

1 **UK public health and ecosystem benefits of currently legislated versus best available**
2 **emission control measures**

3 **Eloise A. Marais¹, Jamie M. Kelly^{1,*}, Karn Vohra¹, Yifan Li², Gongda Lu¹, Naila Hina³, Ed**
4 **C. Rowe³**

5 ¹Department of Geography, University College London, London, UK.

6 ²Reading Academy, Nanjing University of Information Science and Technology, Nanjing, China.

7 ³UK Centre for Ecology & Hydrology, Environment Centre Wales, Bangor, UK.

8 *now at: Centre for Research and Clean Air, Helsinki, Finland.

9
10 Corresponding author: Eloise A. Marais (e.marais@ucl.ac.uk)

11
12 **Key Points:**

- 13 • Efficacy of UK legal and technological control measures tested with emissions projections,
14 GEOS-Chem, and new exposure-harm relationships
- 15 • Both reduce public exposure to particulate matter pollution and avoid 6,800 adult early
16 deaths with legislation and 13,300 with technology
- 17 • Neither is sufficient at lessening sensitive habitat exposure to harmful loads of nitrogen and
18 concentrations of phytotoxic ammonia

19 **Abstract**

20 Past emission controls in the UK have substantially reduced precursor emissions of health-
21 hazardous fine particles (PM_{2.5}) and nitrogen pollution detrimental to ecosystems. Still, 79% of
22 the UK exceeds the World Health Organization (WHO) guideline for annual mean PM_{2.5} of 5 µg
23 m⁻³ and there is no enforcement of controls on agricultural sources of ammonia (NH₃). NH₃ is a
24 phytotoxin and an increasingly large contributor to PM_{2.5} and nitrogen deposited to sensitive
25 habitats. Here we use emissions projections, the GEOS-Chem model, high-resolution datasets, and
26 contemporary relationships between exposure and risk of harm to assess the potential human and
27 ecosystem health co-benefits in 2030 relative to the present day of adopting legally required or
28 best available emission control measures. We estimate that present-day annual adult premature
29 mortality attributable to exposure to PM_{2.5} is 48,625, that harmful amounts of reactive nitrogen
30 deposit to almost all (95%) sensitive habitat areas, and that 75% of ambient NH₃ exceeds levels
31 safe for bryophytes. Legal measures decrease the extent of the UK above the WHO guideline to
32 58% and avoid 6,800 premature deaths by 2030. This improves with best available measures to
33 36% of the UK and 13,300 avoided deaths. Both legal and best available measures are insufficient
34 at reducing the extent of damage of nitrogen pollution to sensitive habitats, as most nitrogen
35 emitted in the UK is exported offshore. Far more ambitious reductions in nitrogen emissions
36 (>80%) than is achievable with best available measures (34%) are required to halve excess
37 nitrogen deposition to sensitive habitats.

38

39 **Plain Language Summary**

40 Particulate matter pollution is detrimental to human health, nitrogen pollution offsets ecosystem
41 balance of sensitive habitats, and ammonia is toxic to plants. Here we determine the potential
42 public health and ecosystem benefits of adopting currently legislated or best available emission
43 control measures. We use state-of-science air quality and exposure assessment models and high-
44 resolution datasets to estimate air pollution abundances and deposition and determine risk of
45 human and ecosystem harm from air pollution in the UK. We find that substantial improvements
46 to public health result from currently legislated measures, estimated as the number of avoided early
47 deaths attributable to decline in particulate matter pollution. The number of avoided deaths doubles
48 with best available emission control measures, as these also target particulate matter precursor
49 emissions of ammonia from agricultural activity not mandated by existing legislation. The benefits
50 to sensitive habitats of either legal or best available measures is near-negligible. Mitigating harm
51 to ecosystems will require major advances in emission mitigation technologies.

52 **1 Introduction**

53 Air pollution is a leading global burden of disease (Cohen et al., 2017; Vohra et al., 2021)
54 and is detrimental to ecosystem balance (Sala et al., 2000; Phoenix et al., 2012). In the UK, about
55 29,000 adult premature deaths have been attributed to long-term exposure to ambient fine
56 particulate matter pollution (PM_{2.5}) in a single year (Gowers et al., 2014) and most (70%) sensitive
57 habitats with low nutrient needs are susceptible to harm from deposition of excessive amounts of
58 nitrogen (Rowe et al., 2022). Anthropogenic emissions of ammonia (NH₃), overwhelmingly
59 (~90%) from agriculture, are a large and often dominant component of PM_{2.5} in densely populated
60 cities (Kelly et al., 2023; Vieno et al., 2016) and of nitrogen deposited to ecosystems (Tomlinson
61 et al., 2021). NH₃ is also a phytotoxin (Dise et al., 2011; van Herk et al., 2007; Krupa, 2003).

62 Measures to reduce NH₃ emissions from agricultural activity in the UK are limited to
63 voluntary implementation of documented best practices (DEFRA, 2018). Over the past 3 decades
64 (1990-2020) anthropogenic NH₃ emissions have declined by just 0.6% a⁻¹; much slower than
65 decline in other PM_{2.5} precursors such as sulfur dioxide (SO₂) at 3.0% a⁻¹ and nitrogen oxides
66 (NO_x) at 2.4% a⁻¹ (Churchill et al., 2022). NH₃ is a semi-volatile, alkaline gas that partitions
67 reversibly to acidic aerosols forming ammonium (NH₄). Due to sustained controls on local and
68 national SO₂ and NO_x sources, concentrations of acidic sulfate aerosols formed from SO₂ oxidation
69 and acidic nitrate aerosols formed from NO_x oxidation have declined, contributing to decline in
70 NH₄ (Tang et al., 2018). Weakening of the NH₃ aerosol sink and absent controls on agriculture
71 have caused an increase in ambient NH₃ concentrations and the amount of NH₃ deposited to
72 terrestrial ecosystems via dry and wet deposition (Hertel et al., 2012). This has been detrimental
73 to habitats sensitive to excessive nitrogen loads and exposure to NH₃. The latter includes
74 bryophytes, a prevalent class of plants in the UK adversely affected by NH₃ at relatively low
75 concentrations (~1 µg m⁻³) (Cape et al., 2009).

76 The UK commits to sustained decline in air pollutant precursor emissions as a signatory of
77 the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range
78 Transboundary Air Pollution (CLRTAP) legislated through the UK National Emission Ceilings
79 Regulations (UK, 2018). The UK met the 2020 targets, according to bottom-up estimates from the
80 National Atmospheric Emission Inventory (NAEI) (Richmond et al., 2020), though top-down NH₃
81 emissions estimated with satellite observations suggest the NAEI may underestimate NH₃
82 emissions by 30-50% (Marais et al., 2021). NH₃ emissions targets, reported as percent reduction
83 relative to 2005, are 8% in any year in 2020 to 2029 and 16% from 2030; modest in comparison
84 to SO₂ (59% in 2020-2029 and 79% from 2030) and NO_x (42% in 2020-2029 and 63% from 2030)
85 (Office of the European Union, 2016a). Readily available emission control measures targeting the
86 largest agricultural sources of NH₃, fertiliser use and livestock and manure management (Churchill
87 et al., 2022), include low nitrogen livestock feed, covered manure storage, low emissions methods
88 of spreading manure, filters and scrubbers of air ventilated from animal housing, and low NH₃
89 alternatives to urea-based fertilisers (Backes et al., 2016). Efficacy of these range from 10-20%
90 decrease in emissions for low nitrogen feed, to 80% decrease for air filtration and covered manure
91 storage, to up to 93% for phasing out urea-based fertilizers (Klimont & Brink, 2004).

92 Past studies that have assessed separately the public health and ecosystem benefits of
93 emissions controls suggest current measures offer greater benefits to public health than the expense
94 to implement (Giannakis et al., 2019), and that mitigating harm to sensitive ecosystems would
95 require large (at least 40%) reductions in NH₃ emissions (Woodward et al., 2022). Here we
96 investigate whether adoption of measures to meet current legislation or of readily available
97 maximum technically feasible technologies across all sectors, including agriculture, are sufficient
98 to offer combined benefits to public health and sensitive habitats in the UK. We do this by
99 implementing emissions scenarios from the EU's Evaluating the Climate and Air Quality Impacts
100 of Short-Lived Pollutants (ECLIPSE) project (Stohl et al., 2015) in the GEOS-Chem model to
101 simulate the response of ambient PM_{2.5} and NH₃ and nitrogen deposition to emissions changes.
102 We apply these to high spatial resolution datasets of contemporary nitrogen deposition to sensitive
103 habitats and models of the relationship between exposure and risk of harm to calculate adult
104 premature mortality in each UK administrative area and the scale of harm to sensitive habitats.

