Supplementary Information (SI) for The effect of surface sink saturation and emission altitude on hydrogen's atmospheric impact

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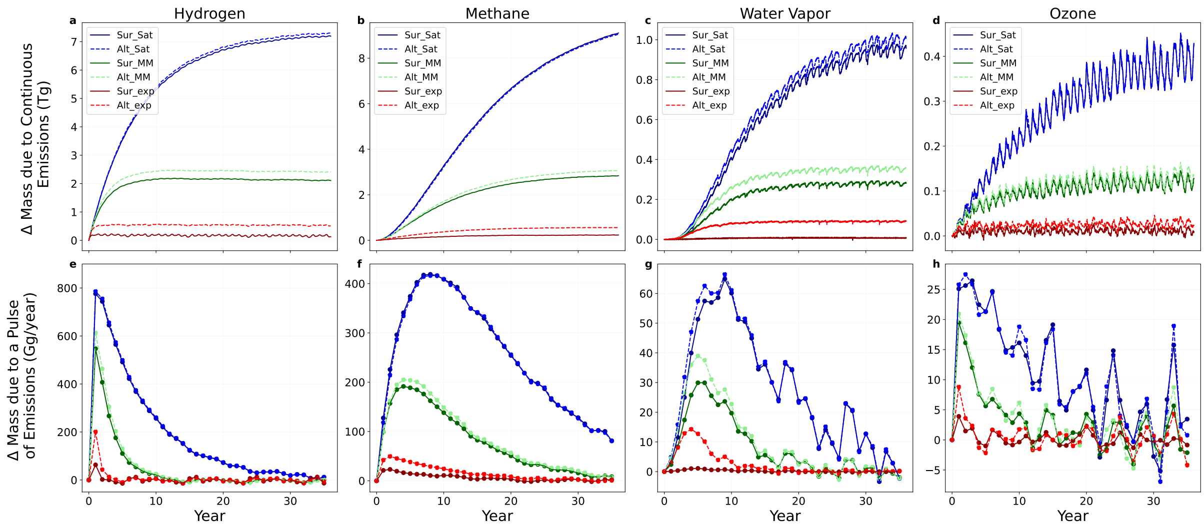
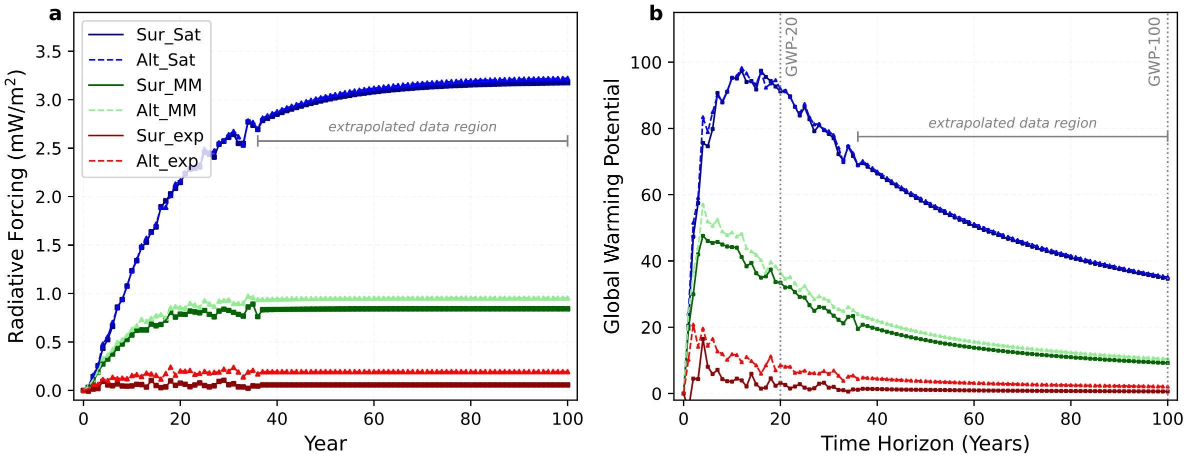
S1 Exponential soil sink uptake model

