Atmos. Chem. Phys. Discuss., 12, C11978–C11993, 2013
www.atmos-chem-phys-discuss.net/12/C11978/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Top-down estimation of carbon monoxide emissions from the Mexico Megacity based on FTIR measurements from ground and space" *by* W. Stremme et al.

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Received and published: 25 January 2013

119781

Response to Referee comments on: "Top-down estimation of carbon monoxide emissions from the Mexico Megacity based on FTIR measurements from ground and space"

25 January 2013

1 General comments

Referee comments are given in black. Answers are given in blue.

1.1 Referee 1

The authors describe a method of combining several years of ground-based total column measurements and IASI space-based column measurements to determine a top-down CO emissions estimate for the Mexico City metropolitan area (MCMA). This analysis takes the spatial and temporal heterogeneity of the MCMA into account in a novel way. The top-down CO emissions estimate suggests that the bottom-up inventory is too low. This paper should be published after major revisions.

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1.2 Referee 2

This paper presents a novel top-down estimation strategy to estimate the CO flux over the Mexico City Metropolitan Area (MCMA). The growth rate of CO around noon, under low ventilation, is calculated from measurements at the UNAM campus. This growth rate allows the derivation of an average surface emission flux at the site. Background CO in the basin is determined from measurements in Tecamac, which is supposedly at the edge of the MCMA. Space based measurements of CO (from IASI) are used to reconstruct the extent of the spatial distribution of CO. The authors come up with a CO emission estimate suggesting that the official inventory may have underestimated the CO emissions from the Mexico Megacity for the year 2008. Prediction scenarios for the future indicate that more people will live in megacities. With the current sluggish departure from fossil fuel dependence, the contribution of megacities to global anthropogenic gas emissions will be even more significant. Therefore, scientific studies such as this paper are important in the effort to quantify the current and future effects of megacities on the Earth's atmosphere and climate. I recommend that this paper be published after the major comments of Reviewer 1 and a few minor comments have been addressed.

2 Major comments (Referee 1)

2.1 Introducing sentence

It would be very helpful if every section (especially subsections in section 2) began with a sentence or two describing the contents of the section and why the data processing step about to be described is important. It is easy to lose the thread of your argument throughout the paper.
Done. we included in all subsections of section 2 an introducing sentence:

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2.1 Ground measurements and retrieval of total columns: We add:
"In this subsection we present the observation sites, instruments, monitoring and retrieval strategies of the solar absorption ground measurements which are used in the following sections."

2.2 Column growth-rate and emission flux: we add:
"The section describes how the mean emission flux at UNAM can be reconstructed from continuous monitoring of the columns. "

2.3 Spatial distribution from satellite measurements:we add:
"In this section we describe a technique to reconstruct the mean CO distribution with high resolution based on satellite data. A high spatial resolution is necessary to account for the strong inhomogeneity in the MCMA and a key ingredient for the top-down estimation in Sect. 2.4."

2.4 Top-down estimation for annual CO emission in MCMA:we add:
"This section presents the top-down emission estimation combining the information provided by the prior sections."

3. Further we add to Section 3 Comparison and validation:
"In this section we compare the results obtained from the ground based with the satellite measurements (Sects.3.1, 3.2). In Sect.3.2 the results from the top-down emission estimation is compared with the bottom-up. The first validation proves a reliability and consistency of the column retrievals and that the concept using Averaging Kernels together with local information about the atmospheric structure works well. Sect.3.3 illustrates how the same results have to be compared to the bottom-up emission inventory and for which area the flux measurements at UNAM are representative. More technical details regarding this subject can be found in the supplement. "

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2.2 Time series analysis / growth rate

I am worried that seasonal cycle or long-term changes may complicate the slope calculated in Fig 3c. Could you address this by subtracting a daily mean (or a daily minimum, or 10am value) from the CO column signal each day to reduce the potential impact of any long-term trends or seasonal cycle changes?