105 2 Materials and Methods

106 2.1 GEOS-Chem Model Description

107 We use the GEOS-Chem chemical transport model (CTM) version 13.0.0
108 (<https://doi.org/10.5281/zenodo.4618180>; accessed 12 December 2021) to simulate present-day
109 and future UK surface concentrations of PM_{2.5} and NH₃ and total (wet and dry) nitrogen deposition
110 due to emissions controls adopted across Europe. The future scenarios are representative of
111 emissions likely in 2030 from adopting either (1) currently legislated emission controls or (2)
112 maximum technically feasible measures. In what follows, the scenarios and associated model
113 results are labelled “PD” for the present-day simulation in 2019, “CLE” for adopting measures to
114 meet current legislation by 2030 and “MTF”, also for 2030, but following adoption of maximum
115 technically feasible measures. The model is nested over the UK (49.25°N to 59.50°N, 9.375°W to
116 3.75°E) at 0.25° × 0.3125° (28 km latitude × 20 km longitude at the centre of the domain) and is
117 updated 3-hourly with boundary conditions from a global GEOS-Chem simulation at 4° × 5°. The
118 model uses GEOS-FP assimilated meteorology from the NASA Global Modelling and
119 Assimilation Office. All simulations are driven with meteorology for the same year (2019) to
120 isolate the effect of changes in anthropogenic emissions only. The year 2019 instead of 2020 is
121 chosen for the PD simulation, as it is unaffected by the brief, but dramatic decline in emissions
122 resulting from lockdown measures in the first wave of the COVID-19 pandemic in Spring 2020
123 (Potts et al., 2021).

124 Anthropogenic emissions in the PD simulation are mostly consistent with those detailed in
125 Kelly et al. (2023) from an earlier model version (v12.1.0) for the same year (2019) used to
126 characterise sources contributing to PM_{2.5} in UK cities. These emissions are from the NAEI for
127 the UK and the European Monitoring and Evaluation Programme (EMEP) for all other European
128 countries. Emissions scaling is applied to terrestrial NAEI and EMEP SO₂ emissions to address a
129 positive bias in modelled SO₂ surface concentrations (Marais et al., 2021) and to NAEI and EMEP
130 NH₃ emissions to resolve differences between bottom-up emissions and those derived with satellite
131 observations of NH₃ column densities (Marais et al., 2021). The scale factors applied are a uniform
132 60% decrease in land-based SO₂ emissions and 50% increase in NH₃ emissions. The NH₃
133 emissions scaling is supported by agreement between the model and surface observations of NH₄
134 concentrations at rural sites throughout the UK with this scaling (Kelly et al., 2023) and, as we
135 show in this work, modelled and observed rainwater concentrations of reduced (NH_x ≡ NH₃ +
136 NH₄) nitrogen.

137 For the future (2030) scenario simulations, we scale anthropogenic emissions in all
138 countries in Europe in the PD simulation using projected changes in emissions derived from
139 integrated assessment models. All anthropogenic emissions, except aviation, are from the
140 ECLIPSE project (Stohl et al., 2015). Emissions due to control measures are predicted in five-year
141 intervals from 1990 to 2050 for multiple major pollutants and precursors (SO₂, NO_x, NH₃, primary
142 PM_{2.5}, non-methane volatile organic compounds (NMVOCs), and carbon monoxide) and sectors
143 (agriculture, residential, commercial, energy, industry, on- and off-road transport, shipping, and
144 waste). We use data from the latest version of ECLIPSE (v6b;
145 <https://previous.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv6b.html>;
146 last accessed 3 August 2022). For the CLE simulation, we use the ECLIPSEv6b ‘CLE’ scenario, as
147 this incorporates changes in anthropogenic emissions due to implementation of mitigation

148 measures legislated as recently as 2018. For the MTF simulation, we use the ECLIPSEv6b
149 ‘MTFR’ scenario that assumes adoption of all available air pollution control measures. The
150 ECLIPSEv6b shipping emissions were recently revised to capture a wider range of emission
151 factors for primary particles. Only the CLE emissions were updated, so the same shipping
152 emissions are used for CLE and MTF scenarios. ECLIPSEv6b does not predict aviation emissions,
153 so we take these from Intergovernmental Panel on Climate Change (IPCC) projections
154 downloaded using the Representative Concentration Pathways Database tool version 2.0.5
155 (<https://tntcat.iiasa.ac.at/RcpDb/>; last accessed 3 August 2022). The IPCC emissions we use are
156 the business-as-usual Shared Socioeconomic Pathways (SSP) 2-4.5 (Fricko et al., 2017) for the
157 CLE scenario and the sustainable pathways SSP 1-2.6 (van Vuuren et al., 2017) for the MTF
158 scenario. All emissions data are provided as gridded annual values at $0.5^\circ \times 0.5^\circ$, so we derive
159 annual scale factors to apply to PD emissions at $0.5^\circ \times 0.5^\circ$ resolution. Hourly, monthly and
160 seasonal scaling factors are the same as those used for the PD emissions. The Harmonized
161 Emission Component (HEMCO) processing package (Keller et al., 2014) is used to apply scaling
162 factors and to grid emissions to the GEOS-Chem boundary condition and nested model resolutions.

163 Natural emissions are the same for all scenarios. These are $0.25^\circ \times 0.3125^\circ$ offline
164 emissions of hourly dust (Meng et al., 2021), biogenic VOCs (Weng et al., 2020), and soil NO_x
165 (Weng et al., 2020), and 3-hourly lightning NO_x (Murray et al., 2012). Natural NH_3 emissions are
166 monthly values from the Global Emissions Initiative (GEIA) inventory for oceans and soils
167 (Bouwman et al., 1997) and annual values from Riddick et al. (2012) for seabirds. GEIA NH_3
168 emissions are halved to address a known overestimate in these (Paulot et al., 2014).

169 GEOS-Chem simulates the individual $\text{PM}_{2.5}$ components sulfate, nitrate, NH_4 , primary and
170 secondary organic aerosols (POA, SOA), black carbon (BC), sea salt, and dust. Formation of
171 sulfate from oxidation of SO_2 , detailed in Park et al. (2004), is dominated by in-cloud oxidation
172 by hydrogen peroxide. Formation of aerosol nitrate from uptake of nitric acid (HNO_3) and aerosol
173 NH_4 from reversible partitioning of NH_3 is determined with the thermodynamic equilibrium model
174 ISORROPIA-II (Fountoukis & Nenes, 2007). SOA is estimated with fixed mass yields applied to
175 anthropogenic and biogenic NMVOCs emissions (Pai et al., 2020).

176 Model representation of other $\text{PM}_{2.5}$ components are detailed in Jaeglé et al. (2011) for sea
177 salt and Fairlie et al. (2007) for dust. The proportion of BC and POA emitted as hydrophobic (80%
178 for BC, 50% for POA) ages to hydrophilic at an e-folding time of 1.15 days (Cooke et al., 1999;
179 Chin et al., 2002). Dry deposition is via the Wesely (1989) resistance-in-series scheme. Wet
180 deposition includes scavenging in convective updrafts and rainfall, as well as washout, entrainment
181 and detrainment as detailed in Liu et al. (2001) for water-soluble aerosols and Amos et al. (2012)
182 for gases. We enable the fast scavenging mechanism of Luo et al. (2020) that addresses a positive
183 cold season bias in modelled surface concentrations of aerosol nitrate and NH_4 across Europe.
184 Chemical initialization of the model is achieved with spin-ups of a year for global boundary
185 conditions and 2 months for the PD, CLE and MTF nested domain simulations.

186 The model is sampled from 1 January to 31 December 2019 to calculate annual mean surface
187 concentrations of NH_3 and components of $\text{PM}_{2.5}$ and of reduced and oxidized gas and aerosol-
188 phase nitrogen wet and dry deposition fluxes. Surface concentrations of $\text{PM}_{2.5}$ are calculated at
189 standard conditions of temperature (20°C) and pressure (1 atm) and associated aerosol water is
190 determined using hygroscopic growth factors of individual $\text{PM}_{2.5}$ components representative of

191 50% relative humidity, as detailed in Kelly et al. (2023). Total nitrogen deposition is calculated as
192 the sum of GEOS-Chem deposition fluxes of reduced and oxidized nitrogen compounds that wet
193 and dry deposit (NH_3 , NH_4 , HNO_3 , aerosol nitrates, peroxy acetyl nitrates, and gas-phase
194 organonitrates) and that only dry deposit (NO_2 , dinitrogen pentoxide, and C1-C3 alkylnitrates).

