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Impact of a hydrogen economy on the stratosphere and troposphere studied in a 2-D model

N. J. Warwick,¹ S. Bekki,² E. G. Nisbet,³ and J. A. Pyle^{1,4}

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[1] A switch from a fossil fuel to a hydrogen-based energy system could cause significant changes in the magnitude and composition of anthropogenic emissions. Model simulations suggest the most significant impact of these emission changes would occur in the troposphere, affecting OH. This impact is dependent upon the magnitude and nature of trade-offs in changing fossil fuel use. In the stratosphere, changes in water vapour resulting from expected increases in surface molecular hydrogen emissions via leaks occurring during production, transport and storage, are found to be significantly smaller than previous estimates. We conclude that the expected increase in molecular hydrogen emissions is unlikely to have a substantial impact on stratospheric ozone, certainly much smaller than the ozone changes observed in the last two decades.

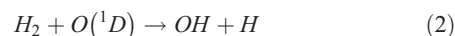
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1. Introduction

[2] Changing to molecular hydrogen as the principle currency of energy is seen as environmentally beneficial both in global and local terms [Prather, 2003]. Hydrogen could replace conventional fossil fuels at the point of use (e.g., in transport vehicles), thus reducing urban pollution. Hydrogen technology is well established in some industries and wider use in transport vehicles and fuel cells is expected within the next few years. The small hydrogen molecule is notoriously difficult to contain and leakage is likely to occur during production, transport and storage, and use as a fuel [Zittel and Altmann, 1996; Schultz et al., 2003].

[3] Today's hydrogen mixing ratio is about 0.5 ppmv in tropospheric regions removed from local sources. Sources of atmospheric hydrogen currently include biomass burning, fossil fuel combustion, and oxidation of methane and non-methane hydrocarbons [Novelli et al., 1999; Hauglustaine and Ehhalt, 2002]. Hydrogen leakage from microbial

production in landfills is poorly quantified, but may also be an important source. Loss of atmospheric H₂ is dominated by deposition to soil surfaces and chemical reactions with hydroxyl radicals and, to a lesser extent, O(¹D):



The magnitude of H₂ leakage in a hydrogen economy is presently very difficult to quantify as it will depend both upon the leakage rate into the atmosphere (which might be cut by engineering improvements) and on the extent to which hydrogen replaces fossil fuels. However, a simple estimate of the magnitude of H₂ emissions resulting from a switch to a hydrogen economy can, of course, be made. Our calculations assume: (1) lower and upper leakage rates of 1% and 12%, (2) that 2.6×10^{15} g H₂ will deliver sufficient energy for a global hydrogen economy, and (3) that hydrogen will supply between one third to all of the present-day energy consumption. The choice of 2.6×10^{15} g H₂ is a best guess scenario derived from a review of the available literature [Zittel and Altmann, 1996; Tromp et al., 2003]. These assumptions imply that tropospheric H₂ mixing ratios will increase by between 10% and 330% (provided there is no substantial change in the sinks of H₂). Today's reported losses of gaseous and liquid hydrogen are below 1% and 10% respectively [Zittel and Altmann, 1996], suggesting that any future increase in H₂ mixing ratios would be in the lower half of the above range. In addition, the high end-use efficiency of H₂ may encourage emission reduction. If better engineering containment and hence smaller leakage rates were to be achieved, or if other sources of hydrogen, such as tropical biomass burning, fossil fuel combustion and oxidation of non-methane hydrocarbons were reduced, then future global anthropogenic H₂ emissions in a hydrogen economy could potentially be less than today.

[4] A preliminary assessment of the potential impact of a hydrogen economy on atmospheric chemistry was performed by Tromp et al. [2003]. This study (hereafter referred to as TR2003) calculated the expected increases in stratospheric water vapour resulting from increased concentrations of atmospheric hydrogen and speculated on the possible implications of these changes on stratospheric temperature and ozone. A fuller assessment of the impact of a hydrogen economy on atmospheric composition needs, at least, (a) to account for the expected trade-offs associated with the switch to a hydrogen economy, (e.g., the concurrent reductions in carbon monoxide, methane and nitrogen oxides emissions from the fossil fuel industry, see the recent study of the tropospheric impact of this shift by Schultz et al. [2003]) and (b) to calculate explicitly the coupling

¹Centre for Atmospheric Science, Cambridge University, Cambridge, UK.

