



Atmospheric composition and climate impacts of a future hydrogen economy

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Abstract. Hydrogen is expected to play a key role in the global energy transition to net zero emissions in many scenarios. However, fugitive emissions of hydrogen into the atmosphere during its production, storage, distribution and use could reduce the climate benefit and also have implications for air quality. Here we explore the atmospheric composition and climate impacts of increases in atmospheric hydrogen abundance using the UKESM1 chemistry-climate model. We find that increases in hydrogen result in increases in methane, tropospheric ozone and stratospheric water vapour, resulting in a positive radiative forcing. However, some of the impacts of hydrogen leakage are partially offset by potential reductions in emissions of methane, carbon monoxide, nitrogen oxides and volatile organic compounds from the consumption of fossil fuels. We derive a new methodology for determining indirect Global Warming Potentials from steady-state simulations which is applicable to both shorter-lived species and those with intermediate and longer lifetimes, such as hydrogen. Using this methodology, we determine a 100-year Global Warming Potential for hydrogen of 12 ± 6 . To maximise the benefit of hydrogen as an energy source, emissions associated with hydrogen leakage and emissions of the ozone precursor gases need to be minimised.

1 Introduction

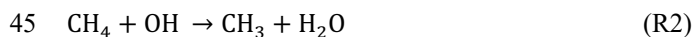
The adoption of low carbon hydrogen (H₂) as an energy source could lead to substantial reductions in carbon dioxide emissions and a significant climate benefit. However, fugitive emissions from a hydrogen economy will increase levels of hydrogen in the atmosphere which can further affect atmospheric composition through chemical feedbacks. It is important that the atmospheric implications of potential changes to hydrogen emissions are investigated in detail before the implementation of widespread hydrogen use.

The hydrogen abundance in the atmosphere increased over the twentieth century (Patterson et al., 2021), and today has a mixing ratio of around 530 parts per billion (ppb), with a small inter-hemispheric gradient (± 20 ppb) (Novelli et al, 1999, Patterson et al., 2021). Fossil fuel combustion and biomass burning account for approximately 50% of the current total global



hydrogen source, with the remainder arising from the oxidation of methane (CH₄) and volatile organic compounds (VOCs) in the atmosphere (e.g. Ehhalt and Rohrer 2009; Pieterse et al., 2013; Grant et al., 2010). Hydrogen removal is dominated primarily by uptake to soils and reaction with hydroxyl radicals (OH). OH is the main atmospheric oxidising agent and if hydrogen emissions were to increase, subsequent changes in the OH concentration could alter the lifetimes of important atmospheric greenhouse gases. Therefore, whilst hydrogen itself is not radiatively active, it can act as an indirect greenhouse gas.

An increase in the tropospheric concentration of hydrogen reduces the availability of OH via reaction R1. Reductions in tropospheric OH will result in increases in the atmospheric lifetime of CH₄ and its abundance (Derwent et al., 2020; Derwent and Field 2021) since the major atmospheric sink of CH₄ is through reaction with OH (R2).



The oxidation of both hydrogen and CH₄ in the troposphere can lead to the generation of ozone, with impacts for both climate and air quality (e.g., Archibald et al., 2020a; Schultz et al. 2003). In the stratosphere, oxidation of hydrogen and methane will lead to increases in stratospheric water vapour with potential implications for climate and stratospheric ozone (e.g., Tromp et al. 2003; Warwick et al., 2004). Increases in atmospheric hydrogen could also influence stratospheric ozone recovery through the production of HO_x radicals (=OH+HO₂), which are involved in ozone destruction cycles.

The extent to which future changes in hydrogen might affect atmospheric composition and climate will depend upon the level of hydrogen leakage and the ultimate size of a future hydrogen economy. In addition, emission reductions in species currently emitted by the production and consumption of fossil fuels, including methane, CO, NO_x (=NO+NO₂) and Volatile Organic Compounds (VOCs) (hereafter referred to as co-emissions), will also induce feedbacks on atmospheric composition (e.g., Jacobson et al. 2008). There is significant uncertainty in the size of a future hydrogen economy and the leakage rates and co-emission reductions are uncertain, depending on the forms of new technology implemented (e.g., Frazer-Nash 2022; Lewis 2021).

Here we report on calculations with the UKESM1 (Sellar et al., 2019) chemistry-climate model on the climate and atmospheric composition effects of a range of different atmospheric hydrogen boundary conditions, consistent with a range of different scenarios for a future hydrogen economy. We determine changes in effective radiative forcing (ERF) and present an improved estimate of the hydrogen Global Warming Potential (GWP), accounting for composition changes in both the troposphere and



65 stratosphere. As part of our GWP calculations, we present a refined methodology for calculating indirect GWPs from steady-state simulations, appropriate for use with both short-lived and longer-lived species.

2 Model and scenarios

2.1 The UK Earth System Model (UKESM1)

70 The U.K. Earth System Model, UKESM1 (Sellar et al., 2019), is a state-of-science Earth system model that couples Earth system modules to the HadGEM3-GC3.1 climate model (Kuhlbrodt et al., 2018; Williams et al., 2018). UKESM1 has a horizontal resolution of 1.875° in longitude and 1.25° in latitude, and 85 vertical levels extending from the surface to 85 km. Atmospheric composition changes are calculated using the UK Chemistry and Aerosols (UKCA) atmospheric chemistry module, which includes a coupled stratospheric-tropospheric chemistry scheme (Archibald et al., 2020b) and interactive 2-moment aerosol scheme (Mulcahy et al., 2020). UKCA includes the emissions of nine chemical species: nitric oxide (NO), carbon monoxide (CO), formaldehyde (HCHO), ethane (C_2H_6), propane (C_3H_8), acetaldehyde (MeCHO), acetone (Me_2CO), isoprene (C_5H_8) and methanol (MeOH), while surface mixing ratios of CH_4 , N_2O , CFC-11 ($CFCl_3$), CFC-12 (CF_2Cl_2), CH_3Br , H_2 and COS are prescribed. The photochemical sources and sinks of H_2 (and CH_4) are fully interactive, but the use of the Lower Boundary Condition (LBC) fixes the atmospheric burden of H_2 (and CH_4). LBCs are widely used in chemistry-climate models as they 1) allow the observed burdens of intermediate lifetime ($t > 1$ year) species to be imposed, bypassing the need to include emissions, and 2) remove the need for a long spin up for longer lived species burdens to reach a steady state. By using a LBC any chemical feedbacks that would affect the burden of these species are over-written. However, the effect of feedbacks on the steady state burden can be computed through quantification of the appropriate chemical feedback factors (see e.g., Heimann et al. (2020) for a discussion on how we have done this for methane).

2.2 Box model

85 Atmospheric box model simulations were performed using a coupled H_2 - CH_4 -CO-OH chemical scheme to link lower boundary condition values of H_2 with fugitive H_2 emission rates and obtain CH_4 lower boundary conditions for use in the UKESM1 experiments. In addition, the box model was used for extensive testing of the framework for GWP calculations.

Our box modelling approach is described in more detail in Warwick et al. (2022) and is similar to that described in Prather (1994) and used in other studies (Heimann et al., 2020; Nisbet et al., 2020). The model was initialised with realistic values for total methane emissions (585 Tg(CH_4) per year), CO (1300 Tg(CO) per year) and H_2 (80 Tg(H_2) per year) (e.g. Saunio et al., 2020; Pieterse et al., 2013; Zheng et al., 2019), and was found to give methane and H_2 levels in broad agreement with present day levels – 1865 parts per billion (ppb) CH_4 , 552 ppb H_2 and 101 ppb CO, as well as a CH_4 lifetime of 9.9 years, which is within the range of observationally-derived values.

