



1 **Interannual variations and future changes in the soil uptake of** 2 **hydrogen**

3 Ye Wang*, Oliver Wild*, Xuewei Hou, Ryan Hossaini

4 Lancaster Environment Centre, Lancaster University, Lancaster, UK

5 *Correspondence to:* Ye Wang (y.wang251@lancaster.ac.uk), Oliver Wild (o.wild@lancaster.ac.uk)

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7 **Abstract:** Hydrogen (H_2) is under consideration as a green energy source for future low-carbon development
8 pathways. However, assessment of the future climate impacts of large-scale H_2 use is currently limited by poor
9 understanding of the dominant atmospheric removal process, the uptake of H_2 by microbial activity in soil. Here
10 we implement a soil dry deposition scheme for H_2 in a global chemistry transport model and conduct a sensitivity
11 analysis to quantify how uncertainties in soil parameters contribute to uncertainty in H_2 uptake. We find that soil
12 moisture dominates the uncertainty (52 %), along with contributions from soil porosity (34 %) and the water
13 threshold for biological activity (13 %). However, the sensitivity of uptake to soil moisture is strongly nonlinear,
14 with increases in uptake with declining soil moisture where it is under biotic control and with increasing soil
15 moisture where it is under diffusivity control. We show that soil uptake dominates the interannual variation in
16 atmospheric H_2 abundance from 2010 to 2022, and the notable decrease in 2015, while enhanced atmospheric
17 production drives the observed long-term increase. Our results suggest that both soil sink and source of H_2 are
18 affected by strong ENSO events driven by changes in soil moisture. Using output from 11 CMIP6 models, we
19 project changes in H_2 soil uptake between 2015 and 2100 of -2.5 % to $+7.9$ % under SSP1-2.6 and $+4.1$ % to
20 $+20.4$ % under SSP5-8.5, suggesting an increase in soil uptake under climate change associated with future
21 changes in soil moisture.

22



23 1. Introduction

24 Hydrogen (H_2) is the second most abundant oxidizable trace gas in the troposphere (Ehhalt and Rohrer, 2009),
 25 and currently has a global average mixing ratio of approximately 530 parts per billion (ppb) (Novelli et al., 1999;
 26 Patterson et al., 2020). The dominant photochemical sink of H_2 in the troposphere is reaction with hydroxyl
 27 radicals (OH). However, reaction with OH is also the major atmospheric sink for methane (CH_4), and hence an
 28 increase in H_2 concentrations leads to a longer lifetime and a higher atmospheric abundance of CH_4 (Bryant et al.,
 29 2024; Derwent et al., 2001; Derwent et al., 2020; Field and Derwent, 2021; Warwick et al., 2004). This causes
 30 increased production of the greenhouse gas ozone (O_3), and affects the formation of a wide range of tropospheric
 31 trace gases and aerosol (O'Connor et al., 2022). In the stratosphere, the oxidation of hydrogen and methane lead
 32 to elevated levels of stratospheric water vapor, which may have significant implications for climate and
 33 stratospheric O_3 (Tromp et al., 2003; Warwick et al., 2004). Moreover, increases in atmospheric hydrogen affect
 34 the recovery of stratospheric O_3 by increasing the production of HO_x radicals (i.e., OH and HO_2), which play a
 35 role in O_3 destruction (Warwick et al., 2023). As a result, changes in H_2 abundance cause a range of indirect climate
 36 impacts though the troposphere and stratosphere. The 100-year global warming potential (GWP100) of H_2 is
 37 estimated to be 11.6 ± 2.8 (Sand et al., 2023), although a recent study adjusting OH concentrations to match the
 38 observationally-derived CH_4 chemical lifetime suggests a 20% lower estimate of 8.8 (Yang et al., 2025).

39 Observations at background sites suggest that global average surface H_2 mixing ratios increased by about 1.4 ± 0.7
 40 ppb yr^{-1} between 2010 and 2019 because of increased H_2 industrial and leakage sources (Paulot et al., 2024). In
 41 a future low-carbon economy, widespread usage of H_2 is considered a promising alternative as an energy carrier
 42 to fulfil increasing energy demand while replacing fossil fuel use. Over recent years, there has been increased
 43 interest and investment in H_2 worldwide (International Energy Agency, 2024), resulting in enhanced H_2 demand.
 44 Global H_2 demand was almost 100 Mt yr^{-1} in 2024 and is predicted to reach 402–660 Mt yr^{-1} by 2050 (Trapani
 45 et al., 2025). Along with the increasing demand for H_2 , leakages across the supply chain are projected to rise from
 46 1.3 Mt yr^{-1} in 2023 to 22.0–45.3 Mt yr^{-1} in 2050. Such a dramatic increase in leakage raises concerns about the
 47 future climate impacts of increased H_2 concentrations, and thus good understanding of the atmospheric H_2 budget
 48 is very important.

49 The tropospheric H_2 abundance is dominated by four processes: surface emissions, atmospheric production,
 50 atmospheric loss, and the soil sink. Photolysis of formaldehyde (HCHO) is the main photochemical source of H_2 ,
 51 and this exceeds direct anthropogenic sources from fossil fuel combustion and leakage. Reaction with OH is the
 52 main photochemical sink of H_2 , but this is less important than uptake by microbial activity in soil, which accounts
 53 for 70–80 % of the total loss. A recent assessment of the H_2 budget for 2010 based on an ensemble of atmospheric
 54 models found sources from chemical production (47 Tg yr^{-1}) and emissions (36 Tg yr^{-1}) balanced by sinks to
 55 photochemistry (25 Tg yr^{-1}) and soil uptake (57 Tg yr^{-1}) (Sand et al., 2023). Another study estimated an annual
 56 average sink of H_2 of 68.4 Tg yr^{-1} during 2010–2020, with soil uptake accounting for 73 % of this (Ouyang et al.,
 57 2025). In contrast to the other processes, uncertainty in assessment of the soil sink of H_2 remains high because of
 58 limited understanding of the governing biological processes (Paulot et al., 2021).

59 Soil uptake of H_2 is typically represented in models through calculation of a two-layer deposition velocity (Brown
 60 et al., 2025; Ehhalt and Rohrer, 2013; Paulot et al., 2021; Yonemura et al., 2000). A new, more comprehensive
 61 scheme was recently developed by Bertagni et al. (2021), coupling water and hydrogen dynamics in 12 soil
 62 textures, with thresholds for estimating the deposition velocity based on the relationship between soil
 63 characteristics and soil water potential. This scheme delivers a lower H_2 soil uptake (47.2 Tg yr^{-1} averaged over
 64 2003–2013) (Stroo et al., 2026) than previous estimates. The soil moisture dependence from this scheme was
 65 subsequently adopted in an existing H_2 deposition scheme (Paulot et al., 2024), which was shown to represent the
 66 global trend in H_2 concentration and the latitude distribution of deposition velocities relatively well. A lower water-



67 activation threshold for the canopy and litter resistance in this scheme was shown to provide a better seasonality
 68 but greatly overestimated the hemispheric gradient.

69 Given the critical role of soil uptake processes in controlling the atmospheric abundance of H_2 , it is important to
 70 understand the uncertainty in representation of these processes. Recent studies have focused on the range of uptake
 71 rates across different models. For instance, [Brown et al. \(2025\)](#) calculated a 20 % spread in H_2 soil uptake based
 72 on results from Coupled Model Intercomparison Project Phase 6 (CMIP6) models due to large differences in soil
 73 moisture and porosity. Parameter perturbation approaches to identify the sensitivity to different processes are
 74 computationally expensive and impractical for complex models, but Gaussian process emulation approaches have
 75 been successfully applied to address this ([Ryan et al., 2018](#); [Wild and Ryan, 2026](#)). [Wild and Ryan \(2026\)](#) used
 76 Gaussian process emulation with a global chemistry transport model to show that the largest contributions to
 77 uncertainty in tropospheric ozone and OH arise from photochemical parameters, and from factors controlling
 78 photolysis, which initiates tropospheric photochemistry.

79 Despite a sustained increase since 2010, the observed atmospheric H_2 abundance exhibited an unexpected drop
 80 during 2014–2015 (Fig. A1). The causes of this interannual variability remain poorly quantified, even though
 81 several studies have reproduced the overall trend. [Paulot et al. \(2024\)](#) simulated H_2 concentrations from 2010 to
 82 2019, and suggested that the failure to reproduce the observed increase in H_2 concentrations at background sites
 83 could be attributed to underestimation of current anthropogenic emissions, including potential leakages from H_2 -
 84 producing facilities. [Surawski et al. \(2026\)](#) performed a long-term budget simulation and reproduced the upward
 85 trend in observed H_2 from 2010 to 2023. [Brown et al. \(2026\)](#) implemented H_2 emissions and an H_2 soil uptake
 86 scheme into the UK Earth System Model and showed that the model captures long-term trends from the pre-
 87 industrial to 2012, and reproduces the observed seasonal cycles of both H_2 and CH_4 . A recent H_2 inversion study
 88 from [Stroo et al. \(2026\)](#) suggested that the growth of H_2 between 2003–2023 was primarily driven by changes in
 89 the photochemical source from CH_4 oxidation, together with a declining global soil sink. However, the role of soil
 90 uptake in interannual H_2 variability in recent years is not well understood and remains a significant research gap.

91 The analytical results of [Bertagni et al. \(2021\)](#) suggest that H_2 diffusion through the soil generally limits H_2 uptake
 92 in humid regions, while biotic limitations occur in hyperarid soils where bacteria suffer from a lack of water.
 93 However, this was based on analysis of a single year, and variation in long-term historical H_2 dry deposition
 94 velocity and its sensitivity to soil parameters has not been fully explored. The influence of intense climate events
 95 such as El Niño on H_2 soil uptake also remains unclear. Using CMIP6 output data from five models with a two-
 96 layer deposition scheme, [Brown et al. \(2025\)](#) showed changes in global mean deposition velocity from 2015 to
 97 2100 under a range of climate scenarios. Results indicated that the deposition velocity increased by 4.8 % under
 98 the SSP1-2.6 scenario between 2015 and 2100 and by 16.7 % under SSP5-8.5. However, they focused on
 99 quantifying the spread of future deposition velocities from different models and assessing inter-model uncertainty,
 100 while the mechanisms underlying these projected increases remain poorly understood.