²Service d'Aéronomie du CNRS, Institut Pierre-Simon Laplace, Université Paris VI, Paris, France.

³Royal Holloway, University of London, Egham, UK.

⁴NCAS Atmospheric Chemical Modelling Support Unit, Cambridge, UK.

Table 1. Surface Emission Variables in the Base-Line Emission Scenario and the Six Hydrogen Emission Scenarios

Model Experiment	H ₂ /ppmv	CO/Tgyr ⁻¹	NO _x /Tgyr ⁻¹	CH ₄ /Tgyr ⁻¹	NMHCs/Tgyr ⁻¹
BASE	0.55	680	51.4	525	160
1	2.3	680	51.4	525	160
2	1.4	680	51.4	525	160
3	1.4	380	51.4	525	160
4	1.4	380	18.4	525	160
5	1.4	380	18.4	440	160
6	1.4	380	18.4	440	90

Except for hydrogen which has a prescribed global mixing ratio in the surface level, all emissions have prescribed global and seasonal variations (for further details see *Bekki and Law* [1997]).

between temperature changes and changes in all the radiatively active gases, not only H₂O, but also CH₄ and O₃. In this study, we make progress towards the fuller assessment using an interactive global 2-dimensional model of the atmosphere.

2. Model Description and Scenarios

[5] The 2D model in this study has been used previously for a variety of atmospheric studies and its basic formulation is described elsewhere [*Harwood and Pyle*, 1975; *Law and Pyle*, 1993a, 1993b]. It extends from pole to pole and from the ground to 60 km with a horizontal resolution of 9.5° and a vertical resolution of half a pressure scale height (~3.5 km). The model contains detailed representations of atmospheric chemistry (including heterogeneous reactions on sulphuric acid aerosols), transport and emissions of CH₄, CO, NO_x, and NMHCs. Atmospheric circulation is calculated from forcing terms which include solar heating in the stratosphere by O₂ and O₃ and longwave heating in the stratosphere by CO₂, H₂O, O₃, CH₄ and N₂O [*Haigh*, 1984; *Bekki et al.*, 1996]. Tropospheric heating rates and surface temperatures are specified.

[6] To investigate the potential impact of switching to a hydrogen economy on atmospheric chemistry, the model is run for a base-line emission scenario (representing today's emissions from the energy industry) and six possible illustrative hydrogen economy scenarios. These scenarios are designed to assess the influence of various trade-offs in fossil fuel and hydrogen emissions on atmospheric composition and are described in Table 1. Each emission scenario is run for a 20 year period to allow the model to reach equilibrium and the results are taken from the final year of this integration. In the first hydrogen scenario, H₂ mixing ratios at the surface level of the model are increased from 0.55 ppmv to 2.3 ppmv, as in the TR2003 study. This quadrupling of H₂ mixing ratios is the expected effect of highly enhanced H₂ emissions resulting from a hydrogen energy economy equivalent to today's total fossil fuel energy and assuming a relatively high H₂ leakage rate (e.g., ~12%). It should be noted that so high a leakage rate is unlikely to occur on a large scale, because it is both unsafe and, tellingly, expensive [*Schultz et al.*, 2003]. There would be powerful pressure for development of better containment. However, it is useful to consider extreme assumptions for an H₂ economy and it aids comparison of our model results with the TR2003 study. The absence of trade-offs in this experiment (fossil fuel emissions remain unchanged) makes it unreasonable to consider it as a plausible worst-case scenario because the rationale for the

development of a global hydrogen economy includes potential reductions in fossil fuel consumption. The results of Experiment 1 therefore correspond to an extreme. We view them as the maximum possible impact of switching to an hydrogen economy.