In addition to the unsaturated sink model presented in the manuscript, we also assess an alternate model for hydrogen uptake. This second dynamic flux boundary condition we implement for hydrogen uses an exponential relationship to vary the scale factor as a function of changes in surface hydrogen concentration. This formulation is parametrized as $F_{\text{new}} = F_{\text{old}}(1+\alpha)^{\beta}$. Here, α represents the relative difference in surface hydrogen concentration between the simulation and the reference case, and β is a tunable exponential parameter. We adopt a value of 25 for β, as an upper bound for uptake of hydrogen that produces the minimum positive response in methane, water vapor, and ozone in response to the hydrogen perturbation experiments. This exponential formulation represents an idealized, extreme case of an entirely unsaturated soil sink, in which biotic hydrogen uptake increases drastically in response to elevated atmospheric concentrations, greatly buffering the net hydrogen climate impact.

While there is currently no empirical evidence to suggest, or reason to suspect, such a response in the hydrogen soil sink, this model serves as a valuable bounding case to contrast with the fully saturated regime. By bounding the extreme cases of sink behavior, we can better understand the potential range of atmospheric responses to hydrogen emissions and evaluate the sensitivity of climate impacts to uncertainties in future sink dynamics.

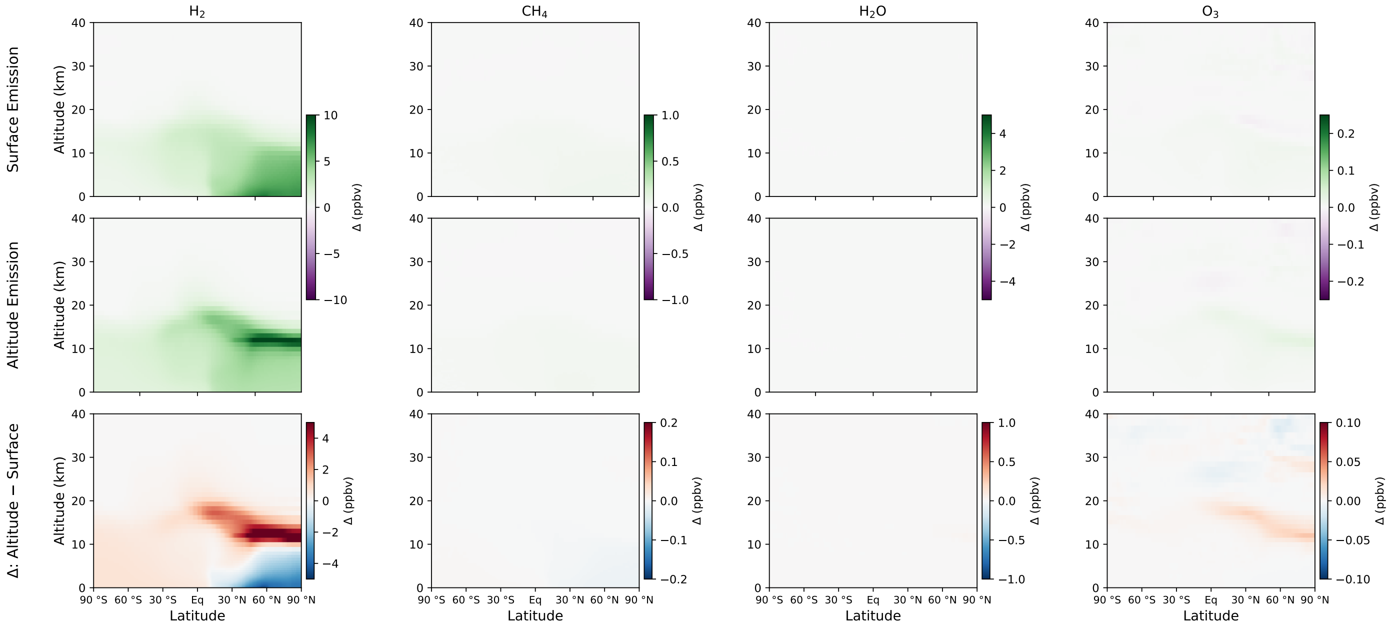