101 This study provides an assessment of the overall uncertainty in H_2 soil deposition velocity from major soil
 102 parameters, quantifies the contribution of the soil sink to interannual variation in tropospheric H_2 abundance, and
 103 projects the future H_2 soil uptake from 2015 to 2100. We introduce the atmospheric model, soil deposition scheme,
 104 and soil parameter data in Sect. 2. We compare the simulated H_2 deposition velocity, tropospheric budget, and
 105 surface mixing ratios with observations in Sects. 3.1 and 3.2. We quantify the uncertainty in deposition from
 106 different soil factors in Sect. 3.3. We then diagnose the spatial and temporal sensitivity of soil deposition velocity
 107 to soil moisture in Sect. 3.4, allowing us to explain the changes in velocity from 2010 to 2022. We further explore
 108 the role of the soil sink in the interannual variation in H_2 abundance in Sect. 3.5, and project the changes H_2 soil
 109 deposition under different future scenarios in Sect. 3.6. We present our conclusions in Sect. 4.



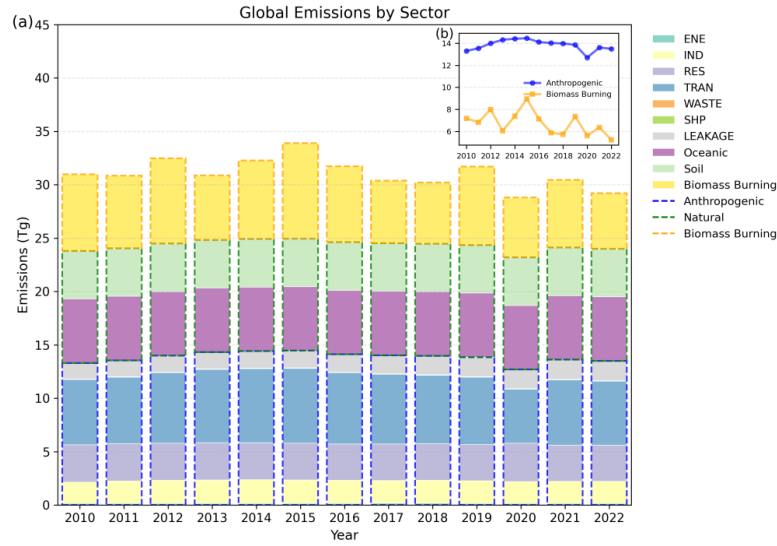
110 2. Data, model, and methods

111 2.1 Global chemical transport model

112 To simulate the global atmospheric H₂ budget we use the Frontier Research System for Global Change version of
 113 the University of California Irvine Chemical Transport Model (FRSGC/UCI CTM) as described in [Wild \(2007\)](#).
 114 This model was developed to represent gas-phase processes and was run at T42 (2.8° × 2.8°) horizontal resolution
 115 with 57 vertical levels between the surface and 64 km driven by meteorological fields from the European Centre
 116 for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast System (IFS). We use an updated version
 117 of the model that includes iodine chemistry. We have also expanded the treatment of volatile organic compounds
 118 to include acetone and methanol chemistry, as these influence the abundance of HCHO which is important as a
 119 source of H₂.

120 Anthropogenic emissions of CO, NO, and VOC species were taken from the Copernicus Atmosphere Monitoring
 121 Service global emissions (CAMS-GLOB-ANTH) v6.2 emission inventory, while biogenic emissions were from
 122 the CAMS-GLOB-BIO dataset. Soil emissions of NO_x were from CAMS-GLOB-SOIL v2.4. Biomass burning
 123 emissions for species including H₂ were taken from the CAMS Global Fire Assimilation System (GFAS) v1.2.
 124 We estimated H₂ anthropogenic emissions from combustion sources using CO-based emission factors (H₂/CO)
 125 from [Paulot et al. \(2021\)](#) for the relevant emission sectors. The oceanic source of H₂ was taken to be 6.0 Tg yr⁻¹
 126 following previous studies ([Ehhalt and Rohrer, 2009](#); [Price et al., 2007](#); [Rhee et al., 2006](#); [Yashiro et al., 2011](#)),
 127 and distributed based on oceanic emissions of CO from the POET inventory ([Granier et al., 2005](#)). Soil emissions
 128 are assumed to be 4.5 Tg yr⁻¹, following [Paulot et al. \(2024\)](#), and are distributed according to NO_x soil emission.
 129 We assume a H₂ leakage rate of 2.0 % each year, taking total H₂ demand from the Global Hydrogen Review 2024
 130 ([International Energy Agency, 2024](#)), and distribute this based on transportation CO emissions. To avoid double-
 131 counting, agricultural waste burning has been removed from the anthropogenic source, as this sector is already
 132 included in GFAS. Overall, annual H₂ emissions are higher in the Northern Hemisphere than in the Southern
 133 Hemisphere, with anthropogenic sources dominating in the former and biomass burning in the latter (Fig. S1).

134 Fig. 1 shows the global total annual emissions of H₂ from anthropogenic (energy, industry, residential and
 135 commercial buildings, transportation, waste, ships, and leakage), natural (soil and oceanic), and biomass burning
 136 sources based on the above estimates during 2010–2022. As variations in anthropogenic and biomass burning
 137 emissions are the primary contributor to the annual variation in H₂ emissions, we show these separately in Fig. 1.
 138 Transportation dominates anthropogenic sources, with emissions from industrial, residential, and leakage also
 139 playing important roles. Driven by high anthropogenic (14.5 Tg) and biomass burning (9.0 Tg) emissions in 2015,
 140 H₂ total emission peaked at 33.9 Tg yr⁻¹ and then decreased to 29.2 Tg yr⁻¹ in 2022. Our estimates are lower than
 141 those of [Paulot et al. \(2024\)](#) due to lower anthropogenic CO emissions and biomass burning from the inventories
 142 we are using, but fall within the range of 27–68 Tg yr⁻¹ in 2010 from [Sand et al. \(2023\)](#), and align well with the
 143 emissions of 31.5 Tg yr⁻¹ averaged over 2010–2020 from the recent H₂ budget assessment of [Ouyang et al. \(2025\)](#).



144

145 Figure 1. H₂ emissions from different sources from 2010 to 2022. Anthropogenic, natural, and biomass burning
 146 sources are outlined separately with blue, green, and orange dashed lines, respectively. Variations in anthropogenic
 147 and biomass burning emissions from 2010 to 2022 are shown in the inset in the upper righthand corner.

148 The standard control experiment (CTRL) simulates the variation in H₂ concentration and associated budgets from
 149 2010 to 2017 under varying meteorology, including interannually varying anthropogenic, biomass-burning, and
 150 natural emissions. We extend these runs from 2018 to 2022 using varying emissions but holding meteorological
 151 conditions fixed at 2017 as we do not have a consistent set of meteorological fields for the following years. We
 152 impose annual mean latitudinally-varying atmospheric mixing ratios for CH₄ following CMIP6 recommendations,
 153 extending these beyond 2015 using NOAA annual mean observations such that the global mean abundance of
 154 CH₄ shows an increase from 1808 ppb in 2010 to 1920 ppb in 2022. We then perform an experiment with a
 155 modified soil source to explore the influence of changes in soil emissions during 2014–2015, and a simulation
 156 with deposition velocities fixed at 2010 values to isolate the contribution of soil uptake to interannual variability
 157 in H₂ from 2010 to 2022. Simulations were started in 2008, and the first two years were used as spin-up.

158 2.2 Deposition velocity calculation

159 The deposition scheme of [Paulot et al. \(2024\)](#) was applied in this study to calculate H₂ dry deposition velocity.
 160 The deposition velocity $v_d(H_2)$ can be expressed as:

$$161 \quad \frac{1}{v_d(H_2)} = \frac{1}{g_i} + \frac{1}{g_s}$$

162 where g_i represents the H₂ conductance through an inactive surface layer of litter or snow and g_s represents the
 163 conductance in the soil. The latter is expressed following [Ehhalt and Rohrer \(2013\)](#) as

$$164 \quad g_s = \sqrt{k_m h(T) f D_s}$$



where $h(T)$ and f are the sensitivity of H_2 biological uptake to soil temperature and soil moisture, respectively. We show the function of $h(T)$ and f for Sand and Loam soil types in Fig. S2. D_s is the moisture-dependent diffusivity of H_2 in the soil. k_m represents the maximum uptake rate of H_2 and is used to constrain the annual mean deposition velocity over land to a reference value of 0.035 cm s^{-1} . Moisture dependencies were calculated following Bertagni et al. (2021) as

$$\frac{D_s}{D_0} = \alpha_1 n^{\alpha_2} (1 - s)^{\alpha_3}$$

where D_0 ($\text{cm}^2 \text{ s}^{-1}$) is the free air diffusion coefficient suggested by Ehhalt and Rohrer (2013), and the values for α_1 , α_2 , α_3 are assigned to be 1, 2, and $2 + 3/b$. Here n and b have specific values for different soil textures (see Table S1 and Bertagni et al. (2021) for details). Further details of the calculation of f are given in the Supplement.

