194/acp-15-2227-2015.]

- The "standard chemical mechanism" of Emmons et al. (2010) (cited on page 14374, line 19) does not include HCN chemistry. An updated reference or a short description of the HCN chemistry implemented in the model version used in the manuscript would be welcome.

We thank the referee for pointing this out. The MOZART-4 tropospheric chemistry scheme is exactly the same as that used in the CAM-chem model which has been fully described in Lamarque et al. (GMD, 2012) including HCN and C_2H_2 chemistry. This reference is now mentioned in Section 3.3.1.

[Lamarque, J.-F., Emmons, L. K., Hess, P. G., Kinnison, D. E.,Tilmes, S., Vitt, F., Heald, C. L., Holland, E. A., Lauritzen, P. H.,Neu, J., Orlando, J. J., Rasch, P. J., and Tyndall, G. K.: CAMchem: description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, Geosci. Model Dev., 5, 369–411, doi: 10.5194/gmd-5-369-2012, 2012.]

- Model output is sensitive to the injection height of biomass burning emissions. How was the issue of distributing these emissions in the vertical addressed in the simulation described in the manuscript?

In the MOZART-4 simulation used here, the fire emissions are injected at the surface, which might indeed result in an underestimation of concentrations at high altitudes and explained the bias observed when comparing with IASI which shows an increased sensitivity in the middle-upper troposphere. This has been now specifically mentionned in the section 3.3.2.

Note, however, that the convection in MOZART is strong enough to transport emissions out of the boundary layer. It has been previously reported for CO that no significant differences between standard simulation and simulation performed with an injection height between the surface and 6 km were observed (Pfister et al., GRL 2005 and Tilmes et al., ACPD 2011).

[Pfister, G., Hess, P. G., Emmons, L. K., Lamarque, J.-F., Wiedinmyer, C., Edwards, D. P., Petron, G., Gille, J. C., and Sachse, G. W., Quantifying CO emissions from the 2004 alaskan wildfires using MOPITT CO data, Geophys. Res. Lett., 32, L11809, doi:10.1029/2005GL0229952005.5946]

- Is it possible to elaborate in what way "the modeled species lifetime could be improved to simulate the impact of the long range transport for these species" (page 14376, line 26; page 14378, line 16)?

The reasons for explaining the biases observed between observations and model outputs are now better developed. The sentence is replaced by:

"However, the low background concentrations in the Southern Hemisphere as simulated by the model, especially for Southern Africa and Australia is possibly due to a mix of uncertainties introduced by the coarse grid of the model producing too much diffusion and problems in the transportation scheme for fine-scale plumes, the fire injection set at the surface and uncertainties in the emissions. The fact that only three representative jacobians are used to perform the global comparison might also play a role."

- The temporal span (three years) of the dataset presented in this paper is relevant information and should probably be mentioned in the abstract.

This is now mentioned in the abstract:

"We present global distributions of C_2H_2 and HCN total columns derived from the first 3 years of the Infrared Atmospheric Sounding Interferometer (IASI) measurements."

- Corrections:
• Page 14359, line 15: Rather than stating that ". . .the model seems to overestimate [. . .] emissions" it would be accurate to say that ". . .the emissions used in the model seem to be overestimated. . ." Same in Conclusions, page 14378, line 14.
• Page 14367, lines 20 & 25: Change ". . .measurements is. . ." to ". . .measurements are. . ."

These corrections have been included in the manuscript.

Response to Anonymous Referee #2

General Comment: This is a well-written, clearly structured article, documenting the fast retrieval and evaluation of HCN and C2H2. I recommend publication after the following (minor) comments have been addressed.

The authors would like to thank the reviewer for reading the manuscript and suggesting changes which have helped us to improve it. The responses to individual comments are developed here below:

- Abstract, lines 8 and 14: Please include the range of correlations coefficients found for the agreement.

The suggested changes have been made in the Abstract (lines 8 and 14).

- Intro, line 19-20: Please include some information on atmospheric levels (background, urban,biomass burning) from literature.

The suggested changes have been made in the Intro (lines 21-23) for background and biomass burning levels (no reference has been found for urban levels).

- Intro: Think including an extra table for C2H2 and HCN summarizing their budgets (sources/sinks etc) would be useful to the reader & improve paper.

The suggested changes have been made in the Intro (lines 21-23) and an extra table has been added (Table 1).

- Page 12363, line 11: Why were the million spectra only chosen from 2009? Surely a sample selected from 2008-2010 would have been better? Were all these observations cloud free? If not, how does the HRI vary with different amounts of cloud cover?

The goal here was to have a set of spectra wide enough to generate a globally representative total measurement error covariance matrix. The choice of the year has no impact on the results. The randomly chosen spectra are cloud free spectra; this has been specified in the text (page 14363 line 11).

- Page 12363, line 24: Here I assume you mean minimal interference from CO2, H2O and O3? Sentence read ok, but could be phrased slightly better.

No, the goal here is not to minimize the interferences from the interfering species: we need to have clear signatures from them to calculate the total measurement error covariance matrix and maximize the contrast with the spectral background. This is why the selected ranges include signatures from the interfering species.

- Page 14364, line 15: Please add the units of B_HCN and B_C2H2.