Supplementary Figure S1: Changes in the masses of atmospheric hydrogen, methane, stratospheric water vapor, and tropospheric ozone relative to the reference scenario, resulting from 1 Tg of annually recurring hydrogen emissions. The top row shows the step response, obtained directly from daily GCHP model output. The bottom row shows the corresponding impulse response, calculated as the numerical derivative of the step response averaged over two years. The blue curves and green curves represent the response to the saturated and unsaturated surface sinks, respectively, as presented in the manuscript. The red curves represent the response when using the exponential parameterization of the soil sink as discussed above. The light and dark shaded curves represent the response to hydrogen emitted at an altitude of 11 km and at the surface, respectively.

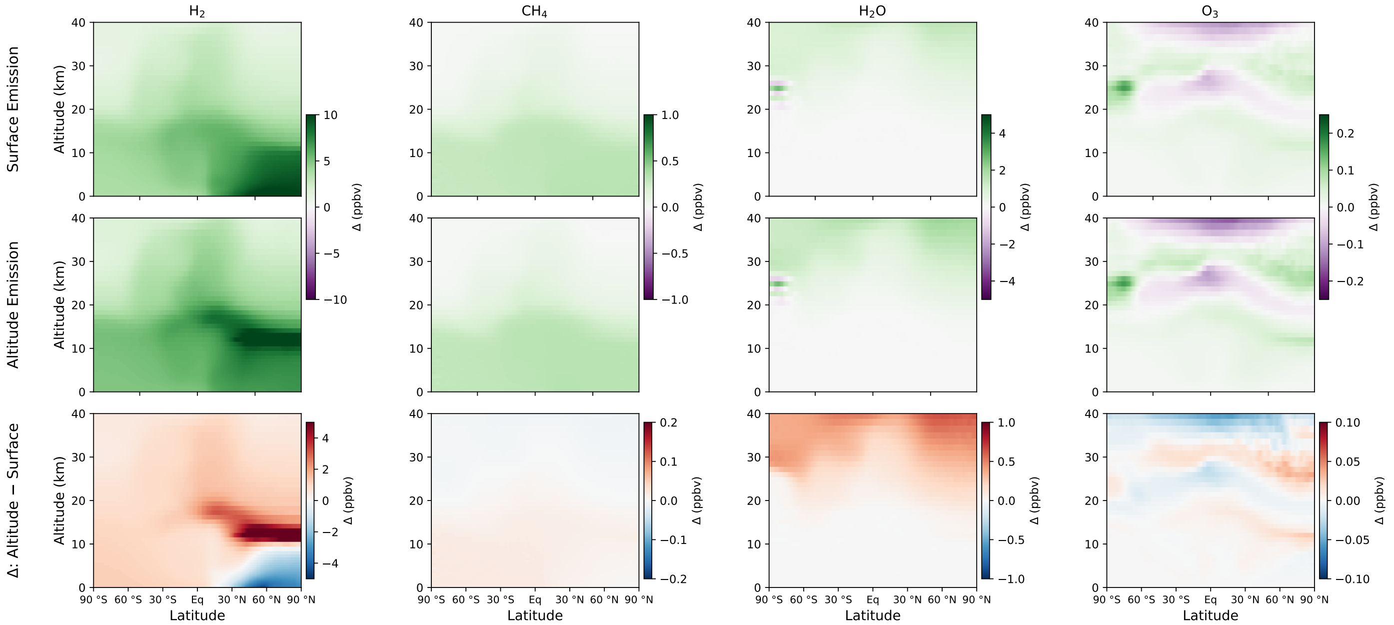


Supplementary Figure S2: Stratospherically-adjusted radiative forcing relative to the reference scenario resulting from 1 Tg of annually recurring hydrogen emissions (a) and the associated GWP of hydrogen over varying time horizons (b). The blue curves and green curves represent the response to the saturated and unsaturated surface sinks presented in the manuscript, respectively. The red curves show the response from the alternate model using the exponential parameterization discussed above. The light and dark shaded curves represent the response to hydrogen emitted at an altitude of 11 km and at the surface, respectively. An exponential fit is applied to the hydrogen radiative forcing curves beyond year 36, after which the GEOS-Chem simulations had concluded.