[7] In the second model scenario, hydrogen mixing ratios at the surface are raised to 1.4 ppmv, based on a smaller leakage rate of 5%. This scenario seems much more plausible than the first one. Further hydrogen economy scenarios (Experiments 3 to 6) include reductions in emissions of CO, NO_x, CH₄ and NMHCs. These emission reductions represent the maximum emission trade-off expected from the complete removal of the fossil fuel industry. Again, we regard these scenarios as extreme end-members of possible scenarios, designed to assess the sensitivity of the model to a range of changes in surface emissions. As production of hydrogen, at least in part, is likely to be from fossil fuels (although nuclear or renewable power represent viable alternatives), no scenario should be considered a complete representation of emissions in a future hydrogen economy.

3. Results and Discussion

[8] The results from these model experiments are summarised in Table 2. It shows the change in concentrations of H₂, H₂O, OH, O₃ and CH₄ for each of the model experiments, averaged over both the troposphere and stratosphere. The model tropopause is at ~7 km at middle to high latitudes increasing to ~14 km in the tropics. Raising surface hydrogen mixing ratios by 1.75 ppmv from 0.55 to 2.3 ppmv, increases average stratospheric mixing ratios by ~214%. In this scenario, stratospheric H₂O increases by up to 20% with maximum enhancements in the upper stratosphere at high latitudes. This percentage increase is significantly less than that found by TR2003, who reported

Table 2. Percentage Changes in Global Annual Mean Hydrogen, Methane, Water Vapour, Hydroxyl Radical and Ozone in the Troposphere (T) and Stratosphere (S) for Each of the Six Hydrogen Emission Scenarios Relative to the Base-Line Emission Scenario

Model Experiment	% Changes in:									
	H ₂		H ₂ O		OH		O ₃		CH ₄	
	T	S	T	S	T	S	T	S	T	S
1	313	214	0	6.9	-10.1	9.9	5.1	0.6	7.7	7.0
2	156	107	0	3.4	-5.5	5.1	2.7	0.3	4.1	3.7
3	156	106	0	3.0	1.4	4.3	-1.7	0.1	-1.6	-1.6
4	156	109	0	4.0	-10.3	6.1	1.8	0.6	11.8	10.6
5	156	105	0	2.3	-5.3	3.1	-1.8	0.1	-8.0	-8.0
6	156	105	0	2.3	-5.0	3.1	-2.2	0.1	-8.3	-8.3

increases of water vapour of up to 35% in the same region based on a similar experiment performed using the Caltech/JPL 2D model. To produce a 35% increase over a background concentration of about 6 ppmv in the upper stratosphere (representing an H_2O increase of at least 2 ppmv), given the magnitude of the 1.75 ppmv surface H_2 increment, would require additional increases in stratospheric CH_4 and complete oxidation of H_2 , stringent requirements. In our calculations, H_2 is certainly not completely converted to water vapour, even in the upper stratosphere. For example, the present-day simulation indicates a more or less constant H_2 mixing ratio throughout the stratosphere (the fraction of H_2 oxidised being somewhat balanced by the production from CH_4 oxidation), which is consistent with previous calculations [Letexier *et al.*, 1988] and the limited amount of observational evidence [Scholz *et al.*, 1970; Ehhalt *et al.*, 1975]. In addition to an increase in stratospheric water vapour, the model predicts significant increases in stratospheric OH and decreases in tropospheric OH ($\sim 10\%$) which does lengthen the lifetime of methane in the troposphere.

[9] If it is assumed that engineering quality is adequate to restrict the hydrogen leakage rate to 5% (Experiment 2), then the modelled average increase in stratospheric H_2 is limited to 107% (see Figure 1). In this case, the average increase in stratospheric water vapour is restricted to 3.4% with a maximum of 10% in the upper stratosphere. These changes are modest in comparison to those reported from balloon and satellite observations which suggest recent increases in H_2O of about 10–20% for the past couple of decades [Nedoluha *et al.*, 1998; Oltmans and Hofmann, 1995; Oltmans *et al.*, 2000; Rosenlof *et al.*, 2001]. It is already acknowledged that even these possible large changes in H_2O are a minor factor in the ozone depletion problem compared to the chlorine loading issue [Chipperfield and Pyle, 1998]. Chemistry-transport model simulations suggest a low sensitivity (less than 2%) of Arctic ozone loss to an increase of 1 ppmv of H_2O in the lower stratosphere (which is still larger than our calculated H_2O increases).