195 **2.2 Surface Observations for GEOS-Chem Evaluation**

196 Kelly et al. (2023) compared UK network measurements of $\text{PM}_{2.5}$ to $\text{PM}_{2.5}$ obtained with
197 an earlier version of the model driven with similar PD emissions to those detailed in Section 2.1.
198 In that comparison, GEOS-Chem annual mean $\text{PM}_{2.5}$ is spatially consistent with the observations
199 ($R = 0.66$), but with an expected model underestimate (of 11%), as many of the network sites are
200 in cities along busy roads. We compare our PD model output to that of Kelly et al. (2023) archived
201 on the University College London Data Repository (Marais et al., 2022).

202 We also evaluate model simulation of nitrogen wet deposition with surface network
203 rainwater concentrations of oxidized and reduced nitrogen. These are from rural and semi-rural
204 UK Eutrophying and Acidifying Pollutants (UKEAP) Precipitation Network (Precip-Net)
205 measurement sites established to determine long-term trends in wet deposition of air pollutants.
206 Data at the forty-one sites operational in 2019 are from the UK-Air data portal ([https://uk-
207 air.defra.gov.uk/data/data_selector](https://uk-air.defra.gov.uk/data/data_selector); last accessed 15 March 2023). Samples at these sites are
208 collected every two weeks to record rainwater volume and measure oxidized (nitrate) and reduced
209 (NH_x) nitrogen rainwater concentrations (Kelleghan et al., 2021). We identify and remove
210 observations contaminated by bird strikes (Conolly et al., 2021) and correct biases in GEOS-FP
211 monthly total precipitation (Neelam et al., 2021; Travis et al., 2016; Paulot et al., 2014). Only three
212 samples are contaminated with bird strikes, identified with phosphate (P) rainwater concentrations
213 $> 0.1 \text{ mg P L}^{-1}$. We correct for biases in GEOS-FP precipitation by multiplying GEOS-FP monthly
214 total precipitation (precip) by the correction factor derived by Paulot et al. (2014)
215 ($[\text{precip}_{\text{observed}}/\text{precip}_{\text{modelled}}]^{0.6}$), where modelled precipitation is for grids coincident with UKEAP
216 sites. We use the corrected modelled rainwater volume and GEOS-Chem wet deposition fluxes to
217 calculate GEOS-Chem nitrate and NH_x rainwater concentrations.

218 **2.3 Public Health Burden Attributable to Exposure to $\text{PM}_{2.5}$**

219 We use the hybrid health risk assessment model from Weichenthal et al. (2022) to calculate
220 adult premature deaths attributable to exposure to $\text{PM}_{2.5}$ in the PD and by 2030 due to CLE and
221 MTF control measures. The hybrid model we use merges two functions to represent the
222 relationship between $\text{PM}_{2.5}$ and relative risk (RR) of adult (25+ years) premature mortality
223 covering a wide $\text{PM}_{2.5}$ concentration range. The functions are known as the extended Shape
224 Constrained Health Impact Function (eSCHIF) and Fusion. eSCHIF was derived with Canadian
225 Census Health and Environment Cohort (CanCHEC) study data covering 2.5 to 17.7 $\mu\text{g m}^{-3}$
226 (Brauer et al., 2022; Weichenthal et al., 2022). Fusion itself is a hybrid of three widely used health-
227 risk assessment models, established to address deficiencies in each (Burnett et al., 2022). As the
228 Fusion function is underconstrained at very low $\text{PM}_{2.5}$ concentrations, Weichenthal et al. (2022)
229 used eSCHIF at 2.5-5 $\mu\text{g m}^{-3}$, choosing the recently revised World Health Organization (WHO)
230 guideline value of 5 $\mu\text{g m}^{-3}$ (WHO, 2021) to transition between the two curves. This requires an
231 artificial ~14% increase in Fusion RRs at concentrations relevant to the UK to achieve a continuous

232 curve. We avoid this by rather adapting the code provided by Weichenthal et al. (2022) to transition
 233 at the intersection of the two curves at $9.8 \mu\text{g m}^{-3}$.

234 The eSCHIF RR function we apply from 2.5 to $9.8 \mu\text{g m}^{-3}$ is:

$$235 \quad eSCHIF(z) = \exp \left\{ \frac{\phi \ln\left(\frac{z-z_{cf}+1}{\alpha}\right)}{\left(1+\exp\left(-\frac{z-z_{cf}-\tau}{\nu}\right)\right)} + \omega \ln\left(\frac{z-z_{cf}}{\delta} + 1\right) \right\} \quad (1),$$

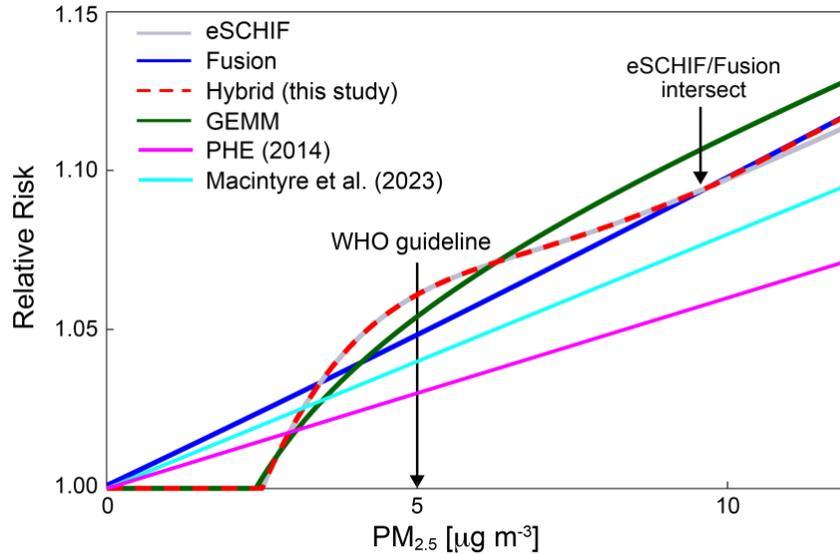
236 where z is annual mean $\text{PM}_{2.5}$, z_{cf} is the counterfactual concentration of $2.5 \mu\text{g m}^{-3}$ at and below
 237 which $\text{RR} = 1$, and $\phi, \alpha, \tau, \nu, \omega$ and δ are best-fit parameters derived from the cohort data. The
 238 Fusion curve we apply for $\text{PM}_{2.5} > 9.8 \mu\text{g m}^{-3}$ is:

$$239 \quad Fusion(z) = \exp \left\{ \gamma \times (\min(z, \mu) + \int_{\mu}^z \left(1 + \frac{1-\rho}{\rho} \left(\frac{x-\mu}{\theta-\mu}\right)^{\frac{\theta-\mu}{\theta(1-\rho)}}\right)^{-1} dx + \rho \theta \ln(\max(z, \theta)/\theta) \right\} \quad (2),$$

240 where γ, μ, ρ and θ are best-fit parameters obtained with the cohort data. $\text{PM}_{2.5}(z)$ in Eq. (1) and
 241 (2) are PD, CLE and MTF annual parameters in each GEOS-Chem grid.

242 Figure 1 shows the shapes of the Fusion and eSCHIF curves and the hybrid of the two for
 243 $\text{PM}_{2.5} \leq 12 \mu\text{g m}^{-3}$, the peak measured and modelled annual mean $\text{PM}_{2.5}$ in the UK in 2019 (Kelly
 244 et al., 2023). Fusion is linear at the range relevant to the UK. eSCHIF is supralinear from 2.5 to ~ 5
 245 $\mu\text{g m}^{-3}$; a shape supported by epidemiological cohort studies conducted in Europe and North
 246 America (Di et al., 2017; Strak et al., 2021).

247



248

249 **Figure 1.** Curves relating relative risk of adult premature mortality to ambient $\text{PM}_{2.5}$. Lines are
 250 eSCHIF (Eq. (1), grey), Fusion (Eq. (2), blue), the hybrid of the two (red dashed), the Global
 251 Exposure Mortality Model (GEMM, green), and curves used in past UK studies by Public Health
 252 England (PHE, 2014) (magenta) and Macintyre et al. (2023) (cyan). Arrows point to the WHO
 253 2021 guideline at $5 \mu\text{g m}^{-3}$ and the intersection of eSCHIF and Fusion at $9.8 \mu\text{g m}^{-3}$.

