

Supplementary Information 2:

Development of a Spatially Explicit Fate Factor Model at the 5 Arcmin Resolution

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31

32 I Methodology

33 I.1 Fate Factor Model Structure

34 FFs express the mass of nutrients in waterbodies (kg) per unit of emission per year (kg year⁻¹), yielding
35 a time dimensional unit (year). As such, FFs describe the persistence fraction of the original nutrient
36 emission (E) in the receiving waterbodies (w)¹.

37 For freshwater receptors (*f_w*), the cumulative fate factor ($FF_{soils E (w)(z)(i)}$) for a soil emission in cell
38 (*i*) of emission pathway, *z* (surface runoff, erosion or leaching) and nutrient type, *E* (Nitrogen (N) or
39 Phosphorus (P)), is the product of nutrient soil emissions ($f_{E(z)(i)}$) to streams and the sum of the
40 instream transportation of nutrients from cell (*i*) to all downstream freshwater receptor grid cells
41 (*j*) ($f_{E(i,j)}$ dimensionless), divided by the rate of persistence in water in receptor cell *j* ($\lambda_{E(j)}$, year-
42 1); the ultimate downstream receptor cell *j* is the mouth (M) of the river system. ($FF_{E(fw)(i,j)}$)²:

$$43 \quad FF_{soils E (fw)(z)(i)} = f_{E(z)(i)} \sum_j FF_{River E(fw)(i,j)} = f_{E(z)(i)} \sum_{j=i}^M \frac{f_{E(i,j)}}{\lambda_{E(fw)(j)}}$$

44 All grid cells downstream of emission cell (*i*) are receptor cells (*j*) within freshwater environments.
45 Therefore, for each downstream receptor cell (*l* being the receptor grid cell of focus) the fraction of
46 downstream removal is multiplied by the product of all fractions of nutrients transported downstream
47 ($1 - R_{E_l}$; *R* = retention) to the first upstream cell of receptor cell (*j*) (when the emission cell is
48 also the receptor cell $f_{E(fw)(z)(i,i)} = 1$):

$$49 \quad f_{E(fw)(i,j)} = \prod_{l=i}^{j-1} (1 - R_{E_l})$$

50 For marine receptors (LME), the cumulative fate factor is not considered and is the product of the
51 fraction of soil emission $f_{E(z)(i)}$ and all subsequent instream transportation $f_{E(i,j)}$ from an emission in
52 cell (*i*) until its receptor marine cell (*j*):

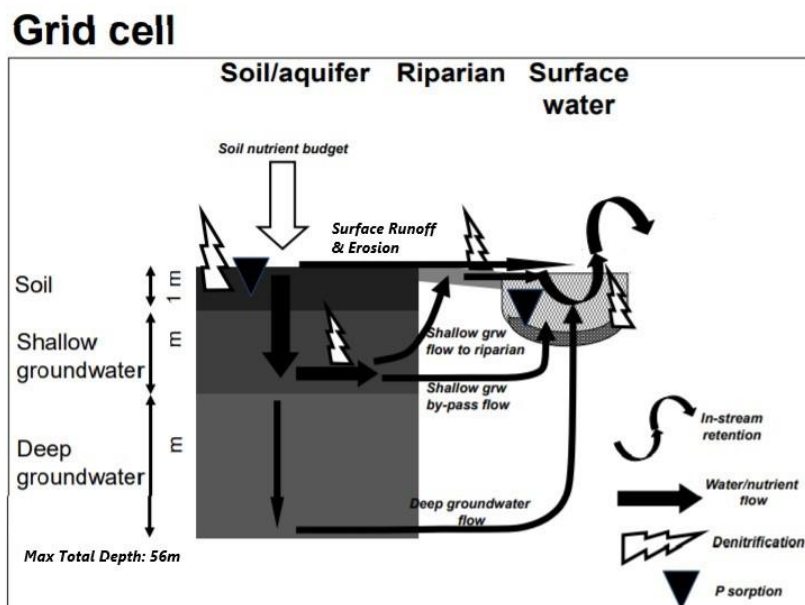
$$53 \quad FF_{soils E (LME)(z)(i)} = f_{E(z)(i)} \frac{f_{E(i,j)}}{\lambda_{E(LME)(j)}}$$

54 I.2 Nutrient Emission Fraction in Soil ($f_{E(z)(i)}$), and water ($f_{E(i,j)}$)

55 The IMAGE-GNM model is the basis for estimating the nutrient emission fraction in soil ($f_{E(z)(i)}$) and
56 water ($f_{E(i,j)}$) compartments of a cell. Studies have identified IMAGE-GNM as the most comprehensive
57 option to develop FFs from soils^{3,4}. IMAGE-GNM provides predictive advantages through detailed
58 modelling of land fate processes rather than previous watershed lumped regression models (e.g. Global
59 NEWS 2⁵), which have been used to develop previous FFs⁶. Additionally, IMAGE-GNM runs at the

60 annual time-step which ties in with the preferred global and temporal scope of LCIA methodologies⁴;
 61 whereas, sub-annual (seasonal) temporal resolution models do not.

62 IMAGE-GNM model is coupled with PCR-GLOBWB⁷ (a global hydrological model) to estimate global
 63 and spatially-explicit nutrient delivery of N and P to freshwater systems, via point and diffusive sources,
 64 at the 30 arcmin resolution⁸. Figure 1 shows a schematic of nutrient emission pathways from soils to
 65 rivers within each grid cell of IMAGE-GNM. Each cell has a land and water compartment; land
 66 compartments consider three emission pathways z (surface water runoff, erosion, leaching). The
 67 fraction of emission reaching receiving waterbodies undergoes processes such as denitrification within
 68 the riparian zone, soils and aquifers. Within water compartments, nutrients are removed under
 69 biological and chemical processes relating to denitrification, sedimentation and uptake by aquatic plants
 70 before being transported downstream to the next cell through advection.



71
 72 Figure 1: Adapted schematic diagram from Beusen et al., (2015) of soil emission flows to rivers.

73 This study uses IMAGE-GNM equations that transport and reduce nutrients within cells ($f_{E(z)(t)}$) and
 74 between cells ($f_{E(i,j)}$, dimensionless) to devise emissions from agricultural soils (Table 1). Whilst
 75 IMAGE GNM is written in Python, the FF model presented here is entirely written in MATLAB. We
 76 updated datasets used within IMAGE-GNM, which are now available for the 5 arcmin resolution (e.g.
 77 soil data⁹, hydrographic information through PCR-GLOWBWB 2¹⁰, aquifer thickness¹¹, porosity and
 78 permeability¹²), to tie in with our preferred 5 arcmin resolution of this study. Where datasets could
 79 not be updated (i.e., nutrient data modelled by IMAGE-GNM) we disaggregated, through an equal
 80 average, IMAGE-GNM datasets from 30 arcmin to 5 arcmin resolution. The datasets can be found in
 81 Appendix I. The IMAGE-GNM equations that support the derived export fractions are described

82 below and further details on the IMAGE-GNM methodology are presented by Beusen et al., (2015)⁸.
 83 All reduction factors can be found in Appendix 2.

84 **Table 1: Inland Fate equations for N and P in receptor j.**

Emission Route	Removal Fraction [-]	Derived equation for exported fraction*
Surface Runoff	$f_{Surface\ E\ (i)}$	$f_{qrs0} \times R_{Subgrid}$
Erodibility	$f_{Erosion\ E\ (i)}$	$f_{ero} \times R_{Subgrid}$
Leaching – Shallow Groundwater flow to riparian	$f_{Leach,Shallow\ E\ (i)}$	$f_{leach,soil} \times f_{shallow,gw} \times f_{non-bypass} \times (1 - f_{den,rip}) \times R_{Subgrid}$
Leaching – Shallow Groundwater by-pass flow	$f_{Leach,Shallow\ E\ (i)}$	$f_{leach,soil} \times f_{shallow,gw} \times f_{bypass} \times f_{N,TS/SS} \times R_{Subgrid}$
Leaching – Deep Groundwater	$f_{Leach,Deep\ E\ (i)}$	$f_{leach,soil} \times f_{deep,gw} \times f_{N,TS/SS} \times R_{Subgrid}$
Leaching - Submarine Shallow Groundwater Discharge	$f_{Shallow,SGD\ (i)}$	$f_{leach,soil} \times f_{shallow,gw} \times f_{Shallow,SGD} \times R_{Subgrid}$
Leaching - Submarine Deep Groundwater Discharge	$f_{Deep,SGD\ (i)}$	$f_{leach,soil} \times f_{deep,gw} \times f_{Deep,SGD} \times f_{N,TS/SS} \times R_{Subgrid}$

85 * $f_{qrs0}, f_{ero}, f_{leach,soil}$ relate to export fractions from surface water, erosion, and leaching; $f_{shallow,gw}, f_{deep,gw}$ indicate to
 86 export fractions within shallow and deep groundwater; $f_{shallow,SGD}, f_{Deep,SGD}$ represent the fraction emitted directly to
 87 marine environments through submarine groundwater discharge; $f_{N,TS/SS}$ represent the reduction or increase fraction
 88 caused by historical fertiliser leaching; $f_{den,rip}$ is the denitrification fraction with riparian zones; $R_{Subgrid}$ is the subgrid
 89 retention factor in rivers solely.

90 **1.2.1 Surface Runoff (f_{qrs0})**

91

$$92 \quad f_{qrs0} = f_{cal}(f_{qrs0}(texture)f_{qrs0}(slope)f_{qrs0}(landuse))$$

$$93 \quad f_{qrs0}(slope) = 1 - e^{-0.00617MAX[1,S]}$$

94 Where f_{qrs0} is the surface runoff factor, f_{cal} is a correction coefficient of 0.3¹³, S is the slope in m
 95 km⁻¹. f_{qrs0} is reduced by $f_{qrs0}(texture), f_{qrs0}(slope)$ and $f_{qrs0}(landuse)$, for which their values can
 96 be found in Appendix 2 and described in Beusen et al., (2015)⁸.

97 **1.2.2 Erodibility (f_{ero})**

98 The fraction of erodibility is determined by Cerdan et al., (2010)¹⁴ to estimate soil loss by rainfall
 99 erosion:

100

$$f_{ero} = f_{ero}(slope)f_{ero}(texture)$$

$$101 \quad f_{ero}(slope) = -1.5 + \frac{17}{1 + e^{2.3-6.1S}}$$

102 f_{ero} is the erodibility of soils influenced by the reduction factors of $f_{ero}(slope)$ and $f_{ero}(texture)$. S
 103 is the slope in degrees.

104 Global soil loss rates within IMAGE-GNM are estimated by adjusting soil erosion rates found within
 105 Europe based on slope, texture and land use¹⁴. Soil texture classes reduce or increase soil erodibility.
 106 Whilst coarse, very fine and peat soils have low erodibility of 0.5, medium and fine soil textures have
 107 increased erodibility. For the FF, slope and texture is considered within the FF whilst land use is
 108 considered within the emission inventory of an LCIA methodology. This is to ensure consistency of
 109 units within the LCIA method, whereby unit mass is considered within the emission inventory whilst
 110 time is considered within the FF.