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2.3 Hydrogen economy scenarios

2.3.1 Changes in the abundance of atmospheric H₂ in a hydrogen economy

The increase in abundance of atmospheric H₂ in a future hydrogen economy is not well constrained due to uncertainties in fugitive emissions, which will depend on the ultimate size of the hydrogen economy and H₂ leakage rates, as well as uncertainties in the amount of H₂ undergoing uptake by soils. To guide our model scenarios, we estimate how emissions of H₂ and other species may change in response to a switch from fossil fuel to hydrogen technologies in an illustrative hydrogen economy scenario. In this scenario, approximately 23% of global energy consumption (about 133 EJ) is supplied by hydrogen (BP Energy Outlook, 2020). We here assume that 100% of the final energy consumption of fossil fuels in the buildings sector switches to hydrogen, along with 50% of the final energy consumption of fossil fuels in the transport sector and 10% of the final energy consumption of fossil fuels in the power generation sector. The lower percentages for transport and power generation reflect a potentially smaller role for hydrogen in these energy sectors due to other low carbon alternatives such as electric vehicles and wind and solar power together with alternative storage options such as pumped hydro, batteries and compressed air (e.g. Staffell et al., 2019). New hydrogen technologies are assumed to have the same energy efficiency as the fossil fuel technology they are replacing (Staffell et al., 2019), except for in the transport sector where we assume diesel and petrol vehicles with an average tank-to-wheel energy efficiency of 30% will be replaced by vehicles using hydrogen fuel cells with an average efficiency of 50%. Using a net calorific value of 1 kg H₂ = 33.3 kWh, approximately 860 Tg H₂ yr⁻¹ would be required to provide the energy consumption outlined above.

Data on H₂ leakage rates is limited and H₂ leakage rates from the infrastructure associated with a hydrogen economy are uncertain (e.g. E4tech, 2019). However, H₂ leakage rates will likely be higher than for natural gas owing to the small molecule size of hydrogen, all else being equal. A recent study looking at the US natural gas supply chain indicated natural gas leaks of around 2.3% of gross gas production, which is ~60% higher than the US EPA inventory estimate (Alvarez et al., 2018). H₂ leakage rates of 1 and 10% (which represent the range of values used in previous studies e.g. Shultz et al. 2003, Warwick et al. 2004), would lead to additional H₂ emissions of 9 and 96 Tg H₂ yr⁻¹ respectively based on a hydrogen economy supplying 23% of total present day energy consumption.

We bypass some of the uncertainty in H₂ leakage rates, the ultimate size of the hydrogen economy and soil uptake rates, by performing a range of hydrogen economy scenarios with fixed H₂ lower boundary mixing ratios. We consider scenarios with H₂ lower boundary conditions ranging from 500 to 2000 ppb (i.e. increases relative to present day mixing ratios of 0 to 1500 ppb), which we believe span much of these uncertainties (see Table 1). For example, based on the size of a hydrogen economy outlined above, and assuming the magnitude of the soil sink increases in line with the increase in H₂ mixing ratios (i.e. a constant deposition velocity), box model simulations indicate H₂ lower boundary conditions of 750, 1000 and 1500 ppb represent H₂ leakage rates of about 3, 7 and 13% respectively (see Section 2.2 for further details on the box modelling).



130 However, these H₂ lower boundary conditions would also be consistent with other fractions of total energy supplied by H₂,
alternative soil sink responses and H₂ leakage rates. Use of this range of H₂ lower boundary conditions provides a clear signal
in the atmospheric response to increased H₂ mixing ratios relative to interannual variability, and allows us to explore the
linearity of the atmospheric response to increasing H₂. We regard our 2000 ppb H₂ scenario as an extreme end member designed
to assess the linearity of the atmospheric response, rather than a projection of potential future atmospheric H₂ levels in a
hydrogen economy.

135 2.3.2 Changes in CH₄, CO, NO_x and VOC emissions in a hydrogen economy

To determine the associated changes in CH₄, CO, NO_x and VOC emissions, further assumptions are required about the new
technology employed, in addition to the percentage of different energy sectors switching to H₂. In the buildings sector, we
assume that H₂ will be combusted and NO_x emissions limited such that they remain unaltered (despite the potential for higher
NO_x emissions from the increased flame temperature), whilst emissions of CH₄, CO and VOCs are eliminated. In the transport
140 sector, we assume a 50% reduction in emissions of CH₄, CO, NO_x and VOCs based on 50% of oil-based transport being
replaced by hydrogen fuel cells. In the power sector, we assume that stored hydrogen will be used to generate 30% of the
electricity currently generated using natural gas via combustion and NO_x will remain unaltered, whilst emissions of CH₄, CO
and VOCs are scaled appropriately. CH₄ emissions associated with energy production are also scaled according to the
decreased use of CH₄ where it has been replaced by H₂. The assumptions above lead to reductions in CH₄, CO and NO₂
145 emissions of 43, 259 and 19 Tg yr⁻¹ respectively (see Warwick et al. (2022) for further details).

Emission reductions for CO, NO_x and VOCs are determined by applying a uniform global scaling factor, based on the assumed
changes in fossil fuel usage, to emissions from the Gidden et al. (2019) energy sectors where hydrogen is assumed to play a
role. The emission reductions for CH₄ are converted to a change in the CH₄ lower boundary condition for the UKESM1
150 simulations using the box model (see Section 2.2). To separate the atmospheric impacts of increasing hydrogen mixing ratios,
the feedback on the CH₄ abundance and changes in the emissions of other species emitted by the consumption of fossil fuels,
four different sets of hydrogen economy model simulations are performed as described in Table 1.

2.3.3 Model simulations

For the BASE simulation, all boundary conditions are taken from the recently developed CMIP6 dataset and we assume an H₂
155 LBC of 500 ppb. Climatological boundary conditions for sea surface temperatures, sea ice extent and anthropogenic and natural
emissions are averaged over the years 2000-2014. In the first set of hydrogen economy scenarios, we consider changes to the
atmospheric H₂ abundance only. In these simulations, the methane abundance is fixed at 1835 ppb (2014 year level), and does
not respond to changes in the methane lifetime. In the second set of hydrogen economy scenarios, we include the methane
response to changes in atmospheric H₂ via the impact of H₂ on OH and the methane lifetime. The methane LBCs in these
160 simulations are determined via a series of box model simulations where H₂ is varied (see Section 2.2). In the third set of



hydrogen economy scenarios, we consider reductions in emissions of the ozone precursors CO, NO_x and VOCs from reduced fossil fuel use, whilst the methane abundance is fixed at 1835 ppb. The fourth set of hydrogen economy scenarios includes decreases in emissions of CH₄, CO, NO_x and VOCs from reduced fossil fuel use, in addition to the feedback of changes in the methane lifetime on the methane abundance. Each simulation was run for 40 years using annually repeating conditions, with the final 25 years of each simulation used for analysis and the initial 15 years treated as a spin-up period.

Hydrogen economy scenario	Experiment	H ₂ LBC (ppb)	CH ₄ LBC (ppb)	O ₃ precursor emissions
Present day conditions	BASE	500	1835	2000-2014 climatology
Present day CH ₄ and present day O ₃ precursor emissions	750H2	750	1835	2000-2014 climatology
	1000H2	1000		
	1500H2	1500		
CH ₄ feedback and present day O ₃ precursor emissions	2000H2	2000		
	1500H2_CH4f	1500	2058	2000-2014 climatology
	2000H2_CH4f	2000	2171	
Present day CH ₄ and reduced O ₃ precursor emissions	500H2_O3pre	500	1835	CO, NO _x and VOC emissions reduced
	1500H2_O3pre	1500		
	2000H2_O3pre	2000		
CH ₄ feedback and reduced O ₃ precursor emissions	500H2_CH4f_O3pre	500	1652	CH ₄ , CO, NO _x and VOC emissions reduced
	1000H2_CH4f_O3pre	1000	1756	
	2000H2_CH4f_O3pre	2000	1961	

Table 1. List of UKESM1 hydrogen economy simulations employing different H₂ lower boundary conditions (LBCs). The CH₄ LBC used in the simulations not including a CH₄ feedback is 1835 ppb (2014 year level). In simulations including a CH₄ feedback (CH₄f), the CH₄ LBC is determined using box modelling. Reductions in O₃ precursor emissions (O₃pre) are described in Section 2.3.2.



3 Results

3.1 Atmospheric composition impacts following changes in hydrogen and methane

175 To determine the indirect radiative impact of hydrogen, we need to understand how CH₄, O₃ and H₂O will change, for unit
emission of H₂. Those changes will depend not just on how H₂ changes but also on the changes of the co-emitted species. The
scenario space is particularly complex and our scenarios should not be regarded as predictions. Instead, here we try to indicate
generally how the composition of radiatively active species might change and which changes are linear in the change in H₂,
looking first at the changes driven by H₂ and CH₄.

