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Interactive comment on "A linear CO chemistry parameterization in a chemistry-transport model: evaluation and application to data assimilation" *by* M. Claeyman et al.

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The authors are very grateful to Dr. A. Geer for the excellent report he has provided. His remarks and comments have been of great importance to improve the quality of the paper. Below, please find the reply to the general comment as well as the minor comments.

This paper presents LINCO, which is probably the first linear CO chemistry parameterization, and is based on an approach that was originally developed for ozone. It will be useful for long chemistry model runs and for data assimilation. The scheme is validated in three ways: against a more detailed chemistry model, against independent

data, and within a data assimilation system. This validation covers the troposphere and stratosphere, making for a good, comprehensive paper. However, there is one major area that requires more work. The validation shows that in the troposphere, LINCO produces CO values that are around 20% too low compared to both the detailed chemistry model and to independent MOZAIC observations, which agree with the detailed chemistry model. This negative bias crops up throughout sections 3 and 4. The explanation given for this bias is simply that there is "a larger destruction of CO in LINCO" on p 7007. This is not sufficient. The causes of the bias need to be properly explained. I think there is a good chance that the cause can be discovered and moreover, that the bias could easily be removed from the LINCO scheme.

There are some very likely causes of this bias:

1) The bias reflects a difference in the CO climatology of MOBIDIC (used to derive the LINCO coefficients) compared to RACMOBUS. If so, this bias will be reflected in the A3 term in LINCO, which represents the CO climatology towards which LINCO will relax. This hypothesis should be easy to check, and must be considered in the paper.

Our reply :

We fully agree with the reviewer. It is easy to check the bias by comparing the A3 term (Fig1a) and RACMOBUS CO distributions (Fig 3 bottom). But the bias between A3 and RACMOBUS is positive whereas LINCO-RACMOBUS is negative. Then, this is not straight forward to infer that CO concentrations calculated by LINCO will be lower than RACMOBUS ones. However, A3 is a term consistent with the physics and chemistry of the MOBIDIC 2D model and the other parameters (A1 and A2). One possibility for decreasing the bias between LINCO and RACMOBUS, is to increase the A3 term, but we lose the consistency between the different terms of the linearization and we increase the difference between A3 and RACMOBUS. As suggested by the reviewer, this remark is added in the manuscript (section 3.1 and 3.2).

2) The bias comes from a systematic difference between MOCAGE model temperature

T and the RACMOBUS zonal mean temperature coefficient A5. This again should be considered in the paper.

Our reply:

The model temperature (from ARPEGE analyses used as atmospheric MOCAGE forcings) and the LINCO zonal mean temperature (A5) are compared. The difference between both parameters are relatively small in the troposphere (between 0 and 1.5K) except in the polar regions where the difference is of the order of $\pm$4K (see Figure R1). Moreover, in order to check the impact of temperature, we follow the same methodology as Geer et al., (2007) (see §2.7 of this paper) and we calculate the temperature term (defined as: A2*(T_ARPEGE – A5)/A3 for the off-line comparison of photochemistry terms. The results confirm the low dependence of the temperature term in the troposphere (between -0.3 and 0.1 %/day) compared to the other terms. The 'CO' term varies from -2.7 to 0. %/day and the 'P-L' term varies between –0.5 to 2.2 percent/day. (See Figure R2). We added the comment in the new version of the paper. We opted not to present Figure R1 however, as the article has already several figures.

3) The bias comes from systematic transport differences between the MOCAGE model and RACMOBUS, meaning that the production - loss rate (the A1 coefficient) is not in balance with the MOCAGE model transport. The authors already explain polar winter stratospheric biases in this way.

Our reply:

We fully agree that the transport scheme of MOCAGE and MOBIDIC models are different since the first one is 3D and the second is only 2D. This may generate an inconsistency between the A1 term and the transport in MOCAGE. Moreover, the approach developed for the LINCO is slightly different from that of linear ozone scheme in the sense that for LINCO, the emissions are taken into account whereas for linear ozone there are no emissions. Consequently, the 'production – loss' term (A1) should also be balanced with the emissions. In the same way, we have to keep in mind that MOCAGE

uses an emission inventory whereas MOBIDIC is relaxed to a zonal averaged climatological concentrations at the surface. This also could be one of the sources of the discrepancy. We clarified this in the new version of the paper (see section 3.1)

The first two biases are most likely where the photochemical relaxation time is relatively fast, and the third is more likely where it is slow. These kinds of problems are common in linearised ozone schemes and are addressed in Geer et al. (2007, ACP, 7, 939-959, particularly sections 2.1 and 2.2) and in Coy et al. (2007, ACP, 7, 2917-2935). Other studies which have investigated these effects are Eskes et al. (2003, QJ, 129, 1663-1681) and McCormack et al. (2006, ACP, 6, 4943-4972). If either of the first two hypotheses explains the bias, it would be easy to remove it by adjusting the climatology parameters A3 or A5 appropriately. This should produce a version of LINCO that is more in agreement with detailed chemistry and with observations. A final possible explanation for the bias would be that the MOBIDIC, MOCAGE or RACMOBUS emission and deposition climatologies may be inconsistent. Again this should be considered.

