Response to referee 1

Interactive comment on “Carbon monoxide climatology derived from the trajectory mapping of global MOZAIC-IAGOS data” by M. Osman et al.

Anonymous Referee #1

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Reviewer’s comments on: "Carbon monoxide climatology derived from the trajectory mapping of global MOZAIC-IAGOS data" by M. Osman, et al.

General Comments

This manuscript by Osman et al. reports a novel use of MOZAIC/IAGOS (or ’M/I’) measurements to construct a global, 3D, time-varying climatology for CO. The method relies on the HYSPLIT trajectory model and NCAR/NCEP reanalysis wind fields to ’project’ measured CO concentrations to regions and altitudes lacking actual measurements.

The resulting climatology is evaluated first by comparing CO climatology maps generated with forward and backward trajectories separately. The method is then validated by comparing M/I CO statistics at a number of airports with trajectory-mapped climatologies calculated after withholding M/I measurements at each of those airports.

Finally, the CO climatology is compared with MOPITT satellite results, where MOPITT Level 3 CO values have been re-gridded to a 5-by-5 degree grid, matching the resolution of the trajectory-mapped climatology.

While the general method described in the manuscript is novel, the significance of the new climatology product is very unclear. The manuscript includes very little in the way of an error analysis that would permit an understanding of the limitations of the climatology. This analysis should be presented separately from the validation of the method. At a minimum, such an analysis should include (1) a clear description of the underlying assumptions of the method, and the impact of these assumptions on the accuracy of the climatology, and (2) a statistical analysis of the ’robustness’ of the climatology (based on the variability of the CO values that are averaged together in each ’bin’). An important assumption that is only vaguely mentioned in the manuscript is that measurements of CO near source regions (e.g., urban regions surrounding airports) are directly useful for estimating CO concentrations large distances both upwind and downwind of the airport. This ’airport effect’ would seem to result in a significant positive bias in the trajectory-mapped CO values. A statistical analysis of the variability of the trajectory-mapped CO values is necessary to distinguish regions where the CO climatology is ’statistically robust’ from those regions where the uncertainty is very large.

Following the reviewer's suggestion, sample plots of number of samples per grid cell and the standard error of the mean associated with the trajectory-mapping have been added. These are available for each month/year/decade and level in the climatology. We have added text discussing these results as well. We thank the reviewer for drawing our attention to this oversight.

The manuscript does not include a true validation section, which would involve comparing the trajectory-mapped CO climatology with an existing product with known error characteristics. The presented methods for evaluating the climatology are mostly qualitative.

These methods have some value but are rather indirect and inconclusive. For example, since all airports are located near major urban centers, the biasing effects of urban sources of CO may well be similar at different airports. So, the trajectory-mapped climatology may generally represent CO profiles near urban regions better than in rural areas far from sources. If so, the experiments in which trajectory-mapped climatologies are calculated after withholding observations at one airport are still not indicative of the accuracy of the climatology away from urban regions. A more conclusive validation would be based on independent in-situ data from aircraft deployed during some field campaign, where the issue of local sources was known to be unimportant.

The effects of urban sources of CO are indeed observed in the boundary layer. This is an important point that should have been better emphasized and we thank the reviewer for drawing it to our attention. However, the urban effect decreases above the boundary layer since the aircraft travels up to 200-400 km before reaching its cruise altitude, which considerably reduces the effect of urban sources of CO. The following figure shows the comparison between NOAA in-situ data (Novelli et al., 1992; Lang et al., 1992; and Conway et al., 1994; Emmons et al.,2004; Emmons et al., 2009; Deeter et al., 2010) and the trajectory-mapped CO climatology. Individual NOAA CO measurements were compared with the monthly mean trajectory-mapped CO climatology, or if the monthly mean was unavailable, the seasonal mean climatology for that particular location. The total number of NOAA aircraft field campaign CO profiles from for the period 2001-2012 is 1940.

