

Reply

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In a comment on the paper by Krol *et al.* [1998], Prinn and Huang [this issue] compare the estimates of the linear OH trends from the Atmospheric Lifetime Experiment/ Global Atmospheric Gases Experiment/ Advanced Global Atmospheric Gases Experiment (ALE/GAGE/AGAGE) methylchloroform data using a recursive weighted least squares (Kalman) filter described by Prinn *et al.* [1995] with the OH trend obtained using the ensemble method by Krol *et al.* [1998]. We refer to these two papers as K98 and P95 hereafter. The comment interprets the difference between the estimated trends ($0.0 \pm 0.2\%$ per year estimated by P95 and $0.46 \pm 0.6\%$ per year estimated by K98) by a combination of different emissions and different treatments of the observations. More specifically, the comment claims that the polynomial fitting by K98 leads to a arbitrary deweighting of (1) the CH_3CCl_3 data at the beginning and particularly at the end of the time series, and (2) the CH_3CCl_3 data in the Southern Hemisphere relative to the Northern Hemisphere. However, the major differences between the two trend estimates are attributed to the different emission scenarios.

The comment is organized in three parts, the second and the third parts being additional comments on our first reply. For clarity reasons, we organize our reply in the same way as the comment.

1. First Reply

We will show in this first reply that our results (1) are not biased by the treatment of the observations, and (2) are not sensitive to the different emission scenarios used by Prinn and Huang. We will show that the differences are caused by differences in the estimation methods that are used by K98 and P95, respectively. It is quite important to differentiate between a zero trend and a positive trend in OH. In their comment, Prinn and Huang [this issue] compare the trend estimates of different models. Models generally calculate a negative trend when simulations of preindustrial times are compared to simulations of the modern atmosphere. Only when recent emission changes and/or stratospheric ozone loss are taken into account has a positive trend been inferred [Karlsdóttir and Isaksen, 2000]. It is therefore misleading to compare the OH trend that is derived from CH_3CCl_3 mea-

surements to the modeled OH change between preindustrial times and modern times.

First, we would like to correct some small inconsistencies in the K98 paper that are also mentioned in the comment. In the work of K98 the measurements and model-simulated mole fractions are fitted by the Legendre polynomials

$$\chi(t) = a_0 + \sum_{k=1}^{k_{\max}} N^k \frac{2^k k!}{(2k)!} a_k P_k \left(\frac{t}{N} - 1 \right), \quad (1)$$

where N is half the length of the time series obtained at the particular station (expressed in years) and t (years) runs from 0 to $2N$. To express the Legendre coefficients a_k in units of ppt year $^{-k}$ (as in the work of P95), we should multiply the coefficients presented in the comment (expressed in units ppt) by a conversion factor $N^{-k}(2k)!/2^k k!$. In the work of K98 we presented the coefficients that were multiplied with the incorrect conversion factor $N^{-k}2^k k!/(2k)!$. Therefore the coefficients given in Table 3 of K98 should be multiplied by $((2k)!/2^k k!)^2$. Note that the a_3 term for the Oregon station is also missing in the work of K98. We emphasize that this does not affect our results.

Prinn and Huang [this issue] also note that we augmented a 5% error to the standard deviation to account for uncertainties in the absolute calibration of the CH_3CCl_3 measurements. They correctly argue that these calibration errors are systematic in nature and should not be treated as random errors. However, the two-dimensional (2-D) model used by Prinn and Huang is tuned with observations of chlorofluorocarbons to reduce errors due to inaccurate large-scale model transport [Prinn *et al.*, 1995]. In contrast, our model has not been tuned toward such observations at the ALE/GAGE stations. Errors arise because the models are not able to simulate interannual variability. Also, the description of other transport processes (such as the exchange between the boundary layer and the free troposphere and the strength of the convection) may introduce errors. Since the nature of these model errors is unknown, we argue that the addition of a 5% random error in the measurements to account for these model errors is certainly not unrealistic. In this reply we present results in which this extra random error is removed, and we show the results for absolute calibration errors of $\pm 5\%$.