We addressed this subject:

We realized the suggestion and subtract a daily mean for each measurement before calculating the slope. If we remove the mean value of each day from the particular measurements on that day before we determine a mean slope for all data, we find a slightly higher (3%) slope corresponding to $\approx$ +5 kg/(km^{2}h) additional emission flux and a smaller error ($\approx \pm$ 15 kg/(km^{2}h)) in comparison to the original error ($\approx \pm$ 20 kg/(km^{2}h)), errors according to the 95% confidence interval.

With other words: a) The change of the slope is small -> no significant change in the emission flux, but b) the change in the error of the slope is visible -> increased precision (random error). Both are expected as a) the random change of the offset CO column at each day should not systematically change the value of the mean slope. However b) the slope of a straight line is obviously less significant in a dispersed dataset. The error (95% confidence interval) changes slightly from $\approx 10\%$ error to $\approx 8.5\%$ error.

Actually we tried and tested various calculation strategies (similar to the suggested ones as obtaining a slope for each day, subtracting the mean value of the day), however we finally prefer to use the data set without subtracting estimated background values for this work, for the simple reason that the method should be as simple and transparent as possible and we are reaching the wanted precision of 10% anyway. There are some arguments against the subtraction of a mean value, as sometimes only few measurements are conducted at the beginning or end of the relevant time-interval and the measurements of these days do not allow for calculation of a representative mean value. So if we do a more sophisticated analysis to lower the "random error", we have

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to do a more sophisticated error analysis, estimating how the error of each daily mean value with more or less measurements might influence the retrieval.

Conclusion:
The referee is correct that a more sophistic time series analysis can improve the CO slope estimation, however the change is not significant.
For the manuscript we do not change too much, as we stick with the simple analysis for the above mentioned reasons, and the discussion of possible but not realized improvements of the analysis would degrade readability, but we add a footnote:
"More sophisticated calculation of the growth rate might improve the analysis; e.g. subtracting an individual mean value for the data used of each day prior to the linear regression decreases the random error from 10% to 8.5% while the result does not change significantly."

2.3 Can you quantify the importance of the heterogeneity in the MCMA?

If you scale up the UNAM FTS fluxes under the assumption that the MCMA is well-mixed, by how much do you overestimate or underestimate the emissions? (That is, can you quantify the importance of the heterogeneity in the MCMA?) **2.8 times.**
The referee addresses an interesting question and the answer shows that the effort of taking heterogeneity into account has to be done. Assuming homogeneity in the MCMA results in an overestimation by a factor of 2.825 (4.286) given by the ratio between the area 5177 km^2 (7854 km^2) of the MCMA and the effective area $A_{eff} = 1832.6\text{km}^2$ reconstructed in this work from CO column measurements. 5177 km^2 is the calculated area of MCMA enclosed by the green line in Fig 6, while (7854 km^2) the value in parentheses is the area estimated by the bottom up inventory (SGDF-INE 2010).)
In the manuscript we add the obtained value near the end of Section 2.4.1 (last but one sentence): *this is* " 2.8 times " *smaller* and " The assumption of an homogeneous CO emission in the MCMA would result in an overestimation by a factor of 2.8."

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3 Minor comments

3.1 Referee 1

The terms in the equations are not always defined, and this can lead to confusion. Please ensure that all variables are defined.
As explicitly pointed out by Referee#2 we defined: prf_1^{VMR}, prf_2^{VMR} .