[10] The potential impact of the removal of fossil fuel use and consequent strong reductions in CO and NO_x is most

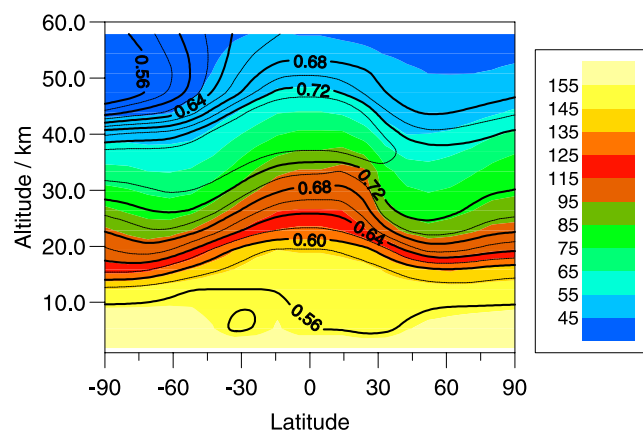


Figure 1. Simulated H_2 mixing ratios in January resulting from the base-line emission scenario (given by contours in units of ppmv) and the increase in H_2 mixing ratios resulting from a 160% increase of H_2 in the surface model level in Experiment 2 (given by colour in % change).

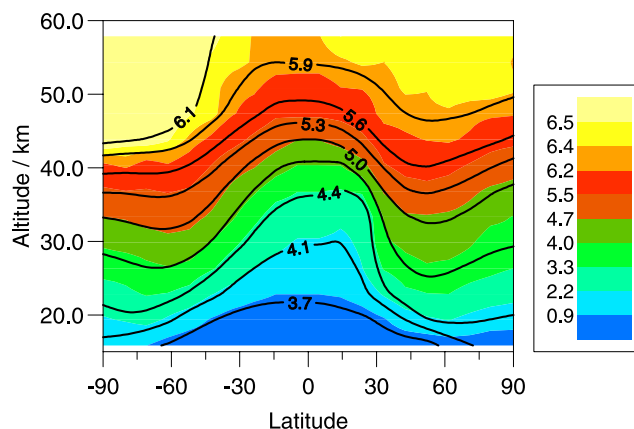


Figure 2. Simulated stratospheric H_2O mixing ratios for January in the base-line emission experiment (given by contours in units of ppmv) and the increase in H_2O mixing ratios in Experiment 5 resulting from a 160% increase of H_2 in the surface model level and the removal of CO, NO_x and CH_4 emissions from the fossil fuel industry (given by colour in % change).

apparent in the troposphere. In Experiment 4 (which includes raised surface H_2 mixing ratios and CO and NO_x emission reductions), the model calculates global mean tropospheric decreases in OH of -10.3% relative to the base-line emission scenario. These results are consistent with OH decreases calculated in a recent study focusing on the tropospheric impact of a hydrogen economy [Schultz *et al.*, 2003]. A decrease in tropospheric OH of this magnitude has important implications for the lifetime of methane and possibly other greenhouse gases. In this scenario, the decrease in OH results in an increase in atmospheric CH_4 of $\sim 10\%$. As methane contributes to stratospheric water vapour, this enhances the stratospheric H_2O increase relative to Experiment 2. The model also calculates a slight net increase in global mean tropospheric ozone, which is the averaged result of local decreases in O_3 throughout much of the Southern and Northern Hemispheres and some model-calculated increases in both OH and O_3 in latitudes north of 40°N , the result of strong NO_x reductions in the polluted model chemical regime there. (Note that the model's coarse resolution and the zonal averaging limit its ability to reproduce more local changes in the chemical regime.)