111 Nutrient loads from erosion are a proportion of the total soil loss. For P, the IMAGE-GNM model
 112 keeps track of all inputs and outputs in the soil P budget. Hence historical tracking of P available in the
 113 soil is tracked from 1900, with initial stocks taken from Yang et al., (2013)¹⁵. We used soil P contents
 114 taken from 2000 within the IMAGE-GNM model. The soil P content for the year 2000 is reduced by
 115 subtracting the average nutrient balance for the year 1999 and 2000 and adding the average agricultural
 116 soil loss for 1999 and 2000; both taken from IMAGE GNM model results. This allowed us to obtain
 117 an initial P soil content to devise our erosional soil for specific crops within the main paper. N soil loss
 118 is solely related to soil loss through soil organic carbon (SOC) and N ratios.

119 For Phosphorus:

$$120 \quad V_{Soilloss} = \frac{0.01Soilloss(Landuse)}{BULK}$$

121 Where $V_{Soilloss}$ is the volume of soil loss in cubic meter per hectare, $Soilloss(Landuse)$ is the soil
 122 loss factors as per IMAGE-GNM in tonne per km² (See Appendix 2 for $Soilloss(Landuse)$ factors)
 123 and BULK is the soil bulk density measured in tonne per cubic metre. 0.01 is a conversion factor
 124 from km² to ha.

$$125 \quad Density_{P\ soil} = Pcontent \times 0.001/Depth_{topsoil}$$

126 Where $Density_{P\ soil}$ is the P soil density in kg P/m³, $Pcontent$ is the remaining mass balance of
 127 fertiliser plus existing P in the soil in grams of P per m². $Depth_{topsoil}$ is the depth of topsoil which is
 128 0.3m. 0.001 converts grams of P per m² to kg P/m². Therefore, soilloss for P is:

$$129 \quad Soilloss_P = V_{Soilloss} \times Density_{P\ soil}$$

130 Where $Soilloss_P$ is the P soil loss in kg P per ha.

131 For Nitrogen:

$$132 \quad Soilloss_N = \frac{SOC \times Soilloss(Landuse) \times 0.01 \times 1000}{C:N(Landuse)}$$

133 Where $Soilloss_N$ is the N soil loss in Kg N per ha, SOC is the soil organic carbon content fraction,
 134 $C:N(Landuse)$ is the carbon to nitrogen ratio which varies for land use (see Appendix 2).

135 1.2.3 Leaching ($f_{leach,soil}$)

$$136 f_{leach,soil} = [1 - MIN[(f_{climate} + f_{text} + f_{drain} + f_{soc}),1]]f_{landuse}$$

137 The fraction of N lost by denitrification ($f_{den,soil}$) complements $f_{leach,soil}$ ($f_{den,soil} = 1 -$
 138 $f_{leach,soil}$). Leaching fraction is related to soil texture (f_{text}), aeration (f_{drain}) and soil organic carbon
 139 (C) content (f_{soc})¹⁶ (Appendix 2). Fine textured soils are more susceptible to reach and maintain
 140 anoxia, which favours denitrification as they are characterised by high capillary pressures and hold
 141 water more tightly than sandy soils. Denitrification rates tend to be higher in poorly drained soils¹⁷.
 142 The soil organic C content is used as a proxy for supply, which can have direct impact on the soil
 143 oxygen concentrations. $F_{landuse}$ is the land use effect on leaching whereby arable land has a value of 1,
 144 and grassland and natural vegetation has a value of 0.36¹⁸. The factor $f_{climate}$ combines the effects of
 145 temperature, water residence time, and NO_3 in the root zone on denitrification rates:

$$146 f_{climate} = f_K T_{RZ}$$

147 Where f_K is the temperature effect using the Arrhenius equation^{19,20}:

$$148 f_K = 7.94 \times 10^{12} \exp\left(\frac{-E_{a,d}}{RmK}\right)$$

149 Where $E_{a,d}$ is the activation energy (78430 J mol⁻¹), K the mean annual temperature (Kelvin) and Rm
 150 is the molar gas constant (8.3144 J mol⁻¹K⁻¹). T_{RZ} is calculated via:

$$151 T_{RZ} = \frac{AWC}{Q_{eff}}$$

152 Where AWC (m) is the available water capacity for the top 1m of soil and Q_{eff} is the total recharge
 153 rate (m/yr). AWC was taken from the IRSIC-WISE Soil database⁹ and Q_{eff} was calculated based on
 154 the PCR-GLOBWB 2¹⁰ Hydrological Model for precipitation input circa 2000.

155 1.2.4 Steady State Shallow Groundwater Fraction ($f_{SS\ shallow,gw}$)

156 The shallow groundwater fraction, $f_{shallow,gw}$, represents the steady state fraction of the total
 157 output from the shallow aquifer into surface water systems⁸:

$$158 f_{shallow,gw} = f_{Qint} \times DC_{shallow}$$

$$159 f_{Qint} = 1 - f_{Qgwb}$$

$$160 DC_{shallow} = \frac{1}{1 + k \times Tr}$$

161 Where $f_{Q_{int}}$ is the fraction of excess water delivered to the shallow aquifer derived from the partition
 162 ratio $f_{Q_{gwb}}$ of excess water flow between the shallow and deep aquifer. DC represents the delivery
 163 coefficient of nutrients under steady state assumptions, derived analytically for a homogenous and
 164 mixed system, where k represents the degradation rate coefficient and Tr represents the time of
 165 residence. The decay rate, k , is obtained via the half-life of nitrate ($dt50_{den}$) due to denitrification²¹:

$$166 \quad k = \frac{\ln(2)}{dt50_{den}}$$

167 Siliciclastic material exhibits low $dt50_{den}$ values of 1yr^{-1} , whereas alluvial material has a $dt50_{den}$ value
 168 of 2yr^{-1} and all other lithology classes have a $dt50_{den}$ value of 5yr^{-1} . Time of residence, T_r (years), is
 169 derived by estimating the (D) depth to groundwater and nitrate velocity. Global groundwater table
 170 depths are estimated by de Graff et al., (2017)¹¹ and implemented in PCR-GLBWB 2 at the 5 arc min:

$$171 \quad T_r = \text{MIN} \left[\frac{pD}{R}, 1000 \right]$$

172 Where R is the recharge rate (m year^{-1}) and p is the effective porosity (dimensionless). Global recharge
 173 rates are estimated by the PCR-GLOBWB 2¹⁰. Within IMAGE-GNM, the shallow aquifer is
 174 represented by a 5m thick layer and is subject to denitrification during transportation along the various
 175 flow paths in a homogenous and isotropic aquifer.

176 1.2.5 Steady State Deep Groundwater Fraction ($f_{SS\text{ deep},gw}$)

177 The deep groundwater fraction again represents the steady state input of nutrients from the deep
 178 groundwater into surface water systems from the total balance of nitrogen left on the field as within
 179 IMAGE-GNM model:

$$180 \quad f_{deep,gw} = f_{Q_{gwb}} \times DC_{shallow} \times DC_{deep}$$

181 Where DC_{deep} is the delivery coefficient for the deep groundwater aquifer. Denitrification is not
 182 considered to occur in the deep aquifer, therefore $DC_{deep} = 1$ ⁸.

183 1.2.6 Submarine Groundwater Discharge (SGD) ($f_{shallow,SGD}$, $f_{Deep,SGD}$)

184 SGD assumes a proportion of nitrogen load in groundwater is discharged directly to marine
 185 environments for cells adjacent to the coastline. The SGD fraction for shallow groundwater
 186 ($f_{shallow,SGD}$) is 0.1, whilst for deep groundwater ($f_{Deep,SGD}$) the value equals 1.0. SGD is considered
 187 on all cells within 0 and 60km of the coastline.

188 1.2.7 Riparian Zone Reduction Fraction ($f_{den,rip}$)

189 A fraction of the shallow groundwater N load travels through the riparian zone ($f_{non-bypass}$). This
 190 fraction is determined by the fraction of the cell without a waterbody. The fraction with a waterbody

191 represents the bypass fraction of the shallow groundwater N load that discharges directly into rivers
192 (f_{bypass}).

193 Geochemical processes in the riparian zone require detailed hydrological and geographical information
194 at very high spatial scales. Even at 0.1km resolution the topography of the riparian area cannot be
195 adequately assessed. IMAGE-GNM therefore uses a conceptual approach whereby only the shallow
196 groundwater input to the riparian zones is subject to denitrification and depends on local pH,
197 temperature, water saturation, NO₃ availability and soil organic carbon availability^{22,23}. As with soil
198 denitrification, riparian zone denitrification is calculated using the dimensionless reduction factor:

$$199 \quad f_{den,rip} = MIN[(f_{climate} + f_{text} + f_{drain} + f_{soc}),1]f_{denpH,rip}$$

200 Where $f_{climate}$ is the product of f_K , temperature effect, water and NO₃ travel time through the
201 riparian zone ($T_{r,rip}$). $T_{r,rip}$ depends on the thickness of the riparian zone (D_{rip}), available water capacity
202 for the top 1m of the riparian zone ($tawc$) and on the flow of water entering the riparian zone from
203 the shallow groundwater aquifer (q_{int}):

$$204 \quad T_{r,rip} = \frac{D_{rip}tawc}{q_{int}}$$

205 1.2.8 Historical Fertiliser Transient State Fraction ($f_{N,TS/SS}$)

206 For the inclusion of historical transient state flows (i.e., historically varying fertiliser inputs) we used
207 IMAGE-GNM historical nutrient inputs disaggregated from 30 arcmin to 5 arcmin resolution.

208 In transient state flows, the vertical flow distribution for the shallow system is uniform, therefore
209 travel time is equal to mean travel time⁸. However, travel times for lateral flows to fluvial systems vary
210 considerably. The travel time distribution for lateral flows is represented through the age of the
211 groundwater at a specific depth:

$$212 \quad g_{age}(z) = -Tr \times \ln \left(1 - \frac{z}{D}\right)$$

213 Where g_{age} (yr) is the age of groundwater at a specific depth (z). The effects of denitrification in N
214 leaching load at time t and depth z ($L_N(t, z)$) is represented through a first order degradation
215 reaction and an exponential decay equation:

$$216 \quad L_{N,TS,River} = L_N(t - g_{age}(z), 0)e^{-k g_{age}(z)}$$

217 Where t is time and k is the decay rate as described in steady state.

218 To develop a reduction or increase fraction that represents the varying historical fertiliser inputs, we
219 divided total transient state loads by steady state loads for each grid cell:

220
$$f_{N,TS/SS} = \frac{L_{N,TS,River/Marine}}{L_{N,SS,River/Marine}}$$

221 **1.2.9 In-Stream Nutrient Retention (R) and Subgrid Retention ($R_{Subgrid}$)**

222 The process of denitrification, sedimentation and uptake by aquatic plants contribute to N retention.
 223 Denitrification is generally the major component of N retention²⁴. P is removed by sedimentation and
 224 sorption by sediment²⁵. The IMAGE-GNM calculates retention as a first order approximation
 225 according to:

226
$$R = 1 - \exp\left(\frac{v_{f,E}}{H_L}\right)$$

227 Where R is the fraction of nutrient load removed, $v_{f,E}$ is the net uptake velocity (m yr⁻¹) relating to
 228 nutrient E (N or P), and H_L is the hydraulic load (m yr⁻¹) obtained from:

229
$$H_L = \frac{D}{\tau}$$

230 Where D is the depth of waterbody (m), τ is the residence time (yr) calculated by:

231
$$\tau = \frac{V}{Q}$$

232 V is the waterbody volume (m³) and Q is the discharge (m³ yr⁻¹) for all waterbodies, except for
 233 river channels and floodplains where the discharge Q is reduced by the water volume in the
 234 floodplains (Q_f):

235
$$\tau = \frac{V}{Q - Q_f}$$

236 The Hydraulic Load (H_L) relates to removal through hydrological processes and the net uptake velocity
 237 ($v_{f,E}$), relates to removal from biological and chemical processes. Net uptake velocity is different for
 238 each element E (N or P). For N, the basic value for all water body types is 35m yr⁻¹, modified by
 239 temperature and N concentration²⁶.