You mention a MOPITT bias in v3 on P29919. Does this statement hold for v5, where the NIR+MIR retrievals were introduced, increasing sensitivity to surface CO? (e.g.Worden et al. JGR, 2011). If not, an analysis using the MOPITT v5 data may beneficial to your analysis (an extra two datapoints per day). Actually we started our analysis with MOPITTv.4. We have not checked for the V5 data, but the problem of MOPITT is not the sensitivity V4 reports reliable columns and averaging kernel. There seems to be a problem with the MOPITT instrument, we believe it has got a pointing bias. This might not affect studies on a $2^\circ \times 2^\circ$ grid, however on a sub-footprint scale ($<$ 10 km). The MOPITTv.4 CO columns show in our plots in average a hotspot almost 100 km WEST of Mexico City near the urban area of Toluca, while the hotspot near Mexico City probably is related with the CO emission originating from Puebla (100 km in the east). If this is a pure systematic error it can be corrected, but it should be evaluated first in a separate detailed study. The huge pointing error of NADIR-sounders (around 150km) was, to our understanding, also recently mentioned by Yurganov et al.(2012), unfortunately they did not give more detailed information about their findings regarding the pointing accuracy of the different NADIR sounders MOPITT, AIRS,... to our knowledge most validation papers have not addressed this topic too much, even though it seems important and even crucial for comparisons on a $1^\circ \times 1^\circ$ or smaller grid. Reconstruction of the mean CO column distribution with sub-footprint resolutions but sacrificing time resolution (as done here in this work)

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might be the best tool to quantify the pointing bias, but it is out of the scope of this work and we have decided not to use MOPITT for this work and not to mention explicitly our not sufficiently proved suspicion. But we agree with the referee, that after a validation and if necessary a correction of its pointing, MOPITT data can be used to improve the analyses. However the improvement is not done by adding the 2 data points and using the averaging kernels, as MOPITT and IASI measure different distributions at different times.
Fortunately the instrument IASI shows an quite accurate pointing, which is implicitly validated by Figure 4 showing the correlation between the main highways, mountains and suburbs and the CO-distribution.
Regarding the MOPITT discussion, we do not want to modify anything in the manuscript.

P29923L27: Please explain what a "cluster analysis" is.
Done. to:" The selection of low ventilation days was realized by cluster analysis, we add:, which defines for a given data set, different groups of elements with similar properties such as a similar ventilation pattern. The set is here given by the days between 2007-2009. The analysis afterwards assigns each measurement day to one of these groups (e.g. the group of low ventilation).

P29942L24: define "sufficient" in this case
we replace "sufficient" by "a" and add, " better than 10 km in the center of MCMA." The true discussion which resolution is sufficient is given in the supplement.
P29943L12: "This indicates that the spatial distribution used in the emissions inventory might be much more inaccurate than the absolute amount reported for the entire region." Could this also mean that your column measurements are more representative of the entire basin than the local emissions?
There is no analogy as there are really measurements at UNAM, but in the bottom-up

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inventory there are not any local measurements of CO emissions. Therefore the answer is probably "no", but "local " is rather relative, the question about "representative" (representation error = smoothing error) is to be answered with the reference to the "Averaging Kernel" and therefore we introduced the "Averaging Kernel for surface emission" **AKE** (in Section 3.3). The whole section 3.3 tries to discuss and answers this question quantitatively with equation (16) conceptually and (18) practically. The result is not given graphically but is slightly larger than the area given by the green contour line which is still smaller than the whole basin. We did not change anything in the manuscript, as we think that Section 3.3 addresses the question sufficiently, and we should not discuss the technical details again in the section "Summary and Conclusion".

P29944L9 : How do you get the 17.3 mass ratio from Wunch et al?

The referee is right. That is an error, the correct mass ratio according to Wunch et al. is 143=$\frac{44 \cdot 1000}{28 \cdot 11}$.

We adapt the discussion to the corrected value: ", a value far smaller than the CO_2 /CO mass ratio (143) derived for the urban area around Los Angeles, CA (USA) by Wunch et al.(2009) from solar absorption FTIR spectra taken between August 2007 and June 2008."

Further we just use the measurements taken in Mexico and change -> "From these ..." to "The emission factor 14.8 ..." and remove the values which have been calculated with the measurements valid for Los Angeles.