A more serious problem would be the deweighting that, according to Prinn and Huang [this issue], influences the trend estimate. First, we note that Prinn and Huang used our polynomial coefficients which they sampled to generate

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Table 1. Results Obtained for Various Methods of Data Treatment

Method	ΔOH , %	Trend, % yr ⁻¹	ΔMCF , %
<i>Krol et al.</i> [1998]	-4.8±0.7	0.46±0.09	5.5±3.7
M1 ^a	-4.9±0.2	0.47±0.03	3.3±1.2
M2 ^b	-5.0±0.2	0.46±0.03	5.8±1.2
M1 × 1.05	-11.1±0.2	0.58±0.03	11.5±1.1
M1 × 0.95	2.5±0.2	0.36±0.03	-4.2±1.3

^aM1 refers to filtering measurement and model with a polynomial (see text).

^bM2 refers to no filtering of measurements and model (see text).

so-called K98 "observations" and associated 1σ uncertainties. These observations are subsequently used in their least squares filtering approach instead of the true measurements. This approach is different from the approach followed by K98 in which the model results were also replaced by a polynomial fit. Therefore we do not use K98 "observations" in the way that is suggested in the comment.

To assess whether different data treatments lead to different estimates of the trend, we use two different methods. In the first method (referred to as M1) we still use the polynomial fit to smooth the data. However, we only adjust the measurement data to the polynomial but retain the 1σ errors of the original data. Thus, instead of the cost function that is given by equation (10) of K98, we now use

$$P = \sum_{s=1}^5 \sum_{t=1}^{N_s} \frac{(\tilde{x}_{c,s,t} - \tilde{x}_{s,t})^2}{2(\sigma_{s,t})^2} \quad (2)$$

in which $x_{s,t}$ denotes the measurements at time t at station s with error σ , x_c denotes the corresponding model estimate, and the tilde denotes a smoothing operation on the model and measurement data. The second method (M2) leaves out all data smoothing and simply uses the original data and 1σ uncertainties. Table 1 shows the results that are obtained with different treatments of the measurement data. The first row shows the results from K98. As in the work of K98, three parameters are optimized: (1) an OH offset ΔOH (%) which is used to scale the OH field used in the three-

dimensional model, (2) the 1978–1993 OH trend (% yr⁻¹), and (3) a scaling factor ΔMCF (%) which is applied to the initial CH_3CCl_3 field in 1975. All new results are calculated without the 5% absolute calibration error and a calibration factor of 1. The last two rows correspond to calibration factors of 1.05 and 0.95, respectively.

Obviously, the parameters are more strongly constrained compared to the K98 results because the 5% absolute calibration error is not included. Table 1 shows that the deduced trend is sensitive to the absolute calibration. Given a 5% calibration error [Prinn *et al.*, 1995], the estimated trend ranges between 0.36 and 0.58 % yr⁻¹. Furthermore, it is clear that the estimated trends are essentially insensitive to the data treatment.

Prinn and Huang [this issue] observe that their deduced trends are sensitive to the emissions used in the model. Using three different emission estimates (K98, P95, and PMK), in combination with the K98 "observations," they derive OH trends of 0.42, 0.06, and 0.34 % yr⁻¹, respectively. Table 2 lists the corresponding estimates that are obtained with our ensemble method without filtering the data (M2). The first two lines are taken from K98, and the last column denotes the minimum value of the cost function (equation (2)). We show the results for all three emission estimates. If we treat ΔMCF as a free parameter (as in the work of K98), we learn that the estimated trend is only slightly sensitive to the emission estimates (0.46±0.03 for K98, and 0.43±0.02 for P95 and PMK). This completely disagrees with the results of Prinn and Huang. How can this be understood?

Table 2. Results Obtained for Various Emission Estimates

Emission Estimate	ΔOH , %	Trend, % yr ⁻¹	ΔMCF , %	Cost Function ^a
<i>Krol et al.</i> [1998]+K98	-4.8±0.7	0.46±0.09	5.5±3.7	
<i>Krol et al.</i> [1998]+PMK	-3.8±0.8	0.42±0.10	3.3±4.0	
P95+M2	-4.2±0.2	0.43±0.02	-5.2±1.0	658
PMK+M2	-4.3±0.3	0.43±0.02	2.5±1.2	659
K98+M2 (Table 1)	-5.0±0.2	0.46±0.03	5.8±1.2	658
P95+M2 (fixed ΔMCF)	-1.8±0.1	0.16±0.02	10 ^b	739
PMK+M2 (fixed ΔMCF)	-3.0±0.1	0.30±0.02	10	680
K98+M2 (fixed ΔMCF)	-4.3±0.1	0.38±0.02	10	666

^aThe minimum of the cost function (equation (2)).