The units have been added in the text (Page 14364, line 15).

- Page 14364, line 20: Where the C2H2 and HCN profiles taken from (i.e. before the 1 km perturbations were applied)? Were they from a model, climatology, or aircraft observations? Page 14365, line 2: what are 'standard absorption profiles'? Please clarify.

To take into account all possible cases of concentration value and altitude of the polluted layer, we artificially built up the C2H2 and HCN profiles, without using any model, observations or climatology. Then, to compute the corresponding spectrum for each of these artificial profiles, we used the forward model of the line by line radiative transfer model Atmosphit together with absorption profiles coming from standard modeled atmospheres (as stated page 14365, lines 6-8) for the other species. This has been clarified in the text (page 14365, line 2).

- Page 14365, line 13: Please add the units of b_HCN and b_C2H2.

The units have been added in the text (Page 14365, line 13).

- Page 14366, line 6-11: Think some discussion of the Jacobians with respect the HCN and C2H2 vertical distribution is warranted, and/or also include or add to the figure some HCN and C2H2 profiles. I would like to see where the HCN and C2H2 is.

HCN and C2H2 vertical distributions in a standard temperate atmosphere are now shown in Figure 3, and discussed with respect to the Jacobians (Page 14366, line 6-20).

- Page 14366, line 14-18: There is no mention of cloud cover/contamination. Is it important? Please add some brief discussion.

We consider only cloud free spectra here. It has been stated in the text (Page 14366, line 14-18).

Page 14366, line 23: Why was a 1.5 km altitude chosen? What is the justification of this level? If a level of 1 km, 2km or 3 km were chosen how would this impact the sensitivity analysis?

In general thermal contrast can be defined as the temperature difference between the surface and the air temperature at some altitude of interest. We chose here to consider the same definition as in Van Damme et al. 2014. This has been added in the text (Page 14366, line 23-25).

- Page 14367, line 18: Sorry, why was a 30% confidence level chosen? Please explain in more detail.

This 30% confidence level for subtropical atmospheres was first chosen as a prudent assumption (considering that subtropical atmospheres share properties with both tropical and temperate atmospheres, and that the confidence in the independence of the retrieval method to the atmospheric parameters for a 0.26 10^16 molecules/cm^2 stability threshold is 10% for a tropical atmosphere and 40% for a temperate atmosphere). We decided to perform simulations in subtropical atmospheres to check our first guess. It results in a 25% confidence level for a 0.28 10^16 molecules/cm^2 stability threshold in subtropical atmospheres. It has been added/corrected in text (but not shown in Figure 5).

- Section 3.1: How do the IASI/FTIR comparisons change if the 0% cloud contamination is used, or if the co-location limits are shrunk or expanded?

IASI spectra are usually considered to be exploitable with a cloud contamination fraction up to 25% (Clerbaux et al., 2009 ; Van Damme et al., 2014). As we deal in this study with species with quiet weak spectral signatures, we chose here a prudent 10% cloud contamination fraction threshold to avoid interferences in the considered spectral ranges. Using a 0% cloud contamination threshold would not change significantly the comparison, but would only decrease the number of spectra useful for it.
These (usual) co-location limits (1°x1°) were chosen as they allow to have a significant number of collocated spectra together with a reasonable representativeness of the ground-based measurement point.

- Also why were these four sites selected, it was not clear in the text (but I assume because they only measured these target gases)?

Indeed, these four sites were selected because they were the only ones to measure the target species during the studied period (to our knowledge). This has been added in the text.

- Page 14369, line 23 and onward: How does the peak-to-peak accuracy of the measurements compare (i.e. how well does IASI actually capture the timing and magnitude of peak events)? Please add this statistic, as it is important given HCN and C2H2 biomass burning sources.

The way IASI capture the timing of the peak events can be evaluated considering Figure 8 and the given correlation coefficients.
The magnitudes of the peak events are now given in the text for both space and ground-based observations (Section 3.1).

- Page 14370, line 3-6: Could the FTIR averaging kernels be applied to the IASI data? Or could the IASI Jacobians be applied to the FTIR data (as done later for the model comparison)?

Unfortunately not. One should always apply either the averaging kernels of the less sensitive instrument on the data measured by the more sensitive one, or a combination of the two averaging kernels on both data sets. In the case of the comparison FTIR vs IASI, one should then apply IASI averaging kernels on FTIR measurements (IASI being obviously less sensitive than the ground-based instruments).
Regarding the Jacobians, they only can be applied on profiles to take into account IASI sensitivity. It was then possible to apply them on model outputs (profiles), but not on FTIR measurements (total columns).

- Section 3.2: Nicely summarised!

Thanks !

- Section 3.3: The key point for me here (which is not discussed) is how well does the model agree with the FTIR data? I think a sub-section addressing this point needs to be added to the paper (& probably an extra figure).

This is right. A subsection (3.3.2) as well as a new figure (Figure 14) have been added to the article to address this point.

- Figure 9: There are some C2H2 enhancements over the China coast in DJF and MAM, simulated by the model that are not observed by IASI. I don't think this was raised in the text. Can you elaborate?

This is right. Discussion has been added in the text on this topic (Page 14377, lines 5-15).