240
$$v_{f,N} = 35 f(t) f(C_N)$$

241 Where t is annual mean temperature (°C) and C_N is the N concentration in the water. $f(C_N)$ describes
 242 the effect of concentration on denitrification as a result of electron donor limitation in the case of high
 243 N loads. We interpolated, as conducted by Beusen et al., (2015), whereby $f(C_N)$ is 7.2 for $C_N = 0.0001$
 244 mg L⁻¹ and $f(C_N)$ is 1 for $C_N = 1$ mg L⁻¹, a further decrease to 0.37 $f(C_N)$ is for $C_N = 100$ mg L⁻¹.
 245 N concentration in water is taken from IMAGE-GNM and equally disaggregated from 30arcmin to 5
 246 arcmin.

247 A temperature effect $f(t)$ is used in the calculation of v_{fE} formed by:

$$248 \quad f(t) = \alpha^{t-20}(t - 20)$$

249 Where $\alpha = 1.0717$ for N²⁷ and $\alpha = 1.06$ for P²⁸. For P the basic value for v_f is 44.5 myr⁻¹ and again
250 this is modified based on the temperature effect²⁸.

251 The subgrid retention fraction ($R_{Subgrid}$) is considered before retention at the grid cell level for all cells
252 which do not have a lake or reservoir. Here we follow the parameterization method of lower-order
253 streams following the approach by Wolheim et al., (2006)²⁶ and presented by Beusen et al., (2015)⁸.
254 Subgrid retention fraction removes nutrient load from rivers below stream orders of 6 found within
255 each grid cell that is not a lake or reservoir. Once subgrid retention is removed, grid cell retention
256 and global load accumulation is devised. The final subgrid retention fraction ($R_{Subgrid}$) of stream
257 orders 1 – 6, is the difference between original input load for the first order stream minus the input
258 from the sixth order stream:

$$259 \quad R_{Subgrid} = \frac{(Load_{in}^{1st} - Load_{in}^{6th})}{Load_{in}^{1st}}$$

260 1.3 Persistence Rate, λ

261 The persistence in a freshwater grid cell j or a Large Marine Ecosystem (LME) is the inverse sum of
262 removal rates by hydrological (advection, $\lambda_{Adv E(j)}$) and biological and chemical processes
263 ($\lambda_{Rent,E(j)}$):

$$264 \quad \lambda_{E(j)} = \frac{1}{\lambda_{Adv E(j)} + \lambda_{Rent,E(j)}}$$

265 For freshwater environments, λ_{Adv} corresponds to the residence time τ of the hydrological load,
266 which is formed by dividing the volume V (m³) of the water body by the discharge Q (m³yr⁻¹), taken
267 from PCR-GLOBWB 2¹⁰ for freshwater environments. For LMEs, Cosme et al., (2018)⁶ provides water
268 residence times for 66 LMEs devised from a literature review.

269 The persistence of net uptake velocities relating to biological and chemical processes (λ_{Rent}) in
270 freshwater is calculated by dividing the net uptake velocity v_{fE} (myr⁻¹) by the depth D (m) of the water
271 body (rivers, lakes and reservoirs).

$$272 \quad \lambda_{Rent} = \frac{v_{fE}}{D}$$

273 For LMEs, nitrogen denitrification rates vary with geography and over time²⁹. This study used Cosme
274 et al., (2018)⁶ results via a modelling approach to devise spatially variable denitrification rates across
275 the 66 LMEs. The effect of time and space may be represented through an empirical relationship

276 between nitrogen removal as a function of water residence time (months) in estuaries, river reaches,
 277 lakes, and continental shelf:

$$278 \quad DIN_{rem,k} = 23.4 \times \tau \tau_l^{0.204}$$

279 The LME(*l*) – dependent denitrification rate constant ($\lambda_{denitr,l}$) is determined using a first order
 280 removal equation, with *t* set to 1 year for the annual time integration, as per Cosme et al., (2018)⁶:

$$281 \quad DIN_{rem,l} = e^{-\lambda_{denitr,l} \times t} \Leftrightarrow \lambda_{denitr,l} = -\frac{\ln DIN_{rem,l}}{t}$$

282 1.4 Assumptions

283 **Groundwater** – IMAGE-GNM assumes a constant groundwater thickness for the shallow aquifer
 284 ($D_{sgrw} = 5m$) and for the deep aquifer ($D_{dgrw} = 50m$) following Meirnardi, (1994)³⁰. In contrast, this
 285 study provides spatially-explicit groundwater thickness using data from PCR-GLOBWB 2¹⁰, following
 286 aquifer thickness and distance to groundwater table researched by de Graff et al., (2017)¹¹.

287 Under the assumptions of IMAGE-GNM, the soil profile exists for 1 meter below the ground level⁸.
 288 The shallow groundwater aquifer exists up to 6 meters below the ground surface. Therefore, we
 289 assume a shallow aquifer is present at depths between 0 - 6 meters below ground level:

$$290 \quad D_{sgrw} = \max(0.0, 6 - d_{gwT})$$

291 The deep aquifer lies between 6 and 56 meters below ground. The deep aquifer thickness is the
 292 difference between the overall ground thickness (D_{gw}) and the shallow aquifer thickness.

$$293 \quad D_{dgrw} = \min(56, D_{gw} - D_{sgrw})$$

294 If the depth to groundwater is greater than 6 meters, only the deep groundwater aquifer exists. If the
 295 depth to groundwater table is greater than 56 meters, neither the shallow nor deep aquifer exists, and
 296 nutrients are assumed to be stored within the vadose zone. For continuity, regions where the deep
 297 aquifer exists, we assume all nutrients flow towards surface water bodies.

298 1.5 Aggregation:

299 Individual cell (i) FFs can be aggregated to larger spatial resolution (e.g., country, basin) to meet
 300 research requirements and address data and model constraints. Here we aggregated our FF data based
 301 on IMAGE-GNM diffusive emissions (DE) of N and P from all soils (arable, grassland and natural land)
 302 using the following equation:

$$303 \quad FF_{i,LSR} = \frac{\sum_{LSR} (FF_{i,LSR} \times DE_{i,LSR})}{\sum_{LSR} DE_{i,LSR}}$$

304 We provide resampled diffusive emissions from IMAGE-GNM and boundary shape files to preform
305 aggregation of the FF to larger spatial resolutions.

306 1.6 Fate Factor Model Emissions Validation

307 For the comparison of the IMAGE-GNM model against our FF model for emissions, we used the
308 percentage root mean squared error (PRMSE) as per Beusen et al., (2015)⁸. PRSME is calculated as:

$$309 \quad PRMSE = \frac{100}{\bar{O}} \sqrt{\frac{\sum_{i=1}^n (O_i - M_i)^2}{n}}$$

310 Where \bar{O} is the mean of the IMAGE-GNM results, O_i is the IMAGE-GNM result at cell i , M_i is the FF
311 model simulated result at cell i . n is the number of grid cells with values greater than 0. As per Beusen
312 et al., (2015), we consider PRSME values of less than 50% acceptable in view of the global scale of the
313 model⁸.

314 To validate our FF emission fractions against IMAGE-GNM model results we calculated the total
315 emission of N & P via each emission pathway using IMAGE-GNM nutrient inputs. To allow for
316 comparability to IMAGE-GNM 30 arcmin resolution, the simulated 5 arcmin emissions were sum
317 aggregated to the 30 arcmin resolution. Where nutrient inputs required classification of landcover
318 (surface runoff and erosional process) we solely validated our emissions from arable (cropland) cells,
319 given our specific interest in crops for this study. Although we have additionally devised the FFs for
320 grasslands, natural lands and direct emissions, we have not validated these against IMAGE-GNM as it
321 is deemed out of scope for this study.

322 We compared simulated nutrient emissions to IMAGE-GNM nutrient emissions via:

- 323 1. Arable field emissions using IMAGE-GNM⁸ nutrient inputs and Earthstat^{31,32} nutrient inputs
324 for surface runoff, erosional soil loss and leaching.
- 325 2. Historical groundwater load and effects from denitrification in the riparian zone using
326 IMAGE-GNM⁸ historical nutrient inputs solely.
- 327 3. Accumulated load after retention in rivers, based on IMAGE-GNM nutrient loads found in
328 rivers after exportation from land cell compartments.

329 Points 2. and 3. used only IMAGE-GNM inputs as EarthStat data only provides arable nutrient inputs
330 for the year 2000, hence the historical groundwater load and accumulated retention river load are
331 incomparable.