^bThese values have been kept fixed to 10%.

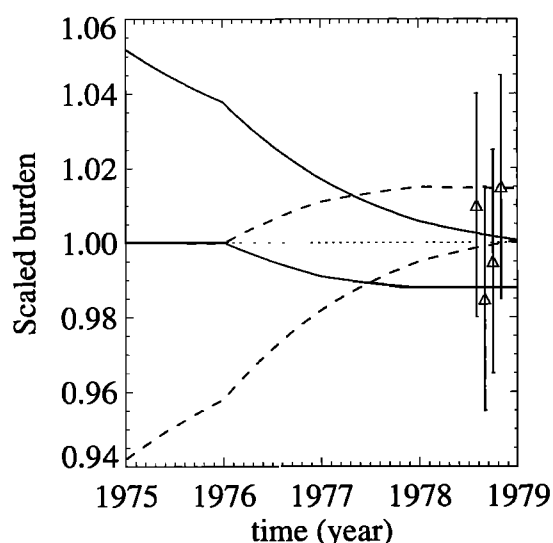


Figure 1. Scaled atmospheric CH_3CCl_3 burden between 1975 and 1979. The dotted line denotes the burden that is used for scaling. The line starts at the 1975 reference burden and refers to optimized emissions. The two dashed lines refer to P95 emissions and start at 1 and 0.948, respectively. The solid lines refer to K98 emissions and start at 1 and 1.058. Note that an initial 11% difference in burden is needed to obtain a comparable 1978 burden. The calculations have been performed with a box model with a CH_3CCl_3 turnover time of 4.5 years and serve only as an illustration.

In our ensemble method we have three free parameters. A scaling factor ΔMCF is applied to a 1975 CH_3CCl_3 field that is obtained in a 1951–1993 reference integration. We start our model integrations in 1975 and optimize ΔMCF . The CH_3CCl_3 concentration in 1978 (when the observations start) is sensitive to the emissions and removal (mainly by OH) before 1978. Since we want to optimize the OH levels and trend in the 1978–1993 period, we effectively avoid sensitivity for the emissions and OH prior to 1978 by optimizing the ΔMCF parameter. In contrast, *Prinn and Huang* [this issue] optimize only the OH level and trend which makes their results sensitive to estimates of emissions and OH prior to 1978. Since the main differences in the emission scenarios occur in the period 1975–1978, the sensitivity of our ensemble method can now be understood. For instance, the PMK emissions integrated over the 1975–1978 period are about 46 kt less than the P95 emissions. If the results for these two emission scenarios are compared, the only difference is found in the ΔMCF parameter, which is optimized to values of -5.2% and 2.5% for P95 and PMK emissions, respectively. The additional emissions in the 1975–1978 period of the P95 scenario are thus accounted for by a 7.7% lower CH_3CCl_3 burden in 1975. The minimum value of the cost function (the last column in Table 2) shows that both sets of optimized parameters compare equally well to the observations.

The agreement between observations and model results is less if the parameter ΔMCF is fixed to a value of 10%, as shown in the last three rows of Table 2. Moreover, in this

case the trend is sensitive to the emission scenario. Importantly, if we allow the system to optimize the initial concentration, the deduced trend remains unchanged compared to the K98 estimate.

Curiously, the matter of optimizing the initial condition has been debated earlier [*Cunnold and Prinn*, 1991; *Spivakovsky*, 1991]. That discussion centered around the optimization of both the OH level and the absolute calibration of the measurements. Similar to the results obtained here, it was shown by *Spivakovsky* [1991] that these estimates are sensitive to the assumed emissions before 1978.

2. Second Reply

In their comment, *Prinn and Huang* [this issue] (P01) discuss the results that are obtained in the first reply. They claim that our calculated corrections to the 1975 CH_3CCl_3 field (-5.2% for P95 and $+5.8\%$ for K98 emissions, see Table 2) are unrealistically high. Moreover, they claim that the optimization of the ΔMCF parameter is ill-conditioned. We will show here that our method is misrepresented by *Prinn and Huang* and that our optimization is not ill-conditioned.