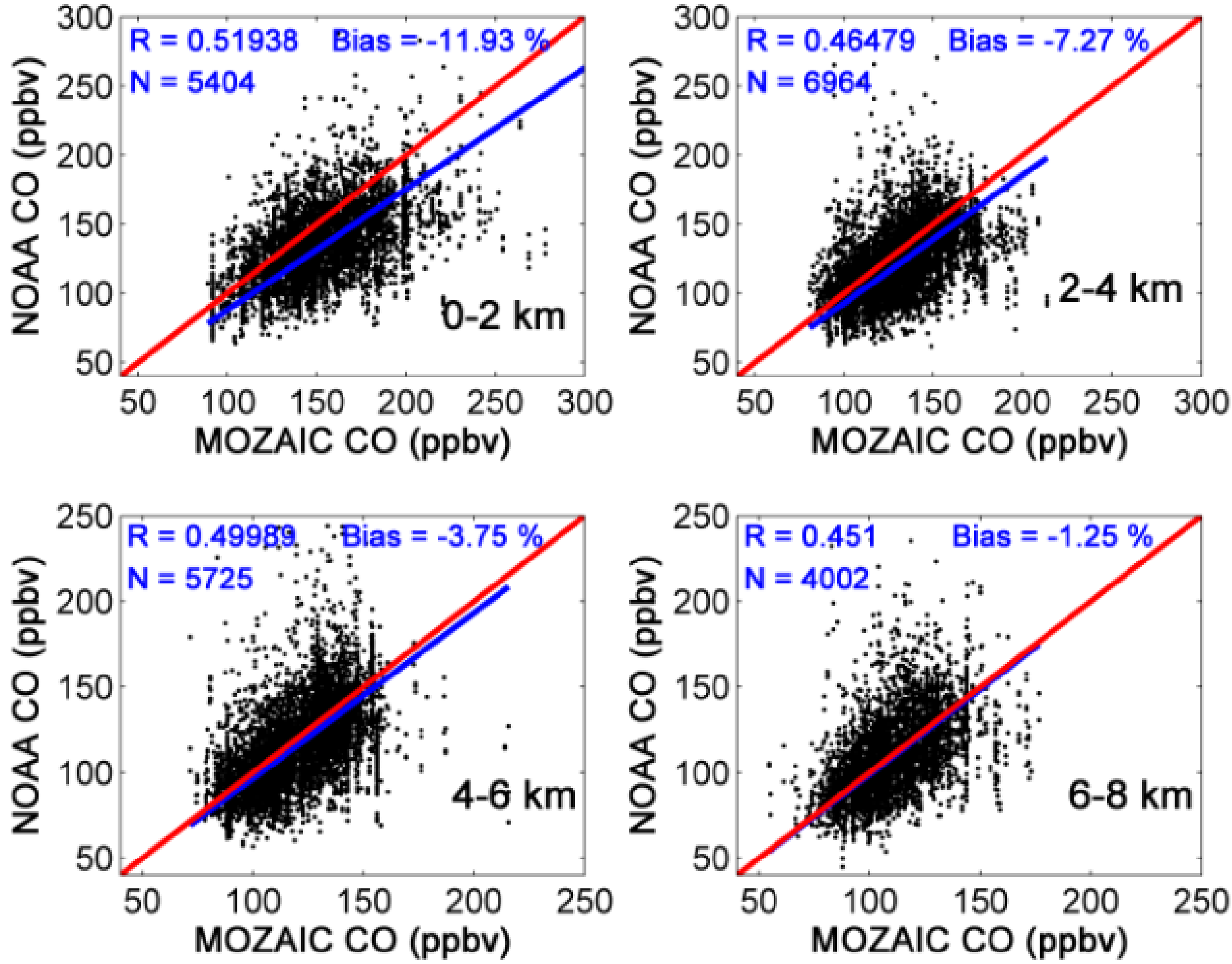
The text has been revised to discuss these points.

Added references:

Conway, T . J., P. P. Tans, L . S. Waterman K, . W. Thoning D, . R. Kitzis, K. A. Masarie, and N. Zhang, Evidence for interannual variability of the carbon cycle from the NOAA/CMDL global air sampling network, J. Geophys. Res.,99, 22,831-22,855, 1994.

Lang, P.M., L. P. Steele, L. S. Waterman, R. C. Martin, K. A. Masarie, and E. J. Dlugokencky NO AA/CMDL atmospheric methane data for the period1 983-1990 from shipboard flask sampling NOAA Tech. Memo., ERL CMDL-4, 88 pp., 1992.

Novelli, P .C., L. P. Steele, and P. P. Tans, Mixing ratios of carbon monoxide in the troposphere. ,J. Geophys. Res.,9 7, 20,731-20,750, 1992.



Comparison between NOAA flask profiles and the monthly trajectory-mapped CO climatology for the period from 2001-2012. MOZAIC CO measurements are biased high in the boundary layer as CO values near airports are typically above background.

The comparisons of the trajectory-mapped climatology with MOPITT Level 3 products re-gridded at 5-degree resolution have only qualitative value due to several effects. First, the trajectory-mapped CO values are based on in-situ measurements near urban regions (esp. in the lower troposphere), whereas MOPITT Level 3 products at 5-degree resolution represent a mix of urban and mostly rural atmospheric conditions. This effect would very likely result in a positive bias in the trajectory-mapped climatology. Second, because of the significant variability of MOPITT retrieval performance with respect to various geophysical parameters (surface type, CO loading, thermal contrast), the validity of MOPITT averaging kernels averaged over large regions is unclear. All previous published MOPITT validation papers have exploited MOPITT Level 2 data averaged over much smaller regions.

