Reply to anonymous referee #2:
We thank the referee for the thorough review of our manuscript. We have added a significant amount of new material and a more robust error estimate in the revised version, as requested by the referee. In this way, incorporating the referee comments has helped us to considerably strengthen the paper. The main conclusions, however, are unchanged. In the following, we give point-by-point replies (in red) to the referee comments (in black).

These experiments add to our knowledge of the deuterium kinetic isotope effects in the photolysis of formaldehyde. A new estimate of KIEmol is provided that is near other recent determinations. However, the authors argue that KIErad and KIEmol are likely not as different as described in the literature. As detailed below, these arguments lack an accounting of sources of statistical uncertainty, and a quantification of the overall effect of these factors on KIEmol and KIErad. It is likely that the error in KIEmol will be adjusted upwards by a significant amount, and, unfortunately, that the error in KIErad
will be so large that the result is not useful.

Reply: This point comes back below in the referee report, but we want to reply already at this stage to clarify the misunderstanding. Already in our present evaluation, KIErad is *not* constrained well by our measurements, and is *not used* for the further evaluation. However, KIEmol is relatively well constrained. Further, we consider the directly measured KIEtot from (Feilberg et al., 2007) the most reliable independent value (all the others were determined indirectly). So when KIEmol from this study is taken together with the KIEtot from (Feilberg et al., 2007), these two values are almost identical, in contrast to the published data. When these data are combined, if follows by mass balance that KIErad cannot be as different from KIEmol *at ~100 kPa* as found previously. This is now clearly stated.

In addition, there is a good mechanistic reason not to expect that KIEmol and KIErad should be the same. The mechanism of photodissociation is very different in these two channels. The quantum yield in the radical channel does not show a pressure dependence, while that in the molecular channel DOES. As described in ACPD 9, 24029, 2009, when the effect of pressure on KIEmol is removed, the low pressure KIEmol is actually equal to the (pressure independent) KIErad. So what is actually going on would seem to be the opposite, and there is no problem with the atmospheric pressure KIErad and KIEmol being different.

As indicated in the answer above, our conclusions are only valid for surface pressure and this has been restated at numerous places in the revised manuscript. New modeling results confirm that the very important observation of the pressure dependence of KIEmol by Feilberg et al, 2010 provides the missing parameter to explain HD in the stratosphere (Groß et al, in preparation). On the other hand, our recent box model results (Pieterse et al., 2009) and new unpublished results with a global CTM (Pieterse et al, in preparation) show that the with the current understanding of the isotope signatures of the H_2 sources, the global δD budget cannot be closed when KIEmol and KIErad at 100 kPa are as different as suggested by (Feilberg et al., 2007) or (Rhee et al., 2008). If the Feilberg values are used, the atmospheric δD is far too light (note that there is a difference of ~200 ‰ in the produced H_2 when KIEmol changes from 1.82 (Feilberg et al., 2007) to 1.63 (this work). However, if values that are based on the data presented here are used, modeled δD values match the measurements much better. Still, the pressure dependence of KIEmol is required to explain the stratospheric observations.

It is important to extract as much information as possible from the experiments, but not to overstate what has been learned. According to the discussion on page 25198 lines 11 to 13, the confidence interval was set at 0.03 'to account for possible differences between KIEmol and

KIErad.' It would be much better to have an error bar that is based on the uncertainties in the experimental data, the fits to the data and the sensitivity of the model. I do not find the article fit for publication; after significant revision this may be possible. Please see further discussion below.

In the original manuscript the error bars are based on the mean squared difference (quantified as CHI^2) between measurements and model. This approach is described in more detail in the revised version. KIEmol and KIErad are free parameters. For a given set of the other parameters (e.g. starting concentration and isotopic composition) KIEmol and KIErad are varied such that the mean squared difference between model and measurements is minimized. These minimized mean squared differences are similar to the square of the estimated measurement uncertainty of $\sigma^2 \sim 25‰^2$, which supports this approach of error estimation.
In response to the referee request, we now perform additional model runs where we vary individual parameters and examine the sensitivity to potential errors in these parameters (amount of HCHO and H_2 used, and initial isotopic composition of the two).