[11] When reductions in surface emissions of methane from the fossil fuel industry are also included in the emission scenario (Experiment 5), the model calculated maximum H_2O increase in the upper stratosphere is reduced to 6.5% (see Figure 2). The averaged stratospheric increase is 2.3% with even smaller increases in the lower stratosphere. In the troposphere, the effects of removing fossil fuel CH_4 emissions include an increase in tropospheric OH and a decrease in tropospheric O_3 relative to Experiment 4. Removing the fossil fuel emissions of NMHCs (Experiment 6) in the model did not have a significant additional effect on the atmospheric concentrations of the species considered in Table 2.

[12] In all model experiments, the changes in stratospheric ozone resulting from the recorded changes in emissions are small (less than 1%) and not significant compared to the magnitude of the ozone changes observed in the last two

decades. Changes in global average stratospheric temperatures are also small and range from -0.1 K to -0.3 K in the six model scenarios. Below 40 km, the changes in temperature are negligible and the greatest cooling (-1.5 K in Experiment 1) occurs in a very limited region at 60 km. The changes in temperature are found to be marginal in the lower stratosphere because the changes in H_2O are small in this region which is critical for the radiative balance of the atmosphere compared to the upper stratosphere. These changes are also significantly smaller than the stratospheric H_2O changes assumed by TR2003 (-0.5°C) to derive enhancements in polar ozone depletions resulting from a fourfold increase of H_2 .

4. Conclusions

[13] The model calculations outlined above predict significantly smaller increases in stratospheric water vapour resulting from expected increases in H_2 emissions than previous estimates. We conclude that a move from a fossil fuel-based energy industry to a hydrogen-based industry would most plausibly have little effect on the concentrations of stratospheric ozone. Leakage rates from a global H_2 energy industry as high as 12% result in a small change in stratospheric O_3 and an average stratospheric cooling of -0.3 K (with the largest temperature change at the top of the stratosphere). However, we find that switching to a hydrogen economy could significantly influence the chemical composition and oxidation capacity of the troposphere, depending on the magnitude of the trade-offs. Although some preliminary work has already been done using a 3-D tropospheric model [Schultz *et al.*, 2003], our results highlight the need for further investigation both by modeling (assuming a wide range of hydrogen economy simulations) and by improvements in global efforts to monitor atmospheric hydrogen. In this study, simulations are performed using a 2-D atmospheric model which has the benefit of allowing a variety of emission scenarios to be explored. Future model studies should include 3D simulations with a radiative feedback and also scenarios where CO_2 is varied. A reduction in fossil fuel burning would lead to reduced atmospheric CO_2 levels which would have a further radiative impact on the stratosphere. This change, and the subsequent impact of temperature changes on stratospheric ozone chemistry, might indeed be the most important factor for the stratosphere of the change to a H_2 economy and needs to be assessed.

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References

Bekki, S., and K. S. Law (1997), Sensitivity of the atmospheric CH_4 growth rate to global temperature changes observed from 1980 to 1992, *Tellus, Ser. B*, 49, 409–416.