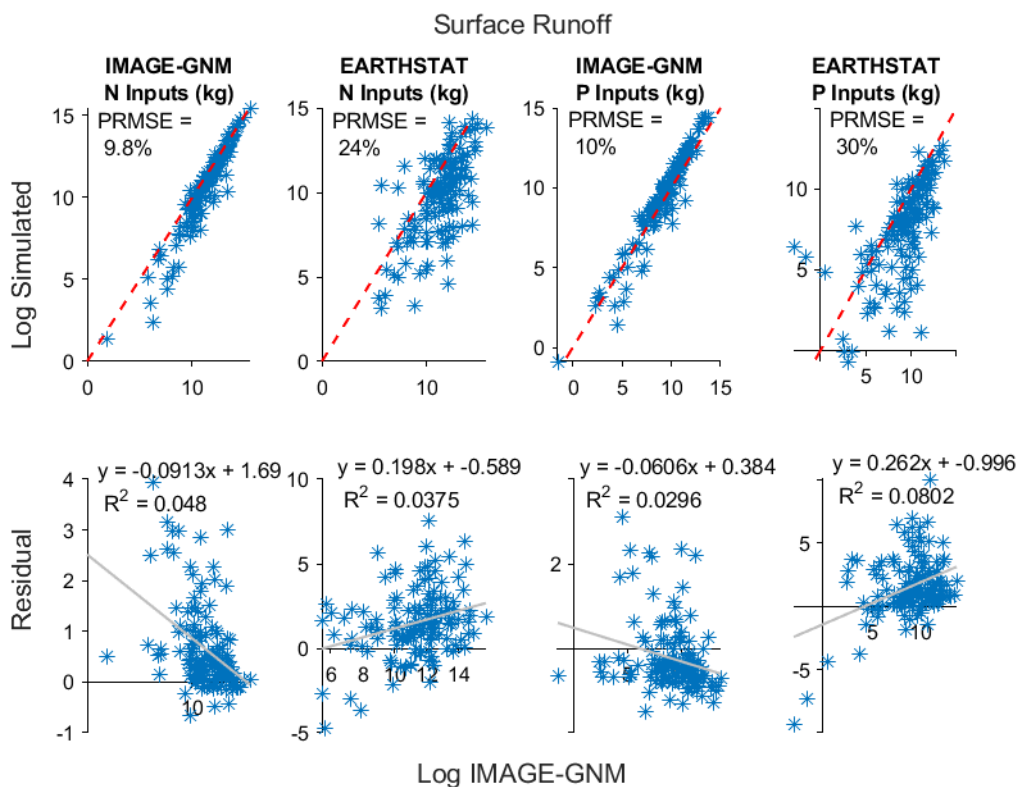
332 **2 Results**

333 **2.1 Comparison of FF Model Nutrient Loads against IMAGE GNM**

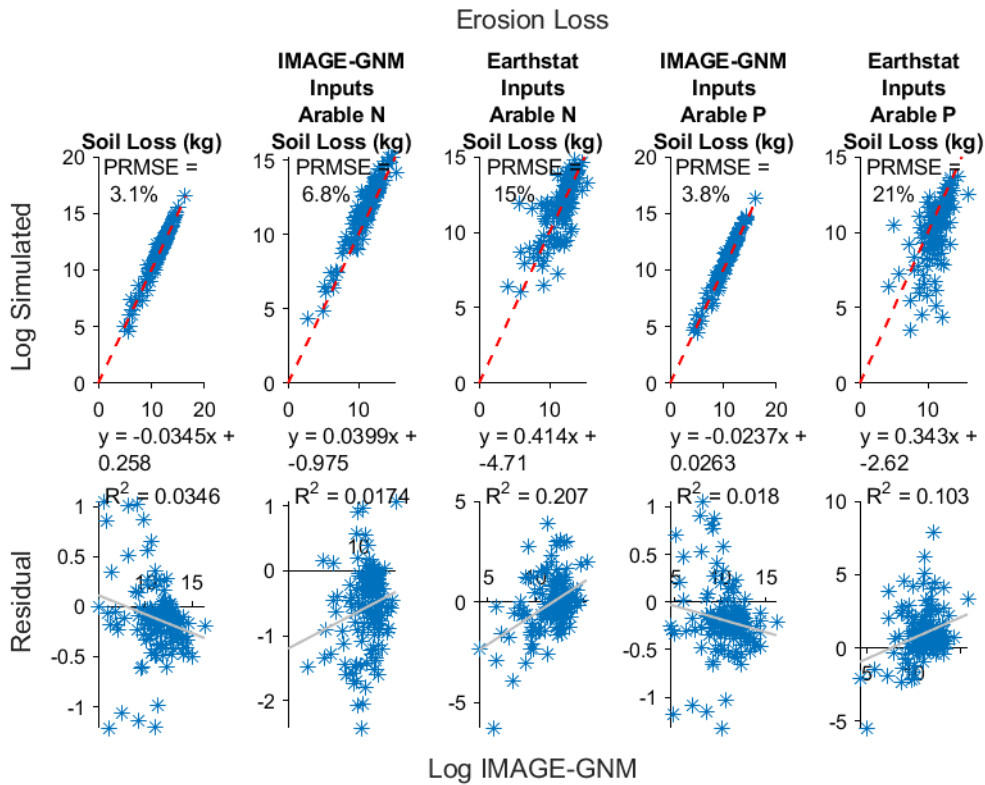
334 **2.1.1 Surface Runoff, Erosion and Leaching**

335 We compared simulated nutrient emissions to IMAGE-GNM model's nutrient emission outputs (figure
336 2 and table 2). We ran our simulation firstly using nutrient inputs from IMAGE-GNM solely and latterly
337 using nutrient inputs for the year 2000 from EarthStat^{31,32}. For pre-existing nutrients found in soils for
338 N and P in groundwater (under steady and transient state modelling), we used nutrient inputs taken
339 from IMAGE-GNM⁸.

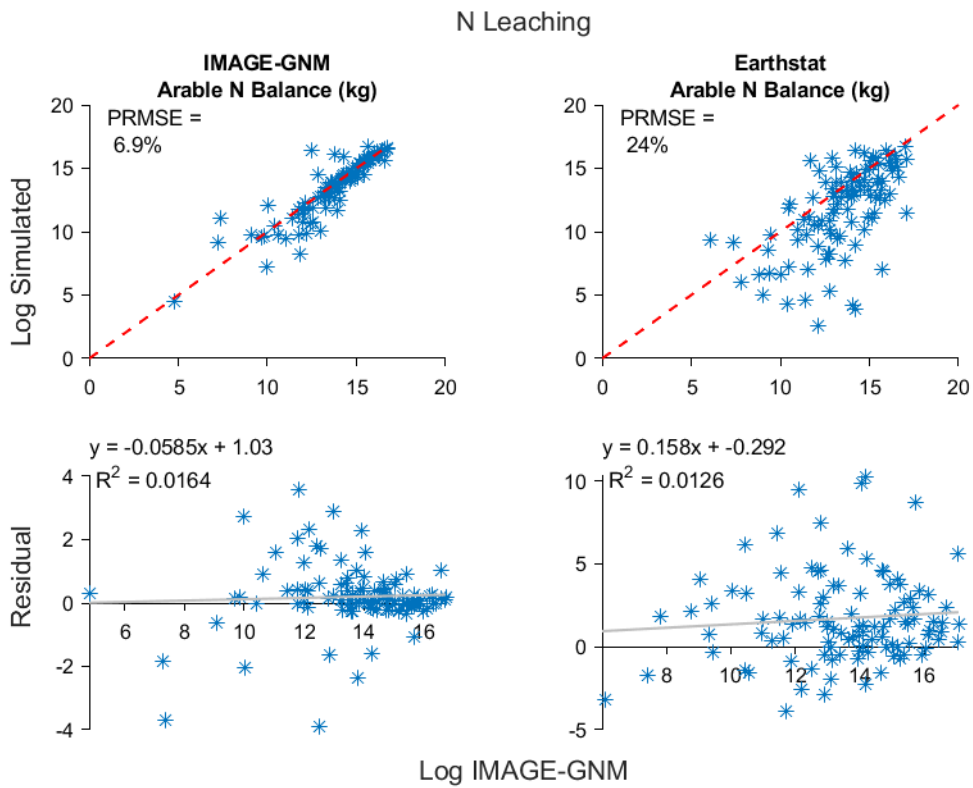
340 Using nutrient inputs from IMAGE-GNM, model performance for erosional nutrient loss showed the
341 best model similarity (PRMSE = 6.8% in N & 3.8% in P), followed by N leaching (PRSMSE = 6.9%) and
342 surface runoff (PRSMSE = 9.8% in N & 10% in P). Erosion for N & P showed very slight overestimation
343 noted in the residual plots clustering below 0. We attributed this slight overestimation due to an
344 increased estimation of soil loss. Using nutrient inputs from Earthstat, all pathways (surface runoff,
345 erosional and leaching) PRMSE performance ranged between 15% and 30%. Naturally, PRMSE values
346 are higher using nutrient input data from Earthstat than IMAGE GNM given the majority of datasets
347 (both nutrient input and model parameter datasets) are from improved data sources at the 5 arcmin
348 resolution. Using Earthstat inputs, surface water runoff and erosion showed underestimation
349 particularly for lower values of N and P. Slight under estimation in leaching is identified with no clear
350 trend.



351 a.



352 b.



353 c.

354 Figure 2: Top graphs for each pathway (a. runoff, b. erosion, c. N leaching) represent the global comparison of log (simulated)
 355 and log (IMAGE GNM modelled) outputs with 1:1 lines. The bottom graph for each pathway represents global residual plots
 356 (log(simulated) minus log (IMAGE GNM modelled)) vs. (log(IMAGE GNM modelled)) with regression lines.

357 Comparing the global totals (table 2.), total nutrient outputs to rivers compared to total nutrient
 358 inputs to fields ratios (TN_{out}/TN_{in} & TP_{OUT}/TP_{IN}) showed an increase in N and reduction in P ratios
 359 using Earthstat nutrient inputs when compared to IMAGE GNM inputs. In comparing the individual
 360 pathways, reduced ratios were seen for surface runoff, however, increased ratios were identified for
 361 erosional and leaching pathways for N and P. Ratio differences are due to differences in crop numbers,
 362 global crop area, reduced nutrient inputs and variations in the local environmental characteristics
 363 found at the 5 arcmin resolution compared to the 30 arcmin.

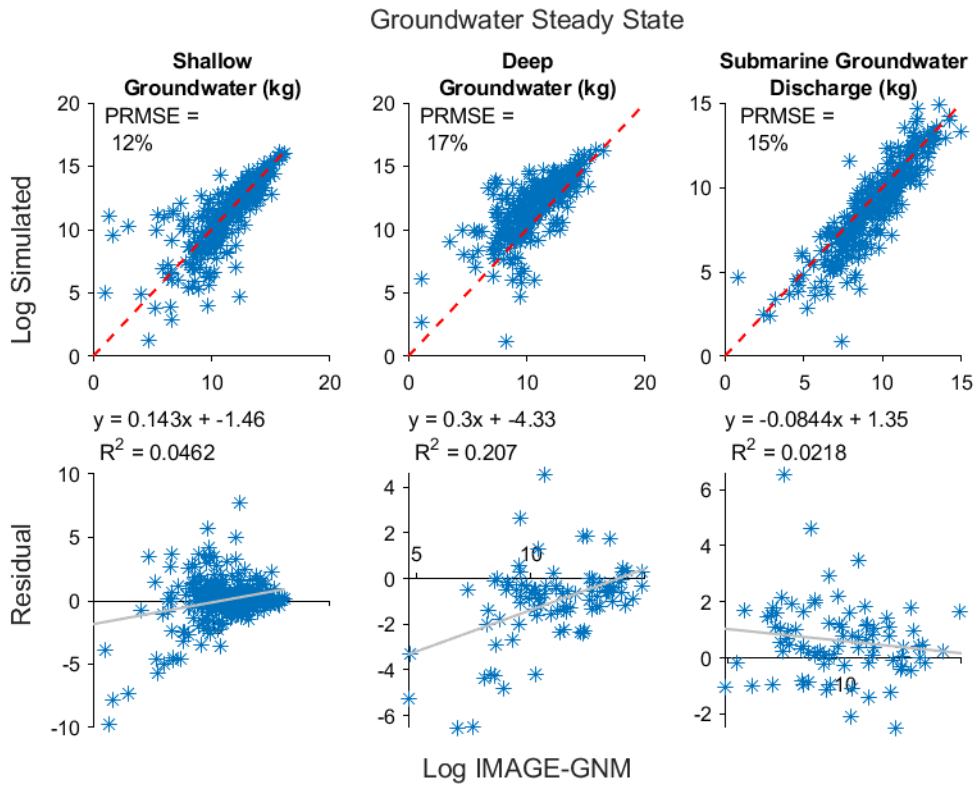
364 Leaching to rivers identifies the quantity of leached N that is delivered to rivers through groundwater.
 365 Here the delivery fractions through the shallow, deep and submarine groundwater discharge (SGD)
 366 are used to estimate the load reaching rivers. We also applied a Transient State/Steady State (TS/SS)
 367 fraction to include the implications of historical fertiliser inputs. Our estimates of leaching to rivers
 368 are slightly higher than in IMAGE GNM. We attribute this to varying groundwater levels which reduce
 369 the time of residence and hence denitrification of nitrogen in the groundwater system. Additionally,
 370 we identified much higher porosity values using Gleeson et al., (2014)¹², which transfers greater
 371 quantities of nitrogen to the deep groundwater where denitrification does not occur, increasing
 372 delivery to rivers over time. Hence, we identified greater deep groundwater loads and higher SGD
 373 values (table 3).