Major Revisions

1. Most readers will find this paper unreasonably long and it should therefore be shortened. I believe the paper should be split into a 'methodology and validation' paper (based on Sections 1-4) and a separate 'analysis' paper (based on Sections 5 and 6). Given that some parts of the methodology need to be lengthened (see below), reducing the paper's length is even more important.

Following the reviewer's comments, the O_3-CO subsection (6.2) has been removed from the paper in order to reduce its size. Other reductions have been made as well.

2. The paper should include a section specifically addressing errors in the trajectory mapped CO climatology product. In addition to other sources of potential error (e.g., chemistry and trajectory errors), this quantitative error analysis should estimate the magnitude of systematic errors due to the fact that the M/I CO measurements in the lower-troposphere (e.g., 800 hPa to 1000 hPa) are likely biased towards CO concentrations observed over urban regions near airports. The error analysis should also provide information on the statistical robustness of the climatology product in relation to the variability of the trajectory-mapped CO values which are averaged together in each climatology bin. This statistical analysis should then be used to assess the expected geographical variability of the uncertainties of the trajectory-mapped CO climatology.

As suggested by the reviewer, we added plots and discussion of the standard errors of the mean and number of sample per grid point associated with the trajectory mapping. These maps are available on the FTP site for each month/year/decade and level in the climatology. We have added some discussion of these to the text. We have also added discussion of the "airport effect".

Standard errors associated with the trajectory mapping:

Figure X2 shows the standard errors associated with trajectory mapping and the number of samples per grid cell for monthly, seasonal and annual averages at 4.5 km. Similar figures for other levels are included with the climatology on the FTP site. The largest number of samples per grid cell and the lowest standard errors are found over North America and Europe as there are more frequent MOZAIC-IAGOS aircraft flights in this region. The standard error is computed using all data points found inside a grid cell. This is probably biased somewhat low since some grid cells may contain more than one value from a particular trajectory. This bias is likely not more than a factor of 2, based on typical trajectory lengths.