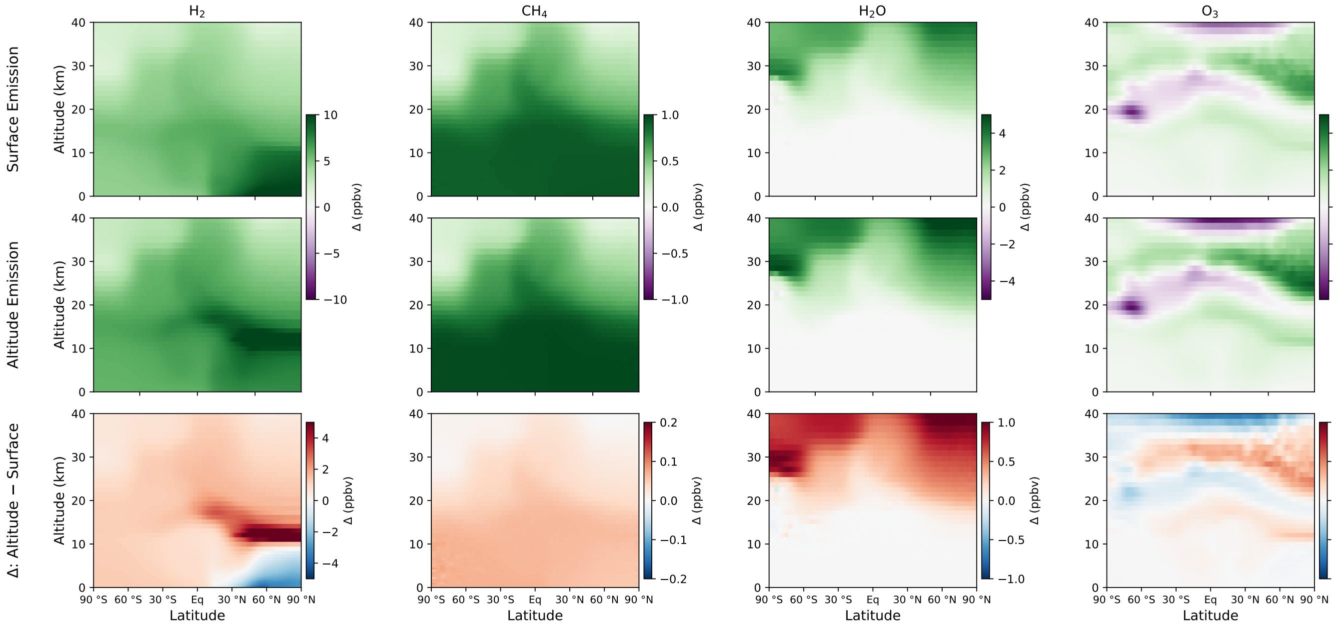
S2 Zonal mean changes in key species over simulation period