374 Table 2: Represents global total comparisons for simulated results against IMAGE GNM modelled results. Here we sum the
 375 global total inputs and outputs for arable soils during the year 2000. Inputs represent nutrient inputs added to the fields and
 376 are not already existing on the field. Nutrient outputs represent the emission from the field edge (i.e. does not consider
 377 historical groundwater).

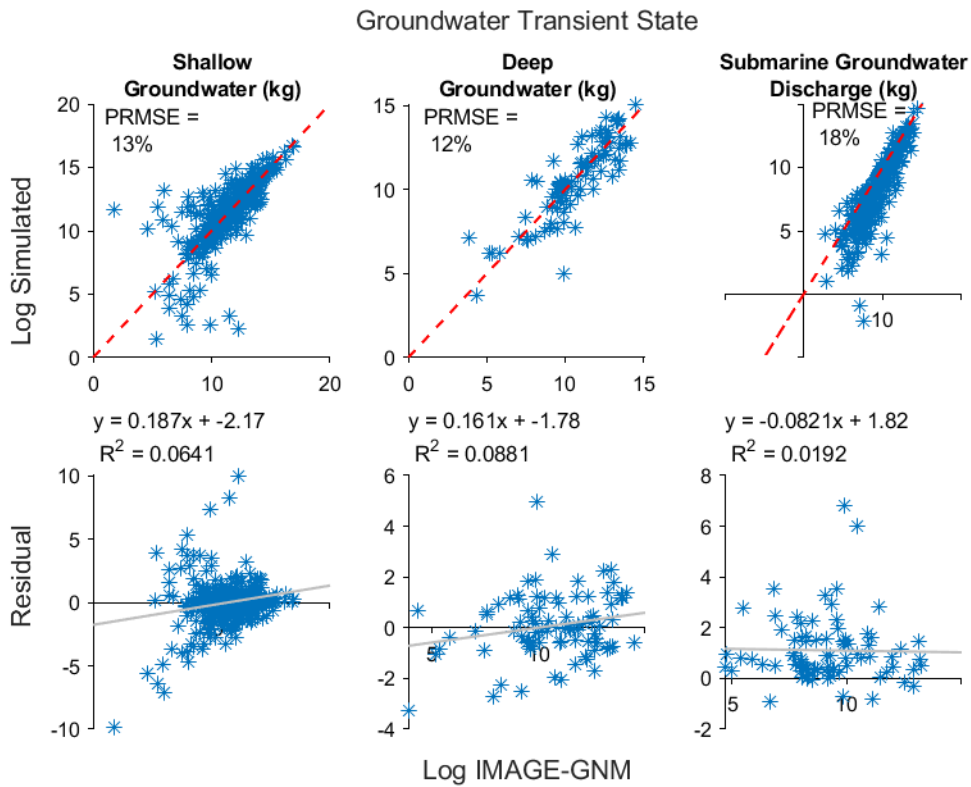
	IMAGE GNM Global Nutrient Totals for Land Pathways reaching rivers from for arable soils					This Study's Global Nutrient Totals for Land Pathways reaching rivers from for arable soils. Using data from Earthstat Nutrient Input data for 2000 and historical data from IMAGE GNM				
Time period	2000					2000				
Number of crops	18					17				
Resolution	30 Arcmin					5 Arcmin				
Global Crop Area in 2000 (ha)	1.52E+09					8.58E+08				
All values are circa 2000	Surface Water Runoff	Erosion	Leaching to groundwater	Leaching to Rivers	Total	Surface Water Runoff	Erosion	Leaching to groundwater	Leaching to Rivers	Total
Soil loss (Tg yr ⁻¹)	~	5.36	~	~		~	3.48	~	~	
Nitrogen										
TN IN (Tg N yr ⁻¹)	129.7	~	51.53	51.53	129.71	105.82	~	41.8	41.8	105.82
TN OUT (Tg N yr ⁻¹)	5.31	5.14	33.58	11.25	21.7	2.81	4.30	27.9	16.38	23.48
TN_{out}/TN_{in}	0.04	~	0.65	0.22	0.16	0.03	~	0.68	0.39	0.22
Phosphorus										
TP IN (Tg P yr ⁻¹)	19.8	3745	~	~	19.8	17.24	2025.4	~	~	17.24
TP OUT (Tg N yr ⁻¹)	0.88	3.27	~	~	4.15	0.51	2.18	~	~	2.69
TP_{OUT}/TP_{IN}	0.04	0.000872	~	~	0.21	0.03	0.0011	~	~	0.16

378

379 2.1.2 Groundwater Steady State and Transient State
 380 N Groundwater under steady state (SS) and transient state (TS) had PRMSE values ranging between
 381 10% and 17% for the shallow, deep and SGD (figure 3).



382

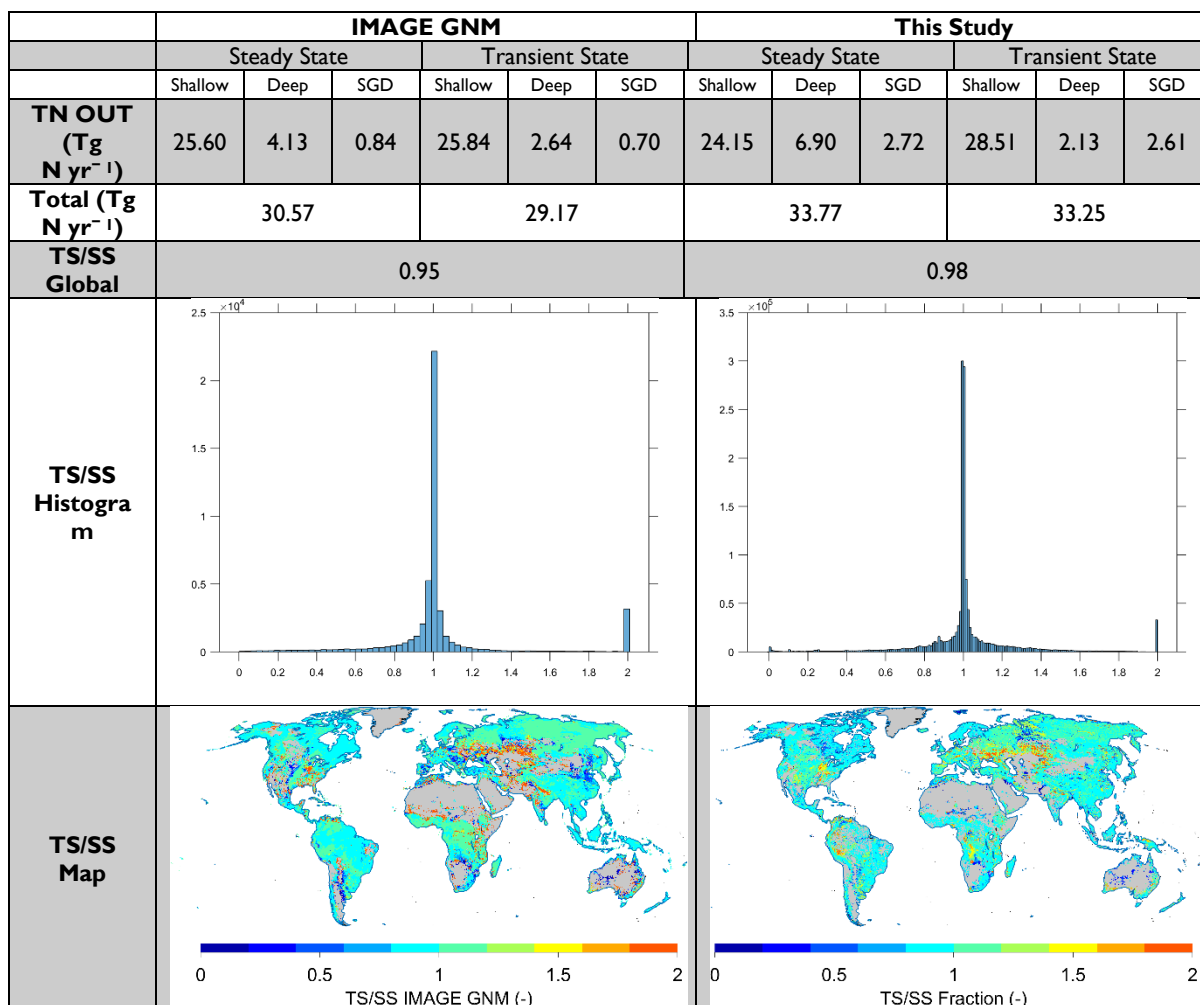


383

384 Figure 3: Top graphs for groundwater under steady (a.) and transient (b.) state assumptions represent the global comparison
 385 of log(simulated) and log(IMAGE GNM modelled) outputs with 1:1 lines. The bottom graph for each pathway represents
 386 global residual plots (log(simulated) minus log (IMAGE GNM modelled)) vs. (log(IMAGE GNM modelled)) with regression
 387 lines.

388 For all landcovers, the total N groundwater flow reaching streams and marine environments is 33.25
 389 Tg, whilst in IMAGE the total quantity was 29.17 Tg under TS (table 3). This is to be expected as
 390 varying groundwater depth reduces the quantity of denitrification occurring in the shallow zone, hence
 391 increasing the nutrient outflow to rivers. TS showed a very slight reduction in the global total
 392 compared to SS in both IMAGE GNM and this study. This is due to varying historical fertiliser use
 393 where some regions have increased fertiliser use and others have decreased use over time. We
 394 devised a historical fertiliser factor by dividing the TS by SS to reduce or increase N loads to rivers
 395 depending on the historical load (table 3).

