

1 **Public water supply is responsible for significant fluxes of inorganic nitrogen in the environment**

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20 **Abstract**

21 Understanding anthropogenic disturbance of macronutrient cycles is essential for assessing risks  
22 facing ecosystems. For the first time, we quantified inorganic nitrogen (N) fluxes associated with  
23 abstraction, mains water leakage and transfers of treated water related to public water supply. In  
24 England, the mass of nitrate-N removed from aquatic environments by abstraction (ABS-NO<sub>3</sub>-N) was  
25 estimated to be 24.2 kt N/yr. This is equal to six times estimates of organic N removal by abstraction,  
26 15 times in-channel storage of organic N and 30 times floodplain storage of organic N. ABS-NO<sub>3</sub>-N is  
27 also between 3-39% of N removal by denitrification in the hydrosphere. Mains water leakage of  
28 nitrate-N (MWL-NO<sub>3</sub>-N) returns 3.62 kt N/yr to the environment, equating to approximately 15% of  
29 ABS-NO<sub>3</sub>-N . In urban areas, MWL-NO<sub>3</sub>-N can represent up to 20% of total N inputs. MWL-NO<sub>3</sub>-N is  
30 predicted to increase by up to 66% by 2020 following implementation of treated water transfers. ABS-  
31 NO<sub>3</sub>-N and MWL-NO<sub>3</sub>-N should be considered in future assessments of N fluxes, in order to accurately  
32 quantify anthropogenic disturbances to N cycles. The methodology we developed is transferable,  
33 using widely-available datasets and could be used to quantify N fluxes associated with public water  
34 supply across the world.

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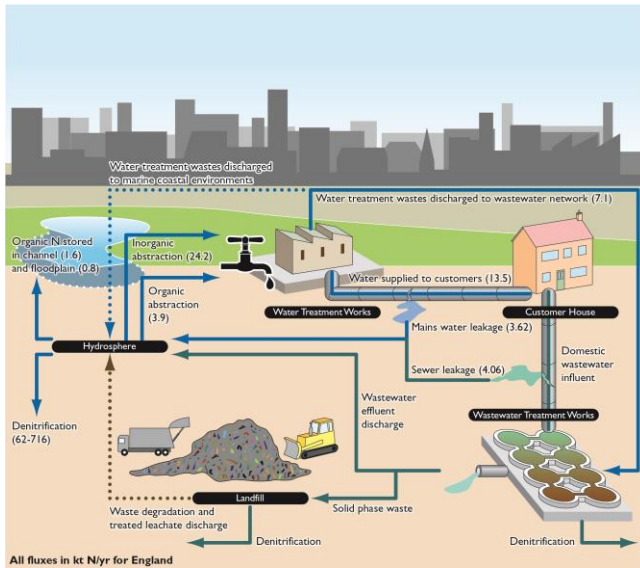
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43 **Abstract Art**

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# 60 1 Introduction

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62 Eutrophication associated with anthropogenic disturbance to nitrogen (N) and phosphorus (P) cycles  
63 within aquatic ecosystems is a widespread environmental challenge that has been recognised for over  
64 50 years<sup>1</sup>. In response, substantial efforts have been made to reduce both point<sup>2</sup> and diffuse<sup>3</sup> source  
65 nutrient inputs to the environment, for example through implementation of national and international  
66 legislation (e.g. the USA Clean Water Act<sup>4</sup>, EU Water Framework<sup>5</sup>, Nitrates<sup>6</sup> and Urban Wastewater  
67 Treatment<sup>7</sup> Directives). Despite such efforts, there are multiple instances in which the reductions in  
68 riverine concentrations and fluxes of N and P after the implementation of mitigation activities have  
69 been lower than expected<sup>8</sup>. The release of nutrients, both from previously unidentified sources and  
70 from legacy stores (e.g. groundwater, soils or the riparian zone<sup>9</sup>), is a potentially important contributor  
71 to unexpected trajectories of catchment nutrient balances. In this context, we sought to constrain the  
72 previously unquantified influence of public water supply on inorganic N fluxes.

73 Recent research has suggested that the water treatment cycle (water abstraction, treatment,  
74 distribution for public water supply and subsequent leakage) may have significant impacts on both  
75 nutrient sources and nutrient retention, particularly in urban areas. For example, previous research  
76 suggests that leakage of phosphate-dosed mains water (MWL-P) can be an important source of P in  
77 urban environments, with MWL-P fluxes now reaching up to 24% of sewage treatment P loads in some  
78 catchments<sup>10-12</sup>. Methods have also been developed to quantify N fluxes to groundwater from mains  
79 water leakage at the city-scale<sup>13-15</sup>, suggesting that approximately 36% of the total N loads in urban  
80 areas could be attributed to this source.

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82 In the context of aquatic ecosystems, retention (also referred to as losses, sinks or removal) can be  
83 conceptualised as transient (e.g. nutrient storage within floodplains or groundwater) or permanent N

84 removal (e.g. denitrification)<sup>16</sup>. Based on the difference between N fluxes from terrestrial to aquatic  
85 ecosystems and N fluxes at the tidal limit of the fluvial system, Worrall, et al. <sup>17</sup> concluded that  
86 substantial in-stream N retention occurs in Great Britain. Whilst detailed understanding of in-stream  
87 retention mechanisms at the national-scale remains to be developed, it has been postulated that  
88 these losses of N are primarily due to permanent N removal via denitrification<sup>17</sup>. However, the  
89 processes associated with public water supply may also contribute significantly to N retention within  
90 aquatic ecosystems. For example, Finlay, et al. <sup>18</sup> estimated the flux of organic N removed by water  
91 abstraction, suggesting that this flux is greater than N retention associated with transient floodplain  
92 storage.