25188, 6, 'the radical channel has only a second order effect' - this lacks precision. Second order in what? Effect on what? The radical channel accounts for 1/4 to 1/2 of formaldehyde photolysis, depending on actinic flux spectrum and choice of absorption cross section/quantum yield, and so at first examination, it is not a second order effect. Please clarify.

This is explained in the manuscript: The radical channel does not directly produce H_2, but it only changes the isotopic composition of the HCHO reservoir that is available to produce H_2 at a later stage via the molecular channel. Therefore, in the beginning of an experiment, the radical channel KIE has absolutely no effect, and in the course of the experiment its effect increases the more time the radical channel has to change the HCHO reservoir. We have changed the wording and called this an *indirect* effect.

25188,10, There are significant uncertainties in the experimental data as described below, and this is compounded by the uncertainty of the model. Therefore it may not be justified to conclude that 'it is likely that KIEmol and KIErad are not as different as described previously in the literature.' What is meant by 'likely'? Instead, please try to quantify this likelyhood and if it cannot be quantified it should not be highlighted as a main conclusion in the abstract.

As described in the first point above, this conclusion is based on combining results of KIEtot from (Feilberg et al., 2007) and the new result of KIEmol from this work. The term likely has been removed and the assumptions are stated clearly.

25188, 16, 'Since H2 production only produces H2O' - note that H2 USE produces H2O, whereas H2 production consumes H2O.
Sorry, corrected

25189, 1-2, water vapor in the stratosphere from H2 will also affect mid-latitude stratospheric aerosol loading, impacting albedo and perhaps more importantly, mid-latitude ozone depletion via hydrolysis of N2O5/heightened sensitivity to HOx and ClOx.
Included in the revised version

25189, 9, reason 2, rather than 'atmospheric oxidation reactions', in situ production is due to one single reaction, the photolysis of formaldehyde.
Correct, the term was chose to indicate that this results from oxidation of various compounds, HCHO photolysis is now specifically mentioned.

25191, 2, please include error in KIEOH
25191, 6, please include error in KIEmol and KIErad
25191, 8, please include error in KIEmol and KIErad
All included in the revised version

25191, 10, ’in none of the experiments this KIE was measured directly’. The argument is good but please be specific about whether this refers to KIEtot, KIEmol or KIErad. Note that Feilberg et measured KIEtot directly.
This has been clarified now by stating KIErad.

25192, 2, It is more usual to use ’matrix’ for solids. ’Bath gas’ is the accepted term.
OK

25192, 9, The starting material was paraformaldehyde. I assume this was vaporised by heating with a flame? (Please describe). It is common that hot formaldehyde recondenses in the inlet, before it reaches the chamber. This can cause two kinds of problems. One is simply that the initial concentration of formaldehyde in the chamber is not as high as would be prediced based on the mass of paraformaldehyde. The second is that the inlet then serves as a hidden source of additional formaldehyde throughout the experiment. Please describe how the gas phase concentration of formaldehyde was determined since this is a central peice of information used to calculate the main results. Was it by FTIR, or UV-DOAS, or ?? If so, what cross sections were used to calibrate the measurements? UV and IR cross sections have recently been redetermined by Gratien et al. JPCA 2007, resulting in some cases in a significant revisions. Due to the difficulty of working with formaldehyde even a very good spectroscopic measurement would be accurate to 5 %, probably more like 10% and possibly more than 20%. How does this error propagate through the model?