254 The RR values obtained with Eq. (1) and (2) are used to calculate the fraction of adult
 255 nonaccidental deaths attributable to exposure to PM_{2.5}, the attributable fraction (*AF*):

$$256 \quad AF = \begin{cases} 1 - \frac{1}{eSCHIF(z)} & 2.5 \mu\text{g m}^{-3} < z \leq 9.8 \mu\text{g m}^{-3} \\ 1 - \frac{1}{FUSION(z)} & z > 9.8 \mu\text{g m}^{-3} \end{cases} \quad (3).$$

258 These are calculated for each GEOS-Chem grid and further used to estimate adult excess deaths
 259 attributable to exposure to PM_{2.5} as the product of *AF*, adult population, and baseline nonaccidental
 260 mortality rates (*BMR*):

$$261 \quad PM_{2.5} \text{ excess deaths} = AF \times \text{Adult Population} \times BMR \quad (4).$$

262 *BMR*_{2.5} excess deaths = *AF* × Adult Population × *BMR*
 263 *BMRs* are from the Global Burden of Disease (GBD) for 2019 to avoid influence from COVID.
 264 These are provided in 5-year age increments (<https://vizhub.healthdata.org/gbd-results/>, last
 265 accessed 30 March 2023), where the nonaccidental premature deaths we use are the sum of non-
 266 communicable diseases and lower-respiratory infections for adults 25 years and older. These
 267 account for 96% of UK total adult premature deaths. *BMRs* are reported for each administrative
 268 area in England and at the national scale for Scotland, Wales and Northern Ireland. UK population
 269 data for 2019 reported in 5-year age increments at 30 arc-seconds (~1 km) resolution are from
 270 WorldPop (<https://dx.doi.org/10.5258/SOTON/WP00671>; last accessed 5 August 2022). The
 271 GBD *BMRs* and cumulative WorldPop data for age-groups 25 years and older are gridded to the
 272 GEOS-Chem grid to collocate population and PM_{2.5} pollution hotspots. These gridded data of *AFs*,
 273 adult population and *BMRs* are multiplied (Eq. (4)) and the resultant gridded excess deaths are
 274 sampled with shapefiles of UK administrative areas (county councils, unitary authorities and
 275 metropolitan counties) from the Database of Global Administrative Areas (GADM)
 276 (<https://gadm.org/>; last accessed 17 March 2023). This yields excess deaths in all 184 administrate
 277 areas: 119 in England, 32 in Scotland, 22 in Wales, and 11 in Northern Ireland.

280 We test sensitivity of premature mortality estimates to health-risk assessment model choice
 281 by comparing to values obtained with the Global Exposure Mortality Model (GEMM) (Burnett et
 282 al., 2018). The GEMM curve, included in Figure 1, is the version obtained without the Chinese
 283 Male Cohort, as detailed in Burnett et al. (2018). GEMM is also for adults 25 years and older and
 284 has the same counterfactual PM_{2.5} as the hybrid model (RR = 1 at 2.5 μg m⁻³). GEMM RRs are
 285 less than the hybrid model at PM_{2.5} < 7 μg m⁻³ and exceed the hybrid model above this.

287 2.4 Harm to Nitrogen Sensitive Habitats

288 We assess the efficacy of emission control scenarios at alleviating harm to sensitive
 289 habitats by quantifying the change in the amount and extent of nitrogen deposited in excess of
 290 sensitive habitat critical loads and the extent of ambient NH₃ > 1 μg m⁻³ indicative of harm to
 291 bryophytes. Recommended habitat-specific critical loads for the thirteen sensitive habitats in the
 292 UK are given in Table 1. These range from 5 kg nitrogen (N) ha⁻¹ a⁻¹ for low resilience habitats
 293 such as bogs and montane habitats to 10 kg N ha⁻¹ a⁻¹ for more resilient habitats such as calcareous
 294 grasslands and all woodlands except Scotts pine. These are revised following a recent review
 295 (Bobbink et al., 2022) and now 2-7 kg N ha⁻¹ a⁻¹ less than previously recommended values

296 compiled by Hall et al. (2015) for all sensitive habitats except the unmanaged oak and other
 297 woodlands habitats that are unchanged. We average the critical loads in Table 1 onto the GEOS-
 298 Chem grid using maps of fractional coverage of sensitive habitats provided at 1 km resolution by
 299 the UK Centre for Ecology and Hydrology (UKCEH). These gridded critical loads are shown in
 300 Figure 2(a). Nitrogen sensitive habitats account for 38% of UK land area, dominated by acid
 301 grasslands, dwarf shrub heath and coniferous woodlands. Most (58%) of the nitrogen sensitive
 302 areas are in Scotland (Rowe et al., 2022). The total coverage in Table 1 of 92,801 km² is 973 km²
 303 less than is in the critical loads trends report compiled by the UKCEH (Rowe et al., 2022), as the
 304 GEOS-Chem UK nested domain excludes Shetland Islands.

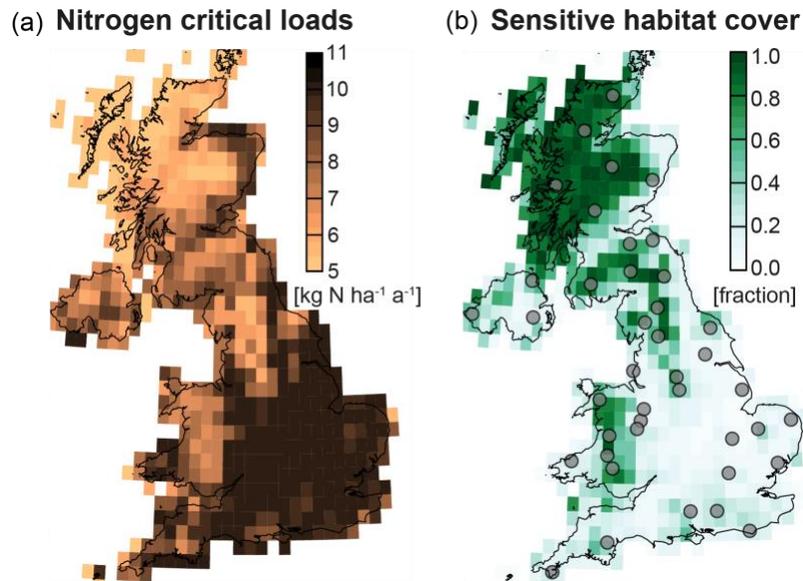
305 **Table 1.** Nitrogen critical loads applied to sensitive habitats in the UK

Sensitive habitat	Critical load [kg N ha⁻¹ a⁻¹]^a	UK coverage [km²]	Contribution [%]^b
Acid grasslands	6	20,342	22
Bog	5	8,514	9
Dwarf shrub heath	5	21,496	23
Calcareous grasslands	10	1,012	1
Coniferous woodland	10	14,452	16
Dune grassland	5	631	<1
Deciduous woodland	10	8,706	9
Unmanaged Beech (<i>Fagus</i>) woodland	10	2,059	2
Montane	5	4,915	5
Unmanaged oak woodland	10	6,958	7
Salt marsh	10	808	<1
Unmanaged Scotts pine woodland	5	1,485	1
Unmanaged other woodland	10	1,423	2

306 ^a Lower end of the range of recently updated values from Rowe & Hina (2023), ^b Percent contribution
 307 to UK sensitive area total coverage of 92,801 km².

308 GEOS-Chem is too coarse to resolve deposition to individual sensitive habitats. This is
 309 particularly apparent across southern and central England where sensitive habitats account for
 310 <20% of GEOS-Chem grid areas (Figure 2(b)). GEOS-Chem wet deposition also does not account
 311 for enhanced nitrogen washout from orographic lifting over upland areas (Dore et al., 1992; Fowler
 312 et al., 1988). Given this, we use UKCEH Concentration Based Estimated Deposition (CBED)
 313 annual multiyear (2018-2020) mean nitrogen deposition rates provided at 5 km resolution to
 314 calculate PD nitrogen deposition exceedances. The derivation of this dataset is detailed in Rowe
 315 et al. (2022). Briefly, CBED wet deposition fluxes are determined with UK Met Office annual
 316 precipitation data and monitoring network observations of ambient reactive nitrogen
 317 concentrations and rainwater ion concentrations interpolated to the 5 km CBED grid and by
 318 accounting for enhanced wet deposition due to orographic lifting and deposition of cloud droplets
 319 to vegetation. Dry deposition is determined with a resistance-in-series model using ammonia
 320 concentrations simulated by the EMEP regional CTM at 5 km resolution and vegetation-specific
 321 deposition velocities. The CBED deposition data includes nitrogen deposition values for forests
 322 and open vegetation. Deposition rates to forests are ~60% more than values for open vegetation,

323 due to greater surface area of forest vegetation. We map CBED deposition data to the GEOS-Chem
 324 grid using UKCEH habitat maps of the spatial coverage of the six woodlands and seven open
 325 vegetation habitats in Table 1 before using the gridded deposition data to calculate nitrogen
 326 exceedances for the PD. This yields maps of nitrogen deposition in $\text{kg N (ha sensitive habitat)}^{-1} \text{ a}^{-1}$
 327 ¹. We then use GEOS-Chem PD, CLE and MTF total nitrogen deposition to calculate changes in
 328 nitrogen deposition due to CLE and MTF emission controls.
 329



330

331 **Figure 2.** UK nitrogen sensitive habitat critical loads and coverage on the GEOS-Chem grid. Maps
 332 are sensitive habitat nitrogen critical loads in $\text{kg N (ha sensitive habitat)}^{-1} \text{ a}^{-1}$ (a) and fractional
 333 cover of sensitive habitats in each GEOS-Chem gridcell. Grey circles in (b) are UKEAP Precip-
 334 Net monitoring site locations (Section 2.2).

