



*Supplement of*

## **Atmospheric composition and climate impacts of a future hydrogen economy**

**Nicola J. Warwick et al.**

*Correspondence to:* Nicola J. Warwick ([nicola.warwick@atm.ch.cam.ac.uk](mailto:nicola.warwick@atm.ch.cam.ac.uk))

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## S1. Derivation of AGWP equations

For our GWP calculations we derive a more universal version of the approach described in Fuglestvedt et al. (2010). Fuglestvedt et al. (2010) consider a mathematical framework in which the forcing is assumed be a result of a one-year, constant emission pulse of the precursor, followed by an exponential decay of the resulting forcing from the end-of-year 1 value after this. The time evolution of the forcing during the pulse, and its subsequent decay following the end of year 1, is dependent only on the perturbation lifetime of the species resulting in the indirect forcing and not the precursor emitted species. Our framework builds on that of Fuglestvedt et al. (2010), by allowing the time-length of the initial constant emission period to be varied, and by considering the impact of the lifetime of the emitted species on the time evolution of the forcing, in addition to the lifetime of the species causing the indirect forcing. As part of our method, we derive three expressions that account for (a) the radiatively active species generated during the precursor emission pulse ( $H_2$  in our work), (b) the radiatively active species generated from the decay of the precursor still present in the atmosphere at the end of the pulse, and (c) the decay of the radiative species generated during the pulse.

### (a) Perturbation to the radiatively active species during the pulse emission

In our framework, the time evolution of the precursor ( $H_2$  in this work) during an initial period of constant emissions (hereafter referred to as the pulse emission) relative to an unperturbed background condition can be represented by

$$H(t) = \alpha_H S_H \left( 1 - \exp \left( \frac{-t}{\alpha_H} \right) \right) \quad (S1)$$

where  $H$  represents the perturbation of the global mean atmospheric  $H_2$  mixing ratio relative to the initial background condition,  $S_H$  represents a constant emission rate of  $H_2$  ( $Tg \text{ yr}^{-1}$ ) forming a pulse of time length  $t_p$ , and  $\alpha_H$  is the atmospheric lifetime of  $H_2$ . When  $t = t_p$ , (SE1) can be used to determine the additional  $H_2$  present at the end of the pulse,  $H_p$ , required below in (b).

The additional  $H_2$  in the atmosphere from the pulse emissions will act to decrease the methane lifetime (which is considered here as an additional methane production term) and lead to the production of ozone and stratospheric water vapour. Assuming the rate of production of these radiatively active species are linearly dependent on the changes in  $H_2$ , then during the  $H_2$  emission pulse their perturbations relative to the background state can be given by

$$\frac{dM}{dt} = a_M \alpha_H S_H \left( 1 - \exp\left(\frac{-t}{\alpha_H}\right) \right) - \left( \frac{M}{\alpha_M} \right) \quad (\text{S2})$$

where  $M$  represents the change in the global mean mixing ratio of the radiatively active species (in this case  $\text{CH}_4$ ,  $\text{O}_3$  or  $\text{H}_2\text{O}$ ), relative to the initial background condition,  $a_M$  is the production rate of  $M$  per unit change in  $\text{H}_2$ , and  $\alpha_M$  is the lifetime of  $M$ . Rearranging and incorporating an integrating factor of  $\exp(t/\alpha_M)$  give

$$\frac{d \left( M \exp(t/\alpha_M) \right)}{dt} = a_M \alpha_H S_H \left( \exp\left(\frac{t}{\alpha_M}\right) - \exp\left(t \left( \frac{1}{\alpha_M} - \frac{1}{\alpha_H} \right) \right) \right) \quad (\text{S3})$$

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which on integrating (assuming  $M = 0$  at  $t = 0$ ), gives the time evolution of the radiatively active species during the  $\text{H}_2$  emission pulse

$$M(t) = a_M \alpha_H S_H \alpha_M \left( \left( 1 - \exp\left(\frac{-t}{\alpha_M}\right) \right) - \left( \frac{\alpha_H}{\alpha_H - \alpha_M} \right) \left( \exp\left(\frac{-t}{\alpha_H}\right) - \exp\left(\frac{-t}{\alpha_M}\right) \right) \right) \quad (\text{S4})$$

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When  $t = t_p$ , (SE4) can be used to determine the additional  $M$  present in the atmosphere at the end of the pulse,  $M_p$ , required below in (c).