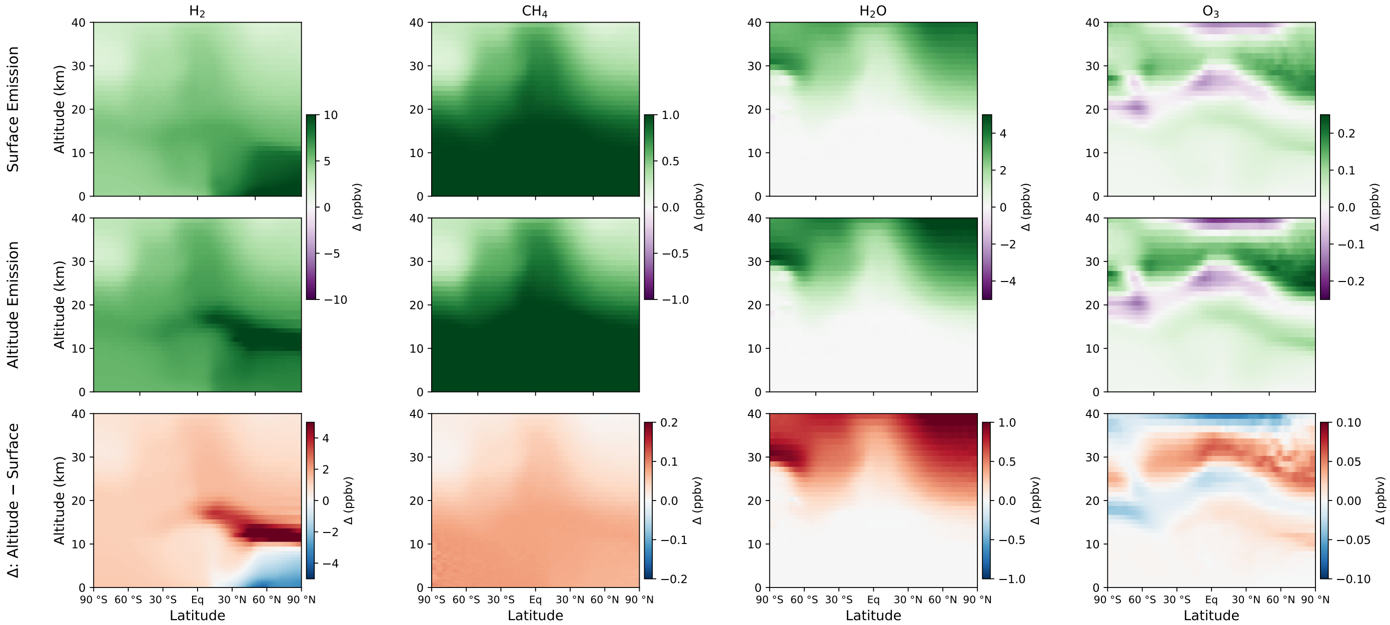
Supplementary Figure S3: Zonal mean abundances of key species after 1 year of simulation time for the unsaturated soil sink scenarios presented in the manuscript. From left to right, the columns show changes in hydrogen, methane, water vapor and ozone, respectively. The top row shows the response to hydrogen emitted at the surface (Sur_MM), the middle row shows the response to hydrogen emitted at altitude (Alt_MM), and the bottom row shows the difference between those two scenarios (Alt_MM – Sur_MM) which represents the additional impact of emitting at altitude compared to at the surface. Results shown here for the monthly mean in August of the specified simulation year.



Supplementary Figure S4: As in Figure S3, except results here are shown after 5 years of simulation.