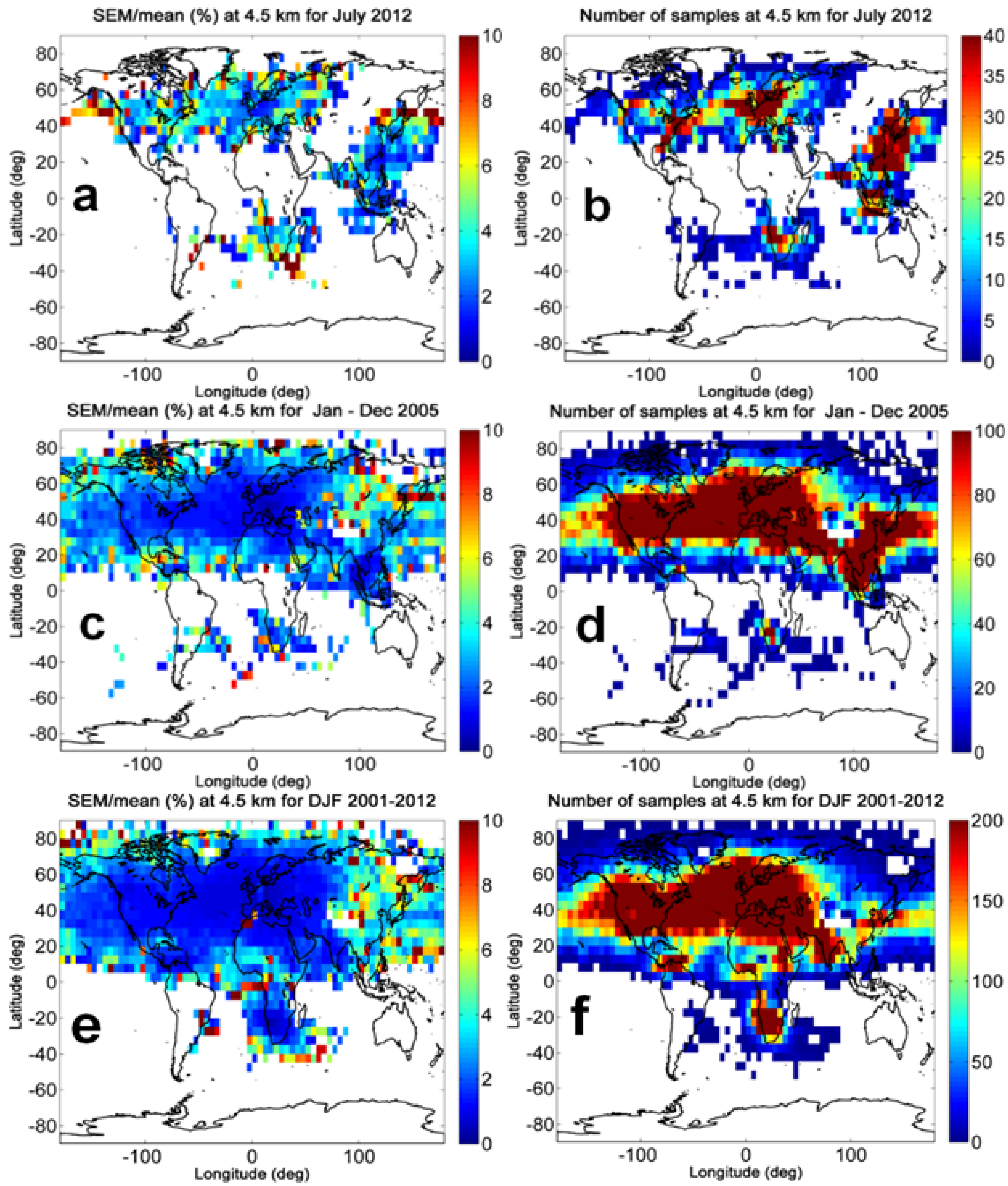


Fig. X2. The standard error of the mean (left panels) and number of samples (right panels) for monthly (July 2012), annual (2005), seasonal (DJF 2001-2012) means at 4.5 km altitude above sea level. The month and year shown are chosen as typical; other months and years show similar patterns. The data are binned on a $5^o \times 5^o$ latitude and longitude grid.

3. The purpose of the comparisons with MOPITT Level 3 products is unclear. Is the intent to use MOPITT products to validate the MOZAIC-based CO climatology, or is it the other way around? Statements on pages 18, 19, and 20 suggest that the trajectory mapped CO climatology reveals biases in the MOPITT V6 product, despite the fact that this product has already been thoroughly validated against in-situ data obtained from NOAA aircraft and the HIPPO field campaign. True validation involves comparisons of a new product with an established product with known error characteristics.

In this case, it seems that the error characteristics of the MOPITT product are better understood and better quantified than the errors associated with the trajectory-mapped climatology. Thus, it seems more reasonable to conclude that the MOPITT V6 product reveals biases in the trajectory-mapped CO climatology (than the other way around).

The main purpose of the comparison of the trajectory mapped CO climatology with MOPITT is simply to show that trajectory mapping produces global maps that look reasonable, and that the mapping product agrees fairly well with MOPITT even far from airports. The difference at higher altitudes is surprising, however, and we felt that it was important to note this in the paper. Since it is not our intent to validate MOPITT, we merely show, by showing that we get similar differences with unmapped MOZAIC data (using Frankfurt as an example), that the differences are not due somehow to the mapping procedure. This, of course, we considered unlikely beforehand, as trajectory errors are found to be typically random; we have observed no biases in our work to date. Such biases would have to stem from the NCEP meteorological reanalyses used as input, which have themselves been extensively validated.

4. The comparisons with MOPITT total column values (Section 4.2) appear to be based on incorrect assumptions regarding the MOPITT layering scheme, and should be repeated and reanalyzed. Specifically, the assumed layer boundaries (at the midpoints between the MOPITT retrieval levels) do not agree with the layering scheme discussed in Deeter et al., (2013) and in the MOPITT V5 User’s Guide.

Indeed. Corrected, and description corrected in the text.

Minor Revisions and Technical Corrections

p. 2, l. 16 - ’comparison’ should be ’comparing’

Done.

p. 2, l. 22 - Are the results really conclusive that MOPITT is biased, or are the authors really just stating that there is some bias between MOPITT and the trajectory-mapped climatology?

No, it is not our intent to validate MOPITT. We have rephrased the text to make our conclusions less definitive. We do see a puzzling difference at higher altitudes, but that could of course be some unknown problem with the aircraft measurements at lower pressures.

p. 2, Abstract - Should be some brief statement about the limitations of the climatology, e.g. primarily for Northern Hemisphere.

We have noted this, and also the urban effect in the boundary layer. In areas where there are few profiles or even none, most of the information comes through relatively long range transport with associated larger uncertainties.

p. 5, l. 9 - The meaning of the sentence "Background CO levels are found ..." is unclear.

We replaced the sentence in p. 5 l. 9-10 by:

Long range atmospheric transport redistributes CO widely due to its relatively long lifetime. Typical tropospheric background CO levels range between 50 and 120 ppbv (WHO, 2000).