When regions are covered by snow, $g_i = g_{\text{snow}}$, and this is calculated from snow depth assuming a snow porosity of 0.645, following Pinzer et al. (2010). The standard scheme (S1) neglects canopy and litter resistances. We therefore additionally explore an alternative scheme (S2), which accounts for canopy and litter conductance and adopts a lower water-activation threshold (-10000 kPa). A comparison of the soil moisture thresholds for different soil types supporting biological activity for the standard (-3000 kPa) and the lower (-10000 kPa) water-activation thresholds is provided in Table S1. Based on the study of Makar et al. (2018), the canopy resistance was assigned specific values for different vegetation types. The litter conductance is estimated assuming a litter porosity of 0.62 (Wang et al., 2019). The litter depth is estimated from simulated above-ground carbon from the IPSL Interaction with Chemistry and Aerosols (INCA) model historical simulation (Boucher et al., 2018), assuming a density of 0.03 g cm^{-3} . When we apply scheme S2, k_m is used to adjust the value of annual land mean deposition velocity in 2010 to be the same as that calculated based on the standard scheme (we apply 0.185 for the standard scheme and 0.30 for the alternative scheme). We use the soil parameters described in Sect. 2.3 below to drive an offline calculation of H_2 deposition velocity, and apply the resulting deposition velocity in the CTM to calculate the dry deposition of H_2 .

2.3 Soil parameters

For historical estimation, we use parameters of soil texture, soil porosity, surface atmospheric pressure, snow depth, upper 10 cm soil temperature and soil moisture content from the Global Land Data Assimilation System, GLDAS (<https://disc.gsfc.nasa.gov/datasets?keywords=GLDAS>) Noah v2.1, with a horizontal resolution of $1.0^\circ \times 1.0^\circ$. We adopt monthly parameters for calculation of deposition velocities, with invariant values for soil texture and porosity. The soil moisture content (kg m^{-2}) is converted to soil volumetric water content ($\text{cm}^3 \text{ cm}^{-3}$) using water density in the upper 10 cm.

Table 1. CMIP6 models and soil parameters selected to project future H_2 deposition velocity.

Models	Soil moisture content (kg m^{-2})	Soil temperature (K)	Snow depth (m)
ACCESS-CM2	✓	✓	-
BCC-CSM2-MR	✓	✓	✓
CanESM5	✓	✓	✓
CMCC-ESM2	✓	✓	✓
CNRM-CM6-1	✓	✓	✓
EC-Earth3-Veg-LR	✓	✓	✓



FGOALS-g3	✓	✓	✓
FIO-ESM-2-0	✓	-	-
GFDL-ESM4	✓	✓	✓
GISS-E2-1-G	✓	-	-
IPSL-CM6A-LR	✓	✓	✓
KACE-1-0-G	✓	✓	-
MIROC6	✓	✓	✓
MPI-ESM1-2-HR	✓	✓	-
MRI-ESM2-0	✓	✓	✓
UKESM1-0-LL	✓	✓	✓

For future projections of H_2 deposition velocity we applied monthly upper 10 cm soil moisture content, soil temperature, and snow depth outputs from selected models contributing to CMIP6 under different climate pathways (see Table 1), assuming that soil texture and porosity remained unchanged at 2015 levels. The CMIP6 models used by [Cook et al. \(2020\)](#) for drought projections were adopted as an initial reference set, as drought projections provide a useful reference for evaluating future soil moisture changes. We then selected models for which as many of the required variables as possible were available. The SSP1-2.6 and SSP5-8.5 pathways are chosen to represent scenarios with moderate and extreme climate responses, respectively. Parameter-specific trends were calculated using all available outputs (16 for soil moisture content, 14 for soil temperature, and 11 for snow depth). We employed the multi-model median rather than the mean to characterize trends because it is less influenced by outliers with unusually strong or weak trends, and this is particularly important for hydrological variables which commonly exhibit large inter-model variability. Future H_2 deposition velocity was projected individually for the 11 CMIP6 models that included outputs for all three required parameters. To maintain consistency with the GLDAS data, parameters from CMIP6 models were regridded to $1.0^\circ \times 1.0^\circ$ and restricted to regions north of 60° S before trend calculation and deposition velocity estimation. Soil temperature in the upper 10 cm was calculated as a depth-weighted average, when the upper 10 cm soil moisture is available.

3. Results

3.1 Evaluation of H_2 deposition velocity

Fig. 2 shows the spatial distribution of calculated H_2 deposition velocity and a comparison against observed deposition velocities at three selected sites. The zonal mean deposition velocity shows highest soil uptake in the Northern Hemisphere, reaching a peak of 0.021 cm s^{-1} at $30\text{--}40^\circ\text{N}$, and the mean velocity over land is 0.035 cm s^{-1} , which agrees with previous estimates: 0.033 cm s^{-1} (global average over land during 1997–2005) from [Yashiro et al. \(2011\)](#), 0.035 cm s^{-1} (1989–2014) from [Paulot et al. \(2021\)](#), and lower than the 0.041 cm s^{-1} (multi-model mean over 2005–2014) from [Brown et al. \(2025\)](#). Compared to two-layer schemes, the updated moisture thresholds for different land types supporting biological activity represent the observed high uptake in dry regions better ([Jordaan et al., 2020](#); [Smith-Downey et al., 2006](#)).

Table 2. Site information for the H_2 deposition velocity observations used in this study.

Site	Location	Land type	Observation period	References
Havard Forest	$42.54^\circ \text{ N}, -72.17^\circ \text{ E}$	Forest	Dec 2010–Dec 2011	Meredith et al. (2017)
Helsinki	$60.2^\circ \text{ N}, 25.5^\circ \text{ E}$	Forest	Sep 2005–March 2007	Lallo et al. (2008)



Heidelberg	49.40° N, 8.70° E	Semi-urban	Jan 2005–July 2007	<i>Hammer and Levin (2009)</i>
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We evaluate the calculated dry deposition velocities against observations from three sites, Harvard Forest, Helsinki, and Heidelberg, that span a range of locations and vegetation types, see Table 2. Based on the availability of valid measurements, we choose a year of monthly observations in 2011 and 2006, respectively, for Harvard Forest and Helsinki, and use the full observation period (2005–2007) from Heidelberg to compare with our calculations. Our deposition velocity estimates capture the monthly variations well, with a correlation coefficient (R) of 0.69, 0.65, and 0.59 for the three sites. However, the deposition velocity is underestimated during the warm season, especially at Harvard Forest and Helsinki, as reported in other studies (Brown *et al.*, 2025; Paulot *et al.*, 2021; Paulot *et al.*, 2024). While this may indicate a systematic bias, it may also reflect discrepancies associated with comparison of grid-cell mean estimates with point-scale observations.

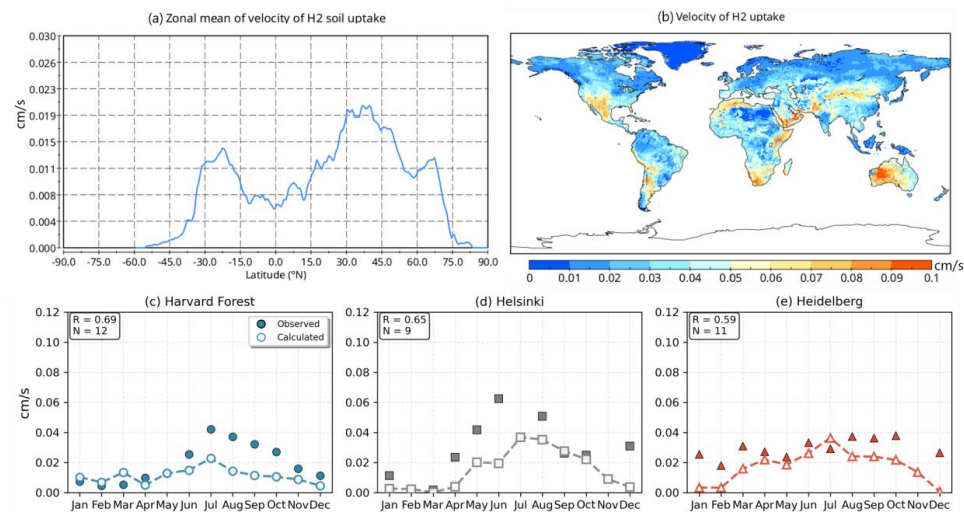


Figure 2. (a) Calculated zonal mean velocity of H₂ soil uptake, (b) distribution of velocity of H₂ soil uptake, and comparison of monthly observed (filled symbols) and calculated (2010, dashed lines) velocities at (c) Harvard Forest (2011), (d) Helsinki (2006), and (e) Heidelberg (2005–2007). See Table 2 for the locations of the observational sites.

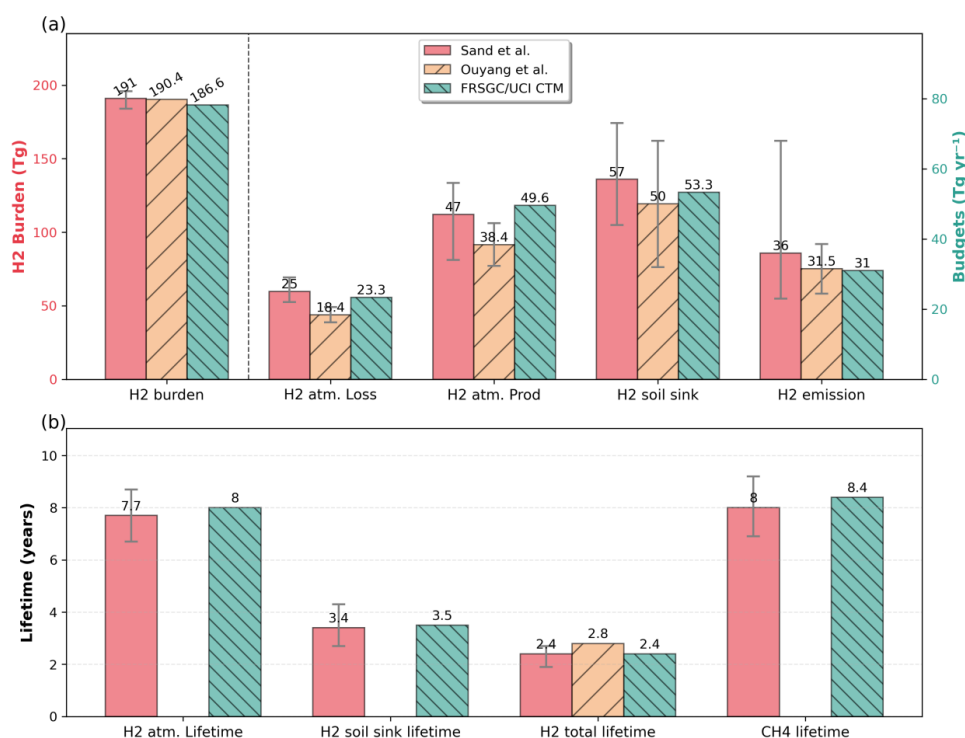
As the magnitude and spatial distribution of H₂ deposition velocity are sensitive to the deposition scheme, we compare the results for 2010 from the two schemes described in Sect. 2.2. Applying lower thresholds for soil moisture and accounting for canopy and litter resistance (scheme S2) leads to greatly overestimated annual mean H₂ deposition velocity in 2010 over Northern Africa, the Middle East, and Southern China (see Fig. S3). The annual mean H₂ soil uptake velocity in the Middle East exceeds 0.15 cm s⁻¹, with monthly mean estimates over 0.18 cm s⁻¹ in arid areas. This distribution appears less realistic, and we therefore choose the standard scheme (S1) neglecting the canopy and litter conductance for further analysis.