2.1. Should We Optimize the Initial Condition?

The ALE/GAGE CH_3CCl_3 measurements started in July 1978. The atmospheric concentrations in 1978 depend on (1) emissions in the period prior to July 1978 and (2) sinks of CH_3CCl_3 , with OH oxidation being the main sink. CH_3CCl_3 concentrations are most sensitive to the emissions that occurred in the period prior to the measurements. The emissions in the 1970s are characterized by a large increase and also by large uncertainties. Figure 1 of P01 shows that the K98 emissions are significantly lower than the P95 emissions in most of the years in the 1970–1978 period. The cumulative effect of these lower emissions in the 1975–1978 period leads to a July 1978 concentration that is almost 3% lower (see Figure 1). It is easy to understand that the optimized OH level and trend will become sensitive to the chosen emission scenario when the initial condition is not optimized. More specific, lower pre-1978 emissions require less OH to obtain the same 1978 concentration. Consequently, a larger positive trend is calculated to maintain a good comparison with the measurements after 1978. These effects can clearly be observed in the work of P01 when the results of the P95 emissions are compared to the lower K98 emissions.

This brings us to the central issue of the current discussion: should we treat the different 1970–1978 emission scenarios as model uncertainty (as in the work of P01) or should we optimize the initial condition as is done by K98? We argue that the large uncertainty in the estimated emissions prior to 1978 justifies the optimization of an extra model parameter that is linked to these emissions. In this way, we avoid sensitivity of the OH trend and level for the uncertain emissions in this time period. Of course, the optimized emissions should fall within a range that can be justified by production and emission data from the industry. This latter issue will be discussed in section 2.4.

2.2. How Can the Pre-1978 Emissions Be Optimized?

Optimizing all annual emission figures prior to 1978 would clearly lead to an underdetermined system. One way to proceed would be the optimization of a factor that is multiplied with the pre-1978 emissions. In that case we would still require an initial condition. A method that is approximately equivalent to the optimization of the emissions is the optimization of the atmospheric burden at a specific time before the start of the measurements. We selected 1975, almost one CH_3CCl_3 turnover time before the start of the measurements in July 1978.

2.3. Is the Determination of ΔMCF Ill-Conditioned?

Prinn and Huang [this issue] mention two reasons why the determination of the ΔMCF parameter would be ill-conditioned. The first reason is that a small percentage difference in 1978 (3%) corresponds to a much larger difference in 1975 (11%), which is illustrated in Figure 1. If we start with the same reference burden in 1975, the difference in July 1978 would amount to 3% when the P95 emissions are applied instead of the K98 emissions. Now, we want to optimize the 1975 burden (ΔMCF) such that the best fit with the measurements is obtained. When this is done, we obtain values of -5.2% and $+5.8\%$ for the P95 and K98 emissions, respectively. The fact that a much larger difference is found in 1975 than in 1978 is caused by the short CH_3CCl_3 lifetime that diminishes concentration differences at a rate of about 5 years. It has nothing to do with an ill-conditioned nature of the optimization itself. Whether the obtained ΔMCF values are reasonable will be discussed in section 2.4.

The second reason is the factor of 14 range in the optimized ΔMCF values for the five ALE/GAGE stations. First, we note that the maximum and minimum scaling factors of the 1975 CH_3CCl_3 fields (see Table 5 of K98) amount to 1.111 (Oregon) and 1.008 (Tasmania). Thus the factor 14 reduces to much smaller proportions. Nevertheless, the more than 10% difference in the optimized ΔMCF values is rather large and has not been discussed in detail by K98. As mentioned earlier, modeling errors influence the results of the optimization. For instance, the Oregon CH_3CCl_3 simulations are systematically lower than the observations (see Figure 3 by K98). As a consequence, the optimized ΔOH is lower, and the optimized ΔMCF parameter is higher compared to the other stations, since both perturbations improve the correspondence with the observations. In contrast, a change in the optimized OH trend would alter the correspondence between model and measurements in a time-dependent way. Since the optimized trends are consistent for all stations, we deduce that the scatter in the optimized ΔOH and ΔMCF values for the different stations are likely caused by modeling errors and not by an ill-conditioned optimization.

As noted by K98, the optimized parameters are correlated. For the optimization presented by K98 the covariance matrix has the following elements: (1) -0.88 between OH trend and ΔOH , (2) 0.75 between ΔOH and ΔMCF , (3) -0.63 between OH trend and ΔMCF . When the 5% additional measurement error is removed, these values are somewhat closer to ± 1 .