The discrepancy of the CO/CO2 ratios between Los Angeles and Mexico City is interesting and should be investigated, but further discussion of the comparison with this is not in the scope of this work. It might be related also with the location of the measurement site (center or outskirts) and if ratios of the concentration in the mixing layer or ratios of the columns, which contain air of different layers including "older" air, is determined.

P29944L23: Gisi et al. (2012) do not report gases other than CO2 (except for O2,which is only used to compute DMFs). we remove "and other gases"

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Figure 7: Is this figure necessary, or can it be combined with Figure 3 or 5?
Figure 7 illustrates how easy emission flux is measured by solar absorption under favourable conditions and we would like to keep it, but we follow the recommendation and combine figure 7 with figure 5. In addition we remove the panel Fig.3c and include the graphic in figure 3b.

Technical Comments:
Spelling and grammatical errors significantly detract from the flow of this paper. I have tried to list as many of the errors that I can, but this paper should be read through carefully before resubmission. done

There is a table of acronyms, but not all acronyms used in the paper are listed. I think they should all be defined in the text once as well. It should be complete now: We added a lot of acronyms and variables and we ordered the acronyms alphabetically.
Abstract: Define UNAM and IASI Done.
P29917L23: on the basin –> in the basin Done.
L24: According to the 2010 census (INEGI, 2011) Done.
P29918L18: Remove "as will be presented in this study" Done.
P29919L13: Define UNAM Done. UNAM is now defined earlier and in the table of acronyms.
P29920L8 : Define MILAGRO Done. We do not mention MILAGRO and Molina et al. here, but just a littel bit later, wher it is also defined, see next comment.
P29921L17: Define MIRAGE Done. - do you mean MILAGRO? Yes, MIRAGE http://mirage-mex.acd.ucar.edu/index.shtml was just the name in the initial planing phase.
We changed MIRAGE to MILAGRO. Here we define Milagro and cite Molina et al.
P29922L3 : course –> coarse Done.
L5 : and the Hase et al. (2006) sol Done.
L6 : This is confusing. Are there two solar trackers on this instrument? Yes, steering tracker and feedback-tracker using quadrupole-diodes. We changed the sentence to:

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"Observations were made throughout the day using a computer guided mirror followed by a dynamic tracker controlled by a quadrupole diode feedback system to lock the solar image on to the instrument entrance aperture."
L8 : remove "varying" and "retrieved" Done.
L19: can be used as reference –> can be used as a reference Done.
P29923L8 : Define SMA-GDF SM-GDF is already defined on page 29917 line 27 and 29918 line 1. We did not changed anything here.
L14: associated with new emissions mainly –> associated mainly with new emissions. Done.
L16: should be avoided, thus low ventilation (add the ',') Done.
L26: "clean the city" seems a bit strange, consider rewording "clean the city" was replaced by:"refresh the air in the city"
L27: We selected low ventilation days using cluster analysis. Done. We use the suggested wording
P29925L17: remove "rather"Done.
P29926L1 : lager-> largerDone.
L2 : systematic-> systematicallyDone.
L17: This effect prevents the data from being suitableDone.
L18: remove "not a single but"Done.
P29927L1 : remove "interferometer"Done.
L10: profile retrievals from the ACE-FTS spaceborne instrument (Clerbaux et al., 2008b) Done.
L11: aircraft based profile measurements from MOZAIC Done.
L12: define MOZAIC Done.
L20: remove "realized" Done.
L21: explain what you mean by ill-posed problem We add "mathematically". I think "mathematically ill-posed problem" should be known by anyone of the community and it is the basic knowledge about inversion in remote sensing but anyway we add in parentheses: (Mathematical problems or equations which do not have a unique