3.2 Evaluation of simulated H₂ budgets and concentrations

Before using the CTM to explore the changing atmospheric abundance of H₂, we evaluate the model performance in simulating current H₂ budgets and mixing ratios. Fig. 3 shows a comparison of simulated tropospheric H₂



burden, budgets, and lifetime, along with the CH₄ lifetime to OH, with multi-model mean results for 2010 from [Sand et al. \(2023\)](#), and with decadal average values from 2010 to 2020 from [Ouyang et al. \(2025\)](#). Our simulations have a longer CH₄ chemical lifetime (8.4 yrs), indicating a lower tropospheric OH burden, which in turn leads to a slightly longer H₂ atmospheric chemical lifetime of 8.0 yrs. Our estimates of H₂ atmospheric loss (23.3 Tg yr⁻¹) and atmospheric production (49.6 Tg yr⁻¹) are within the ranges reported (22–30 Tg yr⁻¹ for atmospheric loss, 34–56 Tg yr⁻¹ for atmospheric production). [Ouyang et al. \(2025\)](#) provides lower estimates of chemical production and loss, suggesting a lower OH abundance and consistent with the longer H₂ lifetime (2.8 yrs). Our soil sink (53.3 Tg yr⁻¹) and H₂ burden (186.6 Tg yr⁻¹) are very slightly lower than other studies but fall within the multi-model ranges, suggesting that our model performs well in simulating the emission-driven H₂ budget.



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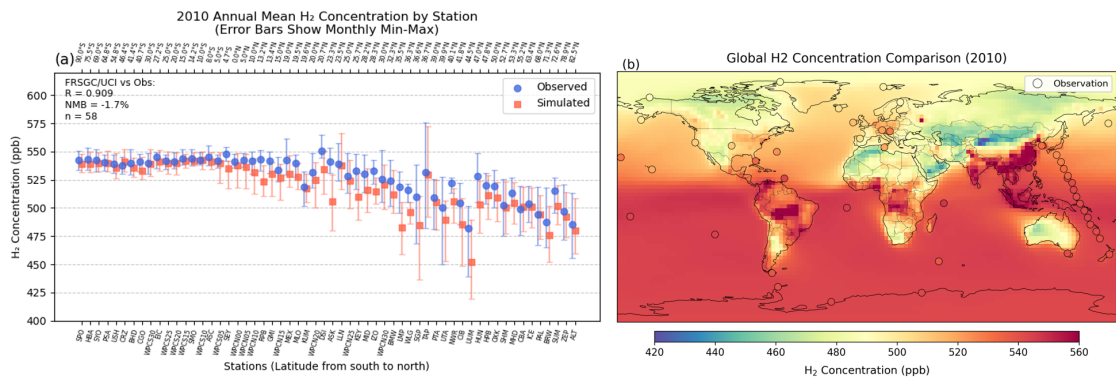
Figure 3. Comparison of simulated (a) H₂ burden (Tg) and annual flux through key production and loss processes (Tg yr⁻¹), and (b) lifetimes from the FRSGC/UCI CTM (green) with those from [Sand et al. \(2023\)](#) (red) and [Ouyang et al. \(2025\)](#) (orange). Results from [Ouyang et al. \(2025\)](#) are mean values over 2010–2020, while others are for 2010. Error bars from [Sand et al. \(2023\)](#) represent the minimum and maximum estimates across different models, and those from [Ouyang et al. \(2025\)](#) indicate one standard deviation uncertainty.

We evaluate the model performance in simulating the global distribution of H₂ using surface concentration measurements from the NOAA flask sample network ([Pétron et al., 2024](#)). Spots in Fig. A1 represent the sites we selected to compare simulations with observations in 2010, which has the greatest number of sites with valid measurements, including those over the ocean. We calculate the annual mean H₂ concentration at a site only if it has at least eight valid monthly records in the year. Fig. 4a shows the observed and simulated annual mean H₂ concentrations at each site. The model successfully captures the higher H₂ concentrations observed in the Southern Hemisphere than the Northern Hemisphere, and annual mean mixing ratios are in good agreement with



268 observations. The model underestimates mixing ratios at some northern hemisphere sites, notably those at coastal
269 sites or at high elevations where the model is unable to adequately resolve local influences on air mass origin.
270 Statistical evaluation yields a correlation coefficient of 0.91 and a normalized mean bias of -1.7% across annual
271 mean samples ($n=58$), confirming that the model can reproduce both the north–south gradient and annual mean
272 abundance, and can capture the observed monthly characteristics of H_2 across most sites.

273 Comparison of the simulated surface annual mean H_2 mixing ratios with observations at sites below 500 m altitude
274 shows that the model can reproduce the spatial distribution of H_2 well (Fig. 4b). The north–south gradient and
275 local characteristics over the continents are captured by the model, including high concentrations in Europe and
276 low concentrations in the Arctic. The seasonal cycle is also well reproduced (Fig. S4), with higher H_2 in warm
277 seasons (spring and summer). Simulated hotspots at high latitudes with extremely high H_2 concentrations in
278 summer are attributed to intense biomass burning emissions. Overall, the model successfully captures the spatial,
279 seasonal, monthly, and annual mean features of observed H_2 in 2010.



281 Figure 4. Comparison of simulated H_2 mixing ratios with observations in 2010. (a) annual mean H_2 mixing ratios
282 from NOAA observation (blue) and the model (orange) with bars showing maximum and minimum monthly H_2
283 mixing ratios over the annual cycle. The correlation coefficient and normalised mean bias are shown in the top
284 lefthand corner. (b) Comparison of the annual mean distribution of H_2 (shading) with observations (spots).

285 **3.3 Quantifying uncertainty in the soil uptake of H_2**

286 The uptake of H_2 by soil is the largest term in the atmospheric H_2 budget, and the least well characterized, and as
287 such it may account for much of the bias in the model simulation of surface H_2 mixing ratios. Much of this
288 uncertainty is associated with incomplete knowledge of the biological processes that underpin H_2 uptake, which
289 are represented here through dependence on soil moisture, porosity, temperature and other properties. To
290 characterize the uncertainty in soil uptake and quantify the contribution from different parameters, we conduct a
291 global sensitivity analysis focusing on the six most important parameters governing uptake. To make this
292 computationally efficient, we use Gaussian process (GP) emulation to emulate the uptake rates from the deposition
293 scheme and then apply a Monte Carlo approach using the emulator to explore the full uncertainty associated with
294 each parameter, following *Ryan et al. (2018)* and *Wild and Ryan (2026)*.

295 Table 3. Parameter uncertainty ranges and scaling approach.

Order	Parameter Definition	Uncertainty Range	Scaling Approach	Reference
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1	Surface pressure (PS)	$\pm 1 \%$	linear ($\pm X \%$)	(<i>Salstein et al., 2008</i>)
2	Soil moisture content (SM)	$\pm 25 \%$	linear ($\pm X \%$)	(<i>Spennemann et al., 2020</i>)
3	Soil temperature (ST)	$\pm 3 \text{ K}$	linear ($\pm X$)	(<i>Gottschalck et al., 2005; Zhao et al., 2022a</i>)
4	Soil porosity (SP)	$\pm 20 \%$	linear ($\pm X \%$)	(<i>Zhao et al., 2022b</i>)
5	Snow depth (SD)	$\pm 30 \%$	linear ($\pm X \%$)	(<i>Qiao et al., 2022</i>)
6	Water threshold for biological activity (WAT)	[-10000, -3000]	logarithmic (absolute)	(<i>Paulot et al., 2024</i>)

296 Firstly, we estimate uncertainty ranges for the key parameters controlling the deposition velocity based on
 297 previous studies, see Table 3. For soil moisture, $\pm 25 \%$ is adopted from the average non-systematic error
 298 (unbRMSE = 24.9 %) reported by *Spennemann et al. (2020)*. For soil temperature, we adopt $\pm 3.0 \text{ K}$, following
 299 *Gottschalck et al. (2005)*, who found considerable spread across GLDAS runs and *Zhao et al. (2022a)*, who
 300 reported a $-3 \text{ }^{\circ}\text{C}$ underestimation in China. Soil porosity uncertainty was set at $\pm 20 \%$, based on the study of *Zhao*
 301 *et al. (2022b)*, which showed differences of -0.2 to 0.6 between GLDAS and SMAP in normalized porosity. For
 302 snow depth, we estimated an uncertainty of 30% by combining the snow-depth ranges from *Sturm et al. (1995)*
 303 with the GLDAS standard deviations for different snow classes reported by *Qiao et al. (2022)*; these are
 304 summarized in Table S2. There is relatively low uncertainty in surface atmospheric pressure, and we take a
 305 conservative estimate of $\pm 1 \%$ based on the study of *Salstein et al. (2008)* which suggested RMS differences
 306 between analyses and observations of around 2–3 hPa.