Our reply:

According to what we said in the 3 previous points and in order to reduce the negative LINCO bias in the troposphere, we should modify the A1 coefficient. However, doing this, we introduce an inconsistency in the set of coefficients and we obtain coefficients only valid for the studied period. Indeed CO interannual variability is typically of 10-20% in the free troposphere, and nothing indicates that the tuning for a specific period could improve results for integrations of several years. Then, in order to reduce the bias between LINCO and RACMOBUS (of about 20% in the troposphere), we removed a value of 20% of A3 *A2 from the original A1 and we obtain this new equation :

dCO/dt =A1-A2*A3*0.2+A2*(rco-A3)

=A1'+A2(rco-A3)

The LINCO results are globally improved compared to RACMOBUS in the troposphere (see Figure R3). Nevertheless, this improvement is not uniform: one can see clearly the improvement of the bias in the troposphere whereas it increases in the stratosphere. Therefore, it is more complicated to adjust the A1 term to fit RACMOBUS fields and we think it is beyond the scope of the paper since the objective of the paper was to implement a very fast linear scheme for CO based on a 2D model (MOBIDIC) and by assimilating the MOPITT data to obtain realistic CO fields. However, we add a comment in the paper that if we change artificially the coefficient A1, we can expect to reduce the bias in the troposphere at the expense of introducing inconsistency in the coefficients set (see section 3.1 and 3.2).

Minor and technical corrections

p.6996 l.6, l.11: "on the one hand .. on the other hand..." suggests the balancing of opposing concepts, which is not the case here.

Our reply: corrected

p.6996 l.10: Differences in ppbv - could the % figures also be supplied?

Our reply: We change the original sentence as suggested by the reviewer by the following one:

“The mean differences between both schemes are about ±25 ppbv (part per billion by volume) or 15% in the troposphere and ±10 ppbv or 100% in the stratosphere.”

The abstract could be compressed. 250 words is often recommended, and helps readers quickly assess a paper.

Our reply : The abstract has been compressed.

p.6997 l.20 "along with photochemistry production". Do you mean "photochemical production"?

Our reply: yes and it is corrected

p.6999 l.2 "consistent 4-D fields" - please say what these fields contain (e.g. chemical species, dynamical quantities?).

Our reply: We change the text as suggested by the reviewer :

"It consists of providing consistent chemical species 4-D fields by combining in an optimal way observations and model fields"

p.6999 l.12 "makes possible long run" -> "makes possible long model runs?"

Our reply: corrected

p.7001 l.25 "in nh" -> "in the nh"

Our reply: corrected

p.7001 l.26 "complemented with surface emissions and deposition" - please add some details or references

Our reply: Now, we refer to the section 2.2 and we add in this section some complementary comments on the deposition scheme.

p.7002 l.5 "hybrid (sigma, p)" - "sigma" and "p" need to be defined.

Our reply: We explain better in the text the hybrid vertical levels :

“MOCAGE uses hybrid vertical levels from the surface up to 5 hPa with a resolution of about 150 m in the lower troposphere (40 m near the surface) and up to 800 m in the lower stratosphere. Hybrid vertical levels are designed so that 135 lowermost levels follow the terrain while upper levels are isobaric. “

p.7002 l.20-22 - this long list of references is not particularly useful to the reader. It does not seem necessary to give more than one or two representative examples. MOCAGE is used for several applications: operational chemical weather forecastingÌĄ in Meteo-France (Dufour et al., 2004) and data assimilation research (e.g., Cathala et al., 2003; Pradier et al., 2006; Clark et al., 2007; Semane et al., 2007; El Amraoui et al., 2008a,b;

Semane et al., 2009).

Our reply: As suggested by the reviewer, we simplified the sentence:

"MOCAGE is used for several applications: operational chemical weather forecastingÌĄ in Meteo-France (Dufour et al., 2004) and data assimilation research (e.g., El Amraoui et al., 2008b; Semane et al., 2009)."

p.7002 l.28 - what were the conclusions of Ricaud et al. (2009)? How do they impact the current work?