To address this comment, we have added the following text in the manuscript:
A weighted amount of solid para-formaldehyde was gently heated with a heat gun until it was pyrolyzed completely into a stream of high purity N_2 (>99.999%), which flushed the gaseous HCHO into the chamber. The inlet was heated and flushed with N_2 after the pyrolysis was complete to achieve a quantitative injection.
A commercially available instrument (AL4001, Aerolaser GmbH, Garmisch-Partenkirchen, Germany) was used for HCHO measurement. The applied technique transfers gaseous HCHO quantitatively into the liquid phase and derivatizes it via the Hantzsch reaction to yield a dye which is fluorimetrically detected (e.g.(Wisthaler et al., 2008)). The instrument was calibrated using liquid HCHO standards. The accuracy of the HCHO measurement was 5 % (accounting for the individual 1-σ errors of the slope of the calibration curve, the flow measurements and the stripping efficiency of HCHO). Zeroing signals were obtained by passing the sampling air through a filter cartridge containing a Hopkalite catalyst.
The HCHO injection procedure has been successfully used, tested and described in previous studies (Brauers et al., 2007;Wisthaler et al., 2008). These studies show that the absolute value and temporal evolution of the HCHO concentration as measured in the SAPHIR chamber can be reproduced very well by the model using the weighted amount of paraformaldehyde added and the known dilution rate (e.g., Fig. 2 of(Brauers et al., 2007)). This applies also for other trace gases as shown in (Apel et al., 2008)). In the present study, the black model curves in Figure 1 are based on the weighted amount of HCHO injected, and this yields good agreement with the measurements.

25193, 6, Did these measurements include the actinic flux spectrum? (Is this what is meant by photolysis frequencies?) That would be very valuable, in order to predict the quantum yields in the molecular and radical channels. Otherwise this partitioning is based on assumptions. The actinic flux spectrum will change from day to day due to the solar zenith angle and atmospehric conditions such as cloudiness. The double layer of teflon will very likely change the spectrum, especially in the UV. What then is the error in the predicted quantum yields in the molecular and radical channels, and how does this error affect KIErad and KIEmol?

Yes, actinic flux spectrum was monitored throughout the experiments in order to calculate photolysis frequencies, which are used as input to the model, as explained in (Bohn and Zilken, 2005). This is now explicitly stated in the text. We test for sensitivity to the photolysis frequencies for the molecular and radical photolysis channel by using different recommendations of the quantum yields. The results in the Table show that the model results are not very different when either the JPL or the IUPAC quantum yields are used. This suggests that the experiments are not strongly dependent on the choice of the precise values for the quantum yields. The influence of the Teflon wall is negligible.

25193, 6, Please present the data for the temperature, RH, pressure, photolysis frequencies, ozone, nitrogen oxides, carbon monoxide, HCs and HCHO, as supplementary information. This is important for evaluating the model. For example the chamber will produce OH and HO2, and these radicals will interfere with the experiment by consuming HCHO. How much will depend on UV flux, ozone and NOx, etc.

We have included and discussed additional figures with relevant measurement and model results. This includes examples of actinic flux spectra, photolysis frequencies, OH and HO2, as well as the relative removal strengths of the two photolysis channels and reaction with OH.

25193, 24, change ’5A’ to ’5 Å’.
OK

25194, 13 - 17, the error in determining [H2] is 2%, and the error in determining dD of the formaldehyde is 2%. How do these errors propagate?
In response to the referee comment, the individual errors that originate from various uncertainties have been assessed and are discussed in the revised version.

25194, 23, what is the error in the value 1.28 and how does this impact the results?
± 0.01 (now included), according to Feilberg et al, but since this channel is effectively suppressed, it does not affect the results.

25194, section 2.1 The model is central to this paper because it allows the main results, KIEmol and KIErad, to be estimated based on the experimental determinations. Therefore more detail must be provided concerning the model (as table of reactions or reaction diagram, and including model output, in supplementary information if necessary), most importantly the reactions concerning formaldehyde. Without this list it is not possible to check independently if the chemistry is correct. It is also important since additional reactions have been added to the model that these appear in the published paper so other scientists can build on this work.
In the revised version we include a table with the chemical scheme used in the model.

The authors state that the model includes the KIE of the reaction of formaldehyde with OH. What concentration of OH is predicted in the chamber by the model, and what fraction of the formaldehyde is being removed by this reaction? How accurate is the models estimation of OH - perhaps 50%? What is the uncertainty in KIEmol introduced by uncertainty in [OH] and KIEOH?

This is addressed in the revised version and the additional figures. The model results show that OH levels are strongly suppressed in the experiments by the addition of excess CO. Therefore, the removal by OH is less than 0.5% of the total removal of HCHO.