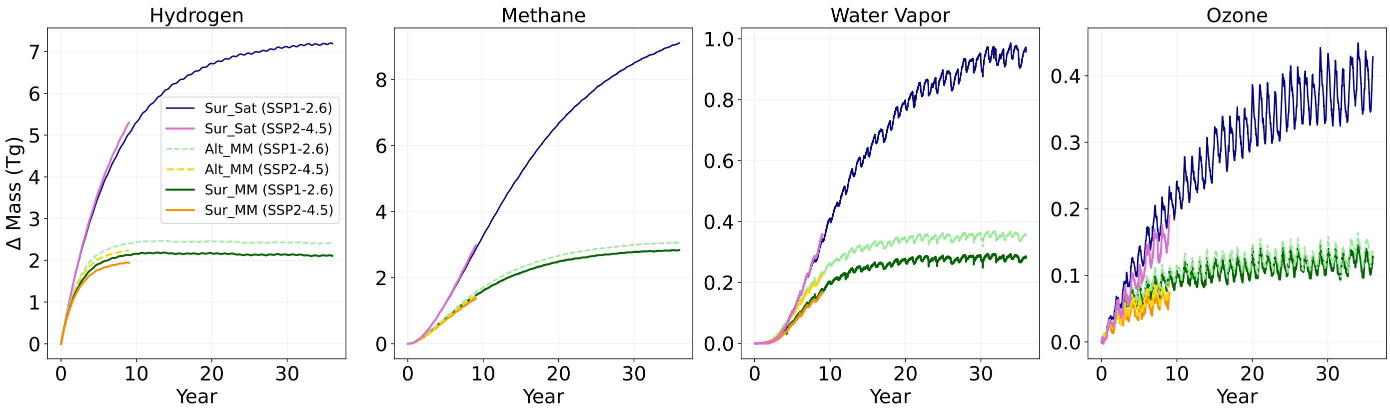


Supplementary Figure S5: As in Figure S3, except results here are shown after 20 years of simulation.



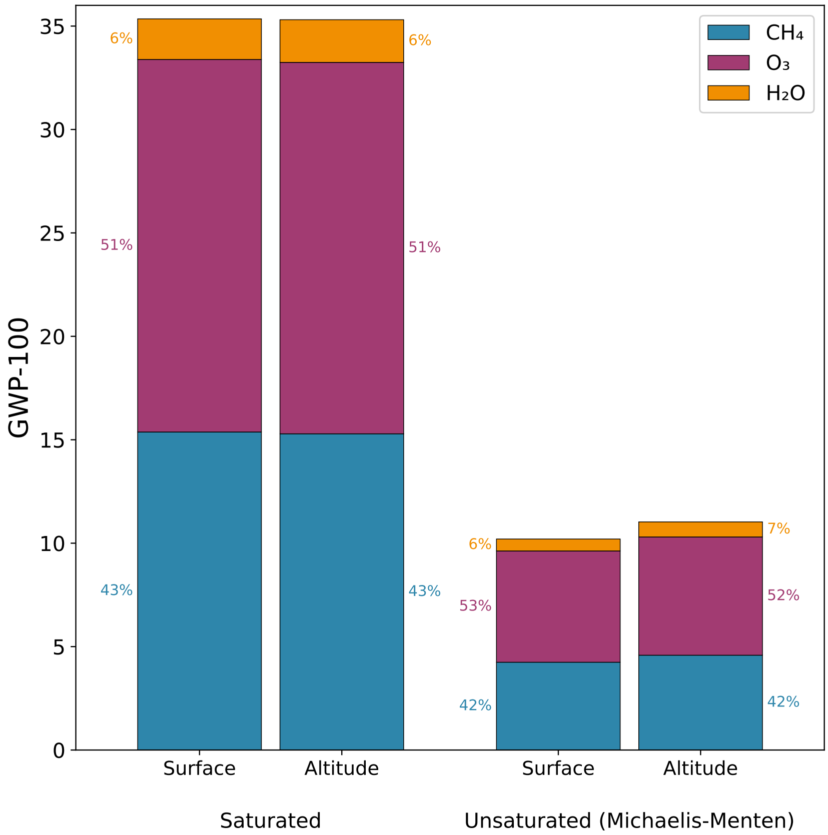
Supplementary Figure S6: As in Figure S3, except results here are shown after 30 years of simulation.

S3 Sensitivity to the Background Atmosphere



Supplementary Figure S7: Step response of the changes in the masses of atmospheric hydrogen, methane, stratospheric water vapor, and tropospheric ozone relative to the reference scenario, resulting from 1 Tg of annually recurring hydrogen emissions. The green curve shows the Sur_MM scenario using an SSP1-2.6 background atmosphere (exactly as presented in the manuscript), and the yellow curve shows the same scenario but using the SSP2-4.5 background atmosphere.

S4 Breakdown of Species Contributions to H_2's GWP-100



Supplementary Figure S8: Contributions of methane, tropospheric ozone, and stratospheric water vapor to the GWP-100 of hydrogen, calculated using published radiative efficiencies. The left bars show the GWP contributions for the saturated sink (_Sat scenarios) and the right bars show the contributions for the unsaturated sink (_MM scenarios).