93 However, important gaps remain in our understanding of how macronutrient fluxes are perturbed by  
94 the processes associated with public water supply. Whilst Finlay, et al. <sup>18</sup> calculated organic N removal,  
95 the mass of inorganic N removed during abstraction and the significance of this N retention process  
96 relative to other forms of in-stream retention have not been quantified to date. Further, previous  
97 methods developed for nutrient source apportionment either do not consider mains leakage<sup>19, 20</sup>, or  
98 use simplistic approaches<sup>13-15</sup> based on export coefficients and literature values rather than observed  
99 leakage and concentration data. Finally, potential changes in inorganic N fluxes due to mains water  
100 leakage associated with future treated drinking water transfers have not been quantified. Water  
101 transfers are considered essential if future water demand is to be met, but the water quality  
102 implications of water transfers remain poorly constrained<sup>21</sup>. If effective, integrated approaches to the  
103 management of nutrient sources and fate are to be developed, it is essential that future nutrient fluxes  
104 associated with water transfers are quantified and the locations where these fluxes are significantly  
105 enhanced in the future are identified. Within this context, in the research reported here we  
106 hypothesized that:

- 107 • Abstraction of raw untreated water removes a significant, but spatially heterogeneous, mass  
108 of nitrate (ABS-NO<sub>3</sub>-N) from the aquatic environment (defined here as groundwater, rivers  
109 and coastal marine environments);
- 110 • A considerable proportion of ABS-NO<sub>3</sub>-N is returned to the environment through leakage of  
111 treated mains water containing nitrate (MWL-NO<sub>3</sub>-N), particularly within urban areas;
- 112 • Treated drinking water transfers will substantially alter the future magnitude and spatial  
113 distribution of MWL-NO<sub>3</sub>-N fluxes.

## 114 **2 Materials and Methods**

### 115 **2.1 Study Area**

116

117 The research reported here was developed at the national-scale for the country of England (Figure 1).  
118 Water is supplied by 21 water companies across England. Water company areas are divided into water  
119 resource zones (WRZs) within which water supplies are largely self-contained<sup>22</sup>. These WRZs are  
120 further divided into water supply zones (WSZs) which correspond to areas with uniform water quality,  
121 supplying up to 100,000 people<sup>23</sup>. WSZs in England derived in this research (see supporting  
122 information) and principal regions referred to in this research are shown in Figure 1.

123

### 124 **2.2 Quantification of ABS-NO<sub>3</sub>-N and MWL-NO<sub>3</sub>-N**

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#### 126 **2.2.1 ABS-NO<sub>3</sub>-N**

127

128 We derived ABS-NO<sub>3</sub>-N using estimates of the rates at which treated water enters the distribution  
129 network from water treatment works (referred to herein as “distribution input”) and raw water quality  
130 data for each WSZ. Distribution input rates are not reported nationally at the WSZ level, so these were

131 derived as follows using estimates of treated water supplied to customers (referred to herein as  
132 “water supplied”). The total distribution input,  $DI$  ( $m^3/day$ ) into a WSZ was divided into water supplied,  
133  $S$  ( $m^3/day$ ), and leakage,  $L$  ( $m^3/day$ ):

$$134 \quad DI = S + L \quad (1)$$

135  $S$  for each WSZ was extracted from published data submitted by the drinking water regulator for  
136 England to the European Commission<sup>24</sup>. For 12% of WSZs, values of  $S$  were not reported by the  
137 drinking water regulator. For these WSZs,  $S$  was estimated using WRZ-level estimates of per capita  
138 water consumption ( $PCC$ , L/h/d) published by Water UK<sup>21</sup> and the total population ( $P$ , unitless) for  
139 each WSZ area, based on 1 km gridded population density mapping<sup>25</sup>:

$$140 \quad S = \frac{PCC \cdot P}{1000} \quad (2)$$

141 Because WSZ-level leakage rates were not available, these were derived based on published WRZ-  
142 level leakage rates<sup>21</sup>. Assuming that the percentage leakage rate in a WSZ is equal to the percentage  
143 rate in the corresponding WRZ,  $L$  was derived as a fraction of the  $DI$ :

$$144 \quad L = DI \cdot f_{WRZ} \quad (3)$$

145 where  $f_{WRZ}$  (unitless) is the fractional leakage rate of the WRZ. Combining equations (1) and (2),  $DI$   
146 was derived as:

$$147 \quad DI = \frac{S}{1 - f_{WRZ}} \quad (4)$$

148

149 As no national scale datasets on WSZ raw water quality were available, mean raw water nitrate  
150 concentrations for each WSZ were estimated using a heuristic approach based on data held in the  
151 English environmental regulator’s water quality monitoring database. Following previous studies of  
152 national pollutant transport<sup>11</sup> and groundwater use maps<sup>26</sup>, WSZs were classified as either  
153 groundwater or surface water sources using national-scale hydrogeological mapping<sup>27</sup>. If a WSZ is  
154 underlain by a moderate or highly productive aquifer, then the WSZ was considered to be a