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solution.)
L22: remove "are taken as measurements" and put (TC_{IASI}) in parentheses Done
P29928L1 : CO distribution mainly depends on the constraint done
L7 : I'm not sure what you mean by "on the base of an average" we changed to: by a linear transformation of
L10: define epsilon Done. We add just before the equation:" The vector $\vec{\epsilon}$ contains the difference between measurements (column retrievals) and simulations. Each component describes the deviation of a retrieved CO column from the estimated mean value at each measurement (smoothed by the IASI kernel) plus the measurement noise error and other errors of the IASI retrievals."
L13: are prf1 and prf2 supposed to have VMR as a superscript to match the text? No. They have the unit "column density" and summed up total column 1cm^{-2}. We added the definition of $prf_{1/2}^{VMR}$, which should clarify please see Ref#2.
L21: operated on the profiles done
P29929L4 : remove "actually" done
L7 : remove the comma between "here" and "that". done
P29930L20: an –> adone
L20: visible as CO hotspots from the daytime measurements done
P29931L9 : contains information about the ith grid point. done
L16: by an empirically adjusted done
L19: and how consistent the solution is with the a priori done
L21: describes the weighting done
P29932L1 : replace "realized" with "applied" done
L5 : A diagonal matrix constrains the background layer, avoiding problems arising when the background CO column differs... done
P29933L18: we iteratively optimized the constraints done
L21: while the CO anomaly done
L24: Sensibility –> Sensitivity done
P29934L5 : The latter can be estimated by done

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P29935L11: in –> at; no comma after "column" done
P29936L1 : mean wind velocity at the ground done
P29940L8 : write out "wind direction" and "wind speed" instead of WD/WS done
P29941L4 : This sentence is awkward. I'm not sure how to fix it. It is not necessary and we just remove it. Done.
L6 : in –> on done
L10: of –> on This sentence was removed (please see. response to prior comment
P29942L7 : slightly –> slight done
L8 : slightly –> slight done
L19: CO column growth-rate under low ventilation conditions, and during a reliable time interval to prevent contamination from inhomogeneity (11:15-13:15 LT). done
P29943L1 : anual –> annual done
P29944L9 : mass ratios –> mass ratio The sentence was changed, please see minor comments above.
L19: under –> in done
Figure 2 caption: remove "the" before Mexico City.
done
Figure 4 caption: line5: circle –> diamond, and add "line" after horizontal
done
Figure 6 caption: The NCAR measurements during the MILAGRO campaign. done
The supplementary material has many grammatical errors as well, but given that there are no line numbers, it will be too difficult to write them down usefully for you. we corrected various errors
My main concern about the supplementary material is section 1.3, which I had trouble following. We removed section 1.3 of the supplement. There are no results from the error estimation in section 1.3 supplement which are used. A description of how the measurement noise error can be calculated is not necessary as the variables are described in the main article and the equations can be found in text book Rodgers (2000). The analogy of the retrieval to profile retrieval is straight forward so that

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the error analysis can be transferred to the estimation of the reconstruction average distribution of columnar CO from the space measurements.

3.2 Referee 2

line numbers refer not to the online version, but to the manuscript which have been uploaded for the Editor at (19 Oct 2012) . Two different SFIT versions are used for the ground based CO retrievals, why and what is the difference? There is no much difference between the versions, the results are almost identical. The use of a huge spectral window was just possible by changing one parameter in the original sfit2v.393, program (Ortega,2009; Stremme,2009) and we expected that the official release v.394 is able to do so. After changing the operating system of our server to Ubuntu and from the Digital-Intel compiler to the GNU-compiler, we found it easier to get the newer version to be compiled. We planed the reanalysis of the spectra with the sfit2v.394, but the similarity of the results found by the cross-validation of the CO-column results of both versions showed that this effort is not necessary. As the retrieval of one spectrum takes a few minutes and there are around 47457 spectra to be processed (we have recorded around 1 spectra every 20 seconds during monitoring), processing the whole dataset takes several months.
We add:
(Both versions have been compared and show almost identical results for CO.)
Fig. 3b: From the scattered red points, it is difficult to see that the average (black curve) could be obtained. Perhaps a different marker type/size would be more appropriate? Done, we changed the plot, now the lines are stronger and therefore we could also remove the plot fig.3c, which is now included in Fig.3b.
Fig. 4: the figure and the annotations are too small to read (unless zoomed in from the PDF). I think this can be as big as Fig 6. done
L230:P.29925 L.20 From the way the paragraph is written, it is not immediately clear