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Figure 1 summarises the key composition changes seen for a range of different hydrogen lower boundary mixing ratios when
emissions of other species from the fossil fuel industry are held constant (blue and red circles) and when they are reduced
(orange and green circles). Decreases in OH, the main atmospheric oxidant, are modelled throughout the troposphere with
larger decreases in scenarios with higher H₂ abundances (Figure 1a), consistent with R1 being the main atmospheric sink for
185 H₂. The change in OH is linear in H₂ both when the surface CH₄ mixing ratio is held constant (blue circles) and when it is
increased to account for the feedback of changes in OH on the methane abundance (red circles). When including this methane
feedback, tropospheric mean OH decreases by about 0.90×10^5 molecules cm⁻³ (or about 10%) for every 1000 ppb (1 ppm)
increase in H₂ (Figure 1a, red circles). These modelled changes in OH go on to cause a cascade of further composition changes
by altering the methane lifetime, production and destruction rates of tropospheric ozone, as well as aerosol nucleation rates
190 and cloud condensation nuclei (CCN) concentrations.

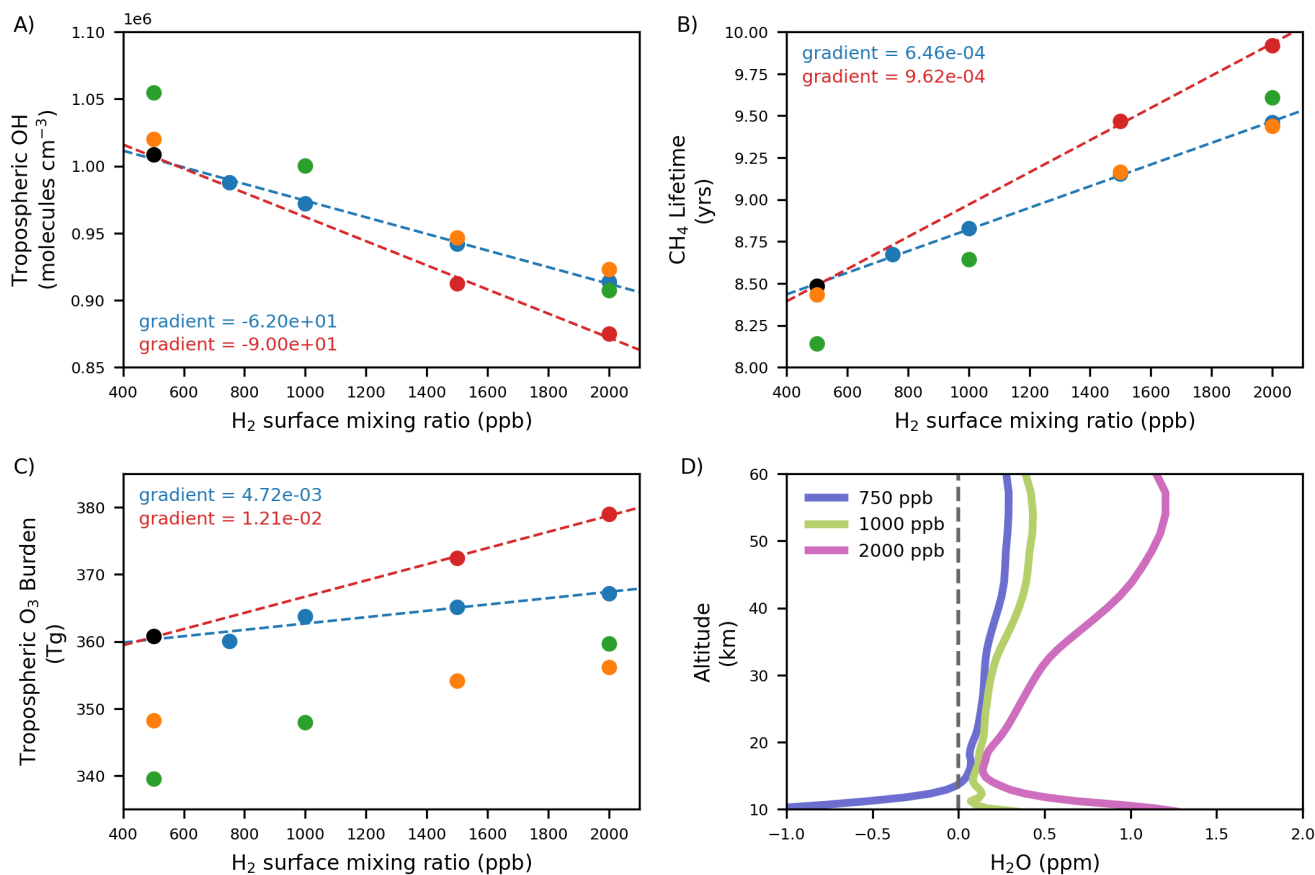


Figure 1. The response of (a) mass weighted tropospheric mean OH, (b) CH₄ lifetime, (c) tropospheric O₃ and (d) stratospheric water vapour to increasing H₂ mixing ratios in UKESM. In Figures 1a-c, the black circle represents the BASE scenario, blue circles represent scenarios where the CH₄ LBC remains fixed at 2014 levels, red circles are scenarios where the CH₄ LBC is adjusted to account for the change in CH₄ lifetime, orange circles are scenarios including changes in emissions of ozone precursors, and green circles include both changes to emissions of CH₄ and ozone precursors, as well as adjusted CH₄ LBCs to account for the response of the CH₄ abundance to changes in the CH₄ lifetime. The blue dashed line represents the fit through experiments where the CH₄ LBC remains fixed at 2014 levels, and the red dashed line is the fit through experiments which include the response of CH₄ to changing H₂ (and OH). Figure 1d shows the change in the vertical profile of H₂O relative to the BASE scenario where the H₂ LBC is increased to 750, 1000 and 2000 ppb (but CH₄ remains fixed at 2014 levels).

Figure 1b shows the CH₄ lifetime as a function of atmospheric H₂ and tropospheric OH. In the BASE simulation, the modelled CH₄ lifetime is 8.5 years, which is in good agreement with other studies (e.g. Stevenson et al. 2013, Prather et al. 2012) and with the lifetime simulated with our box model. As atmospheric H₂ increases but all other factors remain constant, there is a linear increase in the CH₄ lifetime. The methane lifetime increases by 0.64 years for every 1000 ppb (1 ppm) increase in H₂



when the methane abundance is held constant (blue circles), or 0.96 years for every 1000 ppb increase in H₂ when the methane abundance is increased in response to the modelled changes in its lifetime (red circles).

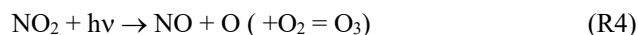
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The oxidation of H₂ in the troposphere can also affect the levels of ozone (O₃) as the HO₂ produced through R1 can catalyse the interconversion of NO_x which drives tropospheric ozone production (e.g., as reviewed in Archibald et al., 2020a):



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followed by photolysis of NO₂ to form ozone



220 In addition, changes in OH and HO₂ produced via R1 influence the destruction of tropospheric ozone by altering the flux through the reaction of O₃ with both of these species. Overall, the tropospheric ozone burden increases approximately linearly with atmospheric H₂ increase, driven primarily through increases in the reaction HO₂ + NO (R3), which increases by ~7% when atmospheric H₂ increases to 2000 ppb (Figure 1c). Changes in the ozone budget are documented in Table S1.

225 Increased H₂ mixing ratios at the surface can also influence stratospheric composition. Increases in H₂ and CH₄ abundance in the troposphere result in an increased flux of these species to the stratosphere, where their oxidation leads to the production of water vapour. Figure 1d shows the increase in the stratospheric water vapour profile relative to the BASE scenario when tropospheric H₂ increases from 500 ppb to 750, 1000 and 2000 ppb, but the tropospheric methane abundance remains unchanged. When H₂ increases to 2000 ppb, stratospheric H₂O increases by up to 25% (> 1 ppm). If the tropospheric methane
230 abundance is increased in line with modelled changes in the CH₄ lifetime, larger increases in stratospheric H₂O are simulated.

