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Interactive Comment

# *Interactive comment on* "What does the global mean OH concentration tell us?" *by* "M. G. Lawrence et al."

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#### **General comments**

The paper focuses on global OH, its calculation and how well this single number represents the oxidizing capacity of the atmosphere. It addresses an important issue of atmospheric chemistry modeling (how should one average OH?). Four different weightings are considered and the weighting with the reaction of  $CH_4$  is selected as being most representative of the oxidizing capacity of the atmosphere. Mass or volume weighted [OH] is considered less adequate for comparing models or as an indicator of the oxidizing capacity. The paper is clearly organized and the results are clearly presented. The authors finish with clear recommendations.

One might ask, however, if the lengthy discussion is necessary to back up the point



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the authors want to make. The amount of detail in which the matter is presented weakens the paper considerably since it makes the paper less accessible. Nevertheless, the paper is worth reading and more importantly, the recommendations should be followed by modelers. In order to stimulate further discussion (or possibly to modify these recommendations) I include in the next section some specific comments.

#### **Specific comments**

The terms "lifetime" and "turnover time" are used alternating. Formally, this is not correct. As outlined in Prather (J. Biogeochem. Cycles, 11, 393-400, 1997) (reference is missing), these two definitions have a different meaning. A "lifetime" is normally based on budgets (total content divided by loss rate), whereas the turnover time is related to loss frequency only. When output of global models is used, the lifetime can be calculated as the yearly averaged burden divided by the yearly integrated loss (a kind of bulk approach). Alternatively, the turnover time in a grid box can be calculated by averaging the loss rate (normally k.OH). From the paper (page 45, lines 23-24) I understand that the relevant time scale is calculated as the "rate of removal relative to the amount", i.e. the lifetime definition, see also eq. (3). However, I think that the authors in fact used the bulk approach mentioned before, since these values are normally standard monthly mean output (see page 52, line 24). Thus, although LX/X formally equals L (L being loss frequency and X being the burden) spatial and temporal averaging might lead to different answers. With respect to spatial averaging equation (3) shows that the global lifetime with respect to removal by OH equals the inverse loss frequency, weighted by the mass of X. Note that here the mass of X is used for the weighting in eq. 3, while the integrated loss (in the 'bulk' approach) is calculated by integrating k.OH.X with X and OH in molecules/cm3. The temporal averaging is mentioned only in Section 8. More specifically, the remark that a proper calculation is "beyond the storage capacity of many computing environments" shows that the authors failed to investigate this in detail. With a few lines of programming code and rerunning the computations, the relevant numbers can be easily calculated. For instance, the

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effect of co-variations of temperature and OH can easily be investigated by calculating the relevant co-variance in the model and comparing this term to the average. Storage capacity will not be the limiting factor. The authors should have admitted that they just did not take the effort to investigate the effects of co-variations. I am highly interested in the results of these additional calculations, although I expect the effects to be small.

Another effect related to the lifetime of the gases is the change in lifetime over time. The MCF distribution changed considerably in the last decade due to the phasing out of the emissions. Currently, MCF is rather well-mixed. However, in the early 1990's this was not the case and the higher concentrations were found near the surface where the emissions took place. What is the effect on the lifetime? What have been the MCF emissions in the model calculations?

A disturbing factor of the paper is the way the authors define the oxidizing efficiency of the atmosphere. Although the authors are probably aware of the fact that most OH is consumed by CO, this species is not mentioned in the paper. Instead, the oxidizing capacity is linked to  $CH_4$  only. Of coarse,  $CH_4$  is well mixed, which has certain advantages. But this gas does not tell the whole story! The rate constant of the CO + OH reaction is only pressure dependent and thus puts more weight on OH in the extratropical boundary layer. I wonder why the authors did not present  $[OH]_{GM}(CO)$ ? Probably model results differ considerably, due to effects of higher hydrocarbons, convection, and direct CO emissions. Recommendation 1 on page 59 ('global mean OH should be computed by weighting with the reaction with  $CH_4$ ') is therefore put too strongly in my opinion. Moreover, in combination with recommendation 2 it would suggest that methane oxidation fully explains the oxidation efficiency of the troposphere.

In my opinion, the oxidizing capacity is more properly defined as the rate of OH production (in molecules/year). Whether the OH molecules react with CO, methane, NO<sub>2</sub>, or hydrocarbons does not matter in this definition. Therefore, it forms a proper metric to express the capacity of the atmosphere to oxidize pollutants. Since transport of OH is unimportant due to its short lifetime, the OH production rate can also be defined locally. Therefore, I would like to add as a recommendation that the OH production rate is used

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as an additional quantity to compare different models. The production can be broken down in the same atmospheric compartments as suggested by the authors.

In section 6, the authors claim that the vertical distribution of  $CH_4$  oxidation has never been quantified clearly. In that respect, the reference to Krol and van Weele (Implications of variations in photodissociation rates for global tropospheric chemistry, Atmos. Environ., 31, 1257-1273, 1997) is missing. This paper shows that the lifetime of  $CH_4$  changes from 4.4 years near the surface to 48 years at 200 hPa.

In the same section the authors fail to mention the role of the ITCZ in the northsouth asymmetry of OH. In a two box analog of the troposphere, the OH maximum is located on average south of the ITCZ. As mentioned in the work of Montzka (2000) the mean location of the ITCZ north of the equator probably plays a role in the OH asymmetry that is inferred from MCF measurements.

In section 7, the authors define 12 atmospheric sub-domains. The horizontal borders are located at 30 N/S and the equator. This leads to domains of equal mass. In that respect, the vertical divisions could conveniently have been placed at 1000, 750, 500, and 250 hPa. With these division lines, the troposphere is divided into 12 boxes of equal air-mass.

The authors mention in section 8 the possible sensitivity of OH trends for the method of weighting OH. The methods of Prinn et al. (1995, 2001) and Krol et al. (1998) both use a scaling approach. This means that the spatial distribution of the MCF loss rate (governed by k xOH) is scaled with a time dependent (i.e. linear) factor until a best fit with the MCF observations is obtained. An OH trend is governed by the trend in this scaling factor. The method of OH weighting can therefore not be an explanation for the different trend estimates obtained by Prinn et al. (1995) and Krol et al. (1998). Note that Prinn et al. (2001) report now also a positive trend in the 1980s, due to changes in emission estimates and absolute calibration.

#### **Technical Corrections**

CH<sub>3</sub>CCl<sub>3</sub> has an official name: 1,1,1 trichloro-ethane, which should have been in-

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cluded at page 46, line 3.

Page 48, line 12: Sect. is used instead of Section.

Interactive comment on Atmos. Chem. Phys. Discuss., 1, 43, 2001.

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