155 groundwater WSZ. If underlain by a low productivity aquifer or rocks with essentially no groundwater  
156 resource, then the WSZ was considered to be a surface water WSZ. For each WSZ, raw water nitrate  
157 concentrations were estimated using the following rules: (1) only samples taken within a WSZ; (2) only  
158 samples that correspond to the derived WSZ source water (i.e. groundwater or surface water); (3) only  
159 samples taken in 2015 for direct comparability with drinking water quality data for the same year (see  
160 section 2.2.2 and supporting information); and (4) only samples for routine environmental monitoring,  
161 to avoid samples taken specifically for pollution investigations unduly impacting the analyses. Where  
162 no water quality samples were present within a WSZ (4.5% of WSZs in England by area) the mean raw  
163 water nitrate concentration of the nearest groundwater or surface water sample point was used.  
164 Where WSZs are fed by existing water transfers, the receiving WSZ was assigned the raw water nitrate  
165 concentration of the donor WSZ. After estimating DI and raw water nitrate concentrations ( $C_R$ , mg  
166 N/L) for each WSZ, the mass of nitrate-N removed from the aquatic environment through water  
167 abstraction, ABS-NO<sub>3</sub>-N (kg N/day), was calculated as:

$$ABS-NO_3-N = \frac{C_R \cdot DI}{1000} \quad (5)$$

## 170 2.2.2 MWL-NO<sub>3</sub>-N

171

172 We calculated MWL-NO<sub>3</sub>-N at the WSZ level using existing datasets for nitrate concentrations in  
173 treated drinking water and the water supply rates and WRZ-level leakage rates detailed in section  
174 2.2.1. Combining equations (3) and (4), L was calculated as:

$$L = \frac{S \cdot f_{WRZ}}{1 - f_{WRZ}} \quad (6)$$

176 Nitrate concentration data were extracted from national-scale databases of WSZ drinking water  
177 quality reports for 2015 (see supporting information). All water company laboratories are required to  
178 be accredited by the UK Accreditation Service to the Drinking Water Testing Specification<sup>28</sup>. This



179 includes interlaboratory proficiency testing for all parameters including nitrate concentrations, and  
180 thus any differences in nitrate concentrations between water companies associated with different  
181 laboratory methods were assumed to be minimal. Using the mean treated drinking water nitrate  
182 concentration for each WSZ for 2015,  $C_T$  (mg N/L),  $MWL-NO_3-N_{WSZ}$  (kg N/day) was calculated as:

$$183 \quad MWL-NO_3-N_{WSZ} = \frac{C_T \cdot L}{1000} \quad (7)$$

184

### 185 **2.2.3 Changes in $MWL-NO_3-N$ associated with future transfers of treated drinking water**

186

187 Transfers of treated drinking water at the WRZ-level that are planned to be implemented by 2020  
188 were provided by the environmental regulator for England (Figure S1). These transfers are all located  
189 in South East and Eastern England, where current water stress is greatest<sup>21</sup>. To assess the impact of  
190 future transfers on the spatial distribution of  $MWL-NO_3-N$ , the WSZ-level  $MWL-NO_3-N_{WSZ}$  estimates  
191 derived in section 2.2.2 were aggregated to the WRZ level ( $MWL-NO_3-N_{WRZ}$ , kg N/day) to obtain a  
192 baseline flux for 2015:

$$193 \quad MWL-NO_3-N_{WRZ} = \sum MWL-NO_3-N_{WSZ} \quad (8)$$

194 The impact of aggregation of WSZ  $MWL-NO_3-N$  fluxes to WRZ level, in addition to impacts of using  
195 WRZ percentage leakage rates at the WSZ level and the impacts of estimating raw water nitrate  
196 concentrations using national environmental regulator water quality databases, are considered in  
197 section 4.3.

198

199 For each water transfer, the future leakage rate for the transferred water in the receiving WRZ was  
200 assumed to equal the existing leakage rate in the receiving zone. This is a reasonable assumption  
201 because all water companies in England have current leakage rates at or below the sustainable

202 economic level of leakage<sup>29</sup> and leakage rates are not forecast to change substantially over the next  
 203 25 years<sup>21, 30</sup>. The volumetric leakage rate for a transfer,  $L_T$  (m<sup>3</sup>/day), can therefore be estimated as:

$$204 \quad L_T = DI_T \cdot f_R \quad (9)$$

205 where  $f_R$  is the leakage fraction (unitless) for the receiving WRZ and  $DI_T$  (m<sup>3</sup>/day) is the additional  
 206 distribution input associated with the transfer. The nitrate concentration in the transferred water in  
 207 the future was assumed to equal to the current drinking water nitrate concentrations in the donor  
 208 WRZ. Nitrate concentrations in drinking water in England are consistently below the European drinking  
 209 water standard, with 99.99% of test samples compliant<sup>31</sup>. Consequently, there is unlikely to be any  
 210 additional treatment for nitrate removal associated with future treated water transfers. The flux of N  
 211 from mains water leakage associated with an individual planned transfer,  $MWL-NO_3-N_T$  (kg N/day),  
 212 was estimated as:

$$213 \quad MWL-NO_3-N_T = L_T \cdot C_{T-D} \quad (10)$$

214 where  $C_{T-D}$  is the mean treated drinking water nitrate concentration in the donor WRZ (mg N/L). Some  
 215 WRZs are expected to receive a number of different water transfers in the future. In these cases, the  
 216 individual transfers were combined to derive the total additional flux of N from mains leakage resulting  
 217 from all transfers,  $MWL-NO_3-N_{WRZ-T}$  (kg N/day):

$$218 \quad MWL-NO_3-N_{WRZ-T} = \sum MWL - N_T \quad (11)$$

219 Using the baseline WRZ-level estimates of  $MWL-NO_3-N_{WRZ}$  for 2015, the percentage change in  $MWL-$   
 220  $NO_3-N_{WRZ}$  expected after the implementation of the planned transfers by 2020,  $\Delta MWL-NO_3-N_{WRZ}$   
 221 (unitless) was calculated as:

$$222 \quad \Delta MWL-NO_3-N_{WRZ} = \frac{MWL-NO_3-N_{WRZ-T} + MWL-NO_3-N_{WRZ}}{MWL-NO_3-N_{WRZ}} \cdot 100 \quad (12)$$

223

224 **2.3 Comparison between ABS-NO<sub>3</sub>-N, MWL-NO<sub>3</sub>-N and estimates of other N**  
225 **sources and N retention in the aquatic environment**

226

227 Our estimates of ABS-NO<sub>3</sub>-N were compared with previous, national-scale estimates of the retention  
228 of organic N via water treatment for public supply, floodplain storage and in-channel storage<sup>18</sup>. We  
229 also compared ABS-NO<sub>3</sub>-N with estimates of N removal via denitrification in the entire English  
230 hydrosphere (groundwater, surface water and the coastal marine environments)<sup>32</sup> and total in-stream  
231 losses (assumed to be dominated by denitrification)<sup>17</sup>. Organic N retention by abstraction was  
232 reported as a per-capita flux which was transformed to a total flux for England using the current  
233 population of England<sup>33</sup>. All other fluxes were reported either as an absolute flux or flux per unit area.  
234 Where necessary, fluxes at the scale of England were calculated using the percentage of the UK land  
235 area which England comprises (54%).

236

237 To quantify the significance of MWL-NO<sub>3</sub>-N, this flux was compared with other datasets aggregated to  
238 both the national and the WRZ level. To compare with previous estimates of urban N fluxes, we used  
239 estimates of N fluxes to groundwater at 1 km grid scale from water mains and the sewer network  
240 derived by the UK Environment Agency<sup>34</sup> following the export coefficient approach of Lerner<sup>14</sup>. We  
241 also used previous catchment-scale estimates of both urban diffuse (roads, residential zones, open  
242 urban spaces, industrial and commercial areas) N fluxes to rivers and from all N sources provided  
243 under the SEPARATE framework<sup>20</sup>. We calculated ratios of both current and future (post-transfers)  
244 MWL-NO<sub>3</sub>-N made in our research to these estimates of N inputs from other sources.

## 245 **3 Results**

246

### 247 **3.1 ABS-NO<sub>3</sub>-N**

248

249 Figure 2 reports the spatial distribution of ABS-NO<sub>3</sub>-N across England. Blank areas denote locations in  
250 which no public water supplies are present (typically in sparsely populated upland areas in the north  
251 of England) or where data were unavailable (a small number of WSZs in southern England). Substantial  
252 spatial variation in the distribution of ABS-NO<sub>3</sub>-N is revealed, with ABS-NO<sub>3</sub>-N broadly greater in the  
253 Midlands and the South East than in the far North and Southwest of England. There are also additional  
254 hotspots of high ABS-NO<sub>3</sub>-N in Yorkshire and the Humber, the West and East Midlands and near to  
255 London. The broad spatial trends and the hotspots are primarily related to variations in ABS-NO<sub>3</sub>-N  
256 associated with high nitrate concentrations in some raw groundwaters and surface waters.

257

258 Table 1 reports national-scale estimates of ABS-NO<sub>3</sub>-N for England in comparison to previous studies  
259 estimating N retention in aquatic environments in England. For 2015, we estimated total ABS-NO<sub>3</sub>-N  
260 for England to be 24.2 kt N. Finlay, et al. <sup>18</sup> suggest that the total mass of organic N removed during  
261 water abstraction for public supply in the UK is 0.07 kg N/h/yr, equating to a total flux in England of  
262 3.9 kt N/yr. ABS-NO<sub>3</sub>-N therefore removes approximately six times more N from the environment as  
263 nitrate than is associated with abstraction of organic N. ABS-NO<sub>3</sub>-N is also 30 and 15 times greater  
264 than N storage in floodplains and in channels as organic N respectively. The range of estimates for the  
265 magnitude of denitrification within aquatic environments in England is substantial, from 62 kt N/yr for  
266 the complete hydrosphere (surface water, groundwater and marine coastal environments)<sup>32</sup> to 716 kt  
267 N/yr within the stream network alone<sup>17</sup>. Based on this range of available estimates, ABS-NO<sub>3</sub>-N is  
268 equivalent to between 3 – 39% of current estimates of denitrification in aquatic environments.