3.2 Atmospheric composition impacts when hydrogen, methane and ozone precursor emissions all change

The impacts discussed above arise from changes in atmospheric H₂ abundance and subsequent changes in methane abundance arising from the feedback of OH on the methane lifetime. Expected reductions in ozone precursor emissions of CO, NO_x (which may also increase locally), VOCs and CH₄ following a shift to a global hydrogen economy also have the potential to
235 further impact atmospheric composition. The green and orange circles in Figures 1a-c show the impact of including these other emission changes, with and without the feedback on the methane abundance included respectively. The relationship remains linear for the air mass weighted tropospheric mean OH, methane lifetime and tropospheric ozone burden plotted as a function

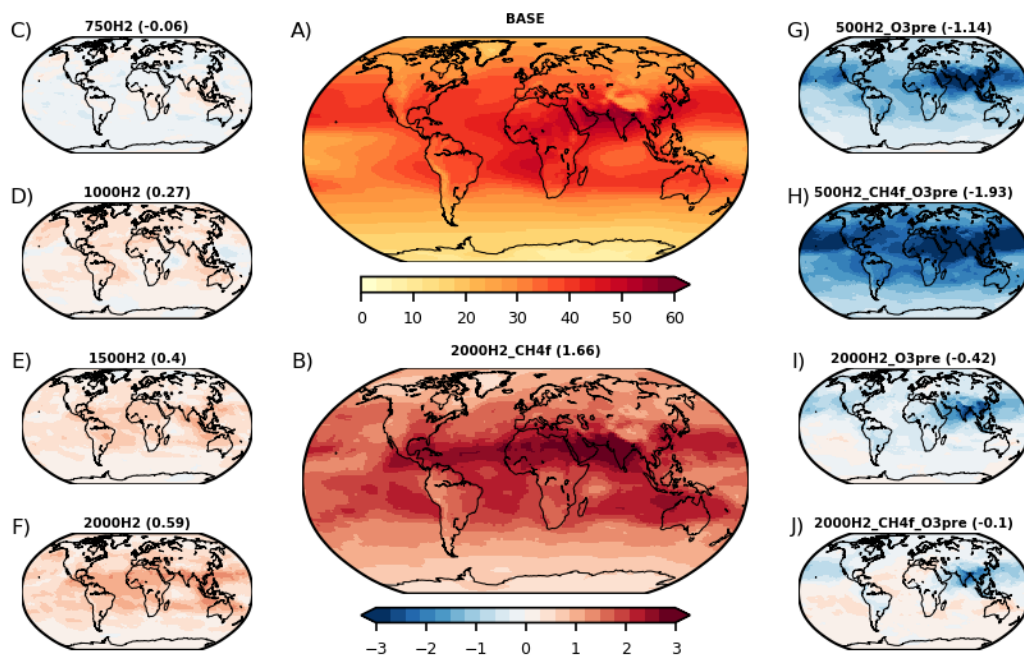


of H₂ LBC. In all cases, the gradients remain similar to the corresponding scenarios not including the co-emission reductions (note the ozone precursor emission changes are the same across all scenarios).

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The linear relationships shown in Figure 1 allow the global methane and ozone changes for a range of different hydrogen economy scenarios to be estimated, giving the response to different realisations of a future hydrogen economy. They can also be employed to derive a GWP for hydrogen (see Section 3.4).

245 Some detail on geographical changes in tropospheric ozone in a hydrogen economy is provided in Figure 2. As the photochemical production of ozone is driven by oxidation of carbon monoxide (CO), methane (CH₄) and volatile organic compounds (VOCs) in the presence of the oxides of nitrogen (NO_x), the net regional change in ozone depends not only on how H₂ changes, but also on changes in emissions of these species. This sensitivity is demonstrated in Figure 2 which shows tropospheric ozone column for the BASE scenario, as well as the change in tropospheric ozone column relative to BASE for
250 different hydrogen economy scenarios, where the tropospheric ozone column is defined as the vertically integrated ozone column between the surface and modelled tropopause. In these simulations we use a blended isentropic-dynamical tropopause combining the 380 K and 2 PV surfaces (following Hoerling et al., 1993). When H₂ increases, so does tropospheric ozone (Figures 2c to 2f, see also Figure 1c). Similarly, when atmospheric methane increases, the effect is to enhance the tropospheric ozone column increase (compare Figure 2b with Figure 2a). In contrast, reductions in ozone precursor and CH₄ emissions
255 would avoid the significant O₃ increases seen in Figure 2b (see Figures 2g, 2h, 2i and 2j). For example, if there is no H₂ leakage, reductions in the other emissions (Figure 2g and 2h) lead, as expected, to reductions in tropospheric ozone. When surface H₂ reaches 2000ppb, reductions in the other emissions still lead to modest net decreases in ozone (Figures 2i, 2j). For example, in the UKESM1 simulation which assumes a large H₂ leakage, with surface mixing ratios of H₂ reaching 2000 ppb, and emissions reductions in CO, NO_x, non-methane hydrocarbon and CH₄ emissions, the global mean tropospheric column ozone response
260 is found to be small (-0.1 DU) due to the competing effects outlined above (Figure 2j). The tropospheric ozone response to a global shift towards a hydrogen economy is therefore strongly influenced by the amount of hydrogen added to the atmosphere through leaks, and the co-benefit reductions achieved in CO, NO_x, non-methane hydrocarbon and CH₄ emissions. Tropopause height differences between the simulations explain only a tiny fraction of the global mean difference in tropospheric ozone column seen (up to 0.06 DU), confirming that the majority of the changes seen here are driven by modelled composition
265 changes.



270 Figure 2. The effect of increases in atmospheric H₂ on tropospheric ozone column: (a) tropospheric ozone column for the
BASE scenario (DU), (b) the change in tropospheric ozone column relative to BASE when H₂ is increased to 2000 ppb,
including the response of the methane abundance to changes in the methane lifetime (DU), (c) to (f) changes in tropospheric
ozone column relative to BASE when H₂ mixing ratios are increased, but all other factors including the methane abundance
remain fixed, and (g) to (j) changes in tropospheric ozone column relative to BASE for scenarios including emission reductions
275 in ozone precursors and methane. The numbers above each figure give the change in global mean tropospheric column in DU
relative to BASE.

As shown above, the changes in H₂ and methane affect OH, which can in turn also influence aerosols in our model simulations.
By reducing the flux through the OH+SO₂ reaction, there is a shift in the oxidation of SO₂ in the gas phase (which contributes
280 to new particle formation) and oxidation in the condensed phase (which grows existing particles). Such a shift leads to a
reduction in the aerosol number and increase in aerosol size with impacts on clouds and radiative forcing (see O'Connor et al.,
2022 for further details). We simulate a linear decrease in tropospheric air mass weighted OH (Figure 1a), and thus the OH+SO₂
reaction flux, for increasing H₂ in the atmosphere. This results in a reduction of the gas phase oxidation of SO₂ and a reduction
in the nucleation of new particles. This shifts the oxidation of SO₂ to be favoured by in cloud oxidation, which favours the
285 growth of cloud condensation nuclei. This decrease in OH is coincident with an increase in HO₂ (R1 leads to the production
of H atoms which in the troposphere near instantaneously form HO₂); thus, H₂ acts to decrease the OH-to-OH₂ ratio, similar
to CO. The increase in HO₂ outweighs the decrease in OH such that the HO₂:OH ratio linearly increases with increasing H₂ in



the atmosphere (from 145 at 500 ppb H₂ to 175 at 2000 ppb H₂) and as a result the H₂O₂ concentration increases, further enhancing in-cloud oxidation of SO₂.

