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► **To cite this version:**

N. J. Warwick, Slimane Bekki, E. G. Nisbet, J. Pyle. Impact of a hydrogen economy on the stratosphere and troposphere studied in a 2-D model. *Geophysical Research Letters*, American Geophysical Union, 2004, 31 (5), pp.L05107. 10.1029/2003GL019224 . insu-02118937

**HAL Id: insu-02118937**

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Submitted on 3 May 2019

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

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Received 5 December 2003; revised 23 January 2004; accepted 4 February 2004; published 4 March 2004.

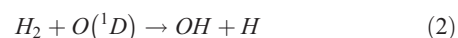
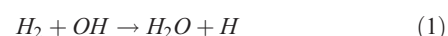
[1] A switch from a fossil fuel to a hydrogen-based energy system could cause significant changes in the magnitude and composition of anthropogenic emissions. Model simulations suggest the most significant impact of these emission changes would occur in the troposphere, affecting OH. This impact is dependent 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. *INDEX TERMS*: 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 1610 Global Change: Atmosphere (0315, 0325). **Citation**: Warwick, N. J., S. Bekki, E. G. Nisbet, and J. A. Pyle (2004), Impact of a hydrogen economy on the stratosphere and troposphere studied in a 2-D model, *Geophys. Res. Lett.*, 31, L05107, doi:10.1029/2003GL019224.

### 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 H<sub>2</sub> is dominated by deposition to soil surfaces and chemical reactions with hydroxyl radicals and, to a lesser extent, O(<sup>1</sup>D):



The magnitude of H<sub>2</sub> 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 H<sub>2</sub> 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  $2.6 \times 10^{15}$  g H<sub>2</sub> 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  $2.6 \times 10^{15}$  g H<sub>2</sub> 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 H<sub>2</sub> mixing ratios will increase by between 10% and 330% (provided there is no substantial change in the sinks of H<sub>2</sub>). 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 H<sub>2</sub> mixing ratios would be in the lower half of the above range. In addition, the high end-use efficiency of H<sub>2</sub> 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 H<sub>2</sub> 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

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

Model Experiment	H <sub>2</sub> /ppmv	CO/Tgyr <sup>-1</sup>	NO <sub>x</sub> /Tgyr <sup>-1</sup>	CH <sub>4</sub> /Tgyr <sup>-1</sup>	NMHCs/Tgyr <sup>-1</sup>
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 H<sub>2</sub>O, but also CH<sub>4</sub> and O<sub>3</sub>. 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 CH<sub>4</sub>, CO, NO<sub>x</sub>, and NMHCs. Atmospheric circulation is calculated from forcing terms which include solar heating in the stratosphere by O<sub>2</sub> and O<sub>3</sub> and longwave heating in the stratosphere by CO<sub>2</sub>, H<sub>2</sub>O, O<sub>3</sub>, CH<sub>4</sub> and N<sub>2</sub>O [*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, H<sub>2</sub> 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 H<sub>2</sub> mixing ratios is the expected effect of highly enhanced H<sub>2</sub> emissions resulting from a hydrogen energy economy equivalent to today's total fossil fuel energy and assuming a relatively high H<sub>2</sub> 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 H<sub>2</sub> 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<sub>x</sub>, CH<sub>4</sub> 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 H<sub>2</sub>, H<sub>2</sub>O, OH, O<sub>3</sub> and CH<sub>4</sub> 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 H<sub>2</sub>O 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

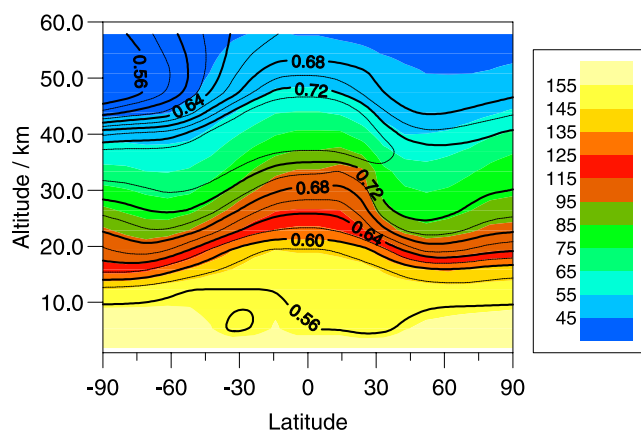
**Table 2.** Percentage Changes in Global Annual Mean Hydrogen, Methane, Water Vapour, Hydroxyl Radical and Ozone in the Troposphere (T) and Stratosphere (S) for Each of the Six Hydrogen Emission Scenarios Relative to the Base-Line Emission Scenario