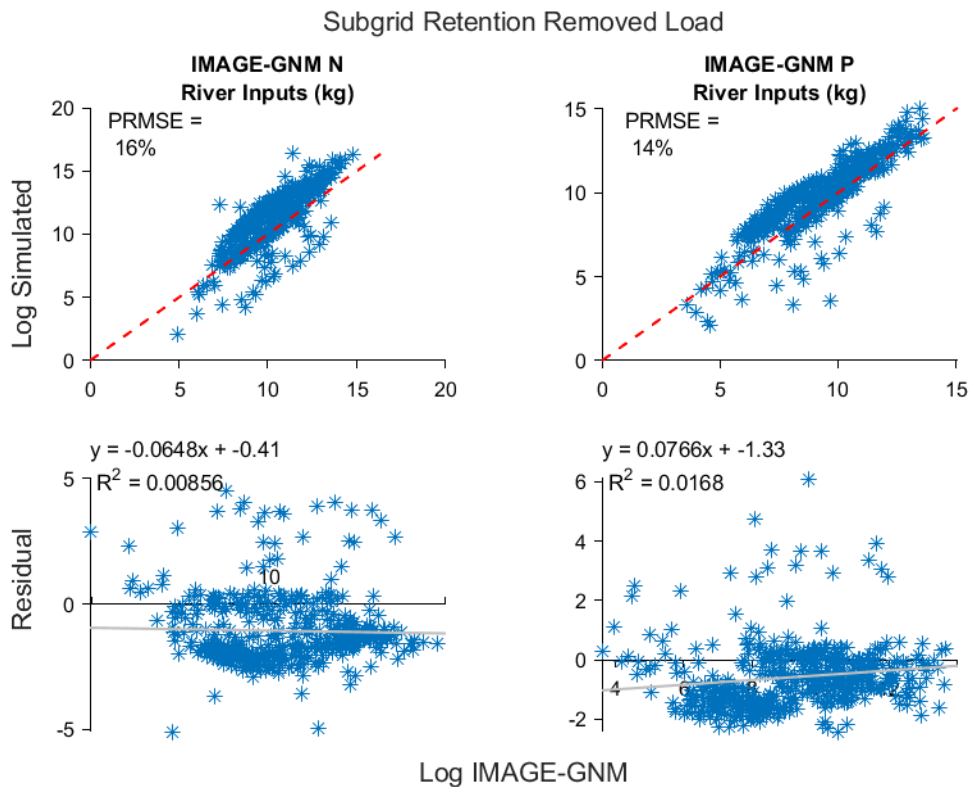
396 Table 3: Represents comparisons of the global total output of N load from groundwater for simulated results against IMAGE
 397 GNM modelled results under steady and transient states. N outputs represent the groundwater load reaching rivers from
 398 the field after leaching has occurred. Steady states consider nutrient leaching is constant over time whilst transient states
 399 consider the variable inputs of fertilisers through time. Nutrient inputs are taken solely from IMAGE GNM. For steady states
 400 N leaching inputs are from the year 2000. For transient states N leaching considers variable leaching rates dating back to
 401 1700. Denitrification is considered to remove all N leaching 50 years after N is added to soils. TS/SS represent the division
 402 of transient state loads by steady state loads to devise the reduction or increase fractions to account for spatial historical
 403 fertiliser input differences. TS/SS histogram shows some regions increase in TS/SS fractions whilst others decrease. We
 404 applied a maximum increase fraction of 2 due to model uncertainty within groundwater.



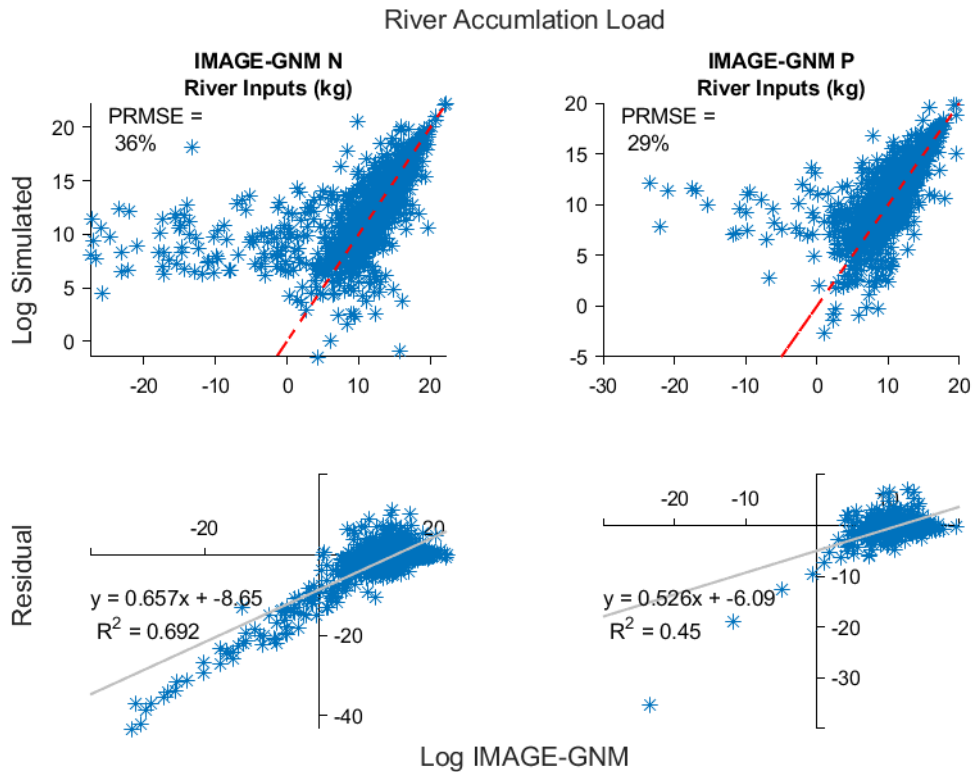
405

406 2.1.3 Subgrid Retention and Accumulated River Load after Grid Retention

407 The subgrid retention load showed model PRSME in N as 16% and in P 14%. Subgrid nutrient retention
408 load was slightly overestimated by our FF model (figure 4a.). Accumulated river nutrient load post
409 grid retention showed the least model similarity (PRSME = 36% for N and 29% for P) (figure 4b.).
410 Extreme overestimation was identified when IMAGE GNM predicts loads of less than 1kg. Spatially
411 these were identified over arid regions where differences in water fractions, water volumes and surface
412 water area at smaller resolutions impacted the hydraulic load; an input parameter to quantify retention.
413 Disregarding simulated accumulation loads that are below 1kg in IMAGE GNM we saw model
414 performance improved to PRSME = 14% for N and 17% for P (figure 4b). Hence, we recommend
415 caution for use of FF model results over arid regions.



416 a.



417 b.

418 Figure 4: Top graphs for subgrid retention removed load (a.) and river accumulation load (b.) representing the global
 419 comparison of log (simulated) and log (IMAGE GNM modelled) outputs with 1:1 lines. The bottom graph for retention loads
 420 represent global residual plots (log(simulated) minus log (IMAGE GNM modelled)) vs. (log(IMAGE GNM modelled)) with
 421 regression lines. Nutrient inputs are taken from IMAGE GNM solely for model validation.

422

423 2.2 Fate Factor Model Results

424 The summary of statistics of the spatial distribution of the fate factor at the global scale is presented
 425 in table 4. The Fate Factor (FF) results for all land covers (arable, grassland and natural) and soil
 426 emission pathways (surface runoff, erosion and leaching) are shown in Figure 5. The results discussed
 427 are for diffusive soil emissions solely for N and P to freshwater and N to marine environments.
 428 Although our results represent diffusive emissions, direct emissions to river and marine FFs can be
 429 found within the raster data files. Direct emissions had to be calculated as part of the diffusive emission
 430 calculation. FFs for all soils at the grid cell are a weighted average (weighted by nutrient emissions
 431 from IMAGE GNM⁸) of the land use fraction (e.g arable, grassland and natural).

432
433

Table 4: Statistics of distribution of fate factors (days) results for all emission routes to freshwater and marine environments

Statistics	Fate Factor (days) per nutrient and receptor waterbody type		
	Freshwater N	Freshwater P	Marine N
Minimum	9.80E-22	6.81E-22	5.11E-43
5th Percentile	7.10E-04	7.34E-04	1.12E-06
Mean	5.24	2.99	26.54
95th percentile	10.50	8.18	119.41
Maximum	70417.88	40126.06	3977.53
Spatial variability	174.03	68.13	96.68

434

435 Under the assumption of the limiting nutrient concept³³, we assessed the inflection point used by
 436 Helmes et al., (2012)² to detect the difference between longer and shorter FF persistence values for
 437 all land cover types (figure 5 b,c,d). The inflection point identifies emissions with short and long travel
 438 times to the river mouth or near marine ecosystem. Inflection for P and N in freshwater is 0.056 days
 439 and 0.084 days, respectively. For marine environments, N inflection increased to 5.45 days.

440

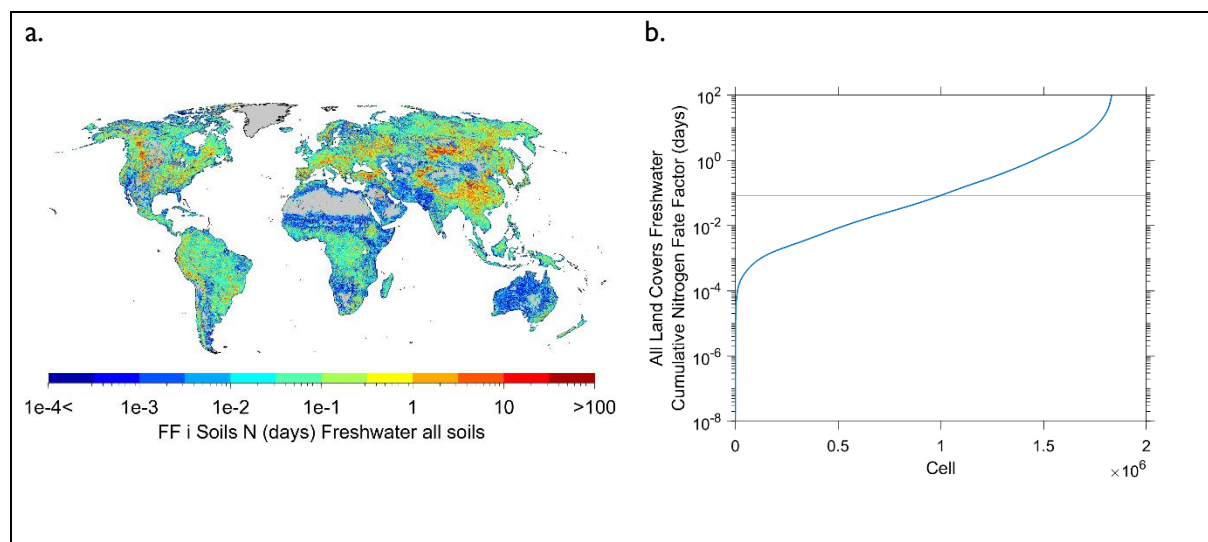
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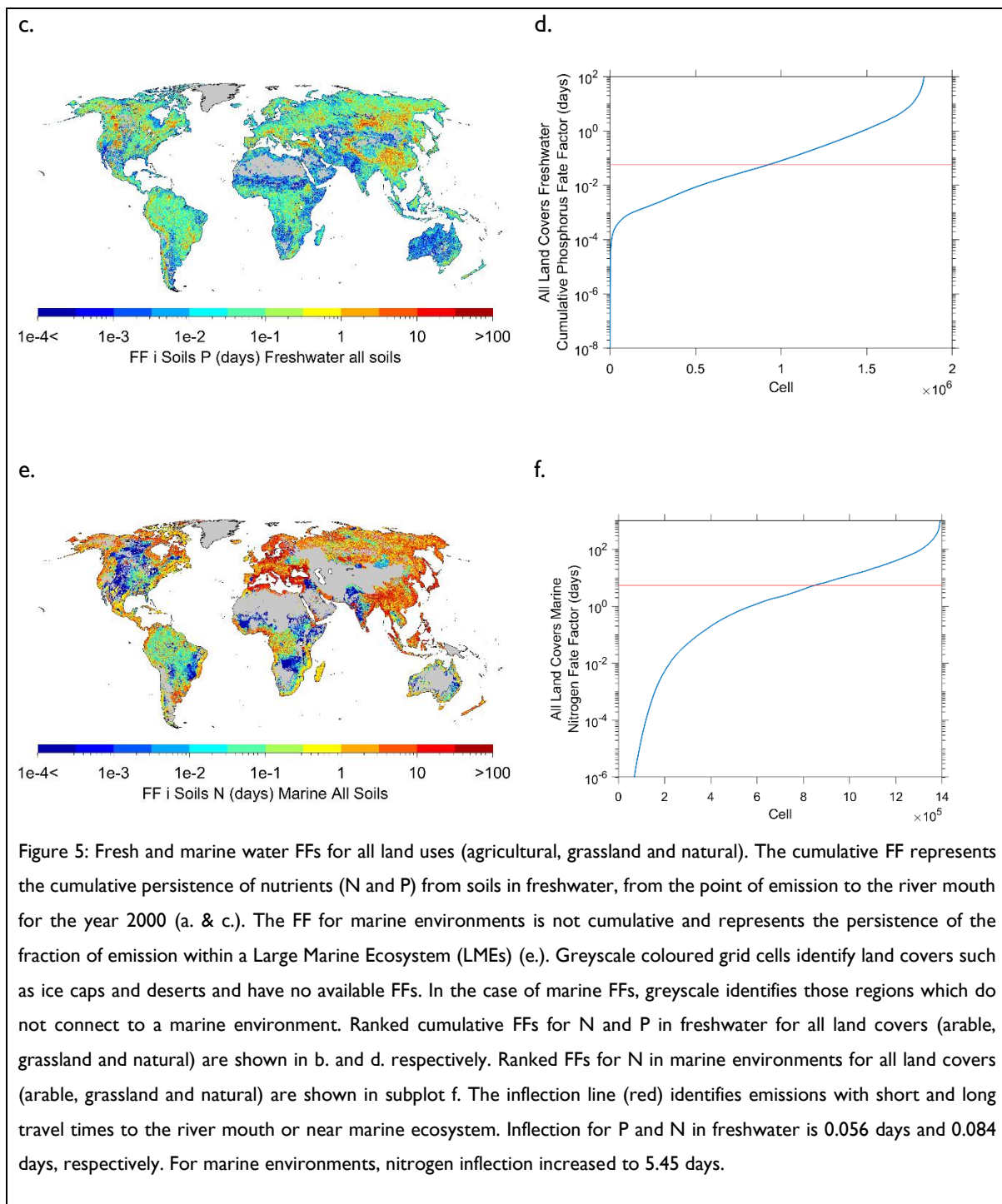
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447 2.3 Comparison with other Fate Factor studies