290 3.3 Effective radiative forcing

Figure 3 shows the effective radiative forcing (ERF) for the scenarios in Table 1. We determined the ERF using Equation (5) from O'Connor et al., (2021), which can be decomposed into the cloud radiative effect (CRE) and the clear-sky forcing. The time-slice experiments using an atmosphere-only model, i.e. using decoupled sea surface temperature (SST) and sea-ice coverage, permit the determination of the top-of-atmosphere radiative forcing including rapid adjustments to cloud and water vapour. The ERF was calculated relative to the 2014 base case. The ERF varies from a cooling (negative ERF) to a warming (positive ERF) tendency depending on the scenario. The ERF increases with H₂ over the range 500 – 2000 ppb. Table 2 shows that these increases are due to both clear-sky and cloud radiative forcing. The clear-sky forcing increases with increasing H₂ presumably due to increased O₃, while the CRE can be ascribed to small decreases in cloud albedo. Figure 4 shows that increasing H₂ from 500 ppb to 2000 ppb leads to changes in cloud droplet number concentration (CDNC) across the globe. The decreased CDNC leads, for the same change in water vapour, to larger cloud droplets and lower cloud albedo. This leads to a CRE of 0.05 Wm⁻². In scenarios where CH₄ is increased, the further suppression of OH leads to a stronger CRE. Whilst the radiative forcing from the CRE in our hydrogen economy scenarios suggests this could form a significant component of indirect forcing from H₂, we do not include forcing from clouds in our GWP calculations. Such couplings between oxidants, aerosols and clouds are relatively unique in UKESM compared with other CMIP6 models and more studies are required to constrain the uncertainties involved.

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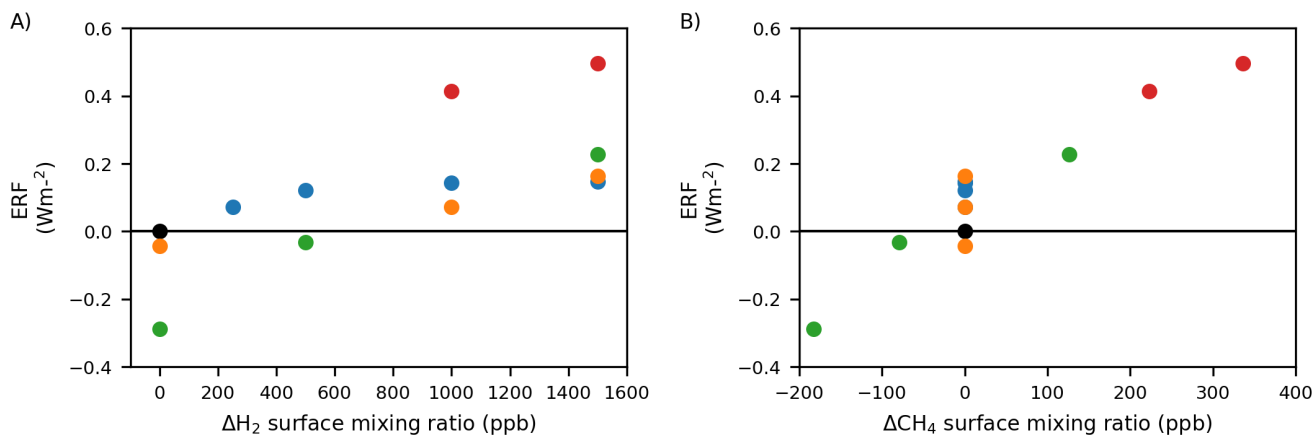


Figure 3. The effect of the simulated changes in atmospheric composition on effective radiative forcing (ERF). Left hand panel shows the ERF as a function of H_2 and other experimental conditions; right hand figure shows the ERF as a function of CH_4 and other experimental conditions. As for Figures 1a-c, the black circle represents the BASE scenario, blue circles represent scenarios where the CH_4 LBC remains fixed at 2014 levels, red circles are scenarios where the CH_4 LBC is adjusted to account for the change in CH_4 lifetime, orange circles are scenarios including changes in emissions of ozone precursors, and green circles include both changes to emissions of CH_4 and ozone precursors, as well as adjusted CH_4 LBCs to account for the response of the CH_4 abundance to changes in the CH_4 lifetime.

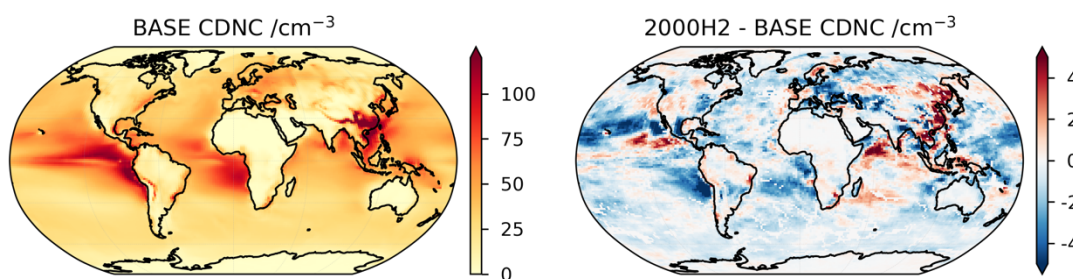


Figure 4. Cloud droplet number concentration (CDNC) in the UKESM1 model at 1 km altitude. The left hand figure shows CDNC in the BASE (500 ppb H_2) scenario, the right hand the change in CDNC when H_2 is increased to 2000 ppb (2000H2 scenario).



Hydrogen economy simulation	ERF Clear Sky LW + SW	Cloud Radiative Effect (CRE) LW + SW	ERF all sky, LW+SW
750H2	0.051 ± 0.041	0.021 ± 0.041	0.0722 ± 0.095
1000H2	0.069 ± 0.054	0.033 ± 0.054	0.102 ± 0.097
2000H2	0.096 ± 0.056	0.051 ± 0.056	0.148 ± 0.076
500H2_O3pre	-0.0021 ± 0.053	-0.051 ± 0.053	-0.053 ± 0.077
2000H2_O3pre	0.110 ± 0.056	0.054 ± 0.044	0.164 ± 0.087
2000H2_CH4f_O3pre	0.186 ± 0.108	0.0404 ± 0.108	0.223 ± 0.202
500H2_CH4f_O3pre	-0.160 ± 0.140	-0.123 ± 0.104	-0.289 ± 0.196
2000H2_CH4f	0.342 ± 0.099	0.155 ± 0.104	0.497 ± 0.179

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Table 2. A summary of the effective radiative forcing (ERF) changes in the hydrogen economy simulations.

3.4 Global Warming Potential

A GWP quantifying the indirect radiative forcings associated with H₂ has so far only previously been determined in a very limited number of studies including Derwent et al., (2020) (an update on Derwent et al., 2006), Field and Derwent, (2021) and Hauglustaine et al. (2022). The first two studies considered only the influence of H₂ on tropospheric composition, whereas Hauglustaine et al. (2022) included both the tropospheric and stratospheric response. In addition to these studies, Ocko and Hamburg (2022) extended the work presented here and in Warwick et al. (2022) to calculate hydrogen's GWP for time horizons between 0 and 100 years, considering both a pulse of H₂ emissions and a constant emission rate.

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We determine a GWP for H₂ based on composition changes in both the troposphere and stratosphere in our UKESM1 simulations, using radiative forcing scaling factors from IPCC (2014) and IPCC (2021) and the equations E1 to E4 presented in Section 3.4.1 (see also Warwick et al., (2022)). Our new estimate of the H₂ GWP considers changes in methane and ozone in the troposphere, as well as changes in stratospheric water vapour and stratospheric ozone. Note that we have not used the model-derived ERFs from Section 3.3 in the GWP calculations below. The model-derived ERFs (for H₂ leakage only, including the methane response) are larger than the radiative forcing due to hydrogen determined from modelled changes in chemical

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composition using radiative forcing scaling factors from IPCC (2021) (see Section 3.4.2). Much of this difference arises due to the large contribution from clouds to the model-based ERFs (see Table 2) that is not accounted for in the radiative forcing scaling factors. For our GWP calculations, we determine the radiative forcing using the modelled changes in composition
345 along with the radiative forcing scaling factors rather than the ERFs. Our reason for this is two-fold. Firstly, it ensures the large uncertainty associated with the aerosol-cloud component of the ERFs (IPCC, 2021) is not propagated into the GWP calculations. Secondly, significant further work would be required to attribute the calculated total ERFs to individual species, so that the appropriate lifetimes could be applied to the different components of the forcing that contribute to the GWP. However, we note that if the model ERFs were to be used in the GWP calculations, the GWPs presented in the next section
350 would be larger.