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to the reader how the time interval is chosen. we changed to:
"Choosing a correct interval for calculating the growth-rate is therefore crucial. There is a trade off between i) low systematic errors which means an interval which is not significantly affected by horizontal inhomogeneity at high sun zenith angles in the mornings or increased ventilation in the afternoons and ii) the random error which means that the time interval is still large enough to obtain a significant result for the mean slope."
L236:P.29925 L.26 . How was the precision derived here? precision=random error therefore we change "precision"->" random error (precision)"
L239: "systematic" > systematically Done.
L302:P.29928 L.14 "prf_1^{VMR} ": It is not shown where this was used. The referee is correct, we replace "where" with "The profiles are initialized with help from prf_1^{VMR} and prf_2^{VMR} , in which....."
L351:P.29931 L.1 "The equations" > Equations
Done.
L352:P.29931 L.3 "(KËEj) belong" > (KËEj) belongs
Done.
L415: P.29933 L.24 "sensibility" > sensitivity
Done.
L423: P.29934 L.8 "error" > errors
Done
L425: P.29934 L.11 the subscript "constrain1" is missing a "t"
Done
L430: P.29934 L.16 put "(Eq. 1)" at the end of the sentence. Done
L460:P.29936 L.5-8 Sentence starting from here up to L464: Restructure and separate into two sentences for clarity. Done.
"The integration of the fresh CO ($TC_{IASI} - 1.51 \times 10^{18}$ molec cm^{-2}) is performed over the area where TC_{IASI} is greater than the background. The division of the integrated mixing layer CO MCMA content by the mixing layer column at UNAM ($TC_{UNAM}^{10:19} - 1.51 \times 10^{18}$ molec cm^{-2}) results in an effective area $\mathbf{A}_{eff}$, Eq. (1). The

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value for $TC_{UNAM}^{10:19}$ is the total column measured by IASI over UNAM which is basically the same as the column measured from the ground at UNAM backward extrapolated to 10:19 LT, Sect. 3.

$$\mathbf{A}_{eff} = \frac{\int_{MCMA}(TC_{IASI} - 1.5 \times 10^{18}\text{cm}^{-2})\text{d}\mathbf{A}}{(TC_{UNAM}^{10:19} - 1.5 \times 10^{18}\text{cm}^{-2})} \quad (1)$$

"

We recognized also an error in Equation (15), which is now corrected.

L485: P.29936 L.24/25 "information of" > information on. "by the" > from the Done

L487: "emissions depends" > emissions depend Done

L529: There seems to be a missing unit for "15", I guess minutes (min) ? Done

L553: "within certain criteria" > within a certain criteria Done

L578: "focuses in" > focuses on Done

L583-589: Sentence construction should be improved for clarity. done

L598: "affect significantly" > significantly affect done

L601&602: "slightly" > slight Done

L651: The derivation of the mass ratio (17.3) from Wunch et al, 2009 is not immediately obvious at first read. A sentence explaining this would be helpful. Their was an error: Wunch et al, 2009 report a CO/CO2 of 11 molecule ratio which reflects a mas ratio (CO2/CO) of 143g CO2 per 1g CO. See also comment above. The sentence is rewritten and the result of Wunch et al. (2009) is just mentioned qualitatively.

Supplement:

Table 4 caption: "meanvalue" > mean value Done.

Table 4 footnote: fonts are too small Done. we changed from tiny to normal fontsize.

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