The reaction mixture included 500 ppm of CO to quench OH, and this may indeed prevent OH from reacting with formaldehyde - but the model will say for sure. The reaction is: CO+OH–>CO2+HfollowedbyH+O2+M–>HO2+MAlso,HCHO photolysis will produce H, and HCO. H will be converted to HO2 as above, and HCO + O2 –> HO2 + CO Therefore significant amounts of HO2 will be generated in the chamber, and HO2 is not quenched by CO. As described for example in Nilsson et al. 2007 (cf. Fig 2), HO2 reacts reasonably fast with formaldehyde giving formic acid as the main product. What is the HO2 concentration predicted by the model, was the HO2 + HCHO reaction included in the model, and how much of the formaldehyde is being removed by this reaction? What KIE is used for the HO2 + CH2O reaction? How will errors in these numbers affect KIEmol?

The role of this reaction is assessed in the revised version. HO2 and HCHO form a HO2-HCHO adduct that can further react with HO2. In order to quantify the loss of HCHO via this mechanism, it has been included in the model. It turns out that the loss is negligible. The important point is that the adduct dissociates with a time constant of 150 s^{-1} into the reactants again, so that the reaction with HO_2 is negligible for our experiments.

It is well known that the walls of reaction chambers produce formaldehyde, in particular when exposed to sunlight. For example the EUPHORE chamber produces ca. 1 ppb per hour (Zador et al., J. Atm. Chem. 2006; Feilberg et al. 2007b). There is no reason to think SAPHIR will be any different. Were measurements made of the background source of formaldehyde? It may not seem large however it does make a significant difference by the end of the experiment, when the integrated emission is larger and the concentration of formaldehyde is at its lowest. Feilberg found that the background source of formaldehyde gave a significant correction to the EUPHORE results. Assuming that the background source is 1 ppb per hour, how much would this change KIEmol? Please add to uncertainty estimate.

The wall source of HCHO is well characterized for the SAPHIR photochemical reactor and is roughly 0.75 ppb/h. This is negligible given the starting concentrations of ~500 ppb HCHO. Nevertheless, in response to the referee comment, the effect of the wall source for the present experiments has been explicitly examined in additional model runs. For this purpose, the isotopic composition of the wall source was varied between δD = 0 ‰ and δD = 300 ‰, which covers the entire range of (modeled) δD(HCHO) values as shown in Figure 1. The values are included in the extended table of model results and show that this leads to variations in values of KIEmol which are smaller than 0.002.

25196, 16, the authors note that 'the system is underdetermined, since we only have one measureable, dD(H2), but two unknowns, KIEmol and KIErad.' In addition, at 25197, 8 - 14, it is noted that the model is relatively insensitive to changes in KIErad, but somewhat sensitive to changes in KIEmol, the difference in sensitivity being a fac- tor of five. Further, at 25198, 7 it is argued that KIErad must be >= 1 because the ZPE of HCDO is lower than that of HCHO. While it is true that the ZPE of HCDO is lower it does not follow that KIErad must also be lower, for a few reasons. The ZPE argument rests on the reflection principle, see for example Yung and Miller's article in Science 1997. A key assumption for the reflection principle is that the system undergoes direct photodissociation. This is not the case for formaldehyde which undergoes an indirect dissociation confounded by multiple electronic surfaces and exit channels. A key piece of evidence for this is the highly structured UV absorption band. In addition formaldehyde has complicated dissociation dynamics with curve crossing and a competition between radical channel, molecular channel and collisional deactivation. Jumping over the discussion about how