## 269 3.2 MWL-NO<sub>3</sub>-N

270

271 The distribution of MWL-NO<sub>3</sub>-N across WSZs in England is reported in Figure 3. Substantial spatial  
272 variation is also revealed in the MWL-NO<sub>3</sub>-N fluxes, reflecting both the spatial distribution of leakage  
273 rates and drinking water nitrate concentrations. Significant N fluxes from mains leakage are present  
274 in urban areas with high leakage rates such as London, the West Midlands and the Humber. Relatively  
275 high MWL-NO<sub>3</sub>-N fluxes are also observed in Eastern England and along parts of the south coast, where  
276 particularly high nitrate concentrations have been observed in drinking water.

277

278 Table 2 reports the total MWL-NO<sub>3</sub>-N flux for England alongside previous national-scale N flux  
279 estimates. The estimate of national-scale MWL-NO<sub>3</sub>-N in our research (3.62 kt N/yr) is significantly  
280 below previous estimates of this flux made by the Environment Agency <sup>34</sup> and relatively similar to  
281 estimates of fluxes from sewer leakage (4.07 kt N/yr). Based on previous N source apportionment  
282 studies (SEPARATE<sup>20</sup>), the return of MWL-NO<sub>3</sub>-N to the environment is small (c. 1%) at the national-  
283 scale relative to all other N sources, but much more significant (c. 160%) relative to urban diffuse N  
284 sources.

285

286 Figure 4 reports our estimates of MWL-NO<sub>3</sub>-N as a percentage of the total N flux from all sources  
287 derived from Zhang, et al. <sup>20</sup>, aggregated to the WRZ level. MWL-NO<sub>3</sub>-N is generally small relative to  
288 total N fluxes, at 2 – 5% of the total flux in the majority of WRZs in Southeast England and the Midlands.  
289 MWL-NO<sub>3</sub>-N is <1% in large WRZs in Northern, Southwest and Eastern England, where agricultural  
290 land is extensive and urban areas are small relative to the total land area. However, in London and the  
291 surrounding area where urban land area is more extensive, MWL-NO<sub>3</sub>-N grows in significance. For

292 example, MWL-NO<sub>3</sub>-N fluxes are estimated to be c. 15%, 16% and 20% of total N sources in WRZs in  
293 London, the south coast and Kent (east of London) respectively.

### 294 **3.3 Changes in MWL-NO<sub>3</sub>-N associated with future treated water transfers**

295

296 Figure 5 reports the percentage change in the MWL-NO<sub>3</sub>-N flux in receiving WRZs after  
297 implementation of planned treated water transfers by 2020. In some WRZs on the south coast of  
298 England, MWL-NO<sub>3</sub>-N fluxes are predicted to increase by up to 66% compared to 2015 levels, although  
299 implementation of water transfers is generally predicted to increase MWL-NO<sub>3</sub>-N fluxes by a relatively  
300 small amount (5 – 15%) compared to the 2015 baseline. MWL-NO<sub>3</sub>-N remains small relative to other  
301 N sources in the majority of WRZs where transfers are implemented. However, in WRZs to the west  
302 of London, MWL-NO<sub>3</sub>-N is expected to rise after the implementation of planned water transfers to the  
303 equivalent to up to 20% of all N sources (Figure S2).

## 304 **4 Discussion**

### 305 **4.1 ABS-NO<sub>3</sub>-N: A significant sink for N within the environment?**

306

307 Nitrogen retention within the aquatic environment contributes to the differences between the N flux  
308 from the terrestrial biosphere and the fluvial flux of N to the oceans<sup>35</sup>. Assuming net growth and decay  
309 of riverine biota at the national-scale is negligible<sup>17</sup>, denitrification has previously been considered the  
310 primary mechanism for N removal in aquatic ecosystems<sup>36, 37</sup>. Only recently has research begun to  
311 quantify direct N removal from the environment through anthropogenic processes such as water  
312 abstraction<sup>18</sup>. In the context of N retention, a significant unique contribution of our research is the  
313 quantification of nitrate removed from aquatic environments by water abstraction, which is a  
314 significantly greater N flux than the estimate of organic nitrogen removal by abstraction, storage  
315 within floodplain and in-channel environments by Finlay, et al. <sup>18</sup>. The wide range of estimates for

316 the magnitude of aquatic denitrification means that quantifying the relative importance of ABS-NO<sub>3</sub>-  
317 N is challenging. Further research is needed to better constrain these highly variable estimates to  
318 more accurately evaluate the significance of ABS-NO<sub>3</sub>-N. Nevertheless, given that the upper limit for  
319 ABS-NO<sub>3</sub>-N represents c. 40% of denitrification in the hydrosphere of England, including ABS-NO<sub>3</sub>-N in  
320 future catchment N budgets appears to be important.

