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Atmospheric Chemistry and Physics Discussions

Interactive comment on "What do ^{14}CO measurements tell us about OH?" *by* M. C. Krol et al.

Anonymous Referee #1

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The paper addresses an important issue in atmospheric chemistry and uses a sound state-of-the-art methodology (adjoint modelling) in order to investigate the relationship between the global OH distribution and measurements of isotopic carbon monoxide (14CO). The text is generally well written and clear, but there are a number of statements which can easily be misinterpreted or which overemphasize certain minor aspects of the problem. The analysis of the adjoint OH concentrations obtained from pulses added to the 14CO concentrations at 5 measurement locations is good, but the paper remains inconclusive with respect to the promising title. Unless the authors add some discussion on the uncertainties and the potential improvement for estimating global OH compared to the much simpler approach used by Volz and Ehhalt, 1981, the paper is not of great interest to the community. Therefore I would reject this paper

in its present form and encourage the authors to go a step further and resubmit the manuscript after major revisions.

The major concern I have with this paper is that it appears to make matters much more complicated than they are. From a simple-minded “forward” view, the loss of 14CO is given by the integral over time of k * OH * 14CO * dt. It is therefore evident that high OH concentrations lead to a fast degradation of 14CO so that the distance that a particular air parcel has to travel until a given reduction in 14CO is achieved is smaller than in the case of low OH concentrations. Unless I missed an important point, this is essentially the message that can be taken from the adjoint OH field presented in the paper. Obviously, tropical OH concentrations are higher and therefore the OH “signal” that is contained in a 14CO measurement at a tropical location is generally more localized than at high latitudes where the air mass has to travel much longer before the integral loss rate reaches the same value. Do we need an adjoint model for this?

Detailed comments: (1) Abstract: after a promising start the abstract looses its momentum and trails off by listing some detailed results and a very vague conclusive sentence “may offer the possibility to constrain (local) OH”. Unless the paper demonstrates this capability (or disproves it) I don’t see why it deserves publication. (2) Abstract: “14CO measurements” - unclear if these are existing measurements (the authors should then say “the existing 14CO measurements at five locationsĚ”) or if these are hypothetical measurements which if they existed would allow to fulfil the claim the paper makes. (3) Introduction contains too much discussion and should be more tailored to reporting the facts. (4) Introduction: 14CO formation - a symbol seems to be missing in between 14N and 14CO. (5) p. 10407, l10ff: language “the action of OH” (6) structure: p. 10407 mentions the measurements but the reader is left unclear until much later what kind of observations exist. Instead of discussing the usefulness of 14CO in the second paragraph of the paper, the authors should first describe the (bio)geochemical cycle of 14CO and what observations exist of this compound. (7) p. 10409, l. 7 “relatively

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limited distance". This is misleading because it assumes a 1-dimensional point if view. It would be better to say "smaller region of influence." (8) p. 10410, l. 18 figure 1 indicates a production range of 1.2-1.9 molecules cm-2 s-1, but the text says "scaled to a global production of 1 molecule cm-2 s-1. Then again the source strength seems to be modulated according to Lowe and Allan. What is the reason for scaling it to 1? (9) While the discussion on the source strength and variability is generally good, I am missing some summarizing statement about the uncertainties related to the source strength or rather the amount of 14CO entering the troposphere. Is this model dependent? Is there a resolution dependence? Are there independent constrains? If these issues are discussed elsewhere (e.g. Jöckel et al) then the major findings should be summarized and referenced here. (10) p. 10411: as above: structural deficit. Description of observation is scattered throughout the text. (11) p. 10412, l.13ff: I don't like to see "measurement values generated" or "corrected" because of model assumptions. This needs to be rephrased. For example: "In order to estimate the cosmogenic part of the observed 14CO signal, we Ě" (12) p. 10413, l.4 Uncertainty of the source - this needs to be quantified (see comment 9 above) - also: the source distribution (e.g. the real one) must be independent of the calculation method (13) p. 10413, l.10: "may be too weak" (instead of "can") (14) p. 10415, eq. 2b: this is a steady state equation which implies dOH/dt = 0. While I believe I understand the reasoning behind this, it may look puzzling to the reader unless this boundary condition is explained (especially because the paper later discusses changes in OH). (15) p. 10417, l.7 ff: unnecessary detailed discussion about the necessity for scaling or mass-weighting the adjoint field. Shorten to 1-2 sentences. (16) figure 3 may be pretty but does not contain very useful information beyond what is presented in figs 4 and 5. Suggest to remove this figure. (17) p. 10418, l.8 Delete sentence "Higher OH also impliesĚ" This duplicates the statement from the sentence before. (18) p. 10418, l.19: that sentence is misleading. The concept of "transported upward" applies to a forward tracer and is not applicable to an adjoint variable. "Propagated" may be a better term. (19) Figs 4 and 5: I don't quite understand the reasoning for leaving out the first 20 days of the backward integration.

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Why 20 and not 1, 5 or 100? (20) p. 10419, l. 11 it is not the integral that reflects the lifetime but the plot. Perhaps figure 6 should be shown with a log y axis? (21) p. 10419 ff. The term “sensitivity to OH” is misleading (see major comment above). The interesting question how much one can learn from the individual station measurements about global OH or average OH concentrations in certain regions is not really addressed. (22) p.10421 The beginning of the Discussion and conclusions section is more descriptive and would belong into the results section (variability patterns). (23) p. 10421, l.15 “is explained by Ĕ” An important part of the explanation is missing: without (occasional) transport into tropical regions the high-latitude pool would be irrelevant. (24) the three bullets drawn as conclusions are somewhat disappointing (see major comment above). The statement that the current measurements are more sensitive to high latitude OH than to tropical OH is misleading. As explained above, the loss of 14CO depends on the integral OH, but it doesn’t matter where this OH is located. It is only because of a longer residence time that a small perturbation in high latitudes causes a large 14CO signal. (25) labels in Figure 2 are too small

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 10405, 2007.