335 3 Results and Discussion

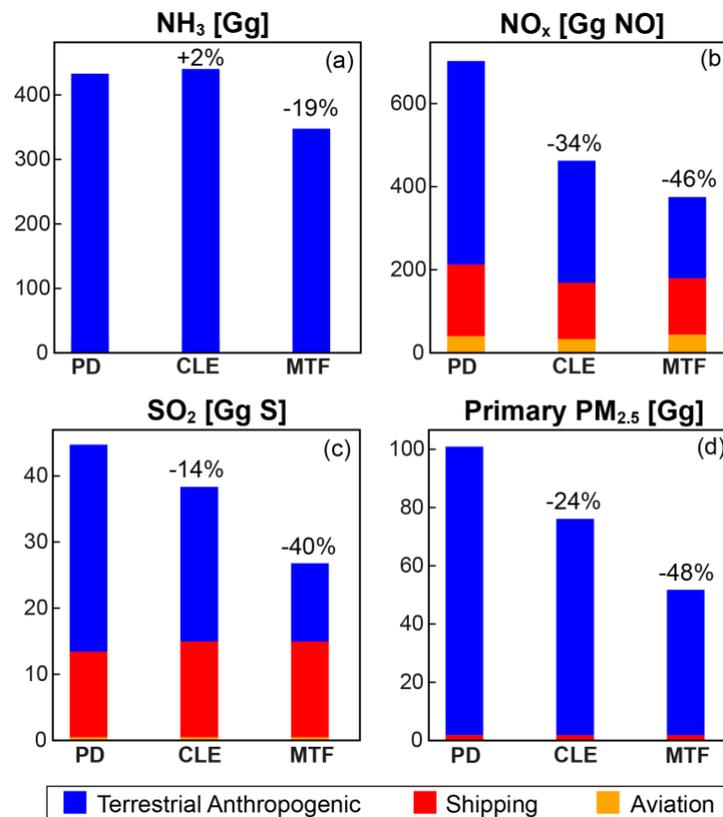
336 3.1 Emission Control Scenarios and GEOS-Chem Evaluation

337 Figure 3 shows annual anthropogenic emissions of NH_3 , NO_x , SO_2 and primary $\text{PM}_{2.5}$ used
 338 in GEOS-Chem PD and future (CLE, MTF) simulations. These are totals for the NAEI domain, so
 339 include shipping emissions in UK territorial waters. PD emissions total 452 Gg NH_3 , 712 Gg NO_x
 340 as NO , 45 Gg SO_2 as S , and 102 Gg primary $\text{PM}_{2.5}$. Agriculture accounts for ~90% of
 341 anthropogenic NH_3 , dominated by fertiliser use and livestock and manure management. NO_x is
 342 mostly from land transport and shipping and is the only $\text{PM}_{2.5}$ precursor with non-negligible
 343 contribution from aviation of 5% in the PD. SO_2 is mostly from industrial processes, energy
 344 generation and shipping (Churchill et al., 2022). Primary $\text{PM}_{2.5}$ is mostly from residential and
 345 commercial sectors and industry.

346 All air pollutant precursors except NH_3 decline in the CLE scenario by 14% for SO_2 , 34%
 347 for NO_x , and 24% for primary $\text{PM}_{2.5}$. According to ECLIPSEv6b emissions projections, the UK
 348 fails to meet its CLRTAP emissions ceilings commitment of 8% decrease in NH_3 emissions with

349 current measures. Adoption of maximum technically feasible measures surpasses this
 350 commitment, achieving a 19% decrease in NH₃ emissions. This is still less than half the minimum
 351 decline that Woodward et al. (2022) estimate is needed to significantly mitigate harm to sensitive
 352 habitats using less strict critical loads than the updated recommended values (Table 1). The 2%
 353 increase in CLE NH₃ emissions in Figure 3(a) is due to limited adoption of suggested measures to
 354 offset increase in NH₃ emissions from intensification of agriculture to meet growing food and
 355 market demands (Alexandratos & Bruinsma, 2012). NO_x emissions from shipping for both the
 356 CLE and MTF scenarios decrease by 21% due to NO_x emissions regulations adopted in the Baltic
 357 and North Seas since 2021 (IMO, 2019). Shipping emissions of SO₂ increase by 12% relative to
 358 the PD, increasing its contribution to anthropogenic SO₂ emissions from 29% for the PD to 38%
 359 for the CLE and 54% for the MTF. According to ECLIPSEv6b, large (>70%) reductions in SO₂
 360 emissions from switching to low-sulfur fuel occur a decade earlier in 2010-2020 (Office of the
 361 European Union, 2016b). Aviation NO_x decreases by 19% in the CLE scenario and increases by
 362 10% in the MTF scenario, due to trade-offs between emissions of NO_x and the greenhouse gas
 363 carbon dioxide (Freeman et al., 2018; Skowron et al., 2021). The relative contribution of aviation
 364 to total NO_x emissions is 7% for CLE and 11% for MTF.

365

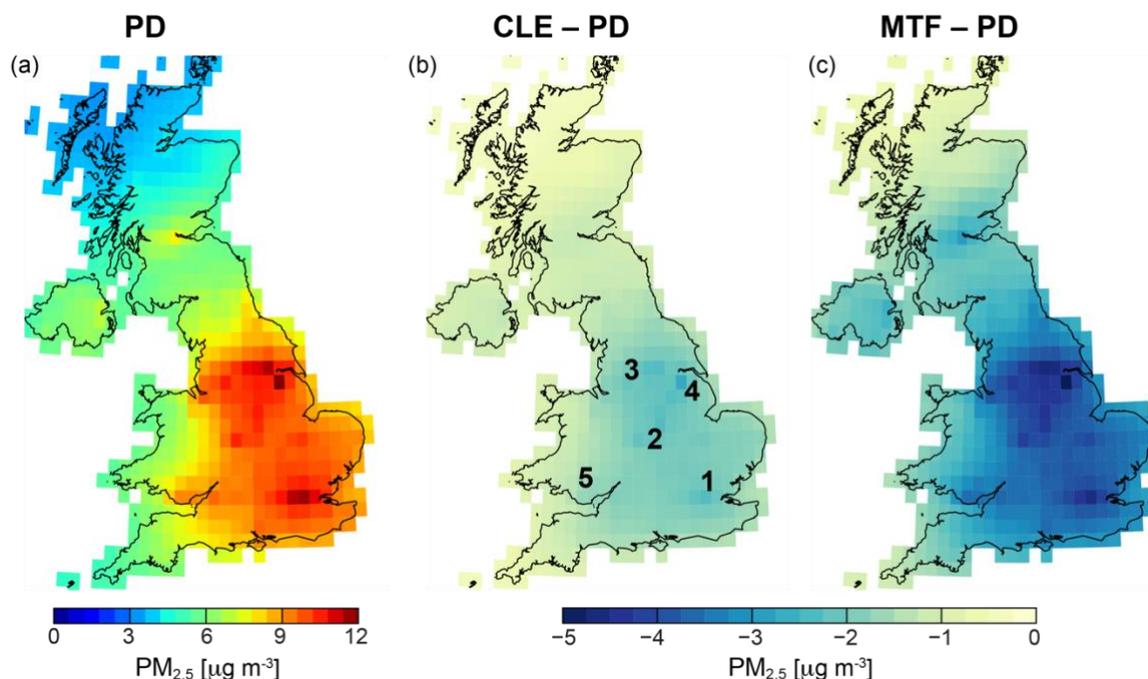


366

367 **Figure 3:** UK present and future anthropogenic emissions of PM_{2.5} and nitrogen pollution
 368 precursors. Panels show total annual emissions of NH₃ (a), NO_x (b), SO₂ (c), and primary PM_{2.5}
 369 (d) for PD and future (2030) CLE and MTF emissions scenarios. Primary PM_{2.5} is the sum of BC,
 370 POA and dust. Inset values are percent change in CLE and MTF relative to PD. Bar colours
 371 distinguish surface-bound terrestrial anthropogenic sources (blue), ships (red), and aircraft
 372 (yellow).