Our reply: We removed this sentence.

p.7004 l.10-12 "altitude" -> "altitudes"; "paper is" -> "paper are"; "profile" -> "profiles"

Our reply: corrected

p.7004 l.15 "noise generated by the galaxy" - could you give just a few more words on what is meant by this?

Our reply:

Pumphrey at al (2009) show that the signal generated by the Milky Way can affect the earth CO retrieval especially at 22 hPa for some specific days and latitudes. In the MLS CO data used in this study, the impact of the Milky Way signal is taken into account in the retrieval of the terrestrial MLS CO data. We rewrote this in the text:

“In the version used in this study, the impact of extra terrestrial signal generated by the Milky Way and affecting the terrestrial CO retrieval mainly at 22 hPa for some specific days of year and latitudes has been eliminated according to Pumphrey at al (2009).”

p.7004 l.16 "MLS CO" -> "The MLS CO"

Our reply: corrected

p.7005 l.10-11 - this sentence doesn’t make sense to me.

Our reply: We replace this sentence by :

“We selected MOZAIC data from flights over Europe, North America and Eastern Asia.”

p.7006 l.24 - If Fig. 2 shows differences in the troposphere up to -40%, how can Fig. 3 show differences of no more than -10%? There should be a consistent reference used in each case, probably RACMOBUS as this appears the more accurate.

Our reply: As suggested by the reviewer, the reference is now RACMOBUS and we obtain the same difference in the troposphere (-40% in Fig 2) whereas we obtain -35% for Figure 3. We changed Figure 3 and the corresponding text which is consistent with Figure 2.

p.7006 l.25 - "region with the intense convective activity" -> ITCZ?

Our reply: We took into account this remark in the new version of the paper :

“The maxima of the difference between both schemes in the troposphere, are located in the region with the intense convective activity, namely the inter-tropical convergence zone.”

p.7007 l.13 - "April maximum in NH" -> "The April maximum in the NH"

Our reply: corrected throughout the paper

p.7007 l.15-16 - "in SH" -> "in the SH"

Our reply: corrected throughout the paper

p.7007 l.17 "However, LINCO scheme" -> " However, the LINCO scheme"

Our reply: corrected

p.7009 l.4 "this behaviour has already been reported by van Noije concerning ERA-40". Are there any relevant results concerning the ARPEGE fields used in MOCAGE?There are many more papers on this phenomenon available, in particular Monge-Sanz et al. 2007, GRL 34, L04801, which evaluates several different versions of the ECMWF system. Perhaps one of these provides a closer parallel to the ARPEGE system you use?

Our reply:

This behaviour was also reported by Teyssèdre et al (2007) for ARPEGE. We add this reference in the text and we modify section 3.3 in this sense.

p.7009 l.27-28 "This indicates that LINCO does not present a global systematic bias" (versus MOZAIC) - this seems to contradict Fig. 10, which shows a clear bias between LINCO and MOPITT and MOZAIC.

Our reply:

This sentence is relative to Figure 8 which is a comparison between MOZAIC and LINCO for levels between 300 hPa and 180 hPa and not for the complete troposphere which is the case in Figure 10. In Figure 10, the bias is also very low around 250 hPa. To avoid any confusion we modified the text in this sense. :

"This indicates that LINCO does not present a systematic bias at the levels between 300 hPa and 180 hPa. However, as expected, LINCO is not able to represent the variability observed by aircraft in situ measurements"

p.7012 l.16-18 - I don't understand this - perhaps rewrite?

Our reply:

We rewrote the corresponding text :

"However, compared to MOZAIC, MOPan overestimates CO at 850 hPa in October-November-December 2004 and January-February-March 2005 over Europe and North America. Nevertheless, 850 hPa is probably not the most MOPITT reliable level for MOPITT data according to Emmons et al. (2004)."

p.7015 l.15 "because of the too rapid downward transport" - this remains a hypothesis

rather than a proven fact for MOCAGE, as indicated on p.7014 l. 26: "we suggest that this deficiency is related to a too rapid downward transport..."

Our reply:

As suggested by the reviewer, we replaced the sentence by :

"However, at extratropical latitudes, MOPITT CO analyses underestimate CO concentrations compared to MLS CO (∼50%). This bias may be related to a too rapid downward transport of the meridional circulation".

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C3857/2010/acpd-10-C3857-2010-supplement.zip

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 6995, 2010.

