Supplement of

Atmospheric composition and climate impacts of a future hydrogen economy

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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

$$H(t) = \alpha_H S_H \left(1 - \exp\left(\frac{-t}{\alpha_H}\right)\right) \tag{SE1}$$

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 yr^{-1}) forming a pulse of time length t_p, and α_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

$$\frac{dM}{dt} = a_M \alpha_H S_H \left(1 - exp\left(\frac{-t}{\alpha_H}\right)\right) - \left(\frac{M}{\alpha_M}\right) \tag{SE2}$$

where M represents the change in the global mean mixing ratio of the radiatively active species (in this case CH_4, O_3 or H_2O), relative to the initial background condition, α_M is the production rate of M per unit change in H_2, and α_M is the lifetime of M. Rearranging and incorporating an integrating factor of $exp(t/\alpha_M)$ give

$$\frac{d\left(M\, exp\left({}^{t}/_{\alpha_M}\right)\right)}{dt} = a_M \alpha_H S_H \left(exp\left(\frac{t}{\alpha_M}\right) - exp\left(t\left(\frac{1}{\alpha_M} - \frac{1}{\alpha_H}\right)\right)\right) \tag{SE3}$$

which on integrating (assuming $M = 0$ at $t = 0$), gives the time evolution of the radiatively active species during the H_2 emission pulse

$$M(t) = a_M \alpha_H S_H \alpha_M \left(\left(1 - exp\left(\frac{-t}{\alpha_M}\right)\right) - \left(\frac{\alpha_H}{\alpha_H - \alpha_M}\right)\left(exp\left(\frac{-t}{\alpha_H}\right) - exp\left(\frac{-t}{\alpha_M}\right)\right)\right) \tag{SE4}$$

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).

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

$$\int_0^{tp} M\,dt = a_M \alpha_H S_H \alpha_M \left(t_p - \alpha_M \left(1 - exp\left(\frac{-t_p}{\alpha_M}\right)\right) - \left(\frac{\alpha_H}{\alpha_H - \alpha_M}\right)\left(\alpha_H \left(1 - exp\left(\frac{-t_p}{\alpha_H}\right)\right) - \alpha_M \left(1 - exp\left(\frac{-t_p}{\alpha_M}\right)\right)\right)\right) \tag{SE5}$$

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

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, α_H. Taking t' to be the time since the end of the pulse, i.e. where $t' = t - t_p$,

$$\frac{dH}{dt'} = H_p\, exp\left(\frac{-t'}{\alpha_H}\right) \tag{SE6}$$

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

$$\frac{dM}{dt'} = a_M\, H_p\, exp\left(\frac{-t'}{\alpha_H}\right) - \frac{M}{\alpha_M} \tag{SE7}$$

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

$$M(T) = \frac{a_M\, H_p}{{}^{1}/_{\alpha_M} - {}^{1}/_{\alpha_H}} \left(\exp\left(-\frac{T}{\alpha_H}\right) - \exp\left(-\frac{T}{\alpha_M}\right)\right) \tag{SE8}$$

Where Hz is the time horizon considered, integrating (SE8) between $T = 0$ and $T = Hz - t_p$ 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)

$$\int_0^{Hz-t_p} M\, dT = \frac{\left(a_M\, \alpha_H^2\, \alpha_M\, S_H \left(1 - \exp\left(\frac{-t_p}{\alpha_H}\right)\right)\right)}{(\alpha_H - \alpha_M)} \left(\alpha_H \left(1 - \exp\left(-\frac{(Hz - t_p)}{\alpha_H}\right)\right) - \alpha_M \left(1 - \exp\left(-\frac{(Hz - t_p)}{\alpha_M}\right)\right)\right) \tag{SE9}$$

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

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

$$M(t') = M_p \exp\left(\frac{-t'}{\alpha_M}\right) \tag{SE10}$$

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.

$$\int_0^{Hz-t_p} M\, dt' = a_M\, \alpha_M^2 \alpha_H\, S_H \left(\left(1 - \exp\left(-\frac{t_p}{\alpha_M}\right)\right) - \left(\frac{\alpha_H}{\alpha_H - \alpha_M}\right)\left(\exp\left(-\frac{t_p}{\alpha_H}\right) - \exp\left(-\frac{t_p}{\alpha_M}\right)\right)\right)\left(1 - \exp\left(-\frac{(Hz - t_p)}{\alpha_M}\right)\right) \tag{SE11}$$

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.

Hydrogen economy simulation	H_2 (ppb)	CH_4 (ppb)	O3_pre Emiss.	[OH] GM (10^6 molec cm^{-3})	[HO_2] GM (10^6 molec cm^{-3})	τ_{CH4} (yr)	O_3 Burden (Tg)	$\tau_{H2(OH)}$ (yr)	O_3 Prod. (Tg yr^{-1})	O_3 Loss (Tg yr^{-1})	HO_2+NO (Tg yr^{-1})	MeO_2+NO (Tg yr^{-1})	HO_2+O_3 (Tg yr^{-1})	OH+O_3 (Tg yr^{-1})	OH+SO_2 (Tg yr^{-1})
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_x, 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_4, CO, NO_x, 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 S1. Ozone budget reaction tendencies, along with global mean OH and HO_2 concentrations, O_3 burdens and CH_4 and H_2 lifetimes with respect to OH, under the different hydrogen economy scenarios explored.