Atmos. Chem. Phys. Discuss., 7, S5787–S5793, 2007
www.atmos-chem-phys-discuss.net/7/S5787/2007/

Atmospheric Chemistry and Physics Discussions

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

M. C. Krol et al.

Received and published: 9 October 2007

In 1981, Volz et al. for the first time simulated the ^{14}CO distribution and derived that the OH fields used in their 2D model had to be scaled by 1.66 to explain the observed ^{14}CO mixing ratios. Seasonal cycles of the measurements were well reproduced. The main concern of reviewer 1 is that our paper does not bring new insights compared to the Volz et al. 1981 paper. We argue here that our 3D approach and adjoint methodology for the first time show quantitatively the sensitivity of ^{14}CO measurements to OH at various locations. This opens the way to quantitatively optimize regional OH by using ^{14}CO measurements. Although this latter step is not taken in the paper, we outline the methodology and discuss in detail the sensitivity of ^{14}CO measurements for changes in the 3D OH distribution.

One quote from the 1981 paper illustrates the many correct insights that existed already at that time: "the14CO concentration at any location depends more or less on the 14CO production and destruction everywhere in the hemisphere". The authors thus concluded that 14CO measured in the southern hemisphere do not "see" northern hemisphere OH and 14CO sources. Our paper goes a big step further in quantifying the "more or less".

Although many more 14CO measurements have become available since 1981, the use of these measurements in constraining OH has been relatively limited compared to methyl chloroform measurements. One of the prime reasons for this is the shorter lifetime of 14CO compared to methyl chloroform, which makes the interpretation of the measurements more difficult.

In the recent paper of Manning et al. (Nature, 2005), the variability of 14CO measured at Scott Base, Antarctica, and Baring Head, New Zealand, is interpreted as OH variability. Given the short lifetime of 14CO, this OH variability should be interpreted as variability in high latitude OH. Nevertheless, the analysis of Manning et al. still uses a single box model and thus ignores possible variability due to transport. It would be very useful to repeat the Manning et al. study with a full 3D model that takes into account variability in the transport including the variability in stratosphere-troposphere exchange. Our paper aims at the development of the mathematical framework for such a study.

Unfortunately, the usefulness of this study is strongly questioned by reviewer 1, who states: "Unless the authors add some discussion of the uncertainties and potential improvement for estimating global OH compared to the much simpler approach used by Volz et al. (1981), the paper is not of great interest to the community". Furthermore, the reviewer states that we make matters much more complicated and

ACPD
7, S5787–S5793, 2007

that the whole problem can be understood simply by a "forward" view and analyzing $k \times OH \times^{14} CO \times dt$. Here the reviewer indeed misses the important point that forms the backbone of our paper and which opens the way to exploit the available 14CO measurements in a more quantitative way than has been done up to now.

This unfortunate misinterpretation of our results is best illustrated by the last few sentences in the review: "The statement that the current measurements are more sensitive to high latitude OH than to tropical OH is misleading. ... the loss of 14CO depends on the integral OH, but is 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". Using the described adjoint methodology, our study directly calculates the sensitivity of single or multiple measurements for the full 3D OH distribution. Our central result (figures 4 and 5, based on the five station network) thus directly illustrates that it does matter where the OH is located! An OH perturbation in the tropics is hardly "seen" in the observational network. In contrast, an OH perturbation in the 14CO source region leads to a large effect on the measurements. Of course, this behavior is strongly linked to the short residence time of 14CO in the tropics as argued by the reviewer. Our formal adjoint method thus confirms and quantifies the long established knowledge that 14CO measurements tell more about high latitude OH.

Furthermore, we show that the sensitivity of a 14CO measurement for the 3D OH field is a strong function of time, or more precisely the meteorological situation, especially at tropical measurement locations. Finally, we contrast the zonal average sensitivity for OH with the methyl chloroform sensitivity for OH. Such a sensitivity calculation never appeared in literature, although the gross picture can be derived with simple arguments as stated by reviewer 1.