These rather large covariances do not imply that the system is ill-conditioned. A system is ill-conditioned if the inverse of the covariance matrix cannot be calculated. A covariance of ± 1 between two parameters produces such an ill-conditioned matrix and implies that many combinations of the two parameters produce the same value of the cost function. In the work of K98 (Figure 6) we showed that the three parameters have a distinctly different effect on the modeled CH_3CCl_3 concentrations and that our optimization is therefore not ill-conditioned. The existence of the inverse covariance matrix emphasizes the validity of that statement.

2.4. Are the Obtained ΔMCF Values Reasonable?

Prinn and Huang [this issue] claim that our ΔMCF values cannot be explained by uncertainties in the emissions reported by the industry and uncertainties in pre-1975 OH values. Indeed, this point deserves more attention than given by K98. First, we note that the accuracy of the pre-1975 emissions should not be exaggerated. Although we are not specialists in estimating emissions, we observe that a mere 6 month shift in the pre-1975 emissions suffices to explain a -6% shift in the 1975 CH_3CCl_3 burden. Moreover, the two equally valid emission scenarios P95 and K98 differ by more than 5% in 1972, 1973, and 1976.

We noted earlier in this reply that the optimization of the ΔMCF parameter is equivalent to the optimization of the pre-1978 emissions. In other words, the ΔMCF parameter is also sensitive to the 1975–1978 emissions. Moreover, we noted with reference to Figure 1 that the forward difference in 1978 burden of 3%, caused by differences in the 1975–1978 emissions, is reflected in a 11% difference in the 1975 ΔMCF value.

The physical interpretation of the ΔMCF parameter of P01 is therefore a misinterpretation. This parameter also reflects the uncertainty in the emissions between 1975 and 1978, and the resulting 1975 concentration cannot be interpreted as a value that depends solely on the pre-1975 emissions and OH. One can easily select an arguably realistic 1975–1978 emission scenario (emissions that correspond to a ΔMCF value of 0) that is in perfect agreement with the pre-1975 emissions and OH levels. From Figure 1 it is clear that these optimized 1975–1978 emissions are higher than the K98 emissions, but lower than the P95 values.

3. Third Reply

In a third comment, *Prinn and Huang* [this issue] raise four additional points regarding our interpretation of the ΔMCF parameter. We deduced in our second reply that a realistic 1975–1978 emission scenario can be inferred for which the ΔMCF value equals to zero. Such a scenario would have higher emissions than the K98 scenario and lower emissions than the P95 scenario. We disagree with the interpretation of *Prinn and Huang* of our previous reply. Since further explanation may lead to further misinterpretations, we recalculated the OH trend and offset. Instead of optimizing the 1975 CH_3CCl_3 concentration field, we optimized a scaling factor (ΔEM) that is multiplied to the 1970–1978 emissions, as outlined in section 2.2. Further-

Table 3. Results of 1951–1993 Optimization

Method	ΔOH , %	Trend, % yr ⁻¹	ΔEM , ^a %
K98 (no 5%)	-5.2±0.2	0.45±0.02	0.6±0.3
K98 (5% error included)	-5.2±0.9	0.45±0.11	0.5±1.1
K98 (5% error + no ΔEM)	-5.4±0.6	0.49±0.09	— ^b
P95 (no 5%)	-4.1±0.3	0.40±0.03	-2.5±0.4
P95 (5% error included)	-4.1±0.9	0.42±0.10	-2.7±1.0
P95 (5% error + no ΔEM)	-2.0±0.6	0.21±0.09	— ^b

^a ΔEM denotes the percent change that is applied to the 1970–1978 emissions.

^b ΔEM has been kept fixed to 0%.

more, we carried out all integrations from 1951 onward to avoid further discussion concerning OH discontinuities and initial conditions. The scaling factor ΔOH is applied to the 1951–1993 OH field, and the OH trend refers to the 1978–1993 period. The Monte Carlo optimization technique is described by K98 and will not be discussed further. We use method M2 outlined in the first reply to compare the model to the measurements. In Table 3 we show the optimized parameters for all stations grouped together. Results are shown for P95 and K98 emissions, with and without optimizing the emissions, and with and without a 5% random error added to the measurements to account for model errors.

