**REVIEW 2**

*General comment:*

The manuscript is generally well written, easy to follow and the subject matter falls well within the scope of ACP. The affects on O3 RF are indeed significant and merit further work on this subject, but the experiments themselves are, in general, not novel (the same parameterization for this reaction already being used in the study by Cariolle et al. [2008]). My major criticism of this work is in the parameterization of the HNO3 forming channel of the HO2 + NO reaction, which (as stated in the on-line comment by Dr J.-F. Müller) does not take into account the H2O dependence that the reaction has been shown to exhibit (Butkovskaya et al. 2009). The importance of the HO2-H2O adduct on reactions such as the HO2 + HO2 reaction has been included in models for many years now. In this manuscript the authors should also investigate the importance of the HO2-H2O adduct on the HO2 + NO reaction. This work would also greatly benefit from detailed analysis of the global (tropospheric and stratospheric) O3 and NOy budgets. The affects that the O3 reductions have on the methane lifetime also need to be more quantitatively discussed. After consideration of these comments and those below, I feel this work will be suitable for publication in ACP.

**Reply:**

**The comment by Dr J.-F. Müller is well taken. As pointed out by G. Le Bras (online comment SC C5228), the parameterisation by Butkovskaya et al (2009) is strictly speaking valid only at 298K and 200 Torr. As stated in that comment, the pressure dependence is likely small, but it is not yet published. And the temperature effect is not yet documented. According to the group who did the measurements, the parameterisation is only valid near the surface, and we therefore leave the H2O-adduct for later studies. To meet the comments of the reviewer, we have included a discussion on the topic in the revised manuscript.**

**With respect to the novelty, the type of model experiments is similar to Cariolle et al (2008), however we extend that work significantly by looking at the effect of the reaction on changes since pre-industrial times, examining the radiative forcing effects and by using a three dimensional stratosphere/troposphere model – Cariolle et al used a 2D stratosphere/ troposphere model while their 3d model was limited to the troposphere.**

**Regarding the O3 budgets, this has been included as global numbers (see reply to reviewer 1).**

**We have also included budget numbers of tropospheric and stratospheric NOy, along with the O3 budget. The effect of reaction (2) is most pronounced in the troposphere, with small reductions in the stratosphere. It reduces tropospheric NOy by 4.5% in 2000 and 7.4% in 1850 simulations. The tropospheric reductions are due to increased HNO3 washout, as discussed in the text, while the small stratospheric reduction mainly comes from reduced upward transport. The increased tropospheric effect in the pre-industrial simulation comes about because lightning is the main source of NOy, and has the same source strength as in our 2000 simulation: Since the lightning source is located where the reaction has the largest effect, the relative change will be larger than for 2000.**

**As for the O3 effects on CH4; separating the effects leading to a change in CH4 is interesting; however, the main interesting point in this study is the total change in CH4 and OH. Whether the change in CH4 is primarily from the change in OH due to reduced NOx or through reduced O('D) from reduced O3 does not change the conclusion on the effect of the reaction.**

*Minor comments:*

p 14802 line 2: Change atmosphere to tropospheric.

**Reply: Done.**

p 14804 line 13: By reaction modelling I presume they mean quantum chemical modelling?

**Reply:**

**They call it quasiclassical trajectory calculations (QCT), which according to the IUPAC is "a procedure for calculating trajectories in which the quantization of the reactants is taken into account, but in which the course of the reaction is treated classically". We have specified this in the revised text.**

p 14807 line 2: I presume that the pre-industrial simulation was driven using the year 2000 wind fields. Climate models predict different temperature/humidity profiles between the preindustrial and the year 2000. Can the authors suggest what impact this might have?

**Reply:**

**The pre-industrial simulation was driven with 2000 wind fields. We have clarified this in the manuscript (section 3). 2000 was a year with colder than usual upper troposphere and lower stratosphere (Randel etal, 2004). A colder stratosphere is also expected in a future climate due to global warming. It may be that the uppermost troposphere also was colder than usual. This may suggest that 1850 should have been warmer, and the effect on reaction (2) would be smaller in 1850. If so, reaction (2) would be relatively more important for the change in O3 trend.**

p 14808 line 7: Has the latest kinetics of the NO2 + OH reaction been included in this study? Mollner et al. [2010] have shown that inclusion of updates to the kinetics of the NO2 + OH reaction can have important consequences for tropospheric O3. Inclusion of this update would be valuable or at least a comment on the potential impacts.

**Reply:**

**Our kinetics are in general taken from JPL 2006 when available, and otherwise from IUPAC, and we have added this information to the model description. The reported values from Mollner et al (2010) have not been considered in this study. It can be noted that JPL 2011 keeps the 2006 values.**

**We have included a comment on the Mollner study in the discussions. As they show, a lower reaction rate would increase OH and thereby O3 in polluted areas. Presumably, this would increase NOx, and could increase NO available for reaction (2). A thorough study is needed to make this clear.**

p14808 line 20: By how much has the flux changed? A table of key fluxes for both sets of simulations would be very useful.

**Reply:**

**The point is well taken. Although such studies are interesting, they are not necessary for understanding the effect of reaction (2) and its effect on the trend and radiative forcing. Also, producing the table would require all simulations to be done again, which is unfortunately not possible due to computational limits. We plan an update of previous studies of the chemical production and loss terms in the Oslo CTM2.**

p 14809 line 27: Again you should be able to diagnose what is going on here. Is the change in methane lifetime driven by changes in the tropical lower troposphere/upper troposphere/extra tropics? Some more diagnostics would be useful here.

**Reply:**

**Yes, it is driven by changes in OH in the tropical UTLS. The text is rewritten to make this clearer.**

p 14810 line 6: remove second “the”.

**Reply: Done.**

p 14810 line 7: remove first “and”.

**Reply: Done.**

**References:**

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Butkovskaya, N., Rayez M. T., Rayez J. C., Kukui A. and Le Bras G. : Water dependence effect on the HNO3 yield in the HO2 + NO reaction: Experimental and theoretical evidence, J. Phys. Chem. A, 113, 11327-11342, 2009.

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