Model Experiment	% Changes in:									
	H <sub>2</sub>		H <sub>2</sub> O		OH		O <sub>3</sub>		CH <sub>4</sub>	
	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

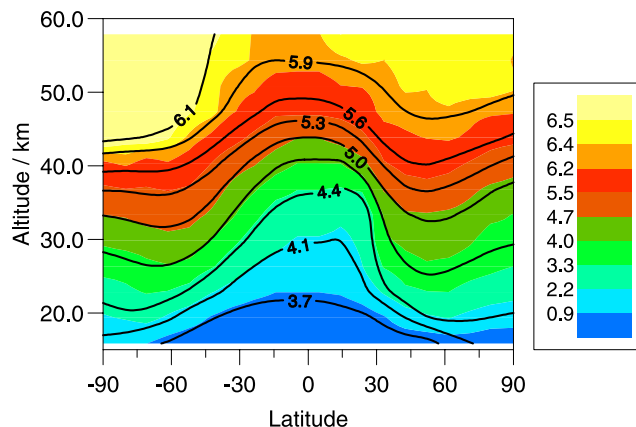
increases of water vapour of up to 35% in the same region based on a similar experiment performed using the Caltech/JPL 2D model. To produce a 35% increase over a background concentration of about 6 ppmv in the upper stratosphere (representing an  $\text{H}_2\text{O}$  increase of at least 2 ppmv), given the magnitude of the 1.75 ppmv surface  $\text{H}_2$  increment, would require additional increases in stratospheric  $\text{CH}_4$  and complete oxidation of  $\text{H}_2$ , stringent requirements. In our calculations,  $\text{H}_2$  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  $\text{H}_2$  mixing ratio throughout the stratosphere (the fraction of  $\text{H}_2$  oxidised being somewhat balanced by the production from  $\text{CH}_4$  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  $\text{H}_2$  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  $\text{H}_2\text{O}$  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  $\text{H}_2\text{O}$  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  $\text{H}_2\text{O}$  in the lower stratosphere (which is still larger than our calculated  $\text{H}_2\text{O}$  increases).

[10] The potential impact of the removal of fossil fuel use and consequent strong reductions in CO and  $\text{NO}_x$  is most



**Figure 1.** Simulated  $\text{H}_2$  mixing ratios in January resulting from the base-line emission scenario (given by contours in units of ppmv) and the increase in  $\text{H}_2$  mixing ratios resulting from a 160% increase of  $\text{H}_2$  in the surface model level in Experiment 2 (given by colour in % change).



**Figure 2.** Simulated stratospheric  $\text{H}_2\text{O}$  mixing ratios for January in the base-line emission experiment (given by contours in units of ppmv) and the increase in  $\text{H}_2\text{O}$  mixing ratios in Experiment 5 resulting from a 160% increase of  $\text{H}_2$  in the surface model level and the removal of CO,  $\text{NO}_x$  and  $\text{CH}_4$  emissions from the fossil fuel industry (given by colour in % change).

apparent in the troposphere. In Experiment 4 (which includes raised surface  $\text{H}_2$  mixing ratios and CO and  $\text{NO}_x$  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  $\text{CH}_4$  of  $\sim 10\%$ . As methane contributes to stratospheric water vapour, this enhances the stratospheric  $\text{H}_2\text{O}$  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  $\text{O}_3$  throughout much of the Southern and Northern Hemispheres and some model-calculated increases in both OH and  $\text{O}_3$  in latitudes north of  $40^\circ\text{N}$ , the result of strong  $\text{NO}_x$  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  $\text{H}_2\text{O}$  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  $\text{CH}_4$  emissions include an increase in tropospheric OH and a decrease in tropospheric  $\text{O}_3$  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  $\text{H}_2\text{O}$  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  $\text{H}_2\text{O}$  changes assumed by TR2003 ( $-0.5^\circ\text{C}$ ) to derive enhancements in polar ozone depletions resulting from a fourfold increase of  $\text{H}_2$ .

#### 4. Conclusions

[13] The model calculations outlined above predict significantly smaller increases in stratospheric water vapour resulting from expected increases in  $\text{H}_2$  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  $\text{H}_2$  energy industry as high as 12% result in a small change in stratospheric  $\text{O}_3$  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  $\text{CO}_2$  is varied. A reduction in fossil fuel burning would lead to reduced atmospheric  $\text{CO}_2$  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  $\text{H}_2$  economy and needs to be assessed.

[14] **Acknowledgments.** This research was supported by NERC and the EU THALOS project. The authors would like to thank P. Braesicke, NCAS-ACMSU, Cambridge for helpful discussions.

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