3.4.1 Methodology

For our GWP calculation, we derive a more universal version of the approach presented in Fuglestvedt et al. (2010) for calculating GWPs for species whose emissions result in indirect radiative forcings, which was also implicitly adopted for Global Temperature Change Potentials in the IPCC Sixth Assessment Report (IPCC, 2021). The Fuglestvedt et al. (2010)
355 method assumes that the time evolution of the radiative forcing depends only on the lifetime of the species causing the forcing and not the lifetime of the species being emitted. In their method, the time evolution of the radiative forcing during a one-year constant emission is described by an approach to steady-state with a time constant according to the lifetime of the species causing the forcing. The peak forcing is assumed to occur directly at the end of the one-year emission, and subsequently to decay with the same lifetime as in the growth phase. Although this assumption may be a good approximation for short-lived
360 emission species such as NO_x, it may not be for species with longer lifetimes such as H₂. Our extension of their method (see Warwick et al., 2022) accounts for both the lifetime of the emitted species and the lifetime of the species causing the forcing; it also allows the time period of the constant emission to be varied.

In our method for deriving indirect GWPs from steady-state simulations, we include the indirect forcing due to perturbations
365 in radiatively active species arising (a) during the constant emission (AGWP1, E1), (b) from the decay of the perturbation subsequent to the end of the constant emission (AGWP2, E2) and (c) perturbations arising from the emission species remaining in the atmosphere after the end of the constant emission (AGWP, E3). Derivations for E1 to E3 are presented in Appendix 1.

$$AGWP1 = \frac{R a_M \alpha_M \alpha_H C}{t_p} \left(tp - \alpha_M \left(1 - \exp\left(\frac{-t_p}{\alpha_M}\right) \right) - \left(\frac{\alpha_H}{\alpha_H - \alpha_M} \right) \left(\alpha_H \left(1 - \exp\left(\frac{-t_p}{\alpha_H}\right) \right) - \alpha_M \left(1 - \exp\left(\frac{-t_p}{\alpha_M}\right) \right) \right) \right) \quad (E1)$$

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$$AGWP2 = \frac{\left(R \alpha_M \alpha_H^2 \alpha_M C \left(1 - \exp\left(\frac{-t_p}{\alpha_H}\right) \right) \right)}{t_p (\alpha_H - \alpha_M)} \left(\alpha_H \left(1 - \exp\left(-\frac{(Hz - t_p)}{\alpha_H}\right) \right) - \alpha_M \left(1 - \exp\left(-\frac{(Hz - t_p)}{\alpha_M}\right) \right) \right) \quad (E2)$$

$$375 \quad AGWP3 = \frac{R \alpha_M \alpha_M^2 \alpha_H C}{t_p} \left(\left(1 - \exp\left(-\frac{t_p}{\alpha_M}\right) \right) - \left(\frac{\alpha_H}{\alpha_H - \alpha_M} \right) \left(\exp\left(-\frac{t_p}{\alpha_H}\right) - \exp\left(-\frac{t_p}{\alpha_M}\right) \right) \right) \left(1 - \exp\left(-\frac{(Hz - t_p)}{\alpha_M}\right) \right) \quad (E3)$$

Where:

$AGWP$ = absolute global warming potential ($W \text{ m}^{-2} \text{ kg}^{-1} \text{ yr}$)

380 M = species resulting in the indirect radiative forcing: CH_4 , O_3 and H_2O (ppb or DU for tropospheric O_3)

R_M = radiative forcing efficiency for M ($W \text{ m}^{-2} \text{ ppb}^{-1}$ or $W \text{ m}^{-2} \text{ DU}^{-1}$ for tropospheric O_3)

α_M = production rate of M [ppb yr^{-1}] per ppb H_2 change at steady-state (yr^{-1})

α_M = Atmospheric lifetime of M (yr)

α_H = Atmospheric H_2 lifetime (combined chemical and deposition lifetime) (yr)

385 Hz = the time horizon considered (yr)

C = conversion factor for converting H_2 mixing ratio (ppb) into H_2 mass (kg)

t_p = length of step emission (yr)

The associated indirect GWPs for each radiative species perturbed by hydrogen are then given by:

$$390 \quad GWP_{H2} = \frac{AGWP_{H2}}{AGWP_{CO2}} \quad (E4)$$

A comparison of the time evolution of the methane perturbation resulting from a change in H_2 mixing ratio based on the
 395 standard Fuglestvedt et al. (2010) equations and our updated equations is shown in Figure 5. Accounting for the atmospheric
 lifetime of hydrogen in the updated equations results in a slower predicted rate of increase in methane during the one-year
 constant emission relative to the standard Fuglestvedt et al. (2010) equations, a delayed methane peak (peaking at 5 years
 rather than 1 year, using the parameters specified in the caption) and a slower rate of methane decrease subsequent to that
 peak; as methane is still being impacted by the remaining H_2 . This difference occurs as the equations from Fuglestvedt et al.
 400 (2010) assume that the emitted species instantaneously reaches a new steady-state atmospheric concentration at the start of the
 one-year constant emission. In addition, their assumption that the decay following the peak forcing is controlled only by the



lifetime of the radiatively active species neglects any subsequent perturbations to atmospheric chemistry as a result of the emitted species still present in the atmosphere after the end of the one-year constant emission.

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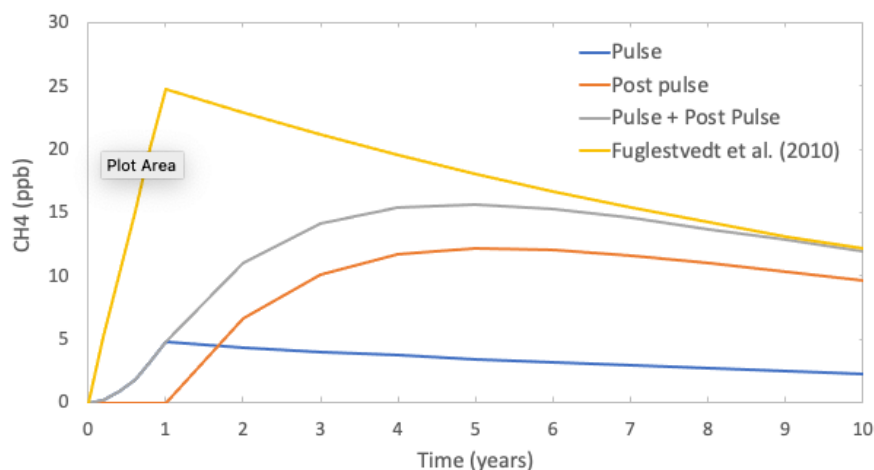


Figure 5. The time evolution of excess methane during and after a one year step hydrogen emission (based on a steady-state H_2 excess of +1500 ppb, an H_2 lifetime of 1.9 years and a methane perturbation lifetime of 12.6 years) as predicted by the
410 Fuglestvedt et al. (2010) equations (yellow) and our updated equations. For our updated equations, the excess methane is split into contributions from methane generated during the step emission (blue), methane generated from excess hydrogen remaining in the atmosphere after the one year step emission (orange), with the total methane excess shown in grey.

3.4.2 Calculation of a hydrogen GWP

In our calculations, $AGWP_{CO_2}(100)$ and $AGWP_{CO_2}(20)$ are taken to be 9.17×10^{-14} ($\pm 26\%$) and 2.49×10^{-14} ($\pm 18\%$) $W m^{-2} kg^{-1}$
415 yr (IPCC, 2014) respectively. $AGWP_{H_2}$ is taken as the sum of $AGWP_{1-3}$. The values for $AGWP_{CO_2}(100)$ and $AGWP_{CO_2}(20)$ change slightly to 8.95×10^{-14} ($\pm 26\%$) and 2.43×10^{-14} ($\pm 18\%$) $W m^{-2} kg^{-1} yr$ in IPCC (2021): the impact of this on our H_2 GWP calculation, along with other IPCC (2021) updates, is shown in Table 3. The length of the step emission, t_p , is 1 year, H_z is taken to be 20 or 100 years and a_H is 1.9 years (with an uncertainty range from 1.4 to 2.2 years). The conversion factor, C , for converting H_2 mixing ratio into H_2 mass based on UKESM1 data is 3.52×10^{-9} ppb kg^{-1} . Values for R_M , a_M , and α_M are
420 dependent on the species causing the indirect forcing (methane, ozone and stratospheric water vapour).