We've added the reference:

WHO, Air quality guidelines for Europe, 2nd ed. Copenhagen, World Health Organization Regional Office for Europe, 2000 (WHO Regional Publications, European Series No. 91), 2000

p. 5, l. 13 - There is insufficient evidence for this claim.

Replaced by: “CO values as high as 1800 ppbv have been reported over Beijing (Zbinden, et al., 2013).

p. 6, l. 25 - What are these ’obvious advantages’?

The trajectory approach takes into account known atmospheric motions from the NCEP meteorological wind fields. The advantage over linear or quadratic interpolation is that it uses additional information about the atmosphere, and therefore can be expected to give a better estimate of the redistribution of CO. Using linear or quadratic interpolation is the default best estimate in the absence of such knowledge. Text revised.

p. 6, l. 28 - This statement is premature, since no data have yet been presented in the manuscript.

The trajectory-mapped MOZAIC-IAGOS CO climatology is publicly available at ftp://es-ee.tor.ec.gc.ca/pub/ftpdt/MOZAIC_output_CO/. We removed the text that discusses about O3-CO correlations in p. 7 to shorten the manuscript.

p. 7, l. 13 - typo in ’transformation’

Corrected.

p. 7, l. 20 - There is no clear reason for dividing the MOPITT-related material into Sections 2.4 and 4; they should be combined in a single section.

We agree and have moved the first part of Section 4 as suggested.

p. 8, l. 21 - what fraction of the airports (or MOZAIC profiles) are located in the Southern Hemisphere?

Nearly 9 % of the airports are located in the SH and 6% of MOZAIC-IAGOS profiles are from the SH.

p. 9, l. 1 - This is incorrect; MOPITT retrievals do not rely on thermal contrast between the surface and atmosphere (but do rely on a temperature gradient within the atmosphere).

Corrected as p.9 l.1-2: "thermal contrast between the Earth's surface and atmosphere" replaced by "a temperature gradient within the atmosphere"

p. 9, l. 6 - Which MOPITT product is exploited in this paper, the TIR-only, NIR-only, or TIR/NIR?

Added p. 9, l. 1- added: TIR/NIR

p. 9, l. 19-26 - MOPITT validation results vary widely from one version to another; only V6 validation results should be listed here since results for other versions are irrelevant.

We agree and have removed the irrelevant sections as suggested by the reviewer. We thank the referee for identifying this.

p. 10, l. 4 - Why were the M/I cruise data not used?

The cruise data are less useful for our purposes since they are all near the tropopause. However, we have used them for validation (see Fig. 6, now Fig. 7).

p. 11, l. 3 - A thorough discussion of the potential effects of source regions on the trajectory-mapped climatology is needed here; It is not conclusive that qualitative comparisons of maps based separately on backward or forward trajectories prove that source regions have an insignificant effect.

The issue with sources is that while forward trajectories should redistribute the CO correctly, backward trajectories will (erroneously) map the CO to places upwind of where it was generated. We have expanded the text on this point.

p. 12, l. 19 - Add reference to Worden et al (Atmos. Meas. Tech., 6(7), 1633–1646, doi:10.5194/amt-6-1633-2013) regarding the variables which affect MOPITT averaging kernels.

Added:

Worden, H. M., Edwards, D. P., Deeter, M. N., Fu, D., Kulawik, S. S., Worden, J. R., and Arellano, A.: Averaging kernel prediction from atmospheric and surface state parameters based on multiple regression for nadir-viewing satellite measurements of carbon monoxide and ozone, Atmos. Meas. Tech., 6, 1633-1646, doi:10.5194/amt-6-1633-2013, 2013.

p. 13, l. 22 - Here the manuscript lacks important details. Exactly how were the CO concentrations at the 'missing' MOPITT levels (above the maximum MOZAIC aircraft altitude) determined? How many levels in the vertical grid are actually affected by this?

Note that the described strategy of using MOPITT a priori profiles is inconsistent with methods used in MOPITT validation papers and might lead to unphysical discontinuities in the CO profile. Does the chosen method of filling in these high levels affect the results?