First, we observe that the optimized OH trend and offset are essentially identical to the earlier estimates. The optimized 1970–1978 emissions are about 0.6% higher when the K98 emissions are used. In contrast, it is calculated that with P95 emissions, the 1970–1978 annual figures have to be scaled down by about 2.5% to obtain the optimal fit between the model and the measurements. When these optimized ΔEM values are compared to Figure 1 of P01, we observe that the calculated difference of about 3% agrees very well with the differences between the K98 and P95 emission estimates. We thus can conclude that the optimization of the ΔMCF parameter is equivalent to the optimization of the pre-1978 emissions, as outlined in the second reply.

The addition of a 5% random error to the measurements to account for model errors results in the same optimized parameters. However, error margins are larger as noted earlier. It must be mentioned, however, that the optimized parameters for the individual stations are less consistent than with the inclusion of a 5% error. For instance, the trend estimates range between $0.23 \pm 0.08\%$ yr⁻¹ for Ireland and $0.49 \pm 0.03\%$ yr⁻¹ for Tasmania. We conclude therefore that an increase of the measurement error to account for model errors improves the consistency of our results.

Finally, when the 1970–1978 emissions are not optimized the results are significantly different. With P95 emissions the optimized trend reduces to 0.21% yr⁻¹, which is much lower than the value that is obtained when the 1970–1978 emissions are optimized.

4. Conclusion

The differences between P95 and K98 are not caused by different data treatment or emission scenarios, but are

mainly due to a different optimization method. We consistently deduce the same OH trend for all emission scenarios, mainly because we allow the system to optimize the initial CH_3CCl_3 concentration. Moreover, we showed that optimization of the initial CH_3CCl_3 concentration is equivalent to the optimization of the estimated pre-1978 emissions. For both methods the inferred trend and OH level are independent of the emissions and OH levels prior to 1978. All arguments raised against our optimization strategy are therefore invalid. We thus infer a trend that ranges between 0.35 and 0.58% yr⁻¹, depending on the absolute calibration ($\pm 5\%$). We therefore see no reason to modify the K98 trend of $0.46 \pm 0.6\%$ yr⁻¹. The (full range) error was obtained by perturbing the emission trends to extreme values [Prinn *et al.*, 1992].

Finally, the question whether the recursive weighted least squares (Kalman) filter, combined with a 12 box model [Prinn *et al.*, 1995], yields the same results as the ensemble method combined with a full three-dimensional model [Krol *et al.*, 1998] remains unanswered. This question can only be resolved if the optimization methods would be identical. In its simplest form our procedure minimizes the cost function (equation (2)) as a function of three free parameters. The fact that the various data treatments and optimization strategies provide identical estimates of the trend underscores the robustness of our method.

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References

- Cunnold, D. M., and R. G. Prinn, Comment on “Tropospheric OH in a three-dimensional chemical tracer model: An assessment based on observation of CH_3CCl_3 ” by C. M. Spivakovsky *et al.*, *J. Geophys. Res.*, **96**, 17,391–17,393, 1991.
- Karlsdóttir, S., and I. S. A. Isaksen, Changing methane lifetime: Possible cause for reduced growth, *Geophys. Res. Lett.*, **27**, 93–97, 2000.
- Krol, M. C., P. J. Van Leeuwen, and J. Lelieveld, Global OH trend inferred from methylchloroform measurements, *J. Geophys. Res.*, **103**, 10,697–10,711, 1998.

- Prinn, R. G., and J. Huang, Comment on "Global OH trend inferred from methylchloroform measurements" by Maarten Krol et al., *J. Geophys. Res.*, this issue.
- Prinn, R. G., et al., Global average concentration and trend for hydroxyl radicals deduced from ALE/GAGE trichloroethane (methyl chloroform) data for 1978-1990, *J. Geophys. Res.*, 97, 2445-2461, 1992.
- Prinn, R. G., R. F. Weiss, B. R. Miller, J. Huang, F. N. Alya, D. M. Cunnold, P. J. Fraser, D. E. Hartley, and P. G. Simmonds, Atmospheric trends and lifetime of CH_3CCl_3 and global OH concentrations, *Science*, 269, 187-190, 1995.
- Spivakovsky, C. M., Reply to comment by D. M. Cunnold and R. G. Prinn on "Tropospheric OH in a three-dimensional chemical tracer model: An assessment based on observations of CH_3CCl_3 ", *J. Geophys. Res.*, 96, 17,395-17,398, 1991.
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