448 To implement a more robust comparison to previous studies, we compared the diffusive and direct
 449 emission FFs formulated in this study to previous research (table 5). Although direct emission FFs are
 450 not a focus of this study, direct emission FFs had to be formulated before estimating diffusive emission
 451 FFs.

452 Direct emission FFs were assumed to be within rivers of stream order six and above. Therefore,
 453 subgrid retention was not considered for direct emission FFs. We assumed that stream orders lower
 454 than six would be too small for effluent nutrient discharge. Additionally, we aggregated FFs by their
 455 respective emission inventories (diffusive emissions for soils and direct emissions to rivers). Emission
 456 inventory data for diffusive and direct were both taken from IMAGE-GNM⁸.

457 The FF model presented here, derives spatially explicit FFs for N and P at the global scale from soils
 458 and direct emissions using a single methodology at the 5 arcmin resolution. Until recently, global FFs
 459 had only been estimated individually for P in freshwater² and N in marine environments⁶. However,
 460 Payen et al., (2021) recently developed global FFs for N and P for freshwater environments at the
 461 basin scale³⁴. Additionally, Zhou et al., (2022) developed global FFs for N to freshwater for direct and
 462 diffusive emissions based on IMAGE GNM model³⁵. Here, our research is the first to derive
 463 comparable FFs to all previous global FFs, devised for N and P, in freshwater and marine environments,
 464 for direct and diffusive sources, within a single study (table 5).

465 Table 5: Globally aggregated weighted-emission Fate and Characterisation Factors. Land cover fractions are taken from
 466 PCR-GLOBWB 2 for the purpose of continuity for modelling within the supplementary material. Within the main paper we
 467 took land cover fractions from Monfreda et al., (2008)³⁶.

Fate Factors (days)

	Zhou et al., (2022) ³⁵		Payen et al (2021) ³⁴		Helmes et al. (2012) ²	Cosme et al. (2018) ⁶	This study					
Resolution	30 arcmin		Basin		30 arcmin	Basin	5 arcmin					
Emission Source	Diffusive (excluding erosions)	Direct	Soils (Diffusive Sources)	Point Sources	Point Sources	Diffusive	Direct	Arable Soils	Grassland Soils	Natural Soils	Diffusive (All Soils)	Direct
Marine (Nitrogen)	~	~	~			43.8	96	19.53	40.60	28.33	34.46	94.30
Freshwater (Phosphorus)			23	247	130			4.78	4.95	5.05	5.38	6.53
Freshwater (Nitrogen)	6.7*	29.3	125	257	~			8.42	6.43	6.30	7.40	23.25

468 * Note: diffusive weighted average. Just the average was provided across all cells for diffusive sources.

469 Our FF results were lower than all previously predicted FFs for direct and diffusive emission sources.
 470 For direct emissions, whilst N marine FFs were within a 5% range predicted by Cosme et al., (2018)⁶,
 471 N and P freshwater FFs were an order of magnitude lower than values predicted by Payen et al.,
 472 (2021)³⁴ and Helmes et al., (2012)². However, our results compared well and were within range with
 473 the Zhou et al., (2022)³⁵ study for N FFs in freshwater. For diffusive emissions, whilst previous LCIA

474 methodologies have suggested diffusive emissions are roughly 10% of direct FF emissions^{2,37}, some
475 LCIA methods have not made any deductions for diffusive emissions³⁸. Our results show for marine
476 environments, diffusive emission FFs are 36.5% of direct N emission FFs and for freshwater are 31.8%
477 for N and 82.4% for P. Cosme et al., (2018)⁶ showed N diffusive emission FFs to marine environments
478 were 45.6% of direct emission FFs. Payen et al., (2021)³⁴ showed diffusive emission FFs were 9.3% of
479 direct emission FFs to freshwater for P and 48.6% of N. Zhou et al., (2022)³⁵ showed diffusive N
480 emission FFs to freshwater were 22.8% of direct emission FFs. Our results showed strong similarities
481 for the comparison of N diffusive to direct emissions with Cosme et al., (2018)⁶ and Zhou et al.,
482 (2022)³⁵. However, our study showed a weak percentage comparison of diffusive to direct emissions
483 with Payen et al., (2021)³⁴.

484 We expected the FF for freshwater environments to be smaller than the FF for marine environments,
485 as rivers are natural transmission pathways for water, nutrients and sediments to oceans. Our N FF
486 for freshwater is within a similar range for our P FF freshwater, providing further confidence in our
487 predicted results. Greater FFs were found on arable land uses for N due to greater removal fractions
488 (Appendix 2). However, for P higher FFs were identified on natural lands due to high erosion on steep
489 slopes.

490 Our results varied with previous studies due to different: (1) nutrient models used for the basis of the
491 FF (e.g. GLOBAL NEWS 2 Vs IMAGE GNM), (2) nutrient indicators assessed (e.g. dissolved inorganic
492 nitrogen vs total nitrogen), (3) spatial resolutions (basin scale vs 30arc min vs 5 arcmin) and (4) FF
493 model structures. Payen et al., (2021)³⁴ and Cosme et al., (2018)⁶ predict their FFs at the watershed
494 level using the Global NEWS 2 model for dissolved inorganic nutrients. Higher FFs predicted by both
495 Payen et al., (2021)³⁴ and Cosme et al., (2018)⁶ may be due to differences between Global NEWS 2
496 model and IMAGE GNM meaning lower total nitrogen was found by IMAGE GNM (37 Tg (N/yr)
497 compared to Global NEWS2 (45 Tg N/yr). Fundamental differences exist between IMAGE GNM and
498 GLOBAL NEWS 2 to stimulate N removal and transportation modelled within the terrestrial and
499 aquatic systems. IMAGE-GNM provide predictive advantages by adding detailed modelling of land fate
500 processes, which includes explicit groundwater denitrification in soils, aquifers and riparian zones,
501 rather than regression models (Global NEWS 2⁵) that lump process behaviour within watersheds by
502 export constants. Additionally, different hydrological and terrestrial data inputs and spatial resolutions
503 exist between the two models.

504 Strong comparisons existed between Zhou et al., (2022)³⁵ and this study because the FFs are based
505 on the IMAGE GNM model (either results or equations) and we adopted similar FF model structures
506 for freshwater (cumulative FF). However, this study improved the FF model by:

507 I. Using updated 5 arcmin resolutions datasets for soil properties and hydrological data inputs.

- 508 2. Considering variations in groundwater depth and thickness globally on the N nutrient load
 509 discharged via groundwater.
- 510 3. Considering subgrid and grid level nutrient retention.
- 511 4. Using updated hydrological datasets through PCR-GLOBWB 2¹⁰; which includes water return
 512 flows for irrigation, not modelled in PCR-GLOBWB 1⁷.
- 513 5. Estimating P in freshwater and N in marine environments for direct and diffusive emissions.
- 514 6. Developing an FF model from climate, soil and hydrological data to facilitate analysis of FFs
 515 under climate and land use change scenarios.

516 Overall, our results and previous studies suggest, the assumption that diffusive emissions are 10% of
 517 direct emission is not a sufficiently robust estimate for diffusive emission FFs. Additionally, large FF
 518 variations exist between studies, identifying the strong reliance on robust global nutrient models for
 519 FF model development.

520 2.4 User Information for the Fate Factor

521 Unlike in some previous FF models², here, the actual emission at the field is incorporated within our
 522 FF model ($FF_{Soils E (w) (i)}$) with; E representing the nutrient type, being N or P; (w) representing the
 523 receiving waterbody, either freshwater (fw) or marine (LME); and (i) representing the emission
 524 pathways, either surface runoff, erosion or leaching. Therefore, our emissions inventory within an
 525 LCIA framework incorporates the nutrient inputs, soil loss and mass balance as per IMAGE-GNM.
 526 This makes formulating the emission inventory for LCIA practitioners easier. For surface water, the
 527 nutrient load within a receiving waterbody receptor via surface runoff is fed solely by the initial nutrient
 528 inputs on land for each nutrient type (E):

$$529 \quad Load_{E(w)(Surface\ Runoff)} = Input_E \times FF_{E(w)(Surface\ Runoff)}$$

530 For erosion, P erosional processes consider the initial soil P content and any positive soil P mass
 531 balances left on the field. Within IMAGE-GNM, P soil content is assumed to vary over time due to
 532 historical fertiliser input. The natural P content baseline is taken from IMAGE GNM from the year
 533 1900. The N erosional process considers the C:N ratio of the soil organic carbon content, which is
 534 assumed to be constant over time. To calculate soil loss load, we used natural soil loss as a baseline
 535 to reflect anthropogenic pressures causing a relative change from natural land to grassland or arable
 536 lands. FF differences between land uses are constant (C_{LD}) with values for N as 45.3046 and 2.4138
 537 and for P as 38.6897 and 2.4138 for arable and grasslands, respectively:

$$538 \quad Load_{E(w)(Erosion)} = \frac{(Soilloss_{E, Landuse} - Soilloss_{E, Natural\ 1900}) \times FF_{E(w)(Erosion)}}{C_{LD} Soilloss_E \times FF_{E(w)(Erosion)}}$$

$$539 \quad Load_{P, Fw, Erosion\ New} = P_{budget > 0} \times FF_{P, Fw (Erosion)}$$

540 For subsurface transport only N is considered, as P is easily absorbed by soil minerals⁸. All N positive
541 soil balances are subject to leaching and exclude the N lost by surface runoff:

$$542 \quad N_{budget} = N_{budget} - (Input_E \times f_{qrso})$$

$$543 \quad Load_{N,Fw/LME,Leaching} = N_{budget>0} \times FF_{N,Fw/LME(Leaching)}$$

544 Practitioners of the FF should use FFs for all soils if the land use is unknown or their data represents
545 different land uses with unknown fractions. If land use is known, FFs for the corresponding land use
546 and grid cell should be used. Practitioners should follow the guidance as per Payen et al., (2020) for
547 variations in N limiting, P limiting or co-limiting regions. Payen et al., (2020) suggests if the limiting
548 nutrient is unknown, one should assume a co-limiting system of N_{eq} and P_{eq} and aggregate using the
549 Redfield Ratio to express $algae_{eq}$. If the user wishes to distinctly identify different pathway routes, as
550 within IMAGE-GNM, then the Fate Factor from a specific land use and transport pathway should be
551 used with the corresponding emission inventory requirements. Use of specific transport pathway fate
552 factors should follow emission inventory requirements as per IMAGE-GNM. Due to retention model
553 validity, we suggest users should use the FF model with caution over natural lands with desert and
554 mountainous geomorphic terrain.