307 To fully sample the uncertainties across these six parameters, we use a maximin Latin Hypercube design to select
 308 80 sets of parameter uncertainty values that optimally span the parameter space defined by the ranges in Table 3.
 309 Dry deposition velocities were calculated for all 80 sets of parameter uncertainties, and these were used to train a
 310 Gaussian process emulator. Deposition velocities were calculated for an additional 20 sets of parameter values to
 311 provide independent validation of the emulator. For each set, we emulate the monthly deposition velocities and
 312 then averaged them to derive the annual mean. Following *Ryan et al. (2018)*, we employ a Matérn-3/2 covariance
 313 function which can accommodate less smooth response surfaces, making it more suitable for representing physical
 314 behaviour. The skill of the emulators in representing the full model is evaluated by quantifying how well they
 315 reproduce the results of the 20 additional runs on which they were not trained. As shown in Fig. S5, the trained
 316 GP model reproduces the deposition velocity of H_2 over land well ($R^2 = 0.993$).

317 To quantify the contribution of uncertainty in each parameter to the overall uncertainty in deposition velocity, we
 318 applied a variance-based sensitivity analysis. We calculated sensitivity indices using the Sobol approach (*Sobol',*
 319 *2001*). The emulator enables efficient exploration of the parameter space, allowing Sobol analysis to be conducted
 320 using a large ensemble of evaluations that would be computationally prohibitive with the model. Analysis was
 321 conducted using a Saltelli sampling scheme (*Saltelli et al., 2010; Saltelli et al., 2004*) with a base sample size of
 322 2000. We find that soil moisture accounts for 52 % of the overall uncertainty, soil porosity for 33 %, and the water
 323 threshold for 13 %. Soil temperature plays a smaller role, contributing less than 3 % (Fig. B1). There are small
 324 interactions between these parameters, but these make only a minor contribution. To assess whether emulator
 325 uncertainty could affect the sensitivity analysis, the ratio between the mean GP predictive variance and the



propagated output variance was evaluated. The resulting ratio (0.75 %) indicates that the output variability is dominated by parameter perturbations rather than by emulator uncertainty.

3.4 Sensitivity of H₂ soil sink to soil moisture variation

Given that soil moisture contributes the most to the overall uncertainty in H₂ uptake estimation, we explore the response of calculated H₂ deposition velocity to soil moisture alone. An increase in soil moisture content tends to decrease annual mean deposition velocity over land at most locations, with a greater decrease occurring for larger increments in soil moisture (from 10 % to 60 %), see Fig. 5. In contrast, decreasing soil moisture leads to increases in annual mean deposition velocity in most regions. However, beyond a drying of 30%, deposition velocities decrease again as the soil becomes extremely dry. This can be explained from the behaviour of the soil moisture dependence (f) (Fig. S3b). At locations with soil conditions wetter than the optimal value for the specific soil type, f decreases with rising soil moisture and increases with declining soil moisture. If the soil moisture falls below the optimal condition, f starts to decrease with decreasing soil moisture. For locations with soil moisture below the optimal value, the deposition velocity will increase as the soil gets wetter until the optimum moisture condition is exceeded.

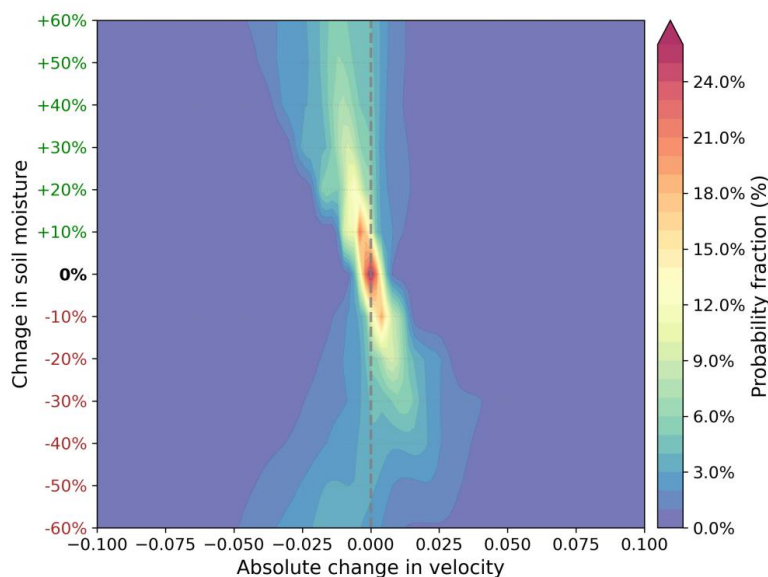


Figure 5. The probability distribution of the absolute change in dry deposition velocities (100 bins) in response to a perturbation in soil moisture content (–60 % to +60 %).

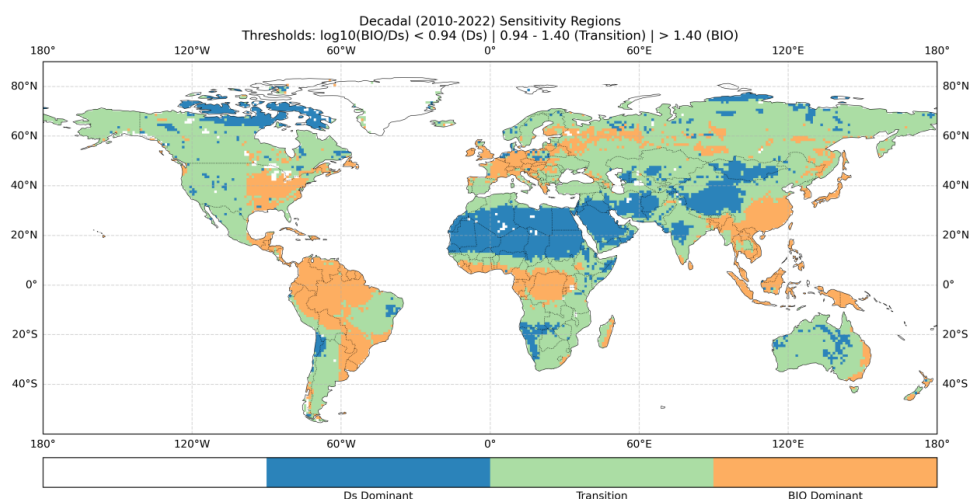
We performed a similar analysis to quantify the sensitivity of deposition velocities to soil temperature alone. We find that the response to soil temperature is much less than that to soil moisture, and that the responses are close to linear. Increasing soil temperature raises deposition velocities while decreasing soil temperature lowers them.

Regional responses of H₂ deposition velocity to soil moisture perturbations in the 2010 simulations are distinct and classifiable. Changes in H₂ dry deposition velocity exhibit opposite responses in North Africa and the Middle East to those in China, Europe and Latin America, see Fig. B2. This indicates the presence of different regimes where soil moisture affects H₂ soil uptake differently. Therefore, we explore the long-term sensitivity of H₂



350 deposition velocity to soil moisture changes during 2010–2022 by quantifying the relative contributions of
 351 diffusivity (D_s ; D_s in Sect. 2.2) and biological processes (BIO: $h(T)f$ in section 2.2). The anomalies in annual
 352 mean velocity, BIO, and D_s relative to the 2010–2022 mean are shown in a quadrant plot (Fig. B3). Higher
 353 deposition velocities occur with higher D_s and BIO (purple regions in upper righthand quadrant), while lower
 354 velocities occur with lower D_s and BIO (green regions in lower lefthand quadrant). This suggests that changes in
 355 deposition velocity are driven by both D_s and BIO. BIO-driven sensitivity is indicated by purple regions in the
 356 upper lefthand quadrant, and D_s -driven sensitivity by green regions in the lower righthand quadrant. These
 357 patterns allow a classification into three regimes: BIO-dominant, D_s -dominant, and Transition zones, which
 358 correspond to sensitivity governed by BIO, D_s , or both.

359 Based on the three target clusters from the above analysis, an Expectation-Maximization method, Gaussian
 360 Mixture Model, was used to classify the deposition responses using gridded long-term mean BIO and D_s values
 361 as input. The Gaussian Mixture Model represents the joint distribution of BIO and D_s as a mixture of three
 362 Gaussian components and estimates the probability that each data point belongs to each component, and it has
 363 been applied to atmospheric studies to classify ozone profiles (Fahrin et al., 2023), aerosol types (Song et al.,
 364 2023), as well as VOC sample clusters (Richards et al., 2020). Fig. 6 shows the distribution of the three sensitivity
 365 regimes characterized by thresholds of $\log(\text{BIO}/D_s) > 1.4$ (BIO dominant), $\log(\text{BIO}/D_s) < 0.94$ (D_s dominant),
 366 and the transition regime in between. The spatial response matches the relationship between the deposition
 367 velocity and soil moisture shown in Fig. B2, with increased velocity in BIO-dominant and Transition regions
 368 under drier soil conditions and in D_s -dominant regions under wetter soil conditions. The seasonal classification of
 369 these regimes is presented in Fig. S6 for January and July, and demonstrate that in winter conditions a greater
 370 number of locations are controlled by BIO processes, whereas D_s control regions expand in summer. These results
 371 align well with those of Bertagni et al. (2021) who presented a similar classification using different criteria. We
 372 additionally tested the sensitivity of our classification to the number of clusters selected. Using four clusters
 373 showed little obvious difference, and five clusters did not provide additional meaningful information. This
 374 suggests that three clusters is sufficient to characterise the sensitivity regimes well.



375

376 Figure 6. Identification of the sensitivity zones of H_2 deposition velocity in response to soil moisture based on H_2
 377 soil sink processes during 2010–2022. The diffusion (D_s) dominated, Transition, and biotic (BIO) dominated
 378 regimes are indicated by blue, green, and orange, respectively, and the dynamic thresholds used to classify the
 379 regions are labelled above the figure.