For methane, R_{CH_4} is taken to be 0.000363 ($\pm 10\%$) $W m^{-2} (ppb CH_4)^{-1}$ (IPCC, AR5) or 0.000389 $W m^{-2} (ppb CH_4)^{-1}$ (IPCC, AR6). The methane perturbation lifetime, α_{CH_4} , in UKESM1 is derived to be 12.6 years in this study, which falls within the range given by IPCC, AR5 (12.4 ± 1.4 years). The value for a_{CH_4} , an additional methane production rate term per unit increase



425 in H_2 , represents the positive CH_4 tendency resulting from an increase in H_2 and corresponding decrease in OH and is defined as:

$$a = CH_4 k_{CH_4+OH} \frac{dOH}{dH_2}$$

where CH_4 is the methane lower boundary condition (1835 ppb in UKCA), k_{CH_4+OH} is the global mean airmass-weighted rate
430 constant for the reaction of CH_4 with OH and dOH/dH_2 is the rate of change of OH with respect to H_2 . Values for these parameters derived from UKESM1 simulations are as follows: $k(CH_4 + OH) = 2.8 \times 10^{-15} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$, $dOH/dH_2 = 3.68 \times 10^{-9}$ (dimensionless). $k(CH_4 + OH)$ is determined from the gradient of methane lifetime against $1/(\text{global airmass-weighted mean OH})$, and dOH/dH_2 from the gradient of the mass-weighted tropospheric mean OH concentration against H_2 surface mixing ratio for the UKESM1 simulations including the feedback on the methane lifetime via an adjusted CH_4 lower
435 boundary condition determined by the box model (see Figure 1a). It is also possible to derive dOH/dH_2 using the gradient of the mass-weighted tropospheric mean OH concentration against H_2 surface mixing ratio from the UKESM1 simulations where only the H_2 LBC is varied (whilst the CH_4 LBC is held constant) in combination with the equation below from Stevenson et al. (2013), and the methane feedback factor of 1.49 derived for UKESM1,

$$440 \quad CH_{4 \text{ new}} = CH_{4 \text{ base}} \left(\frac{\tau_{\text{new}}}{\tau_{\text{base}}} \right)^f$$

where f is the methane feedback factor, $CH_{4 \text{ base}}$ and τ_{base} are the lower boundary condition and chemical lifetime for methane in the base scenario, and $CH_{4 \text{ new}}$ and τ_{new} are the lower boundary condition and chemical lifetime for methane in the perturbed H_2 scenarios. The difference between the dOH/dH_2 values derived using the two methods above leads to a 2%
445 difference in the derived hydrogen GWP, which is negligible in comparison to other uncertainties (see Table 3), and our GWP calculation therefore remains independent of box model results.

The indirect forcing from methane-induced changes to tropospheric ozone and stratospheric water vapour are determined by multiplying the calculated direct methane forcing (R_{CH_4}) by scaling factors of $0.5 \pm 55\%$ and $0.15 \pm 70\%$ respectively (IPCC,
450 2014). Equivalent scaling factors from IPCC (2021) are $0.37 \pm 50\%$ and $0.106 \pm 100\%$.

In addition to the methane-induced indirect radiative forcing from ozone and water vapour outlined above, we also consider non-methane induced forcing from H_2 as a source of tropospheric ozone and stratospheric water vapour. The e-folding lifetime of the water vapour perturbation following a change in H_2 , α_{H_2O} , is derived from model spin-up data following a change to
455 the H_2 lower boundary condition and is determined to be 8 years in the lower stratosphere. A value for R_{H_2O} is determined via comparison of our results to that of Myhre et al. (2007). The vertical profile of stratospheric water vapour changes relative to



BASE obtained for our 2000H2 scenario (Figure 1d) shows very similar changes at all altitudes with stratospheric water vapour changes resulting from methane changes between 1950 and 2000 in Figure 2b of Myhre et al. (2007). We therefore adopt the radiative forcing determined by Myhre et al. (2007) of 0.05 W m^{-2} with an estimated uncertainty of $\pm 20\%$ for stratospheric water vapour changes between our BASE and 2000H2 scenario. This water vapour forcing is associated with a change of ~ 500 ppb in stratospheric water vapour at 30 km altitude, giving a value for R_{H_2O} of $1 \times 10^{-4} \text{ W m}^{-2} (\text{ppb H}_2\text{O})^{-1}$ when considering changes at 30 km. The choice of 30 km for determining R_{H_2O} is arbitrary as we assume that the entire stratospheric H₂O profile will scale proportionally with the H₂ lower boundary condition (following results from our UKESM1 simulations using H₂ LBCs of 750, 1000 and 2000 ppb H₂). The production rate of water vapour per ppb change in H₂, a_{H_2O} , is then given by the change in steady-state water vapour at 30 km (500 ppb) divided by the lifetime of 8 years and the change in H₂ mixing ratio of 1500 ppb.

To determine the contribution of changes in tropospheric ozone to the hydrogen GWP, the e-folding lifetime associated with the ozone perturbation, α_{O_3} , is taken to be 0.07 years, calculated as the tropospheric O₃ burden divided by the loss in our UKESM1 BASE simulation. This is shorter than the lifetime used for the short-lived ozone perturbation in Fuglestad et al. (2010) of 0.267 years. However our results are insensitive to the difference between these values and small variations around them. The value for R_{O_3} is 0.042 (0.037 to 0.047) $\text{W m}^{-2} \text{DU}^{-1}$ and is taken directly from IPCC, (2014). No updated value for R_{O_3} was presented in IPCC (2021), although a similar value of $0.039 \text{ W m}^{-2} \text{DU}^{-1}$ can be derived based on the estimated change in tropospheric ozone of $109 \pm 25 \text{ Tg}$ between 1850 and 2005 and a stratospheric-temperature-adjusted radiative forcing due to tropospheric ozone of $0.39 \pm 0.07 \text{ W m}^{-2}$ between 1850 and 2010. The value for α_{O_3} is calculated as the change in the steady-state global mean tropospheric ozone column in our 2000H₂ simulation relative to base (0.59 DU) divided by the lifetime of 0.07 years and the change in H₂ mixing ratio of 1500 ppb.

Changes in H₂ mixing ratio can also influence stratospheric ozone. The change in global mean stratospheric ozone column between our BASE and 2000H₂_CH4f scenarios was $+0.10 \text{ DU}$, relative to a base value of 283.78 DU and interannual variations of up to several DU. Taking a value for R for stratospheric ozone of $0.0054 \text{ W m}^{-2} \text{DU}^{-1}$ (Schwarzkopf and Ramaswamy, 1993), and a range of e-folding lifetimes for the stratospheric ozone perturbation of between 1 and 10 years, this results in a hydrogen GWP(100) due to indirect changes in stratospheric ozone of less than 0.03 using equations E1 to E4. This value is equivalent to approximately 0.3% of the total GWP(100) determined for hydrogen and is negligible in comparison to our estimated uncertainties in the hydrogen GWP due to uncertainties in the radiative forcing scaling factors and the soil sink (see Table 3). We therefore conclude that changes in stratospheric ozone for our range of scenarios do not significantly contribute to the hydrogen GWP.

For a 100 year time horizon we determine a H₂ GWP of 12 ± 6 (see Table 3). This value is larger than previously published studies which do not account for changes in stratospheric composition (Derwent et al., 2020, 5 ± 1 , and Derwent and Field



2021, 3.3 ± 1.4), but similar to the value of Hauglustaine et al. (2022) which includes a tropospheric and stratospheric response (12.8 ± 5.2). Approximately two thirds of our GWP arises from the influence of H_2 on methane and ozone distributions in the troposphere and one third from the influence of H_2 on stratospheric water vapour. Results from our study can also be compared with that of Paulot et al. (2021), who estimated the indirect radiative forcing at steady-state due to an increase in H_2 . Their study accounted for changes in methane, tropospheric ozone and stratospheric water vapour and found an indirect radiative forcing arising from H_2 of $0.13 \text{ mW m}^{-2} \text{ ppb}(H_2)^{-1}$. We obtain a larger value of $0.18 \pm 0.03 \text{ mW m}^{-2} \text{ ppb}(H_2)^{-1}$ (the same value is obtained using both the IPCC (2014) and IPCC (2021) radiative efficiency scaling factors), which may be partly explained by the larger methane feedback factor in UKESM1 (1.49) relative to the chemistry-climate model used in Paulot et al. (2021). The UKESM1 methane feedback factor lies at the high end of the range of feedback factors reported in Thornhill et al. (2021) for the pre-industrial, but there are indications from the literature that this value varies with climate state and methane burden (Holmes, 2018).