50 Integrating (SE4) gives the time-integrated excess of the radiatively active species during the pulse (SE5) (assuming the time horizon considered,  $H_z > t_p$ ), which can be multiplied by the relevant radiative forcing scaling factor to determine the time-integrated forcing.

$$\int_0^{t_p} M dt = a_M \alpha_H S_H \alpha_M \left( t_p - \alpha_M \left( 1 - \exp\left(\frac{-t_p}{\alpha_M}\right) \right) - \left( \frac{\alpha_H}{\alpha_H - \alpha_M} \right) \left( \alpha_H \left( 1 - \exp\left(\frac{-t_p}{\alpha_H}\right) \right) - \alpha_M \left( 1 - \exp\left(\frac{-t_p}{\alpha_M}\right) \right) \right) \right) \quad (\text{S5})$$

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(b) Perturbation to the radiatively active species as a result of the decay of the precursor perturbation following the emission pulse

60 After the end of the pulse emission, the additional  $H_2$  in the atmosphere,  $H_p$ , will decay exponentially according to the lifetime of  $H_2$ ,  $\alpha_H$ . Taking  $t'$  to be the time since the end of the pulse, i.e. where  $t' = t - t_p$ ,

$$H(t) = H_p \exp\left(\frac{-t'}{\alpha_H}\right) \quad (S6)$$

65 The production of  $M$  (in our work,  $CH_4$ ,  $O_3$  or  $H_2O$ ) from the decay of  $H_p$  is then given by

$$\frac{dM}{dt'} = a_M H_p \exp\left(\frac{-t'}{\alpha_H}\right) - \frac{M}{\alpha_M} \quad (S7)$$

70 As above, a solution to this can be found by introducing an integrating factor  $\exp(t'/\alpha_M)$ . Assuming  $M = 0$  at  $t' = 0$  and integrating (SE7) between  $t' = 0$  and  $t' = T$  (the time since the end of the pulse) gives the time evolution of the radiatively active species generated from the additional  $H_2$  present at the end of the  $H_2$  emission pulse,  $H_p$

$$75 \quad M(T) = \frac{a_M H_p}{1/\alpha_M - 1/\alpha_H} \left( \exp\left(-\frac{T}{\alpha_H}\right) - \exp\left(-\frac{T}{\alpha_M}\right) \right) \quad (S8)$$

Where  $H_z$  is the time horizon considered, integrating (SE8) between  $T = 0$  and  $T = H_z - t_p$  gives the time-integrated excess in the radiatively active species built up following the pulse as a result of the decay of the precursor species ( $H_2$  in this work)

$$80 \quad \int_0^{H_z - t_p} M dT = \frac{\left( a_M \alpha_H^2 \alpha_M S_H \left( 1 - \exp\left(\frac{-t_p}{\alpha_H}\right) \right) \right)}{(\alpha_H - \alpha_M)} \left( \alpha_H \left( 1 - \exp\left(-\frac{(H_z - t_p)}{\alpha_H}\right) \right) - \alpha_M \left( 1 - \exp\left(-\frac{(H_z - t_p)}{\alpha_M}\right) \right) \right) \quad (S9)$$

(c) Decay of the radiatively active species built up during the emission pulse

85 Finally, we consider the decay of the radiatively active species that was built up during the pulse ( $M_p$ ), which is given by

$$M(t') = M_p \exp\left(\frac{-t'}{\alpha_M}\right)$$

(S10)

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As before, this can be integrated to determine the time-integrated perturbation to the radiatively active species as a result of the decay of the radiatively active species built up during the pulse.

$$\int_0^{Hz-t_p} M dt' = a_M \alpha_M^2 \alpha_H S_H \left( \left( 1 - \exp\left(-\frac{t_p}{\alpha_M}\right) \right) - \left( \frac{\alpha_H}{\alpha_H - \alpha_M} \right) \left( \exp\left(-\frac{t_p}{\alpha_H}\right) - \exp\left(-\frac{t_p}{\alpha_M}\right) \right) \right) \left( 1 - \exp\left(-\frac{(Hz-t_p)}{\alpha_M}\right) \right)$$

(S11)

The three components of the perturbation to  $M$  resulting from a pulse emission of  $H_2$  (SE5, SE9 and SE11) can then be summed to determine the total time integrated  $M$  excess relative to the initial background condition, which can be multiplied by the relevant radiative efficiency scaling factors to determine the time-integrated forcings. The corresponding absolute global warming potentials (AGWP) can then be derived by dividing the time integrated forcing by the total mass of the pulse ( $t_p S_H$ ), with equations E5, E9 and E11 leading to the equations for AGWP1, AGWP2 and AGWP3 in Section 3.4.1 respectively.