Given the sensitivity of deposition velocity to soil moisture identified here, we examine the implications for the variations in velocity over the past decade. Fig. 7 shows the calculated annual mean dry deposition velocity over land from 2010 to 2022, along with the Niño 3.4 index that provides a measure of global climate variations. There are two pronounced peaks in deposition velocity in 2015 and 2022, and these are likely associated with strong ENSO events. Anomalies in annual mean deposition velocity and soil moisture (Fig. S7) relative to the long-term mean reveal the differing drivers for the two peaks. The 2015 peak can be attributed to drier conditions in most BIO-dominant and Transition regions, driven by El Niño, with additional contributions from increased soil moisture in northern Africa, which is Ds-dominant. In contrast, the 2022 peak reflects a combination of wetter soils in Ds-dominant regions (western China, Australia, the Middle East) and drier conditions in BIO-dominant and Transition regions (South America, eastern US, North America, Europe). The stronger signal during the 2020–2023 La Niña event compared to the 2010–2011 event is due to its longer duration, as it developed into a “triple-dip” La Niña, which cumulatively altered precipitation-induced soil moisture (Tian et al., 2026; Zhu et al., 2025). We show that short-term variations in H₂ soil uptake are closely linked to strong ENSO events, which redistribute global heat and precipitation. The responses are not related in a simple linear manner due to the spatially heterogeneous climate-induced responses of soil moisture.

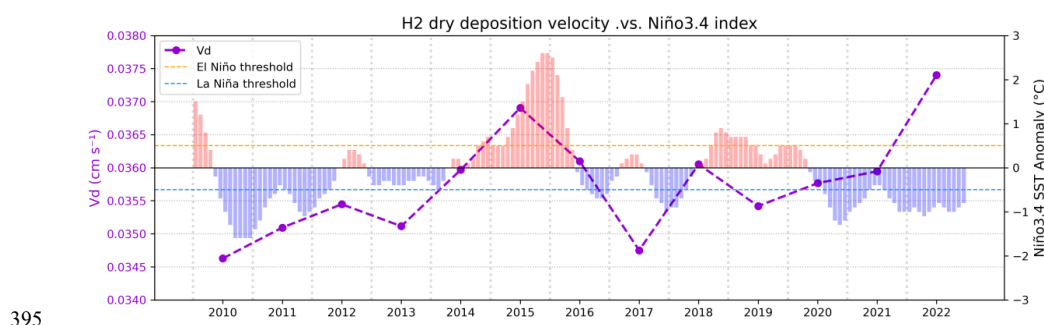


Figure 7. Time series of the annual mean H₂ dry deposition velocity over land (purple line, cm s⁻¹) and the Niño 3.4 index (bars) from 2010 to 2022. Orange and blue dashed lines represent the threshold for El Niño (+0.5 °C) and (-0.5 °C), respectively.

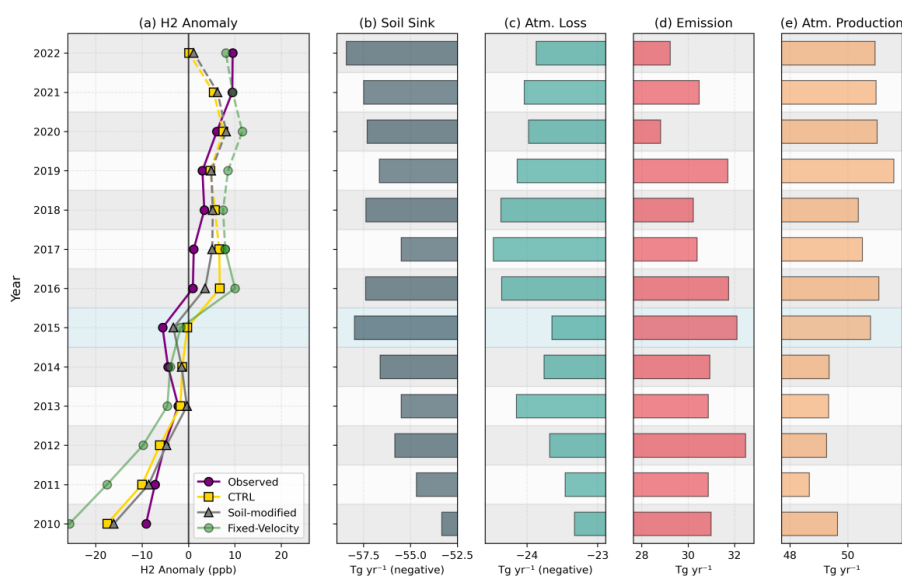
3.5 The contribution of H₂ dry deposition to interannual H₂ variability

Over the past decade, observed surface H₂ mixing ratios have shown an increasing trend, but with a notable drop during 2014–2015 (Fig. A1). Given that soil uptake is the largest term in the atmospheric H₂ budget, and that it is strongly dependent on soil moisture, we explore the extent to which interannual variability in soil moisture may contribute to variations in surface H₂ abundance. The anomalies in H₂ concentration at background sites relative to the 2010–2022 mean from observation and control simulation are shown in Fig. 8. We do not have self-consistent meteorological data after 2017, and we therefore simulate atmospheric transport for the 2018–2022 period using repeated meteorology from 2017, but with surface emissions and H₂ deposition velocity specific to the 2018–2022 period. The comparison suggests that the model can capture the increasing trend in H₂ abundance from 2010 to 2020 quite well, but it fails to reproduce the decline in H₂ abundance during 2014–2015.

As we have shown, the strong El Niño event during 2014–2015 had a substantial impact on soil uptake due to the soil moisture response to the climate signal. However, we assume H₂ soil emission is fixed from 2010 to 2022. H₂ emission from soil is a by-product of the nitrogenase reaction in the process of nitrogen fixation (Flynn et al., 2014). The influence of hydrology on the nitrogenase reaction has been widely explored in previous studies. For example, Kohl et al. (1991) found that nitrogenase activity was inhibited 90 % or more as a result of drought stress.



414 [Aranjuelo et al. \(2011\)](#) showed a decrease in nitrogen fixation in droughted nodules, which was attributed to
 415 increased oxygen resistance. Therefore, we hypothesize that strong climate events also affect the soil emissions
 416 of H_2 . To test this, we assume that drier soil conditions during 2014–2015 associated with the strong El Niño
 417 caused a decrease in H_2 soil emissions of 1.4–1.8 Tg and perform an additional model simulation (labelled “Soil-
 418 modified”) to compare with our control simulation with fixed natural H_2 emissions (10.5 Tg every year). As shown
 419 in Fig. 8a, the simulation with modified soil emission captures the drop in H_2 concentration well over 2014–2015,
 420 suggesting that it would be valuable to develop an interactive soil source of H_2 which is sensitive to climate-
 421 induced soil moisture changes.



422

423 Figure 8. Anomalies of (a) annual mean H_2 mixing ratio averaged across background sites from observations
 424 (purple), CTRL (yellow), Soil-modified (grey), and Fixed-velocity (green) simulations, and annual mean H_2
 425 budget terms from the Soil-modified simulation for (b) soil sink, (c) atmospheric loss, (d) emission, and (e)
 426 atmospheric production from 2010 to 2022. We use dashed lines to show simulated H_2 anomalies after 2017 with
 427 fixed meteorology.

428 To explore the role of the soil sink in governing the interannual variability in H_2 , we further fixed the H_2 deposition
 429 velocity to the level of 2010, retaining the modified soil emissions during 2014–2015. This “Fixed-velocity”
 430 simulation overestimates the increase in H_2 abundance over the period and fails to reproduce the observed decline
 431 in H_2 mixing ratios during 2014–2015. This suggests that the soil sink is an important process controlling
 432 interannual variation in H_2 level over the past decade. It is clear from the tropospheric H_2 budget terms shown in
 433 Fig. 8 that the enhanced soil sink drives the drop in H_2 mixing ratio in 2015, even though emissions are high and
 434 the photochemical loss is low in this year. Similarly, the decrease in soil uptake in 2017 reduces the decline in H_2
 435 abundance from 2016 to 2017 caused by the lower H_2 source and increased chemical loss.

436 The upward trend in atmospheric H_2 production, which aligns with the long-term rise in H_2 abundance, suggests
 437 that increased production is the primary factor driving the trend in abundance, which outpaces the decrease in
 438 direct emissions. This agrees with the conclusions of recent studies ([Ouyang et al., 2025](#); [Stroo et al., 2026](#)).
 439 Without the corresponding meteorology, we overestimate the decline from 2020 to 2022, suggesting that there



may be important meteorological impacts on chemical processes affecting H_2 during this period, especially during the long La Niña event. However, we note that there was a large decline in the observed monthly H_2 in the second half of 2022 that is captured well by the model (Fig. S8), highlighting again the important role played by the higher soil sink.

3.6 Future projection of the deposition velocity of H_2

As H_2 dry deposition is strongly influenced by soil parameters, particularly soil moisture and temperature, it is valuable to explore how soil uptake may change over the coming century under climate change. We show that future trends in soil moisture exhibit large spatial heterogeneity, whereas soil temperature and snow depth increase under both SSP1-2.6 and SSP5-8.5 pathways. Projected changes under SSP1-2.6 show similar spatial patterns to those under SSP5-8.5, but with generally smaller magnitudes. Under SSP5-8.5, the median trends range from -0.80 to $0.47 \text{ kg m}^{-2} \text{ decade}^{-1}$ for soil moisture, 0.0 to $1.68 \text{ K decade}^{-1}$ for soil temperature, and -0.47 to $0.01 \text{ m decade}^{-1}$ for snow depth. Future snow depth does not exhibit uniform changes across all grid cells, and regional increases in snow depth are closely linked to precipitation changes (Fontrodona Bach et al., 2018). Regions with deeper snow show strong spatial consistency with areas characterized by increasing soil moisture in the future (Figs. 9a and 9c). Only 4.8 % of the land area showed robust agreement among models in soil moisture trends under the SSP1-2.6 pathway (Fig. 9a), with 36.6% agreement on SSP5-8.5 (Fig. 9d). Notably, soil conditions under the SSP5-8.5 pathway are wetter in western China, East Middle, India, central Africa, southeastern South America, while most other regions worldwide show reduced soil moisture (Fig. 9d). From the sensitivity analysis of H_2 soil sink to soil moisture change in Sect. 3.4, future increases in H_2 deposition velocity are expected as a result of decreased soil moisture in BIO-dominant or Transition regions and increases in Ds-dominant regimes, especially under the SSP5-8.5 pathway.