Uncertainties in our calculation are based on uncertainties in the hydrogen lifetime with respect to soil uptake, as well as uncertainties in radiative forcing scaling factors and the AGWP for CO_2 . We do not account for uncertainties in chemical lifetimes and other UKESM1-derived quantities which are likely to vary when using different atmospheric models with different chemistry schemes. In addition, we also note that the sensitivity of ERF in UKESM1 to changes in methane is approximately 65% larger than that indicated by the radiative scaling factors used in the empirical relationships applied in the GWP calculation (O'Connor et al., 2022). This suggests a further source of uncertainty than accounted for by the radiative forcing scaling factor uncertainties outlined in IPCC (2104; 2021), associated with the response of both aerosols and clouds, as well as ozone and water vapour to changing methane levels.

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Radiatively active species	H ₂ GWP(100)	H ₂ GWP(100)	H ₂ GWP(20)	H ₂ , GWP(20)
	IPCC AR5	IPCC AR6	IPCC AR5	IPCC AR6
Methane	4.9	5.3	13.4	14.8
Tropospheric ozone (methane and non-methane induced)	3.6	3.2	11.2	9.9
Total Troposphere	8.5	8.5	24.6	24.7
Stratospheric water vapour (methane and non-methane induced)	3.2	3.0	9.9	9.5
Total	11.7 (6 – 18)	11.5 (6 – 18)	34.6 (19 – 51)	34.2 (19 – 51)

525 Table 3. A comparison of the 100 and 20 year time horizon H₂ GWPs (GWP(100) and GWP(20) respectively) determined using radiative efficiencies and the AGWP for CO₂ from IPCC AR5 (2014) and from IPCC AR6 (2021). Uncertainties quoted account for uncertainties in the H₂ lifetime with respect to soil uptake, radiative forcing scaling factors and the AGWP_{CO2} only.

4 Discussion and conclusions

530 Leakage of hydrogen associated with a hydrogen economy will result in an indirect global warming, offsetting greenhouse gas emission reductions made as a result of a switch from fossil fuel to hydrogen. We have presented a methodology for calculating the indirect GWP of H₂, with principal contributions coming from changes in methane, in tropospheric ozone and in stratospheric water vapour. The GWP depends on the lifetime of H₂ in the atmosphere, on the perturbation lifetimes of the radiatively active species, (e.g., CH₄ and O₃) and on their effective production rates per unit change in H₂. We present values for these based on calculations with our chemistry-climate model, UKESM1 and a box model. The 100 year GWP for H₂, based on the above, is 12 ± 6 , a value somewhat higher than some previous estimates. Whilst our GWP uncertainty accounts for uncertainties associated with the size of the soil sink and radiative forcing scaling factors, it does not include uncertainties in model-derived parameters (e.g. chemical and perturbation lifetimes or the chemical production rates per unit change in H₂). In particular, uncertainties associated with the stratospheric response may be significant, as well as the response of aerosols and clouds to changing methane levels. Further simulations using different earth system models and different chemistry schemes would be required to fully assess the impact of uncertainties in these parameters on the hydrogen GWP.

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Determining a GWP for hydrogen allows a change in hydrogen emissions to be compared to an equivalent change in carbon dioxide emissions in terms of time integrated radiative forcing. This increase in equivalent carbon dioxide emissions can be compared with expected reductions in carbon dioxide and methane (as equivalent CO₂) emissions to determine the net impact



on radiative forcing. In our illustrative future global hydrogen economy scenario (Section 2.3) we estimate additional H₂ emissions of between 9 and 95 Tg H₂ yr⁻¹, based on a hydrogen economy supplying 23% of present day energy consumption and H₂ leakage rates of 1 and 10%. Using a H₂ GWP(100) of 12, this is equivalent to the time integrated radiative forcing from carbon dioxide emissions of about 110 and 1140 Tg CO₂ yr⁻¹ respectively. Based on a reduction in fossil fuel use following the fossil fuel energy sectors that are replaced by hydrogen in Section 2.3.1, a low carbon method of hydrogen generation and the sector emissions described in Hoesly et al. (2018), we would expect a reduction in carbon dioxide emissions of ~26,000 Tg yr⁻¹. In addition, expected reductions in methane emissions would lead to a further reduction of ~1200 Tg yr⁻¹ equivalent CO₂ emissions (assuming a GWP(100) for methane of 28). This gives a total reduction of CO₂ equivalent emissions of ~27,200 Tg yr⁻¹ in our hydrogen economy scenario. In this case, the increase in equivalent CO₂ emissions based on 1% and 10% H₂ leakage rate offsets approximately 0.4 and 4% of the total equivalent CO₂ emission reductions respectively, with the benefits from equivalent CO₂ emission reductions significantly outweighing the disbenefits arising from H₂ leakage. However, our calculations clearly demonstrate the climate importance of controlling H₂ leakage within a hydrogen economy.

A switch to a hydrogen economy would also provide the opportunity to reduce emissions of other gases which can themselves directly or indirectly affect both climate and air quality. For example, an immediate impact of increased atmospheric H₂ is to reduce the concentration of OH, the major tropospheric oxidant, which would thus tend to increase the lifetime of methane. Increases in methane would adversely affect climate and also lead to production of tropospheric ozone, impacting both climate and air quality. However, we show that reductions in methane emissions associated with decreased fossil fuel use, along with reductions of CO and NO_x emissions, could lead to a net change in ozone close to (or below) zero with associated climate and air quality benefits, assuming very high H₂ leakage rates (e.g. scenario 2000H₂_CH₄f_O₃pre, Figure 2j). In the case of no leakage of H₂, emission reductions in methane, CO, and NO_x lead to decreases in tropospheric ozone column globally (scenario 500H₂, CH₄f_O₃pre, Figure 2h). To assess whether the ozone response to changes in emissions associated with a hydrogen economy is influenced by the assumed background climate conditions, (i.e. whether the timing of the implementation of a hydrogen economy could be important), we performed a set of additional model simulations using 2045-2055 climatological boundary conditions taken from the CMIP6-SSP2-4.5 scenario. These scenarios assumed a hydrogen economy of the same absolute size and identical leakage rates as our present day (2014) scenarios. We found no discernible difference in terms of the tropospheric and stratospheric ozone response to changes in H₂ and ozone precursors, with emission reductions from decreased fossil fuel use offering similar air quality benefits in both sets of simulations. To maximise the benefit of a switch to hydrogen, not only should hydrogen leakage be minimised, but the emissions of methane, CO, VOCs and NO_x should also be reduced to the maximum extent possible.



575 **Author Contributions**

JAP and ATA designed the research and supervised the analysis. JK performed the UKESM model simulations and atmospheric composition analysis and PTG and FMO performed the ERF calculations. KPS, JAP, PTG and NJW contributed to the H₂ GWP analysis. KPS derived the AGWP equations, with contributions from JAP. NJW performed the UKESM GWP calculations. All box modelling was performed by PTG. NJW compiled the emission changes for the different scenarios. All
580 authors contributed to the writing of the manuscript.

Code availability

Due to intellectual property rights restrictions, we cannot provide either the source code or documentation papers for the UM. The Met Office Unified Model is available for use under licence. A number of research organisations and national meteorological services use the UM in collaboration with the Met Office to undertake basic atmospheric process research,
585 produce forecasts, develop the UM code, and build and evaluate Earth system models. For further information on how to apply for a licence, see <http://www.metoffice.gov.uk/research/modelling-systems/unified-model>.

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