We indeed agree with reviewer 1 that the same information can in principle be

ACPD
7, S5787–S5793, 2007

obtained in a "forward" view. By perturbing the OH concentration in each of our model grid-boxes, we could have calculated exactly the same results. In fact we start the explanation of our adjoint methodology by stating this (section 3). However, as we state on the top of page 10415, the adjoint methodology is much more efficient in calculating the sensitivity of a measurement for the 3D OH distribution. In a full 3D model these forward calculations would take a prohibitive amount of computer resources. The development of adjoint codes for many applications by numerous groups worldwide at the moment illustrates the use for the community. In reply to reviewers 1 question: "Do we need an adjoint model for this" we can therefore answer: in principle no, but in practice yes! To exploit the available 14CO measurements fully, the adjoint approach is convenient and even necessary to calculate the full 3D OH sensitivity for many measurements.

Thus, we feel that, although we do not present a real inversion in this paper, the results presented are new, interesting, and provide a degree of insight that none of the scientists in our team has ever obtained with simple qualitative or semi-quantitative arguments before. The development of a mathematical framework for the interpretation of 14CO measurements is non-trivial and therefore useful to communicate to the scientific community.

In response to the comments from referee 1, in the ACP paper we intend to highlight the new accomplishments more. Furthermore, we aim to present a better quantification of the OH sensitivity on global and regional scales, as requested also by reviewer 2. Reviewer 1 lists a number of useful remarks, which we will take also into account in the revised paper. We thereby adopt the numbering in the interactive comment.

(0) The title: we agree that the title promises more than the paper brings. We will modify the title in "What can 14CO measurements tell us about OH" since this study is

ACPD
7, S5787–S5793, 2007

a preparation for a quantitative study using real 14CO observations.

(1) We will rewrite the abstract.

(2) We used hypothetical measurements (in fact we released unit pulses) at the five measurement stations that are currently operational. This is made clear in the manuscript. Also we will describe the available measurements more clearly.

(3) We will try to shorten the introduction.

(4) We adopted the standard notation ^{14}N(n,p)^{14}C to indicate the that interaction between a neutron and a 14N nucleus produces a proton and 14C. This notation is also used in e.g. the Volz et al. (1981) paper.

(5) We will modify "the action of OH".

(6) See point (2).

(7) Will be changed.

(8) By scaling the 14C production to 1 molecule cm^{-2} s^{-1} we follow the methodology introduced by Jockel et al. (2002). The reason for doing so is that the results can easily be scaled to different estimates of the total production rate. We will add the reference.

ACPD
7, S5787–S5793, 2007

EGU

(9) The amount of 14CO that enters the troposphere is model dependent, and probably also resolution dependent. This issue is indeed studied by Jockel et al. (2002). As requested by reviewer 2 we will perform some sensitivity simulations on higher horizontal and vertical model resolution and include the results.

(10) See point (2).

(11) We agree.

(12) We will add sensitivity experiments with different source assumptions.

(13) OK.

(14) Good point. We will add some text that explains that we use off-line OH that comes as monthly averaged fields and therefore stays constant during the month (hence dOH/dt = 0). We are interested here in the quantity d14CO/dOH, i.e. the sensitivity of 14CO measurements for changes in OH.

(15) We could indeed leave out the mass scaling discussion but are reluctant doing so. It should be noted that such methodological difficulties in adjoint model development may be useful to retain in the paper because other modelers will experience the same difficulties. For our research team, this issue was one of the hardest parts to understand (and explain). We agree that this is a technical issue that is hard to follow for a non-specialist.

(16) We will remove figure 3.

ACPD
7, S5787–S5793, 2007

EGU

(17) OK.

(18) Correct, we will use propagate.

(19) This is another issue that we will modify in the revised paper. The reasoning is that the zonal average will be entirely dominated by the polar station Scott Base. We agree that 20 days is really arbitrary. In the zonal plot we will include the first 20 days and modify the color table.

(20) Good suggestion that we will consider

(21) This will be better quantified in the revised paper.

(22) OK, we consider moving it to the result section.

(23) Correct, we will adapt.

(24) See discussion above.

(25) We will enlarge the labels in figure (2)

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