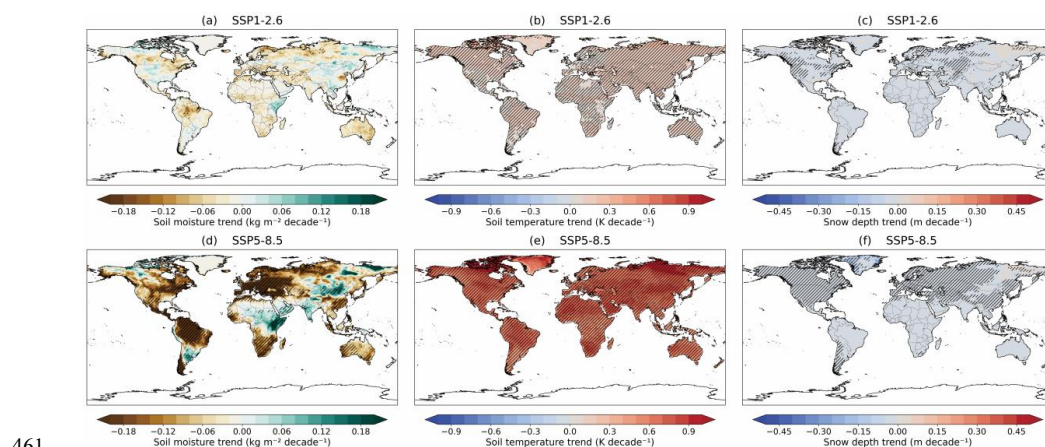
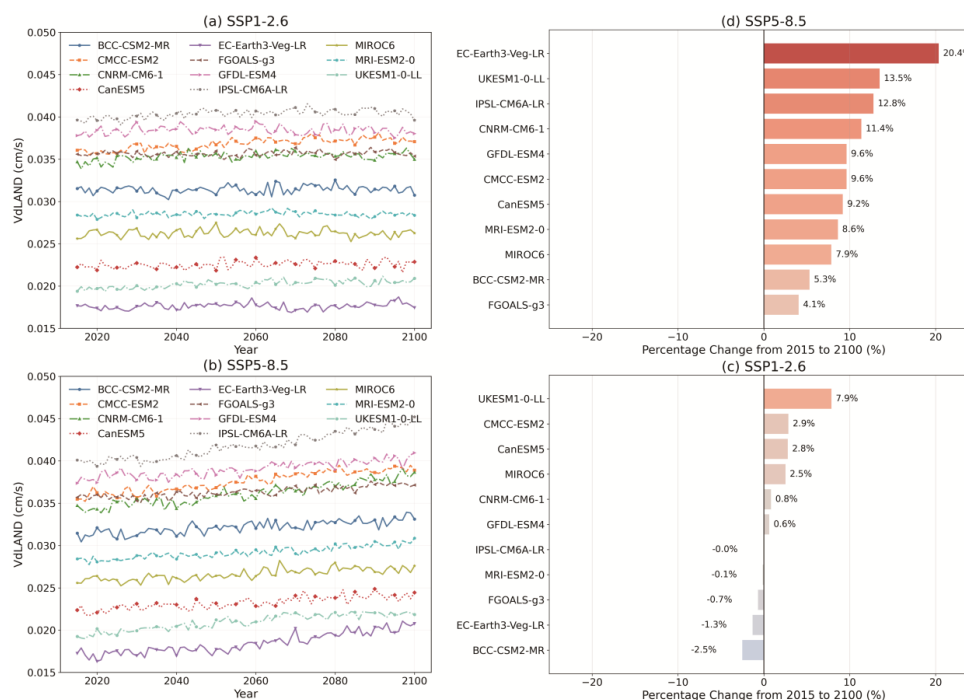


Figure 9. Median trend of upper 10 cm soil moisture ($\text{kg m}^{-2} \text{ decade}^{-1}$), upper 10 cm soil temperature (K decade^{-1}), and snow depth (m decade^{-1}) under (a)–(c) SSP1-2.6 and (d)–(f) SSP5-8.5. The hatching on the plots indicates that nine models (for soil moisture and soil temperature), and six models (for snow depth) agree on the sign.

Fig. 10 shows the projected interannual variation in annual mean deposition velocity to land from 2015 to 2100 for each of the selected CMIP models under SSP1-2.6 and SSP5-8.5 pathways along with the percentage change between 2015 and 2100. Deposition velocities in 2015 were calculated in the range 0.018 – 0.040 cm s^{-1} , with more than a factor of two difference across the models driven by distinctly different levels of soil moisture.



Deposition velocities are projected to show relatively little change (-2.5%–7.9%) from 2015 to 2100 among models under the SSP1-2.6 pathway, although one model shows a larger increase of 7.9 %. Deposition velocities show stronger increases (4.1–20.4%) under the SSP5-8.5 pathway. The multi-model mean deposition velocity is projected to increase 0.8 % under the SSP1-2.6 pathway and 9.7 % under SSP5-8.5 between 2015 and 2100. Our projections suggest a smaller increase in H_2 deposition velocity compared to the estimates of +4.8 % for SSP1-2.6 and +16.7 % for SSP5-8.5 from [Brown et al. \(2025\)](#), which reflects the broader selection of models we have considered and our use of soil porosity from the models, which is higher than in the GLDAS data.



476

477 Figure 10. (a)–(b) Variations in H_2 dry deposition velocity over land from 2015 to 2100 calculated for CMIP6
 478 models, and (c)–(d) percentage change (%) in deposition velocity between 2015 and 2100 under SSP1-2.6 and
 479 SSP5-8.5 pathways.

480 4. Conclusions

481 In this study, we have implemented a soil dry deposition scheme for H_2 in the FRSGC/UCI global chemistry
 482 transport model and have performed emission-driven simulations to investigate interannual variations and trends
 483 in H_2 from 2010 to 2022. The model reproduces the distribution and seasonality of observed surface H_2 mixing
 484 ratios and the underlying increases over the period. We find an atmospheric H_2 burden of 186.6 Tg in 2010 and
 485 an atmospheric lifetime of 2.4 years. Sources of H_2 from emissions (31.0 Tg yr⁻¹) and chemical production (49.6
 486 Tg yr⁻¹) roughly balance losses from chemical destruction (23.3 Tg yr⁻¹) and soil uptake (53.3 Tg yr⁻¹) and these
 487 budget terms align well with those from other recent model studies. Focusing on the largest and most uncertain
 488 term in this budget, soil uptake of H_2 at the Earth's surface, we have conducted a sensitivity analysis to quantify
 489 how uncertainties in soil parameters contribute to uncertainty in H_2 uptake. We find that soil moisture dominates
 490 the uncertainty (52 %), along with contributions from soil porosity (34 %) and the water threshold for biological



activity (13 %). This strong sensitivity of uptake to soil moisture provides a key driver for interannual variations in atmospheric H_2 abundance and highlights the susceptibility of this important term in the H_2 budget to future changes in climate.

To explore the response of soil uptake to soil moisture we examine how uptake varies with global uniform changes in soil moisture. Increased soil moisture leads to lower uptake at most locations, with greater uptake at a limited number of places. Decreased soil moisture leads to greater uptake at most locations, although decreases beyond 30% cause lower uptake at an increasing number of locations, reflecting the non-linear relationships involved. To further characterize these responses in soil uptake we perform a formal classification of regional responses into biotic and diffusivity-dominated regimes, extending the approach taken by [Paulot et al. \(2021\)](#). In the diffusivity-dominated regime, corresponding to dry soil conditions, uptake typically increases with soil moisture, and decreases with soil drying. In biotic-dominated regimes, representing conditions favourable for biological activity, uptake shows the opposite response, decreasing with soil moisture and increasing with drying. This analysis allows us to explain the complex response of soil uptake to soil moisture variations associated with ENSO events, highlighting different responses from different events associated with the spatially heterogeneous patterns of soil moisture change.

We investigate the changes in atmospheric H_2 abundance from 2010 to 2022 and find that much of the interannual variability can be explained by changes in soil uptake, particularly the drop in H_2 abundance in 2015, which cannot be reproduced with an annually invariant uptake rate. However, we are unable to fully reproduce this drop and suggest that incorporating climate-driven changes in the natural soil source of H_2 from nitrogen fixation may also be important. While soil uptake is an important driver of variability, we note that the underlying long-term increase in H_2 mixing ratios over the past decade is largely due to rising atmospheric production, as previous studies have suggested. Exploration of future changes in soil uptake using climate data from CMIP6 models over the period 2015–2100 suggests that changes in soil moisture will lead to increased uptake in future. These changes are small (averaging 0.8 %) on the SSP1-2.6 pathway but more substantial (9.7 %) on the SSP5-8.5 pathway. While this increased uptake may be sufficient to halt the increases in H_2 abundance seen over the past decade, it is unlikely to offset the increased H_2 emissions expected on many future development pathways, and it is thus essential to minimize the increased emissions expected from leakages when developing a future H_2 economy.