373 Figure 4(a) shows GEOS-Chem PD annual mean PM_{2.5}. Grids have the same spatial
 374 distribution ($R = 0.99$) and average only 6% or $0.4 \mu\text{g m}^{-3}$ more than is simulated with the model
 375 version from Kelly et al. (2023) (Section 2.2). Monthly UK mean PM_{2.5} in this work ranges from
 376 $0.6 \mu\text{g m}^{-3}$ less (February) to $1.9 \mu\text{g m}^{-3}$ more (April) than is obtained by Kelly et al. (2023). These
 377 differences are due to more efficient cold season wet deposition of nitrate and NH_4 and less
 378 extreme downscaling of SO_2 emissions in this work. The latter enhances formation of springtime
 379 inorganic sulfate, nitrate and ammonium aerosol. As in Kelly et al. (2023), 79% of GEOS-Chem
 380 grids in Figure 4(a) exceed the WHO 2021 annual mean PM_{2.5} guideline of $5 \mu\text{g m}^{-3}$.

381



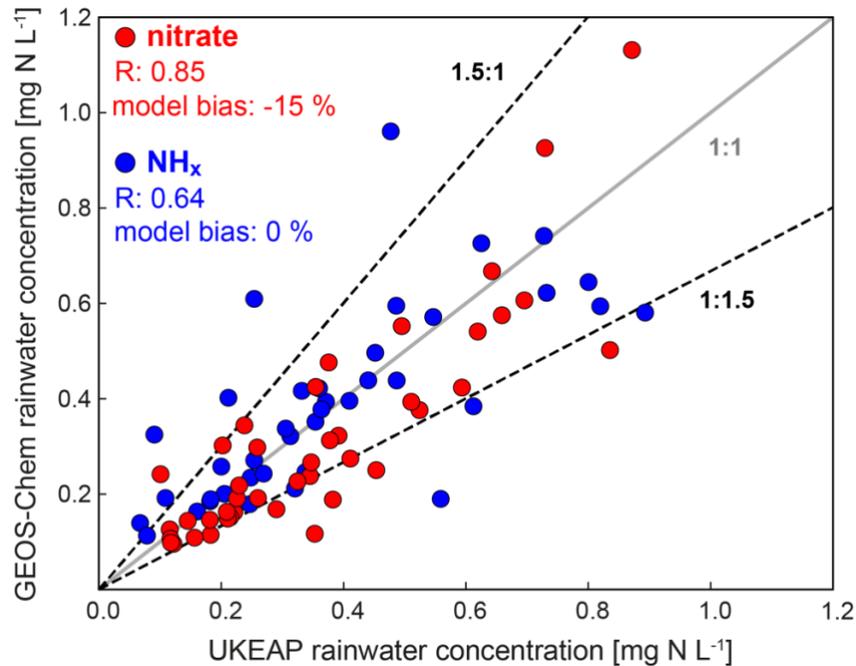
382

383 **Figure 4.** Effect of emission control measures on UK PM_{2.5}. Maps are GEOS-Chem annual mean
 384 PM_{2.5} in 2019 (PD) (a) and changes in PM_{2.5} relative to the PD due to controls on anthropogenic
 385 sources to meet current legislation (CLE minus PD) (b) and adoption of readily available
 386 maximum technically feasible measures (MTF minus PD) (c). Inset numbers in (b) are locations
 387 with greatest decline in PM_{2.5} discussed in the text: 1. London, 2. Birmingham, 3. Manchester and
 388 Leeds, 4. East Yorkshire, and 5. south coast of Wales.

389 Figure 5 compares observed and modelled rainwater concentrations of nitrate and NH_x to
 390 assess GEOS-Chem PD nitrogen wet deposition at UKEAP monitoring sites (Figure 2(b)).
 391 Temporal coverage at all sites exceeds 75%. Site mean UKEAP rainwater concentrations are 0.34
 392 mg N L^{-1} for nitrate and 0.42 mg N L^{-1} for NH_x . The correction applied to GEOS-FP total
 393 precipitation (Section 2.2) leads to changes in GEOS-FP rainwater volume that varies from a 40-
 394 50% increase in December and January, to a 23-26% decrease in April and July, to near-negligible
 395 (4-6%) difference in March and November. The overall effect on total rainwater volume in 2019
 396 is a small 4% increase. Without the correction, the correlation between modelled and observed
 397 NH_x is weak ($R = 0.47$) and there is an apparent underestimate in the model of 33% for nitrate and
 398 22% for NH_x . With the rainwater volume correction, both nitrate ($R = 0.85$) and NH_x ($R = 0.64$)

399 are spatially consistent, modelled nitrate is 15% or 0.05 mg N L^{-1} less than is observed, and
 400 modelled and observed NH_x are equivalent. A 15% difference in oxidized NO_x is more than the
 401 prescribed 8% error in NAEI NO_x emissions (Richmond et al., 2020), but in agreement with a low
 402 bias in NAEI NO_x emissions when compared to top-down emissions derived with satellite
 403 observations of tropospheric columns of nitrogen dioxide (NO_2) (Pope et al., 2022).

404



405

406

407 **Figure 5.** Evaluation of modelled nitrogen wet deposition. Scatterplot compares observed
 408 (UKEAP) and modelled (GEOS-Chem) rainwater concentrations of oxidized (nitrate, red) and
 409 reduced (NH_x , blue) nitrogen at UKEAP sites in 2019. The grey line is the 1:1 relationship and the
 410 black dashed lines bound the 50% difference. Numbers inset are the Pearson's correlation
 411 coefficient (R) and the model normalized mean bias. Site locations are in Figure 2(b).

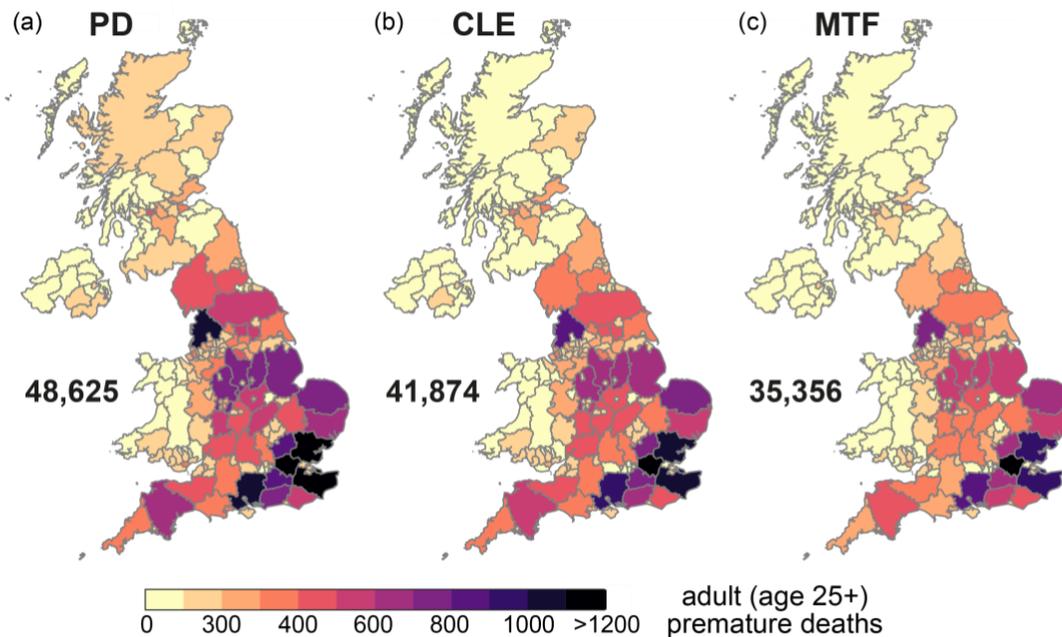
412 The contribution of wet deposition to total nitrogen deposition is 58% for grids coincident
 413 with UKEAP sites. Many of these sites evaluate model grids dominated by sensitive areas,
 414 particularly in Scotland (Figure 2(b)). The 58% contribution to total deposition is similar (57%)
 415 for all UK grids and includes contributions from NH_3 (31%), HNO_3 (28%), NH_4 (25%) and aerosol
 416 nitrate (15%). The 43% of deposited nitrogen undergoing dry deposition in the UK is mostly (64%)
 417 NH_3 . There are no network measurements of dry deposition, though dominance of NH_3 has also
 418 been inferred from derived dry deposition fluxes obtained using concentration and meteorology
 419 measurements and modelled dry deposition velocities at UK monitoring sites (Flechard et al.,
 420 2011; Tang et al., 2009).