If the top of the monthly mean trajectory-mapped MOZAIC-IAGOS CO vertical profile (that is interpolated to MOPITT pressure levels) is below 100 hPa, we use the corresponding MOPITT monthly a priori profile for the missing CO data above the highest trajectory-mapped altitude. However, most of the trajectory-mapped CO profiles reach up to altitude 200-100 hPa, which is about 14 km above sea level. Thus for extending the trajectory-mapped MOZAIC-IAGOS CO profiles to the highest MOPITT level, we have utilized the MOPITT a priori profile mostly for 100 hPa and sometimes for 200 hPa. The same procedure has been used in previous work. de Laate et al. (2012) have shown robust results by using MOPITT a priori profiles for the missing MOZAIC-IAGOS in situ measurements above the maximum MOZAIC-IAGOS aircraft altitude. The authors found the errors as a result of using the MOPITT a priori profile for the missing MOZAIC-IAGOS in situ measurements above the maximum MOZAIC-IAGOS aircraft altitude to be about 5 % or less. Since the maximum altitude of the top of the trajectory-mapped data (i.e., ~14 km) is greater than the maximum altitude reached by the MOZAIC-IAGOS aircraft (i.e., ~12 km), the error in our comparison can be expected to be smaller yet.

p. 14, l. 17 - Why exclude airports in the Southern Hemisphere from the validation study?

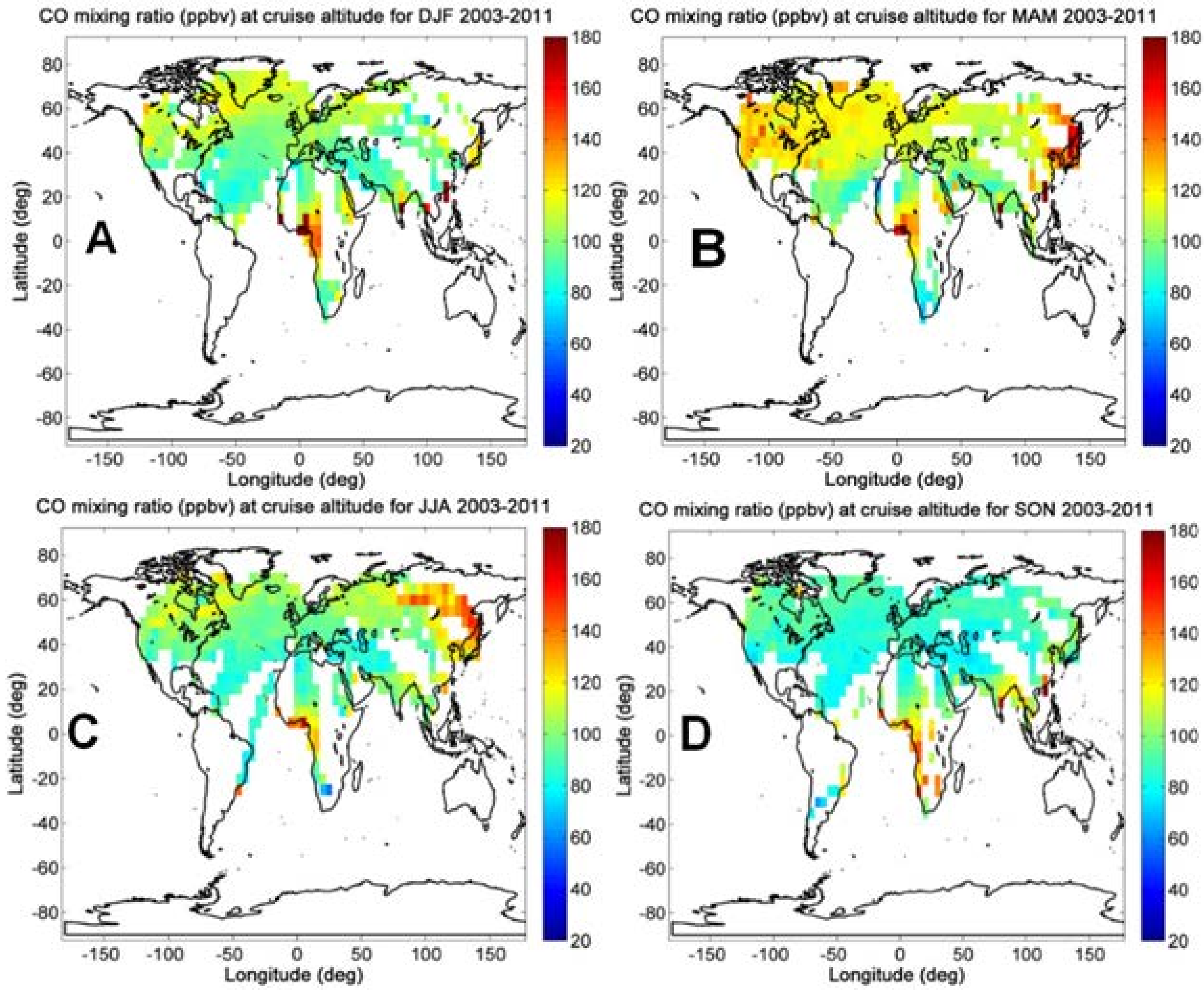
Unfortunately, there are limited MOZAIC-IAGOS data in the SH. Consequently, if an airport in the SH excluded from the trajectory-mapped calculation for the validation, most of the information comes via relatively long trajectories, which are associated with larger uncertainties, and the comparison becomes uncertain as well.

p. 15, l. 25 - All of the listed airports are located in the Northern Hemisphere. Should include several from the Southern Hemisphere, where the climatology might be more challenged.