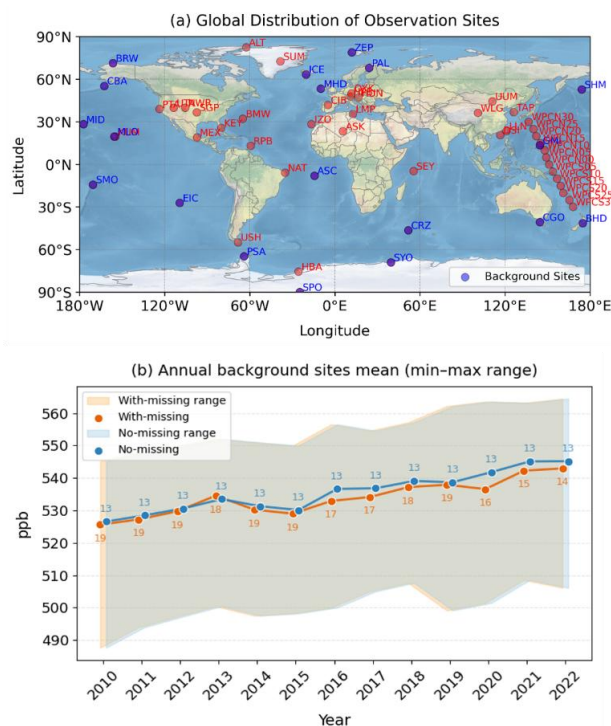
We have demonstrated the major contribution of the soil sink to the interannual variability in H_2 abundance, highlighting the importance of soil moisture in governing uptake and showing the impacts on recent and future H_2 abundance. However, improved understanding of microbial activity in different soil types and the role of soil moisture in governing this is needed to refine estimates of this important term in the atmospheric H_2 budget. Recent laboratory studies have identified surface litter as a strong sink of H_2 , rather than simply a barrier to soil uptake as is commonly assumed ([Drewer et al., 2026](#)). There is also a lack of long-term measurements of soil H_2 uptake across a diversity of biomes, and these are needed to provide a more critical test of uptake schemes. Further refinement of the atmospheric H_2 budget also requires improved estimates of the photochemical production and destruction of H_2 . These terms may be overestimated in current models which typically overestimate OH, reflected in a tropospheric chemical lifetime of CH_4 of 8–9 years compared with an observationally based lifetime of 11.2 ± 1.3 years ([Prather et al., 2012](#)). Addressing this bias will increase the importance of soil uptake for atmospheric H_2 removal and emphasises the need for improved understanding.

530



531 **Appendix A: Observation information**

532 For each background site show in Fig. A1a, we calculated the annual mean H₂ mixing ratio only if valid
533 measurements were available for at least eight months in that year. These site-specific annual means were averaged
534 to obtain the multi-site annual mean for each year, and the results are shown in Fig. A1b (orange symbols), labelled
535 with the number of sites that were included. There is a peak in 2013 (534.6 ppb), and drops in 2015 (529.0 ppb)
536 and 2020 (536.6 ppb) during the long-term rise from 2010 to 2022. A consistent set of annual means from 13
537 background sites, excluding 6 sites with one or more years of missing data, are also shown in Fig. A1b (blue
538 symbols). This smaller set shows a similar pattern, although there is a decline from 2018 to 2019 and the dip in
539 2020 disappears.



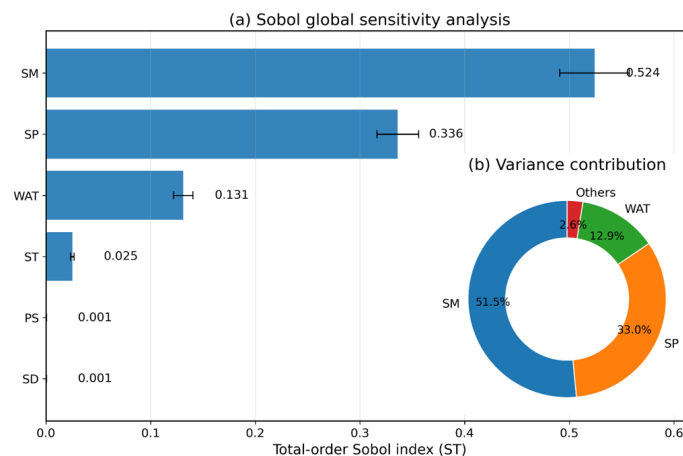
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541 Figure A1. (a) Locations of observation sites providing H₂ measurements selected to calculate and compare with
542 model results; blue symbols represent background sites. (b) Time series of observed annual mean H₂ mixing ratios
543 at background sites. Orange dots: annual means with varying site counts (allowing missing data). Blue dots: annual
544 means from consistent sites (no missing data). The background site selection follows [Paulot et al. \(2024\)](#).

545



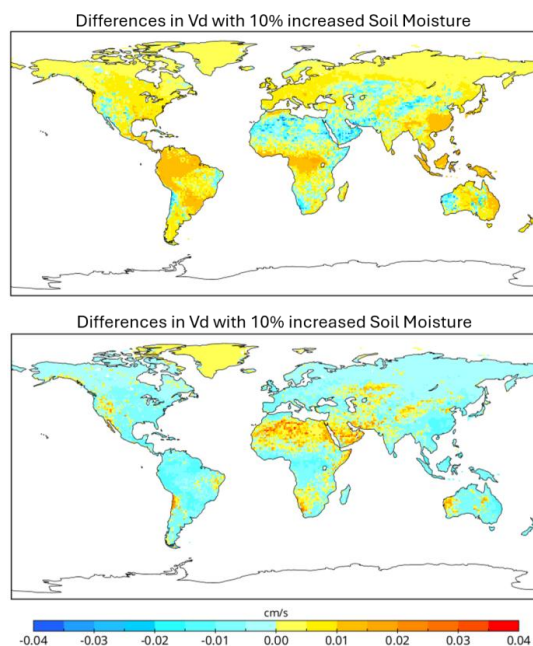
Appendix B: Results of sensitivity analysis



547

548 Figure B1. (a) Total-order Sobol indices showing the overall contribution of each parameter to the propagated
 549 output variance, including both direct effects and parameter interactions. Error bars indicate the statistical
 550 uncertainty associated with the estimated Sobol indices. (b) Percentage contributions normalized from the first-
 551 order Sobol indices, representing the direct contribution of each parameter to the total output variance.

552



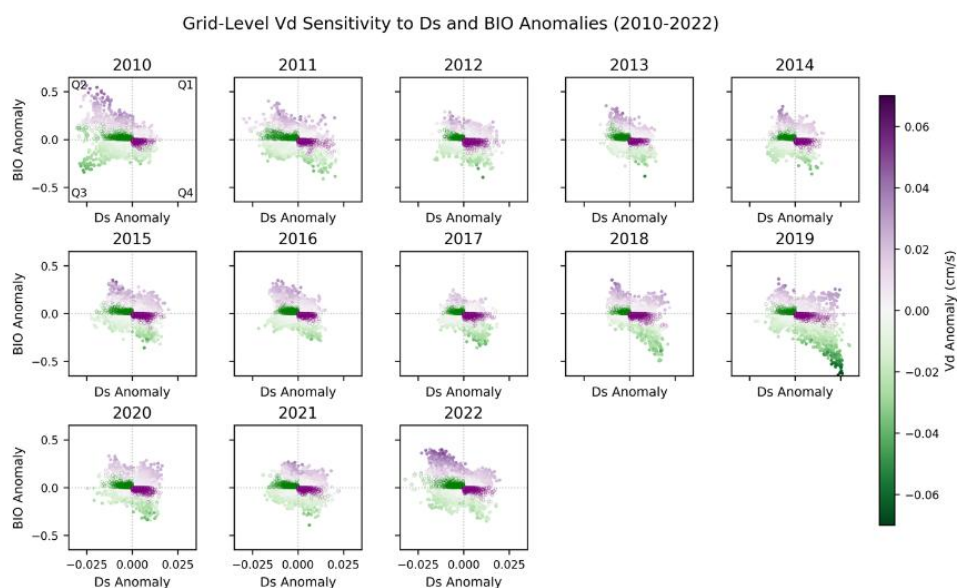
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556

Figure B2. Changes in annual mean H_2 dry deposition velocity (cm s^{-1}) in response to $\pm 10\%$ soil moisture perturbation.



557

558 Figure B3. H₂ deposition velocity anomalies in each model surface grid box (purple: increased Vd; green:
 559 decreased Vd) associated with biotic (BIO) and diffusivity (Ds) anomalies each year relative to the long-term
 560 mean from 2010 to 2022.

561



562 **Code and data availability**

563 Monthly mean results from control simulation and optimal hydrogen deposition velocity maps can be available
564 soon on Lancaster repository. NOAA Global Monitoring Laboratory H₂ observations can be obtained from
565 <https://gml.noaa.gov/ccgg/data/getdata.php?gas=h2>, and CAMS anthropogenic, biogenic, and oceanic emissions
566 can be downloaded from [https://ads.atmosphere.copernicus.eu/datasets/cams-global-emission-](https://ads.atmosphere.copernicus.eu/datasets/cams-global-emission-inventories?tab=download)
567 [inventories?tab=download](https://ads.atmosphere.copernicus.eu/datasets/cams-global-emission-inventories?tab=download) or <https://eccad.sedoo.fr/#/data>. GFAS Biomass burning emissions are available from
568 <https://ads.atmosphere.copernicus.eu/datasets/cams-global-fire-emissions-gfas?tab=download>. Historical soil
569 parameters from GLDAS are from <https://ldas.gsfc.nasa.gov/gldas/soils>. CMIP6 model results of future soil
570 parameters were from CEDA archive (<https://data.ceda.ac.uk/>).

571 **Author contributions**

572 YW and OW designed and conducted this study, and OW provided supervision. YW performed simulations,
573 analysis, and paper writing. OW, XW and RH reviewed and revised the manuscript, and contributed to model
574 development.

575 **Competing interests**

576 The authors declare that they have no conflict of interest.

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581 inventories, NASA Goddard Earth Sciences Data and Information Services Center (GES DISC) for providing
582 GLDAS soil data, and the CMIP6 project for providing future projected data of soil parameters.

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586 development and policies implementation to mitigate possible climate impacts), Horizon Europe project number
587 101137758.

588



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 591 response to drought in alfalfa (*Medicago sativa* L.), *Journal of Experimental Botany*, 62, 111-123,
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