555 We present FF characterisation for all land covers globally (i.e. assuming arable, grass or natural land
556 can be found anywhere across the globe). In doing so, we support LCIA studies that may wish to
557 identify fertiliser use impacts from land conversion. Furthermore, as no global FF factors are developed
558 in transient state, the FFs presented here may in the future provide insight to regions of high fertiliser
559 use impacts under the impacts of climate change. Studies have shown Canada and Russia will be prime
560 agricultural countries under the impacts of climate change in years to come³⁹. Our FF model identifies
561 Russia as one of the highest regions for the transport of nutrients to fluvial and marine environments.

562 2.5 Conclusion and Furtherwork

563 This study presented the opportunity to develop a spatially-explicit FF at the 5 arcmin resolution based
564 on IMAGE-GNM modelling concepts. Given the study's main purpose is to understand fertiliser impact
565 from crop production at the 5 arcmin resolution, there are further developments that can be
566 incorporated to make a more robust FF model using IMAGE-GNM. Here we present an FF model
567 developed from climate, soil and hydrological datasets. As such, this presents an FF model which may
568 be used to predict future FFs under climate and land use change scenarios. Secondly, we only validated
569 our FF model for arable land emissions pathways; validation of grass and natural lands were deemed
570 out of scope for this study. Lastly, the current FF model is complex in a practical sense. To support
571 LCIA practitioners and the use of the FF model, a more simplified model that easily connects emission
572 inventories to FF characterisations may enhance user ability. This will support the growing demand

573 for spatially-explicit LCIA studies in recognition of the UN Sustainable Development Goals and
574 transparency of product and services' impact on the environment.

575 3 Reference List

- 576 1. Rosenbaum, R. K., Margni, M. & Jolliet, O. A flexible matrix algebra framework for the multimedia multipathway
577 modeling of emission to impacts. *Environ. Int.* **33**, 624–634 (2007).
- 578 2. Helmes, R. J. K., Huijbregts, M. A. J., Henderson, A. D. & Jolliet, O. Spatially explicit fate factors of phosphorous
579 emissions to freshwater at the global scale. *Int. J. Life Cycle Assess.* **17**, 646–654 (2012).
- 580 3. Cosme, N. & Hauschild, M. Z. Characterization of waterborne nitrogen emissions for marine eutrophication
581 modelling in life cycle impact assessment at the damage level and global scale. *Int. J. Life Cycle Assess.* **22**, 1558–1570
582 (2017).
- 583 4. Morelli, B. et al. Critical review of eutrophication models for life cycle assessment. *Environ. Sci. Technol.* **52**, 9562–9578
584 (2018).
- 585 5. Mayorga, E. et al. Global Nutrient Export from WaterSheds 2 (NEWS 2): Model development and implementation.
586 *Environ. Model. Softw.* **25**, 837–853 (2010).
- 587 6. Cosme, N. M. D. Spatially explicit fate factors of waterborne nitrogen emissions at the global scale. *Int. J. Life Cycle*
588 *Assess.* **23**, 1286–1296 (2018).
- 589 7. Global monthly water stress: I. Water balance and water availability - van Beek - 2011 - Water Resources Research -
590 Wiley Online Library. <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2010WR009791>.
- 591 8. Beusen, A. H. W., Van Beek, L. P. H., Bouwman, A. F., Mogollón, J. M. & Middelburg, J. J. Coupling global models for
592 hydrology and nutrient loading to simulate nitrogen and phosphorus retention in surface water – description of
593 IMAGE-GNM and analysis of performance. *Geosci. Model Dev.* **8**, 4045–4067 (2015).
- 594 9. Batjes, N. H. ISRIC-WISE derived soil properties on a 5 by 5 global grid (ver. 1.2). 56.
- 595 10. Sutanudjaja, E. H. et al. PCR-GLOBWB 2: a 5 arcmin global hydrological and water resources model. *Geosci.*
596 *Model Dev.* **11**, 2429–2453 (2018).
- 597 11. de Graaf, I. E. M. et al. A global-scale two-layer transient groundwater model: Development and application to
598 groundwater depletion. *Adv. Water Resour.* **102**, 53–67 (2017).
- 599 12. Gleeson, T., Moosdorf, N., Hartmann, J. & Beek, L. P. H. van. A glimpse beneath earth's surface: GLobal
600 HYdrogeology MaPS (GLHYMPS) of permeability and porosity. *Geophys. Res. Lett.* **41**, 3891–3898 (2014).
- 601 13. Velthof, G. L., Oudendag, D. A. & Oenema, O. Development and application of the integrated nitrogen model. 102.
- 602 14. Cerdan, O. et al. Rates and spatial variations of soil erosion in Europe: A study based on erosion plot data.
603 *Geomorphology* **122**, 167–177 (2010).
- 604 15. Yang, X., Post, W. M., Thornton, P. E. & Jain, A. The distribution of soil phosphorus for global biogeochemical
605 modeling. *Biogeosciences* **10**, 2525–2537 (2013).

- 606 16. Drecht, G. V., Bouwman, A. F., Knoop, J. M., Beusen, A. H. W. & Meinardi, C. R. Global modeling of the fate of
607 nitrogen from point and nonpoint sources in soils, groundwater, and surface water. *Glob. Biogeochem. Cycles* **17**,
608 (2003).
- 609 17. Bouwman, A. F., Fung, I., Matthews, E. & John, J. Global analysis of the potential for N₂O production in natural soils.
610 *Glob. Biogeochem. Cycles* **7**, 557–597 (1993).
- 611 18. Keuskamp, J. A., van Drecht, G. & Bouwman, A. F. European-scale modelling of groundwater denitrification and
612 associated N₂O production. *Environ. Pollut.* **165**, 67–76 (2012).
- 613 19. Firestone, M. K. Biological Denitrification. in *Nitrogen in Agricultural Soils* 289–326 (John Wiley & Sons, Ltd, 2015).
614 doi:10.2134/agronmonogr22.c8.
- 615 20. Shaffer, M. J., Halvorson, A. D. & Pierce, F. J. Nitrate Leaching and Economic Analysis Package (NLEAP): Model
616 Description and Application. in *Managing Nitrogen for Groundwater Quality and Farm Profitability* 285–322 (John Wiley &
617 Sons, Ltd, 2015). doi:10.2136/1991.managingnitrogen.c13.
- 618 21. Lithologic composition of the Earth's continental surfaces derived from a new digital map emphasizing riverine
619 material transfer - Dürr - 2005 - Global Biogeochemical Cycles - Wiley Online Library.
620 <https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2005GB002515>.
- 621 22. Denitrification. <https://www.ncbi.nlm.nih.gov/pmc/articles/PMC373209/>.
- 622 23. Šlmeč, M. & Cooper, J. E. The influence of soil pH on denitrification: progress towards the understanding of this
623 interaction over the last 50 years. *Eur. J. Soil Sci.* **53**, 345–354 (2002).
- 624 24. Saunders, D. L. & Kalff, J. Nitrogen retention in wetlands, lakes and rivers. *Hydrobiologia* **443**, 205–212 (2001).
- 625 25. Reddy, K. R., Kadlec, R. H., Flaig, E. & Gale, P. M. Phosphorus Retention in Streams and Wetlands: A Review. *Crit. Rev.*
626 *Environ. Sci. Technol.* **29**, 83–146 (1999).
- 627 26. Wollheim, W. M., Vörösmarty, C. J., Peterson, B. J., Seitzinger, S. P. & Hopkinson, C. S. Relationship between river
628 size and nutrient removal. *Geophys. Res. Lett.* **33**, (2006).
- 629 27. Wollheim, W. M. et al. Global N removal by freshwater aquatic systems using a spatially distributed, within-basin
630 approach. *Glob. Biogeochem. Cycles* **22**, (2008).
- 631 28. Marcé, R. & Armengol, J. Modeling nutrient in-stream processes at the watershed scale using Nutrient Spiralling
632 metrics. *Hydrol. Earth Syst. Sci. Discuss.* **6**, 501–533 (2009).
- 633 29. Seitzinger, S. et al. Denitrification Across Landscapes and Waterscapes: A Synthesis. *Ecol. Appl.* **16**, 2064–2090 (2006).
- 634 30. Meinardi, C. R. Groundwater recharge and travel times in the sandy regions of the Netherlands. (1994).
- 635 31. Mueller, N. D. et al. Closing yield gaps through nutrient and water management. *Nature* **490**, 254–257 (2012).
- 636 32. West, P. C. et al. Leverage points for improving global food security and the environment. *Science* **345**, 325–328
637 (2014).

- 638 33. Harris, G. P. The concept of limiting nutrients. in *Phytoplankton Ecology: Structure, Function and Fluctuation* (ed. Harris,
639 G. P.) 137–165 (Springer Netherlands, 1986). doi:10.1007/978-94-009-3165-7_7.
- 640 34. Payen, S., Cosme, N. & Elliott, A. H. Freshwater eutrophication: spatially explicit fate factors for nitrogen and
641 phosphorus emissions at the global scale. *Int. J. Life Cycle Assess.* (2021) doi:10.1007/s11367-020-01847-0.
- 642 35. Zhou, J., Scherer, L., van Bodegom, P. M., Beusen, A. & Mogollón, J. M. Regionalized nitrogen fate in freshwater
643 systems on a global scale. *J. Ind. Ecol.* **n/a**,
- 644 36. Monfreda, C., Ramankutty, N. & Foley, J. A. Farming the planet: 2. Geographic distribution of crop areas, yields,
645 physiological types, and net primary production in the year 2000. *Glob. Biogeochem. Cycles* **22**, (2008).
- 646 37. ReCiPe2016v1.1. | RIVM. <https://www.rivm.nl/documenten/recipe2016v1.1>.
- 647 38. IMPACT World+. <http://www.impactworldplus.org/en/presentation.php>.
- 648 39. Hannah, L. *et al.* The environmental consequences of climate-driven agricultural frontiers. *PLOS ONE* **15**, e0228305
649 (2020).
- 650