these dynamical processes change with isotopic substitution one could ask how the process of photoexcitation, that is the rate of absorption of light, changes with isotopic substitution. Rather than rely on the ZPE model which is not accurate even for direct dissociations, it is better to simply look at the absorption cross sections of HCHO and HCDO in the UV which have been measured by Gratien et al. JPCA 2007. They find that 'The integrated UV absorption cross-sections of HCHO, HCDO, and DCDO are equal to within the experimental uncertainty.' The bottom line is that there is not sufficient experimental or theoretical knowledge to justify this assumption. This is even more important because this limiting value is what determines the authors best guess result that KIEmol = 1.63 and KIErad = 1. Given the uncertainties in the results and the insensitivity of the model to KIErad, it seems that the error bar of KIEmol must be increased, and that either nothing at all can be said about KIErad, or that it can only be said in connection with a large error bar.
We accept that the reasoning with ZPEs is not conclusive and thank the referee for pointing this out. Nevertheless, none of the prior studies has indicated that KIErad should be *inverse*. Only this case is excluded by the condition KIErad>=1, and we still think that this is reasonable given the published studies. It is true that the error bar for KIEmol would have to be increased if KIErad was assumed to be inverse, but in this case, KIEmol would even be lower.

25197, 10, 'This is because both channels remove HCHO at roughly compatible rates (60%;40%).' Please provide actinic flux spectrum and state which cross sections/quantum yields were used in this calculation. What is the error in this calculation (plus or minus 5%??); how much does the actinic flux change from one day to the next; what is the effect of this error on KIEmol?
As stated above and shown in the table, calculations were performed with two different sets of branching ratios between the molecular and the radical channel (JPL and IUPAC), and the result show that the present experimental setup is not very sensitive to the details of the quantum yields.

25197, 27, I see no reason other than convenience to assume that KIEmol = KIErad. In fact, the mechanisms of the two channels would seem to argue against them being equal.
Yes, this is now stated in the paper, this is indeed assumed for convenience as a starting point, and in view of the fact that the two published datasets disagree strongly. We could start with any other combination. It becomes clear later that the results are so insensitive to KIErad that it is not important with which value we start. We cannot find a value for KIErad that would lead to an agreement with the data for KIEmol values from either Rhee et al or Feilberg et al.

Further, this error estimate does not take into consideration the points that have been raised above which will clearly have an effect.
We provide a robust error estimate in the revised version.

25197, 29 to 25198, 1, 'do not agree on the magnitude or even the direction of the effect', please define what effect - the deviation of KIErad from KIEmol?
This has been clarified. The magnitude of the difference between KIErad and KIEmol or even which one of them is larger.

25198, 25 to 27, It cannot be concluded that 'it is impossible to quantitatively reconcile the results of Feilberg 2007b with the new datase' because quantatative error bars have not been presented in the present work. Given the sources of error detailed above, it may be that the conclusion of this study is for instance KIEmol = 1.63 plus or minus 0.16, and then this work and Feilberg are clearly in agreement.
We provide a robust error estimate in the revised version.
The total error is slightly, but not strongly different from the value given in the original version.

Thus the original argument is still valid.

In addition for the reasons outlined above, I am not yet convinced that anything can be said about KIErad from this study, because of uncertainties in the experimental data, and of the data the model is based on, and even more because as the authors write the model is insensitive to KIErad and therefore we would not expect it to be able to provide a very accurate result.
As stated in the first point of reply, we do indeed *not* use the KIErad values from this study, but derive indirect information about KIErad from our value of KIEmol and the (Feilberg et al., 2007) value of KIEtot.

25199, 19, The authors note that dD(HCHO) was determined by the model, but was not measured in the photoreactor. Another reason to be concerned about the back- ground/chamber wall source of formaldehyde is that there is no reason to assume it has the same dD as the synthetic paraformaldehyde that was used. If by the end of the experiment 1/5 of the formaldehyde comes from the background source, and this formaldehyde has a dD value that differs from the synthetic formaldehyde by say 100 per mil, it is easy to see that this would affect KIErad and KIEmol.
As stated above, the HCHO wall source is of the order of 1% for the present experiments, and additional model runs where the isotopic composition was varied over a very wide range indicate that the effect of the wall source does not lead to a strongly increased error here.