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Variable	Definition	Value	Source
$\alpha_M$	Perturbation lifetime	11.8 yr	UKESM1
$\alpha_H$		1.96 yr	UKESM1
$\alpha_{O3}$		0.07 yr	UKESM1
$\alpha_{H2O}$		8.0 yr	UKESM1
$a_M$	Production rate (ppb yr <sup>-1</sup> )	0.0146 yr <sup>-1</sup>	UKESM1
$a_{O3}$	of species resulting in the	5.62x10 <sup>-3</sup> yr <sup>-1</sup>	UKESM1
$a_{H2O}$	indirect forcing per ppb	0.041 yr <sup>-1</sup>	UKESM1
	H <sub>2</sub> change at steady-state		
$R_{CH4}$	Radiative forcing scaling factor	0.000363 <sup>1</sup> , 0.000389 <sup>2</sup> W m <sup>-2</sup> (ppb CH <sub>4</sub> ) <sup>-1</sup>	<sup>1</sup> Myhre et al., 2013, <sup>2</sup> Forster et al., 2021

$R_{O_3}$		$0.042 \text{ W m}^{-2} \text{ DU}^{-1}$	Myhre et al., 2013
$R_{H_2O}$		$1 \times 10^{-4} \text{ W m}^{-2} (\text{ppb H}_2\text{O})^{-1}$	Myhre et al., 2007
Hz	Time Horizon	20, 100 yr	
C	Conversion Factor (ppb H <sub>2</sub> to kg H <sub>2</sub> )	$3.5 \times 10^{-9} \text{ ppb kg}^{-1}$	UKESM1

Table S1. A list of input parameters for the AGWP equations (E1 to E5).

Hydrogen economy simulation	H <sub>2</sub> (ppb)	CH <sub>4</sub> (ppb)	O <sub>3</sub> _pre Emiss.	[OH] GM (10 <sup>6</sup> molec cm <sup>-3</sup> )	[HO <sub>2</sub> ] GM (10 <sup>6</sup> molec cm <sup>-3</sup> )	τ <sub>CH<sub>4</sub></sub> (yr)	O <sub>3</sub> Burden (Tg)	τ <sub>H<sub>2</sub>(OH)</sub> (yr)	O <sub>3</sub> Prod. (Tg yr <sup>-1</sup> )	O <sub>3</sub> Loss (Tg yr <sup>-1</sup> )	HO <sub>2</sub> +NO (Tg yr <sup>-1</sup> )	MeO <sub>2</sub> +N O (Tg yr <sup>-1</sup> )	HO <sub>2</sub> + O <sub>3</sub> (Tg yr <sup>-1</sup> )	OH+O <sub>3</sub> (Tg yr <sup>-1</sup> )	OH+SO <sub>2</sub> (Tg yr <sup>-1</sup> )
BASE	500	1835	2000-2014	1.01	92.42	8.48	360.764	8.265	5856	4942	3855	1270	1600	689	21.0
750H2	750			0.99	93.79	8.67	360.088	8.400	5874	4954	3906	1252	1621	677	20.8
1000H2	1000			0.97	94.90	8.83	363.739	8.529	5896	4986	3948	1233	1651	668	20.5
1500H2	1500			0.94	97.42	9.15	365.163	8.820	5977	5050	4058	1204	1704	654	19.6
2000H2	2000			0.91	99.63	9.46	367.219	9.102	6050	5096	4150	1175	1750	639	19.9
1500H2_CH4f	1500	2058	2000-2014	0.91	98.65	9.47	372.408	9.130	6071	5135	4095	1269	1761	646	19.7
2000H2_CH4f	2000	2171		0.88	101.40	9.92	378.976	9.551	6202	5246	4209	1275	1843	632	19.2
500H2_O3pre	500	1835	CO, NO <sub>x</sub> , VOC reduced	1.02	90.11	8.44	348.249	8.220	5565	4727	3625	1261	1493	667	21.4
1500H2_O3pre	1500			0.95	95.29	9.17	354.174	8.832	5689	4846	3819	1196	1606	632	20.3
2000H2_O3pre	2000			9.23	97.39	9.44	356.144	9.079	5746	4889	3902	1159	1646	619	19.9
500H2_CH4f_O3pre	500	1652	CH <sub>4</sub> , CO, NO <sub>x</sub> , VOC reduced	1.06	88.73	8.14	339.498	7.922	5469	4629	3573	1203	1434	671	22.0
1000H2_CH4f_O3pre	1000	1756		1.00	92.23	8.64	347.956	8.349	5585	4750	3705	1200	1525	652	21.0
2000H2_CH4f_O3pre	2000	1961		0.91	98.07	9.61	359.668	9.245	5804	4947	3926	1198	1600	617	19.7

Table S2. Ozone budget reaction tendencies, along with global mean OH and HO<sub>2</sub> concentrations, O<sub>3</sub> burdens and CH<sub>4</sub> and H<sub>2</sub> lifetimes with respect to OH, under the different hydrogen economy scenarios explored.