Please see above.

p. 16, l. 15 - For readers' convenience, online M/I CO maps should be reproduced in the manuscript (with permission) to compare against Fig. 6.

The following figure is reproduced and added in the manuscript (now shown in Fig. 7).



p. 17, l. 22 - Both the shapes and magnitudes (or areas) of the averaging kernels are significant.

Done. In p. 17, l. 22 - shapes replaced by shapes and magnitudes (or areas)

p. 17, l. 7 - It is unclear if the analysis of Fig. 2 is included just as an example, or if it supposedly illustrates overall bias in MOPITT. Any conclusions about MOPITT retrieval bias should compare findings to MOPITT validation papers; possible reasons for any discrepancies in validation results (relative to published results) should be discussed.

Figure 2 (now Figure 3) is included as an illustration and it reveals the discrepancy between trajectory-mapped and MOPITT CO. Possible reasons are noted.

p. 18, l. 10 - Which MOPITT product was used: TIR-only, NIR-only or TIR/NIR?

TIR/NIR

p. 18, l. 10 - Emphasize that standard MOPITT L3 retrievals have been regridded to 5-degree resolution for this analysis.

Added in p18, l 11 : The standard MOPITT V6L3 retrievals have been regridded to 5-degree resolution for this analysis.

p. 18, l. 27 - Are the authors suggesting that the trajectory-mapped CO climatology can be used to validate MOPITT, or are the MOPITT data being used to validate the MOZAIC CO climatology? This sentence observes that there is a difference between the two products ("reveals significant biases between MOPITT and the trajectory-mapped ..."), while the first sentence in the next paragraph ("MOPITT seems to underestimate ...") suggests that the trajectory-mapped CO climatology can be used to determine biases in MOPITT products.

No, it is not our intent to validate MOPITT. We have rephrased the text to make our conclusions less definitive. We do see a puzzling difference at higher altitudes, but that could of course be some unknown problem with the aircraft measurements at lower pressures.

p. 19, l. 5 - This is incorrect; all previous MOPITT validation work was performed with MOPITT Level 2 products, not Level 3 products. This may be related to the significant discrepancies in the validation results reported in this manuscript compared to previously
Corrected. We thank the referee for identifying the error.

p. 19, l.9 - It is not true that all published MOPITT validation results have been based on NOAA flask sampling.

Corrected.

p. 19, l. 10-19 - Comparisons of MOPITT L3 data (w/ 5-degree resolution) and M/I profiles for one airport (Frankfurt) do not provide convincing evidence of a general negative bias in the MOPITT retrievals, especially given that MOPITT V6 validation results have been previously reported for a large number of NOAA sites and for the HIPPO field campaign (Deeter et al., 2014).

The comparison of MOZAIC-IAGOS in situ profiles at Frankfurt against MOPITT is intended only to show that the differences are not due somehow to the mapping procedure, since we get similar differences with unmapped MOZAIC data (using Frankfurt as an example),

p. 19, l. 12 - It is unclear if these comparisons are based on standard MOPITT L3 data, or the regridded 5-degree resolution L3 data.

Added: the comparison is based on the regridded 5-degree resolution MOPITT V6L3 data

p. 20, l. 12 - It is unclear why MOPITT retrievals in the lower troposphere would only yield a lower-bound (although it is true that such retrievals are often highly constrained by the a priori).