421 3.2 Influence of Emission Controls on $\text{PM}_{2.5}$ and Public Health

422 Figure 4 includes the decrease in modelled $\text{PM}_{2.5}$ resulting from control measures to meet
 423 current legislation (4b) and from adopting maximum technically feasible measures (4c). Decline

424 in $PM_{2.5}$ in the CLE scenario is at most $2 \mu g m^{-3}$ and typically $0.9-1 \mu g m^{-3}$. MTF measures achieve
 425 up to $5 \mu g m^{-3}$ decline in populated cities (London, Birmingham, Manchester, Leeds) and in
 426 locations with intensive industry (Welsh south coast) and large power plants (East Yorkshire).
 427 Greater decline in the MTF is due to combined decrease in point source emissions of SO_2 ,
 428 vehicular emissions of NO_x , and agricultural emissions of NH_3 (Figure 3). The largest decline is
 429 in April ($3.7 \mu g m^{-3}$ UK mean) and the smallest is in summer months ($1.1-1.5 \mu g m^{-3}$). The percent
 430 UK grids in Figure 4(a) that exceeded the WHO guideline declines from 79% in the PD to 58% in
 431 the CLE and 36% in the MTF. Most of the decline is over Northern Ireland and Scotland north and
 432 south of the Central Belt for the CLE and for the MTF this spreads to the rest of Scotland, all of
 433 Cornwall and most of Wales.

434



436

436 **Figure 6.** Present-day and future UK adult premature mortality attributable to exposure to $PM_{2.5}$.
 437 Maps are administrative area early deaths for the PD (2019) (a) and future (2030) with CLE (b)
 438 and MTF (c) emission controls. Inset values are premature mortality estimated with the hybrid
 439 eSCHIF-Fusion model detailed in Section 2.3.

440 Figure 6 maps adult excess deaths attributable to exposure to $PM_{2.5}$ for all 3 scenarios in
 441 all UK administrative areas in the PD and by 2030 from adopting CLE or MTF measures. The
 442 eSCHIF portion of the hybrid curve (Eq. (1); Figure 1) is applied to 85% of GEOS-Chem grids for
 443 the PD and all grids for the CLE and MTF scenarios. $PM_{2.5}$ in all administrative areas exceeds the
 444 $2.5 \mu g m^{-3}$ threshold for harm in the hybrid model (Figure 1). In the PD, UK annual premature
 445 mortality totals 48,625 (95% confidence interval (CI): 45,118-52,595) and on average 8% of
 446 premature adult deaths are attributable to exposure to $PM_{2.5}$. Greater London accounts for 10% of
 447 these (4,861; 95% CI: 4,549-5,247) and exceeds national totals for Scotland (3,673; 95% CI:
 448 3,214-4,073), Wales (2,462; 95% CI: 2,270-2,660) and Northern Ireland (1,052; 95% CI: 934-
 449 1,156). Administrative areas other than Greater London that exceed a thousand excess deaths in
 450 the PD are Essex, Kent and Hampshire in southeast England and Lancashire in northwest England.

451 Lancashire and Hampshire fall below 1,000 with CLE measures and all except Greater London
452 fall below 1,000 with MTF measures.

453 Annual adult excess deaths for the UK in the PD obtained using GEMM (Figure 1) are
454 4,608 more than the hybrid model at 53,233 (95% CI: 38,526-67,539), due to higher RRs in
455 GEMM for $\text{PM}_{2.5} > 7 \mu\text{g m}^{-3}$ typical of populated cities in England (Figure 2(a)). The excess deaths
456 we obtain for the UK in the PD with the hybrid model exceed previously published estimates of
457 28,969 for 2010 (Gowers et al., 2014) and 40,408 (95% CI: 30,867-45,050) for 2018 (Macintyre
458 et al., 2023). These are both for adults ≥ 30 rather than ≥ 25 years old, but this difference has a small
459 effect on burden of disease estimates, as premature mortality for the 25-30 age range is low (PHE,
460 2014). The curves used in past studies are compared to the hybrid model and GEMM in Figure 1.
461 Incremental changes in risk used in those studies are on average 0.6% (Gowers et al., 2014) and
462 0.8% (Macintyre et al., 2023) per $\mu\text{g m}^{-3}$ $\text{PM}_{2.5}$ or RRs of 1.06 (Gowers et al., 2014) and 1.08
463 (Macintyre et al., 2023) at $10 \mu\text{g m}^{-3}$. The hybrid model change in risk per $\mu\text{g m}^{-3}$ $\text{PM}_{2.5}$ ranges
464 from almost 1% for $\text{PM}_{2.5} > 9.8 \mu\text{g m}^{-3}$ (the Fusion portion of the curve) to 0.5% at $5\text{-}9.8 \mu\text{g m}^{-3}$
465 and 2.5% at $2.5\text{-}5 \mu\text{g m}^{-3}$, due to variability in the eSCHIF curve gradient.

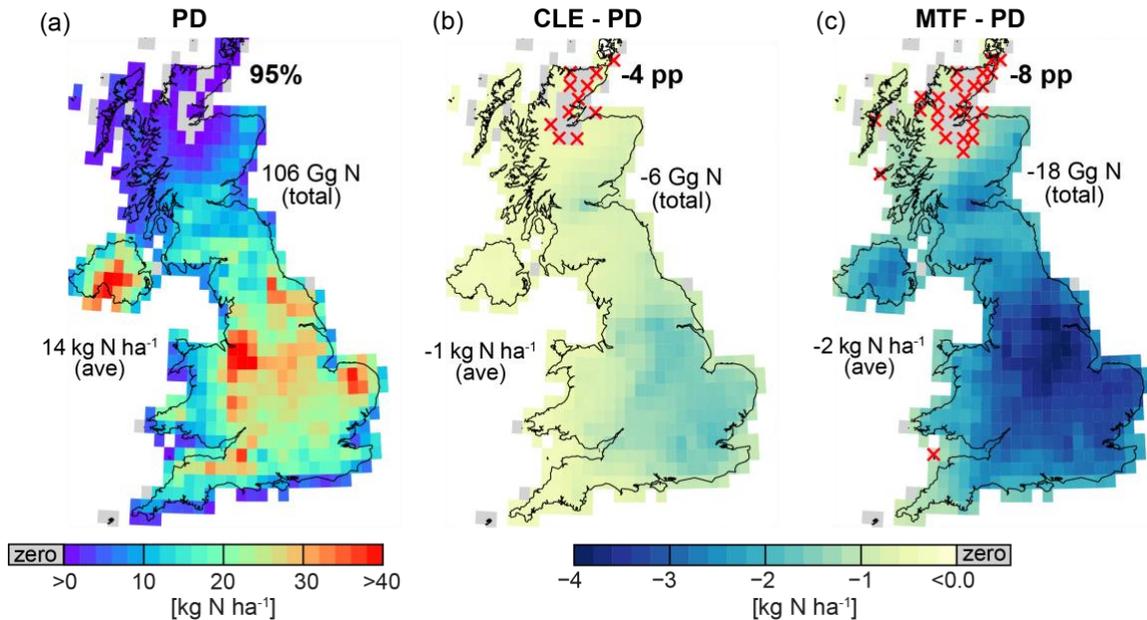
466 Decline in precursor emissions of $\text{PM}_{2.5}$ in the CLE results in a UK mean attributable
467 fraction of 7% and total premature deaths of 41,874 (95% CI: 37,949-45,437) by 2030 or 6,751
468 avoided early deaths. In the MTF, premature deaths attributable to exposure to $\text{PM}_{2.5}$ is 6% of all
469 nonaccidental premature deaths and totals 35,356 (95% CI: 29,882-40,015) by 2030. This is
470 13,269 avoided deaths, representing 27% of PD premature deaths attributable to $\text{PM}_{2.5}$ and twice
471 that achieved with current legislation. The GEMM curve also yields a factor of 2 benefit to public
472 health from adopting best available technologies over what is currently legislated.