321

322 Whilst ABS-NO<sub>3</sub>-N removes significantly more N than is associated with abstraction of organic N, and  
323 may also be significant relative to the N flux associated with denitrification, ABS-NO<sub>3</sub>-N may only  
324 represent a transient retention process. The ABS-NO<sub>3</sub>-N flux can be divided into: (1) N removed by  
325 water treatment processes; (2) N lost through mains water leakage; and (3) N supplied within treated  
326 water that reaches households. Mains water leakage will return N directly into the environment,  
327 although it is likely to be delayed before finally reaching receiving waters. Using drinking water nitrate  
328 concentrations and the WSZ supply rates estimated in this research, we estimate that 13.5 kt N/yr  
329 enters treated public water supply. Based on the difference between ABS-NO<sub>3</sub>-N and the sum of MWL-  
330 NO<sub>3</sub>-N and N in treated public water supply, we estimate that 7.1 kt N/yr is removed by the treatment  
331 of raw water before distribution. Much of the N that enters treated water will ultimately move into  
332 the wastewater network<sup>38</sup>. Waters that are highly enriched in nitrate produced by nitrate removal  
333 from raw water through ion exchange during drinking water treatment will also enter the wastewater  
334 network, although in coastal areas direct discharge to the marine environment may occur<sup>39</sup>. Following  
335 wastewater treatment, N will be partitioned between solid phase waste sent to landfill or returned to  
336 agricultural land and dissolved or particulate N discharged directly to receiving waters. Whilst these  
337 processes are likely to have varying timescales for return of N into the environment, critically they are  
338 all likely to be substantially longer than the mean residence time for water within UK rivers (26.7 hrs<sup>18</sup>).  
339 Therefore, whilst ultimately a transient retention processes, ABS-NO<sub>3</sub>-N will significantly alter the  
340 spatial and temporal distribution of N inputs into aquatic ecosystems.

## 341 **4.2 MWL-NO<sub>3</sub>-N: A significant N source in urban areas**

342

343 At the national-scale, our research suggests that MWL-NO<sub>3</sub>-N is equal to approximately 15% of ABS-  
344 NO<sub>3</sub>-N. This is likely to be reasonable because leakage rates in England have been reported to be up  
345 to 20% of water entering the distribution network<sup>10</sup> and treated waters show substantial variation in  
346 nitrate concentrations. MWL-NO<sub>3</sub>-N is small compared to all other N sources in large WRZs, where  
347 agriculture is the most significant N source. However, in urban areas MWL-NO<sub>3</sub>-N may account for up  
348 to 20% of all N loads. These findings are broadly consistent with initial city-scale studies reported  
349 previously for Nottingham, UK<sup>15</sup>. Previous work that adopts relatively simple estimates of MWL-NO<sub>3</sub>-  
350 N using export coefficient approaches<sup>34</sup> substantially overestimates MWL-NO<sub>3</sub>-N compared to our  
351 approach which uses observed leakage and concentration data. The flux of N associated with MWL  
352 should be considered in more detail in urban N source apportionment studies. The methodology we  
353 report here is transferable and could be applied where the necessary data are available, delivering a  
354 significant increase in accuracy and spatial resolution of estimates of MWL-NO<sub>3</sub>-N compared to  
355 alternative approaches.

356

357 Previous research has suggested that incorporation of the environmental cost of P fluxes from mains  
358 water leakage within methodologies to estimate the sustainable economic level of leakage (SELL)  
359 could result in more stringent leakage targets, and consequently lower P fluxes to the environment<sup>12</sup>.  
360 Extending the SELL approach to consider the environmental impact of MWL-NO<sub>3</sub>-N would also be of  
361 environmental benefit. However, whilst MWL-NO<sub>3</sub>-N represents a substantial N flux to the  
362 environment, concentrations in mains leakage are lower than in sewer leakage<sup>13</sup>. Consequently, mains  
363 leakage to groundwater may dilute high nitrate sewer leakage. Integrating N sources into SELL would  
364 require an evaluation of the tradeoff between the negative environmental impact of MWL-NO<sub>3</sub>-N as  
365 an N source and any environmental benefit associated with dilution of sewer N leakage. Additional  
366 nitrate removal from raw waters to reduce MWL-NO<sub>3</sub>-N fluxes whilst continuing to dilute sewer N



367 leakage is unlikely to be a viable solution, given that concentrations of nitrate in drinking water within  
368 England are already below the European drinking water standard<sup>31</sup>.

369

370 It should be noted that MWL-NO<sub>3</sub>-N, rather than representing a new input of reactive N to the  
371 terrestrial environment (as would be associated with the application of inorganic N fertilisers or N  
372 fixation for example), instead represents a return of N to the environment after water abstraction.  
373 Return of N via MWL therefore represents a potential alteration to the distribution and residence time  
374 of N in different environmental compartments. In this context, it should also be noted that the  
375 ultimate fate of MWL-NO<sub>3</sub>-N currently remains uncertain. The MWL-NO<sub>3</sub>-N flux may be mediated by  
376 the soil microbial community and by plants following leakage. MWL-NO<sub>3</sub>-N may also be subject to  
377 denitrification, although this is unlikely near to the site of a leak as treated water has been shown to  
378 be low in organic carbon<sup>23</sup> and is unlikely to be anaerobic due to oxidation processes during water  
379 treatment (e.g. ozonation producing dioxygen<sup>40</sup>) and extensive contact with the atmosphere<sup>41</sup>. It is  
380 likely that the fate of MWL-NO<sub>3</sub>-N can be broadly divided between fluxes to groundwater, surface  
381 water and the sewer network. Based on the national-scale hydrogeological mapping described in  
382 section 2.2.1, the MWL-NO<sub>3</sub>-N flux on moderate and highly productive aquifers is estimated to be 1.67  
383 kt N/yr. Given the long residence times in the unsaturated and saturated zones<sup>42, 43</sup>, it is likely that  
384 legacy MWL-NO<sub>3</sub>-N from previous decades has accumulated within both groundwater and the vadose  
385 zone, as has been observed for nutrient loads from agricultural land<sup>44-46</sup>. Moreover, historical leakage  
386 rates have been significantly higher than at present, with rates falling by a third between 1994 and  
387 2015<sup>47</sup>. Consequently, the rate of MWL-NO<sub>3</sub>-N accumulation within the vadose zone and groundwater  
388 is likely to have been greater in the past compared to the present day.

