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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heavy hydrogen in the stratosphere" *by* R. Roeckmann et al.

Anonymous Referee #1

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GENERAL COMMENTS

This is a well written paper that presents new research that will be of interest to a wide range of the ACP readership. The key result from this paper is that even though H2 mixing ratios may remain constant as CH4 is converted into H2O in net hydrogen cycling in the stratosphere, the isotopic composition of H2 may change. The analysis shows that the heavy isotope content of molecular hydrogen is increasing with altitude in the stratosphere. The arguments leading to the conclusions drawn are presented clearly and convincingly. This research paves the way for a better understanding of the hydrogen isotope budget of the stratosphere which will be vital when attributing sources of H2 in the stratosphere.

Other than a few specific comments listed below, I have no major criticisms of this paper and it is essentially ready for publication.

SPECIFIC COMMENTS

Page and line numbers referred to below are based on the file acp2003-071.pdf.

Page 2, line 9: I assume that this cycling includes short-term reactive species such as OH, HO2 and HCHO which contain hydrogen?

Page 2, line 14: Is this true? Do the sedimenting particles always evaporate again? Is it possible e.g. in the Antarctic that the ice crystals can fall as low as the surface without evaporating or sublimating?

Page 2, line 19: Your Figure 1 reminds me of Figure 8 in Novelli, P.C.; Lang, P.M.; Masarie, K.A.; Hurst, D.F.; Myers, R.; Elkins, J.W. (1999). Molecular hydrogen in the troposphere: global distribution and budget. Journal of Geophysical Research 104(D23): 30427-30444. Your figure is good but perhaps some value could be added by explicitly including the branching ratios (as in the Novelli figure) so that readers can have some idea of the likelihood of certain reaction pathways.

Page 3, line 1: Refer to Figure 1 here as this statement is well supported by the figure. While on this point, decreases in stratospheric ozone should favour the HCHO + hv -> H2 + CO path over the HCHO + hv -> H + HCO path right? Are there any implications for future stratospheric H2 levels (as we expect stratospheric ozone to recover over the coming decades) that should be commented on?

Page 3, line 8: The stratospheric water vapour increases observed by Oltmans and Hofmann [1995] cannot be explained by oxidation of increased stratospheric methane. This explains only about half of the observed 1%/year trend in lower stratospheric water vapour. I would therefore suggest rewording "has caused an increase in stratospheric water levels" as "is responsible for about 50% of the observed increase in stratospheric water vapour".

Page 3, line 15: Insert "nearly constant WITH ALTITUDE" just in case readers think you mean nearly constant with time.

Page 12: For interpretation of curvature in plots such as those shown in Figure 4, the

ACPD

3, S1465–S1467, 2003

authors are referred to Plumb, R.A.; Ko, M.K.W. (1992). Interrelationships between mixing ratios of long-lived stratospheric constituents. Journal of Geophysical Research 97(D9): 10145-10156.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3745, 2003.