25199 - 25200, Discussion. '..the values of KIEmol, KIErad and KIEtot reported in Feilberg 2007 are not consistent with the recommended relative photolysis yields of the molecualr channel and radical channel.' This is true if it is assumed that 60% of the photolysis is in the molecular channel, and 40% in the radical channel. However, by taking the actinic flux spectrum published in the supplementary information of Feilberg, in combination with the JPL cross sections and quantum yields, it is seen that the split is not 60/40, but rather 72/28. Using the correct numbers it is seen that the numbers do in fact make sense - they are consistent with recommended values and they are self consistent. This underlines the need to publish the SAPHIR actinic flux spectrum with this paper, and shows that any uncertainty in the 60/40 branching will be associated with an uncertainty in KIErad and KIEmol. These uncertainties do not seem to have been included in the analysis.
We have specified the point according to the argumentation of the referee: The values of KIEmol, KIErad and KIEtot reported in (Feilberg et al., 2007) are not consistent with a 60% - 40% distribution between the molecular channel and radical channel.
It is not possible to present all the actinic flux spectra in the paper. They are measured with a temporal resolution of about 2 minutes and therefore hundreds of spectra were measured on each measurement day and are used as input to the model. Three examples are now shown in Figure 1 of the revised manuscript. Of course the actinic flux spectra vary with SZA, clouds, etc. These measurements are then combined with the absorption cross sections and quantum yields for the two channels from JPL and IUPAC, to calculate the total photolysis frequencies. This is described and shown in the revised version.

25201, 3 to 6, it does not seem that a reevaluation is called for.
We have removed this statement and modified the sentence to:
Whereas the discrepancy in the calculated branching ratios between the two studies should be investigate in detail, the effect outlined above may resolve at least part of the deviations between Feilberg et al., (2007) and the present study.

25202, 8, 'Whereas the partitioning between KIEmol and KIErad in Feilberg 2007 may be erroneous..' as written above, there is no evidence to support this statement.
We have modified this statement along the lines of the second last argument (deviation from the

60%-40% distribution). It seems necessary to solve this discrepancy in the future, but we have not been able to obtain the actinic flux spectra from the (Feilberg et al., 2007) study for comparison.

25203, 'Pieterse 2009 already noted that in a simple box model of the isotopic composition of atmospheric H2 it was difficult to close the isotope budget with the original values from Feilberg 2007.' Nor should one expect that that would be the case. It is erroneous to assume that results for KIEtot, KIEmol and KIErad obtained at atmospheric pressure can be used throughout the atmosphere. Please see ACPD 9, 24029, 2009 for a discussion of the pressure dependence of KIEmol.
Our new study using a global CTM (Pieterse et al, in preparation) show the same effect, and there the pressure dependence is included.

25209, Table 1, please include estimate of errors in these concentrations/delta values.
Error estimates for delta values were included, and error estimates for concentrations will be included.

25211, Figure 1, Please provide a better description of the experiments in section 2. What the reader learns is that the experiments were begun at 10.00 and ended at 17.00. By inspecting the figure however it looks like H2 was added already at 7.30 and HCHO at 9.30? Was the chamber open for the entire period from 10 to 17, or only for a portion of this time? What is the cause of the break in the HCHO and H2 concentration data at 16.00? If this is when the chamber was closed to sunlight, I do not understand why HCHO drops just after this event. One would expect rather that the formaldehyde would be lost more slowly in the dark.
The chamber was opened to sunlight at the point that HCHO concentrations start decreasing. And the flushing period starts when the decrease rate increases.

The lower right panel shows that there is very little difference between KIEmol=KIErad=1.63 and KIErad=1 KIEmol = 1.63. Given all the uncertainties, I do not find convincing evidence here for revising estimates for KIErad or arguing that either Feilberg or Rhee are in error. We know already that the two results do not agree - is there any additional information that this study can provide regarding KIErad, given no measurement of dD(HCHO), significant errors that have not been taken into account and a model that is insensitive to variation in KIErad?
The main finding of this study is that the derived value for KIEmol is lower than both the (Rhee et al., 2008) and (Feilberg et al., 2007) results. As stated above, the suggestion for a revision of the value for KIErad is based on a combination of the (Feilberg et al., 2007) KIEtot and the new KIEmol determination. This is clarified in the new version.

References:

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