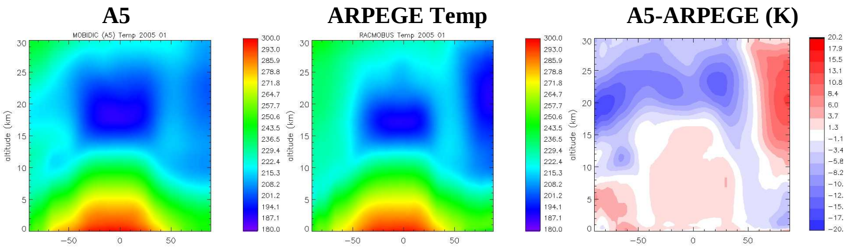

Figure R1: A5 term representing the January climatology zonal mean temperature in MOBIDIC model in K (left); January 2005 zonal mean temperature from ARPEGE used by MOCAGE in K (middle); Difference between A5 term and ARPEGE temperature in K (right).

Fig. 1.

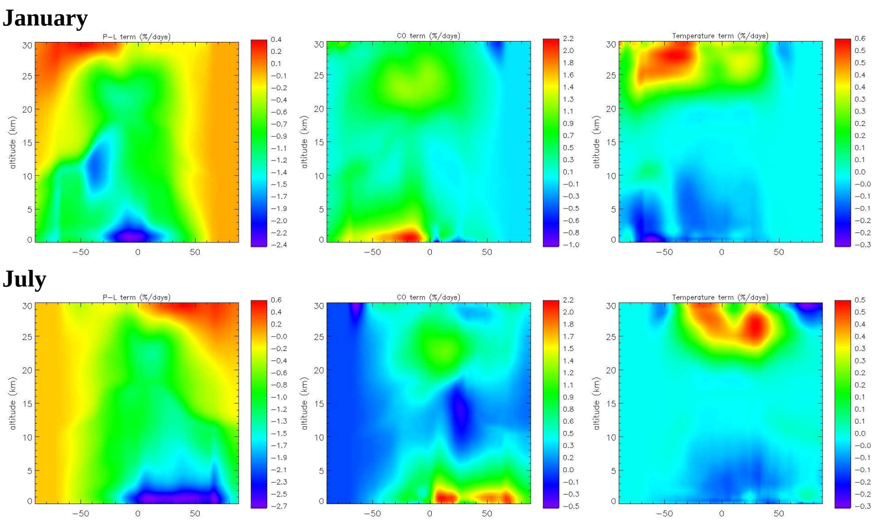

Figure R2 : (P-L) term A1 (left) ; CO term A2(r_{CO}-A3) (middle) ; Temperature term A4(T-A5) (right) in %/day. The top three figures are relative to January and the bottom three figures are relative to July.

Fig. 2.

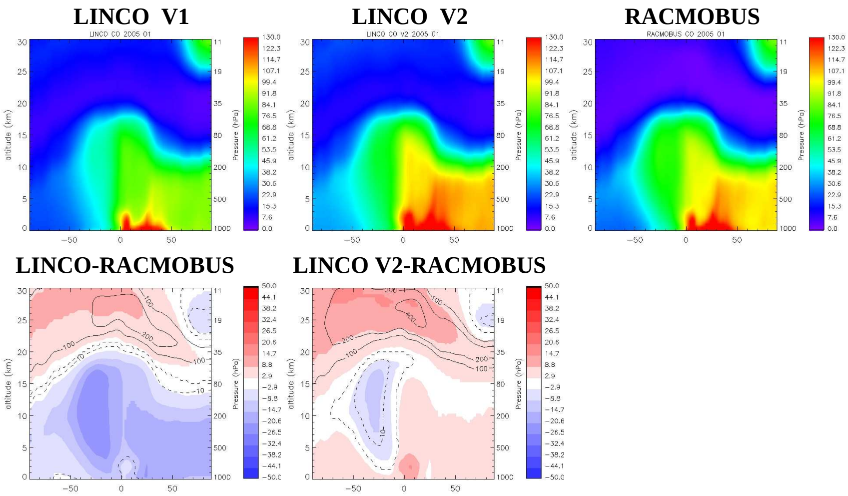

Figure R3 : Top : Zonal mean CO concentration from LINCO V1 (left), LINCO V2 with A1'=A1-0.2*A2*A3 (middle) and RACMOBUS (right). Bottom : difference between LINCO V1 and RACMOBUS (left) and LINCO V2 and RACMOBUS (right).

Fig. 3.