473 The elderly (65+) account for most (86%) early nonaccidental deaths in the GBD data, so
474 an aging UK population and recent epidemiological evidence for much greater health risk than the
475 hybrid curve amongst the elderly exposed to $\text{PM}_{2.5}$ representative of concentrations in the UK
476 would influence our estimates of avoided deaths. The UK Office of National Statistics projects
477 there will likely be 2.5 million more elderly people in the UK in 2030 than in 2020
478 (<https://www.ons.gov.uk/peoplepopulationandcommunity/populationandmigration/populationprojections/bulletins/nationalpopulationprojections/2020basedinterim>; last accessed 2 June 2023),
479 dampening the benefits of emission controls. The causal inference study by Wu et al. (2020) used
480 16 years of US Medicare data to estimate an RR of at least 1.23 at $10 \mu\text{g m}^{-3}$ or on average a 2.3%
481 decline in risk per $\mu\text{g m}^{-3}$ decrease in $\text{PM}_{2.5}$ amongst the elderly for $\text{PM}_{2.5} \leq 12 \mu\text{g m}^{-3}$. This would
482 substantially increase the number of premature elderly deaths attributable to $\text{PM}_{2.5}$ in the PD, but
483 also increase the number of avoided early deaths due to emission controls.

485 **3.3 Harm of Nitrogen Deposition and Ambient NH_3 to Sensitive Habitats**

486 GEOS-Chem nitrogen deposited to all UK land totals 253 Gg N in the PD. This is lower
487 than values from higher-resolution models and derived datasets. GEOS-Chem deposition is 25 Gg
488 N or 9% less than is simulated for 2017 with a 5 km resolution CTM (Tomlinson et al., 2021) and
489 57 Gg N less than gridded mean nitrogen deposition from the CBED dataset (Section 2.4).
490 According to GEOS-Chem, total deposited nitrogen is ~40% of the emitted terrestrial and aviation
491 anthropogenic nitrogen (601 Gg N, Figure 3), suggesting that more than half (~60%) of the
492 nitrogen emitted is exported offshore. As a result, decline in nitrogen deposition by 2030 is only

493 about a third of the decrease in terrestrial and aviation nitrogen emissions from adopting emission
 494 controls. For CLE measures, nitrogen deposited to UK land decreases by 27 Gg N or 31% of the
 495 decline in terrestrial and aviation emissions of 88 Gg N (Figure 3). NH_x deposition increases in
 496 this scenario by 3 Gg N due to a 2% increase in NH_3 emissions. Decline in UK nitrogen deposition
 497 from MTF measures is 75 Gg N or 37% of the 205 Gg N decline in terrestrial and aviation
 498 emissions.



499

500 **Figure 7.** Impact of emission controls on excess nitrogen deposited to UK sensitive habitats. Maps
 501 are PD nitrogen deposition above sensitive habitat critical loads obtained with CBED (a) and the
 502 change in excess nitrogen deposition resulting from CLE (b) and MTF (c) control measures from
 503 GEOS-Chem. All units are $\text{kg N (ha sensitive habitat)}^{-1} \text{ a}^{-1}$. Grey shaded grids are below critical
 504 loads and red crosses identify those that fall below critical loads due to CLE (b) and MTF (c)
 505 controls. Inset numbers in (a) are percent UK land area, and total and average excess nitrogen, and
 506 in (b) and (c) the change in these relative to the PD. “pp” is percentage point.

507 Nitrogen deposited to sensitive habitats in excess of critical loads (Figure 2(a)) is shown in
 508 Figure 7(a) for the PD (2019). Excess nitrogen covers 95% of sensitive habitats, totals 106 Gg N
 509 and averages 14 kg N ha^{-1} . The largest exceedances of $>35 \text{ kg N ha}^{-1}$ coincide with NH_3 emission
 510 hotspots associated with intensive livestock farming (Marais et al., 2021; Hellsten et al., 2008). By
 511 2030, CLE controls decrease total nitrogen deposition to sensitive habitats by 6 Gg N and average
 512 exceedances by 1 kg N ha^{-1} (Figure 7(b)) compared to decline of 18 Gg N total and 2 kg N ha^{-1}
 513 average decline with MTF measures (Figure 7(c)). For both scenarios, decline in the extent of
 514 sensitive habitats exposed to excess nitrogen is modest at 4 percentage points for the CLE and 8
 515 percentage points for the MTF. Almost all this decline is in northern Scotland. A similarly small
 516 decline (6 percentage points) is reported for a decade of emission controls from 2010 to 2019
 517 (Rowe et al., 2022). Given that the decline in excess nitrogen deposition is $<10\%$ of the decline in
 518 emissions, an emissions reduction of at least 500 Gg N may be required to halve excess nitrogen

519 deposition to 50 Gg N. This is ~80% of present-day emissions or more than twice that feasible
520 with best available measures.

521 The extent of UK ambient air with NH_3 concentration $> 1 \mu\text{g m}^{-3}$, indicative of harm to
522 bryophytes, is 73% in the PD. An increase in UK mean ambient NH_3 in the CLE of 8% increases
523 UK area with $\text{NH}_3 > 1 \mu\text{g m}^{-3}$ to 75%. This is due to concurrent increase in NH_3 emissions (Figure
524 3) and decline in the efficacy of the NH_3 acidic aerosol sink (sulfate+nitrate decline by 24%). MTF
525 controls on both NH_3 and acidic aerosol precursor emissions of SO_2 and NO_x (Figure 3) cause
526 decline in both NH_3 (by 13%) and sulfate+nitrate (by 46%), causing a modest decrease in the
527 extent of $\text{NH}_3 > 1 \mu\text{g m}^{-3}$ to 69%. As NH_3 is semi-volatile, its emissions depend on multiple
528 environmental factors. A warmer world may, for example, substantially increase NH_3 emissions.
529 One estimate suggests a 5°C warmer world increases global NH_3 emissions by 30-70% (Sutton et
530 al., 2013).

531 **4 Conclusions**

532 Particulate matter pollution even at the relatively low concentrations typical of the UK
533 adversely affect human health and nitrogen pollution harms habitats sensitive to excess deposition
534 and exposure to the phytotoxin ammonia (NH_3). We assess the potential health and ecosystem co-
535 benefits that could be achieved in 2030 following legislative action or from adopting best available
536 emission control measures using emissions projections, the GEOS-Chem model, high-spatial-
537 resolution datasets, and up-to-date relationships between exposure and harm.

538 Legislative measures are most effective at targeting acidic aerosol (sulfate, nitrate)
539 precursor emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x), causing decline in fine
540 particulate matter ($\text{PM}_{2.5}$) pollution and 6,751 avoided annual early deaths. The extent of the UK
541 above the World Health Organization (WHO) $5 \mu\text{g m}^{-3}$ guideline for long-term exposure to
542 ambient $\text{PM}_{2.5}$ also decreases from 79% to 58%. If instead best available measures are adopted,
543 the number of avoided annual early deaths doubles to 13,269 and less than half (36%) the UK
544 exceeds the WHO guideline.

545 The benefits to habitats are near-negligible for current legislation and modest for adoption
546 of best available measures. Both NH_3 abundance and the amount of nitrogen deposited as NH_3
547 increase with current legislation, as controls on agricultural sources are ineffective at decreasing
548 NH_3 emissions and loss of NH_3 to acidic aerosols declines. Adoption of best available measures
549 decreases NH_3 emissions, as these target major agricultural sources, but these are also insufficient
550 for improving ecosystem health. A far greater (at least 80%) decline in UK nitrogen emissions
551 may be needed to halve harmful nitrogen deposition than the 34% achieved with best available
552 measures.

553 **Author Contributions**

554 EAM conducted the model simulations, analysed the output, calculated the ecosystem harm
555 metrics and led manuscript preparation. JK processed the UKEAP rainwater concentration data
556 and prepared 2050 emissions scalings files. KV calculated premature mortality. YL analysed the
557 emissions data with supervisory support from GLu. GLu also aided in data visualization. NH and

558 ER provided critical load and CBED deposition data and guided EAM through its use. All co-
559 authors provided editorial input.

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565 **Conflict of Interest**

566 The authors declare no conflicts of interest relevant to this study.

567 **Data Availability**

568 Data generated as part of this work and that are used, but not available for public access elsewhere,
569 are available on the UCL Data Repository (<https://doi.org/10.5522/04/23540079>) in NetCDF
570 format. These include GEOS-Chem annual mean ambient PM_{2.5} and NH₃ concentrations and
571 annual total nitrogen deposition, attributable fraction and premature mortality generated with the
572 eSCHIF-Fusion hybrid model, critical loads on the GEOS-Chem grid, UKCEH habitat maps at 1
573 km resolution, and CBED nitrogen deposition to forests and open vegetation at 5 km resolution.
574 GEOS-Chem version 13.0.0 is preserved at <https://doi.org/10.5281/zenodo.4618180> and
575 developed openly on GitHub (<https://github.com/geoschem/geos-chem>).

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