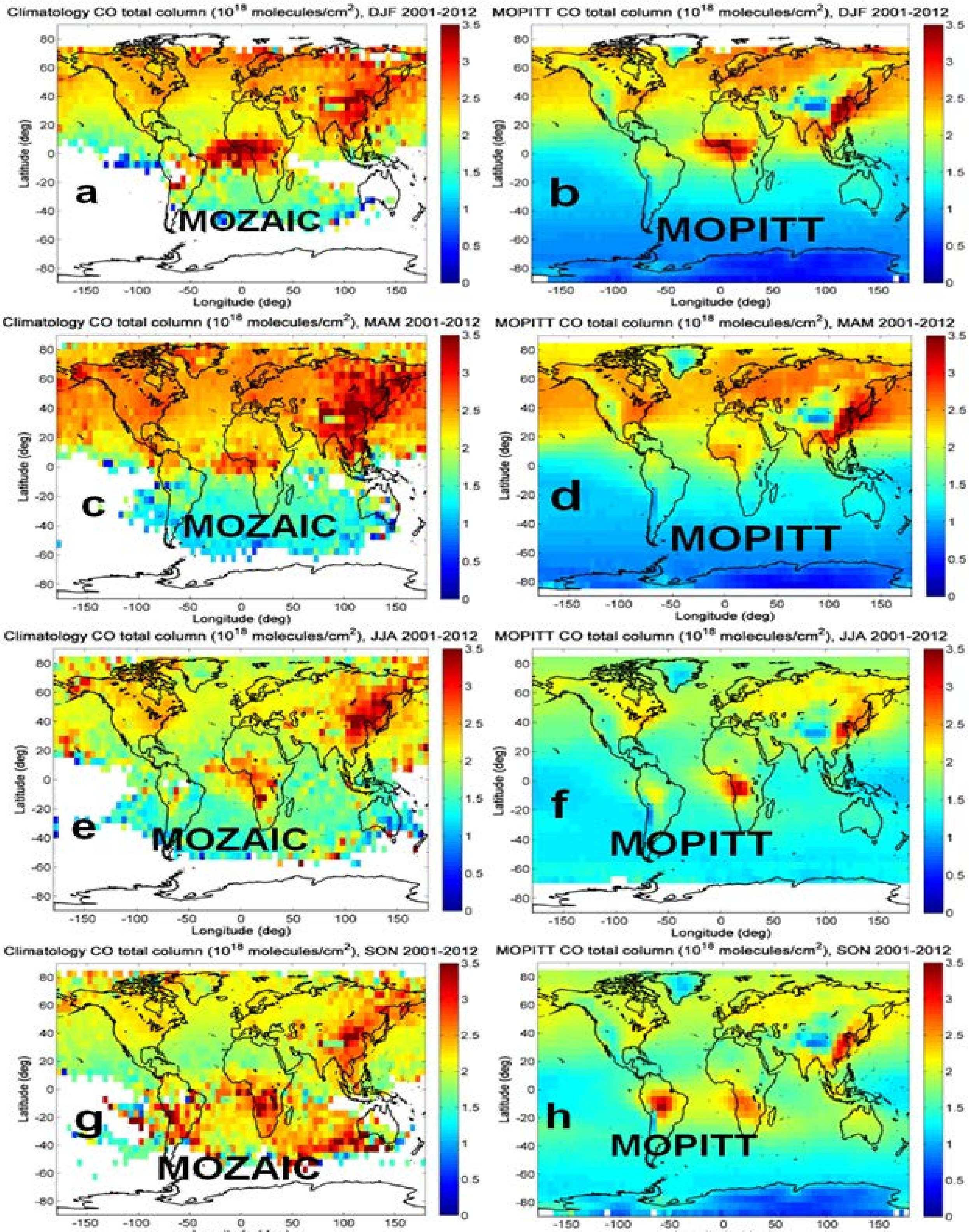
We were citing an observation by Liu et al. (2005), who noted that MOPITT has low sensitivity to CO in the lower troposphere. However, we have removed those remarks in the current version.

p. 21 - References are needed for the text and equations presented in Section 4.2.

added p21. l6:: Deeter 2002 and Emmons et al. (2004)

added p21. l13: Deeter 2002 and Emmons et al. (2004)

added p22 l5: Deeter 2002 and Emmons et al. (2004)



August 2002 to DJF, December 2005 to MAM, December 2011 to JJA and August 2012 to SON 2001-2012. Total column values correcting the MOPITT layering scheme.

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Deeter, M. N. (2009), MOPITT (Measurements of Pollution in the Troposphere) Validated Version 4 Product User's Guide, National Center for Atmospheric Research. Available at http://web3.acd.ucar.edu/mopitt/v4_users_guide_val.pdf.

Deeter, M. N. (2011), MOPITT (Measurements of Pollution in the Troposphere) Validated Version 5 Product User's Guide, National Center for Atmospheric Research. Available at http://www.acom.ucar.edu/mopitt/v5_users_guide_beta.pdf

Deeter, M. N. (2013), MOPITT (Measurements of Pollution in the Troposphere) Validated Version 6Product User's Guide, National Center for Atmospheric Research. Available at https://www2.acom.ucar.edu/sites/default/files/mopitt/v6_users_guide_201309.pdf

Deeter, M. N. (2002), Calculation and Application of MOPITT (Measurements of Pollution in the Troposphere) Averaging Kernels, Available at http://www.acom.ucar.edu/mopitt/data/avg_krnls_app.pdf

We thank the referee for his/her careful review and very helpful remarks.