- Bekki, S., J. A. Pyle, W. Zhong, R. Toumi, J. D. Haigh, and D. M. Pyle (1996), The role of microphysical and chemical processes in prolonging the climate forcing of the Toba eruption, *Geophys. Res. Lett.*, 23, 2669–2672.
- Chipperfield, M. P., and J. A. Pyle (1998), Model sensitivity studies of Arctic ozone depletion, *J. Geophys. Res.*, 103, 28,389–28,403.
- Ehhalt, D. H., L. E. Heidt, R. H. Lueb, and E. A. Martell (1975), Concentrations of CH_4 , CO_2 , H_2 , H_2O and N_2O in the upper stratosphere, *J. Atmos. Sci.*, 32, 163.
- Haigh, J. D. (1984), Radiative heating in the lower stratosphere and the distribution of ozone in a 2-dimensional model, *Q. J. R. Meteorol. Soc.*, 110, 167–185.
- Harwood, R. S., and J. A. Pyle (1975), A two-dimensional mean circulation model for the atmosphere below 80 km, *Q. J. R. Meteorol. Soc.*, 101, 723–747.
- Hauglustaine, D. A., and D. H. Ehhalt (2002), A three-dimensional model of molecular hydrogen in the troposphere, *J. Geophys. Res.*, 107(D17), 4330, doi:10.1029/2001JD001156.
- Law, K. S., and J. A. Pyle (1993a), Modeling trace gas budgets in the troposphere: 1. Ozone and odd nitrogen, *J. Geophys. Res.*, 98, 18,377–18,400.
- Law, K. S., and J. A. Pyle (1993b), Modeling trace gas budgets in the troposphere: 2. CH_4 and CO , *J. Geophys. Res.*, 98, 18,401–18,412.
- Letexier, H., S. Solomon, and R. R. Garcia (1988), The role of molecular hydrogen and methane oxidation in the water vapor budget of the stratosphere, *Q. J. R. Meteorol. Soc.*, 114, 281–295.
- Nedoluha, G. E., R. M. Bevilacqua, R. M. Gomez, D. E. Siskind, B. C. Hicks, J. M. Russell III, and B. J. Connor (1998), Increases in middle atmosphere water vapor as observed by the Halogen Occultation Experiment and the ground-based Water Vapor Millimeter-wave Spectrometer from 1991 to 1997, *J. Geophys. Res.*, 103, 3531–3543.
- Novelli, P. C., P. M. Lang, K. A. Maserie, D. F. Hurst, R. Myers, and J. W. Elkins (1999), Molecular hydrogen in the troposphere: Global distribution and budget, *J. Geophys. Res.*, 104, 30,427–30,444.
- Oltmans, S. J., and D. J. Hofmann (1995), Increase in lower stratosphere water vapour at a mid-latitude Northern Hemisphere site from 1981 to 1994, *Nature*, 374, 146–149.
- Oltmans, S. J., H. Vömel, D. J. Hofmann, K. H. Rosenlof, and D. Kley (2000), The increase in stratospheric water vapor from balloonborne, frostpoint hygrometer measurements at Washington, D. C., and Boulder, Colorado, *Geophys. Res. Lett.*, 27, 3453–3456.
- Prather, M. J. (2003), An environmental experiment with H_2 ?, *Science*, 302, 581–582.
- Rosenlof, K. H., et al. (2001), Stratospheric water vapor increases over the past half-century, *Geophys. Res. Lett.*, 28, 1195–1198.
- Scholz, T. G., D. H. Ehhalt, L. E. Heidt, and E. A. Martell (1970), Water vapor, molecular hydrogen, methane, and tritium concentrations near the stratosphere, *J. Geophys. Res.*, 75, 3049.
- Schultz, M. G., T. Diehl, G. P. Brasseur, and W. Zittel (2003), Air pollution and climate forcing impacts of a global hydrogen economy, *Science*, 302, 624–627.
- Tromp, T. K., R.-L. Shia, M. Allen, J. M. Eiler, and Y. L. Yung (2003), Potential environmental impact of a hydrogen economy on the stratosphere, *Science*, 300, 1740–1742.
- Zittel, W., and M. A. Altmann (1996), Molecular hydrogen and water vapour emissions in a global hydrogen energy economy, *Proceedings of the 11th World Hydrogen Energy Conference*, edited by T. N. Veziroglu et al., pp. 71–82, Schön and Wetzel, Frankfurt, Germany.

S. Bekki, Service d'Aéronomie du CNRS, Institut Pierre-Simon Laplace, Université Paris VI, Tour 15 Couloir 15-14, 4 Place Jussieu, F-75252 Paris Cedex 05, France. (slimane@aero.jussieu.fr)

E. G. Nisbet, Geology Department, Royal Holloway College, University of London, Egham, Surrey TW20 0EX, UK. (nisbet@gl.rhul.ac.uk)

J. A. Pyle and N. J. Warwick, Centre for Atmospheric Science, Cambridge University, Chemistry Department, Lensfield Road, Cambridge CB2 1EW, UK. (john.pyle@atm.ch.cam.ac.uk; nicola.warwick@atm.ch.cam.ac.uk)