389

390 Our research predicts relatively small changes in the spatial distribution of MWL-NO<sub>3</sub>-N fluxes  
391 associated with treated drinking water transfers. These transfers are fully costed, planned and will be  
392 implemented by water utilities by 2020 in England. Larger-scale transfers which may result in larger  
393 differences in water quality (and changes in MWL-NO<sub>3</sub>-N fluxes) are currently being evaluated<sup>21</sup>,  
394 although these transfers are yet to be formally included in the water resources plans. However, it  
395 should be noted that for both planned and speculative transfers of treated water, implementation of  
396 additional treatment for nitrate removal is unlikely because concentrations are already below the  
397 European drinking water limit. With mitigation measures to control other N pollution sources being  
398 implemented<sup>48</sup>, and limited changes planned for mains water leakage rates<sup>30</sup>, it is likely that the  
399 relative significance of MWL-NO<sub>3</sub>-N will increase in the future, as has been reported for MWL-P in the  
400 Thames catchment<sup>30</sup>.

401

### 402 **4.3 Local and global research priorities for ABS-NO<sub>3</sub>-N and MWL-NO<sub>3</sub>-N**

403

404 The unique contribution of this study is the first national-scale estimates of N fluxes from abstraction  
405 and mains water leakage using observed nitrate concentration and leakage data, both before and after  
406 the implementation of future water transfers. There are a number of outstanding research priorities  
407 within this field which remain to be addressed. To date the only published national estimates of  
408 aquatic N retention by abstraction are for organic N compounds<sup>18</sup>, and comparing ABS-NO<sub>3</sub>-N with  
409 other temporary retention processes that influence inorganic N would be helpful. Our research has  
410 quantified the flux of nitrate associated with water treatment processes as the difference between  
411 ABS-NO<sub>3</sub>-N and the sum of MWL-NO<sub>3</sub>-N and N in treated water supplied. Direct quantification of N  
412 removal during water treatment at the national-scale using observed concentration and flow data for  
413 process losses could be used to further validate the approach reported here. Water abstraction for  
414 public supply is also likely to be associated with significant fluxes of other inorganic N species, for

415 example meaning that quantifying the mass of N removed via abstraction of ammonium may also be  
416 important. Further research should also evaluate the ultimate fate of MWL-NO<sub>3</sub>-N and the  
417 timescales for re-release of N to the environment from transient stores associated with the water  
418 treatment cycle.

419

420 The research reported here at the national-scale highlights the particular importance of MWL-NO<sub>3</sub>-N  
421 in urban areas. However, due to the dearth of data at the WSZ level, there are a number of  
422 uncertainties in this study which should be considered when evaluating future research priorities. It  
423 should be noted that WSZ level leakage rates were estimated by applying percentage leakage rates  
424 from the corresponding WRZ. Whilst treated drinking water nitrate concentrations are reported at  
425 the WSZ level, there is likely to be some uncertainty in estimates of raw water nitrate concentrations  
426 as these have been made based on environmental regulator water quality databases and a heuristic  
427 assessment of water sources based on hydrogeological mapping and existing water transfers. Treated  
428 drinking water transfers are reported at the WRZ level and thus the impact of transfers on MWL-NO<sub>3</sub>-  
429 N fluxes has been reported at the WRZ level in this research. As a consequence of these factors, it is  
430 plausible that there may be more detailed local variability in MWL-NO<sub>3</sub>-N, ABS-NO<sub>3</sub>-N and the impact  
431 of treated water transfers in urban areas than reported in this research. Additional local-scale work  
432 should focus on these areas, drawing on datasets such as detailed raw water quality data, District  
433 Metering Area leakage data and information on water transfers.

434

435 Further research to quantify both ABS-NO<sub>3</sub>-N and MWL-NO<sub>3</sub>-N fluxes globally is also likely to be of  
436 importance. For example, water abstraction for all uses at the global-scale has been estimated at c.  
437 4000 km<sup>3</sup>/yr in 2010<sup>49</sup>. Using a conservative assumption that nitrate concentrations in raw abstracted  
438 water are half of the World Health Organisation guideline value (i.e. 5.65 mg N/L), global ABS-NO<sub>3</sub>-N

439 would remove 22.6 Tg N/yr from the aquatic environment. This is equivalent to between 5 and 39%  
440 of denitrification in soils, groundwater and riparian zones globally<sup>50</sup>. Global leakage rates have been  
441 estimated at 32 billion m<sup>3</sup>/yr<sup>51</sup>. Assuming the same nitrate concentration in leakage as in treated  
442 drinking water, leakage of N would contribute 0.525 Tg N/yr into the environment. This equates to  
443 approximately 1% of the global leaching and riverine N flux<sup>52</sup>. Whilst small in absolute terms, this ratio  
444 at the global-scale is similar to that reported above for England. Given the significance of MWL-NO<sub>3</sub>-  
445 N in urban areas in England we report above, it is plausible that MWL-NO<sub>3</sub>-N may be a significant N  
446 source in urban areas across the world. Further, both ABS-NO<sub>3</sub>-N and MWL-NO<sub>3</sub>-N may be more  
447 significant N fluxes in developing countries where nitrate concentrations and leakage rates<sup>51</sup> are likely  
448 to be higher than those reported here for England<sup>53</sup>. Therefore, accurately quantifying N fluxes  
449 associated with public water supply represents an important challenge to be addressed across the  
450 globe.

451

452

453

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455

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459

## 460 6 Supporting Information

461

462 *Methodology for Water Supply Zone mapping and extraction of treated drinking water nitrate*  
463 *concentration data*

464 *Figure S1 Location of planned treated drinking water transfers by 2020 and receiving water resource*  
465 *zones (WRZs) in England*

466 *Figure S2 MWL-NO<sub>3</sub>-N as a percentage of all N sources (as derived from) in receiving WRZs following*  
467 *implementation of treated drinking water transfers by 2020*

468

469

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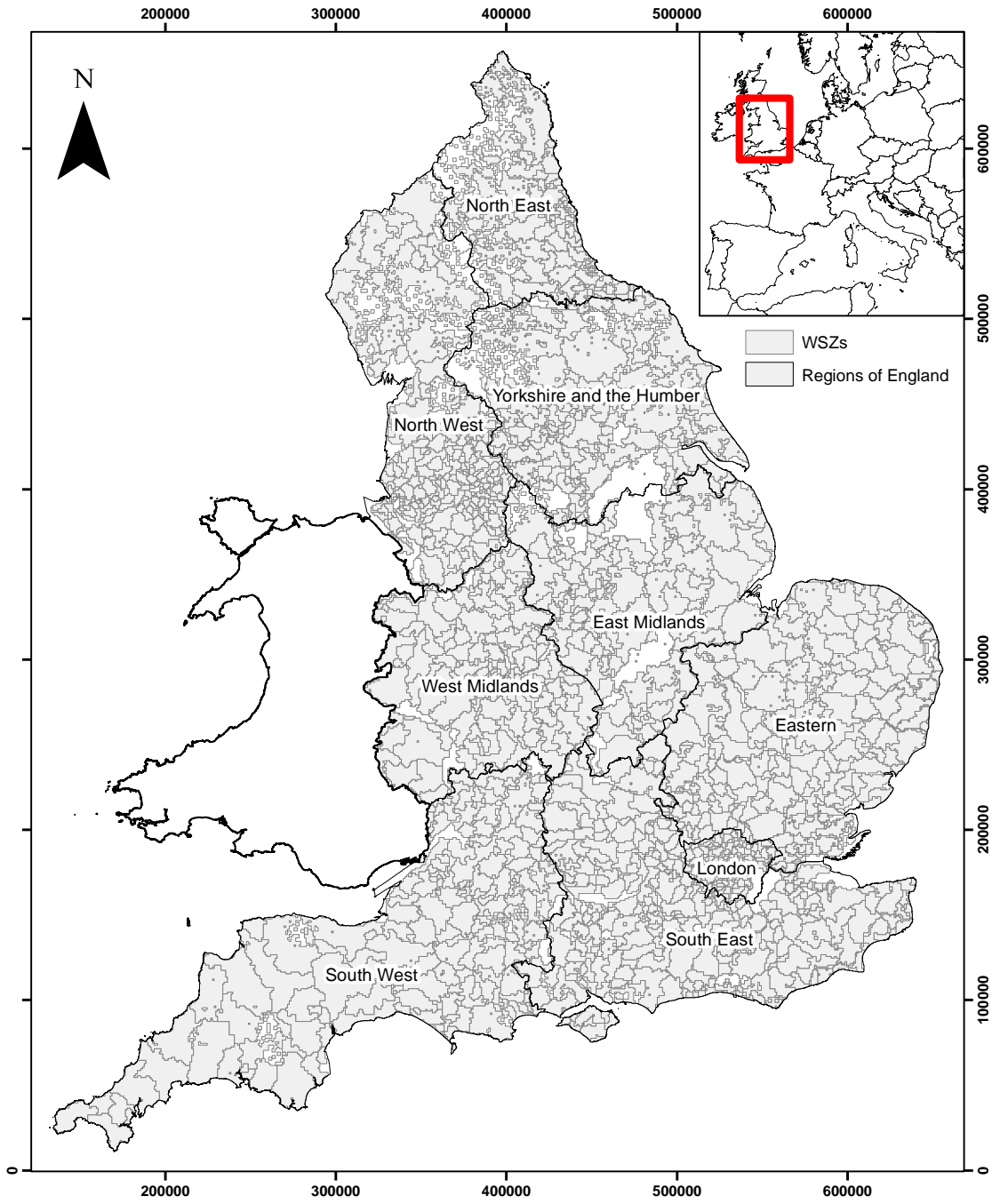
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652 **Figures**

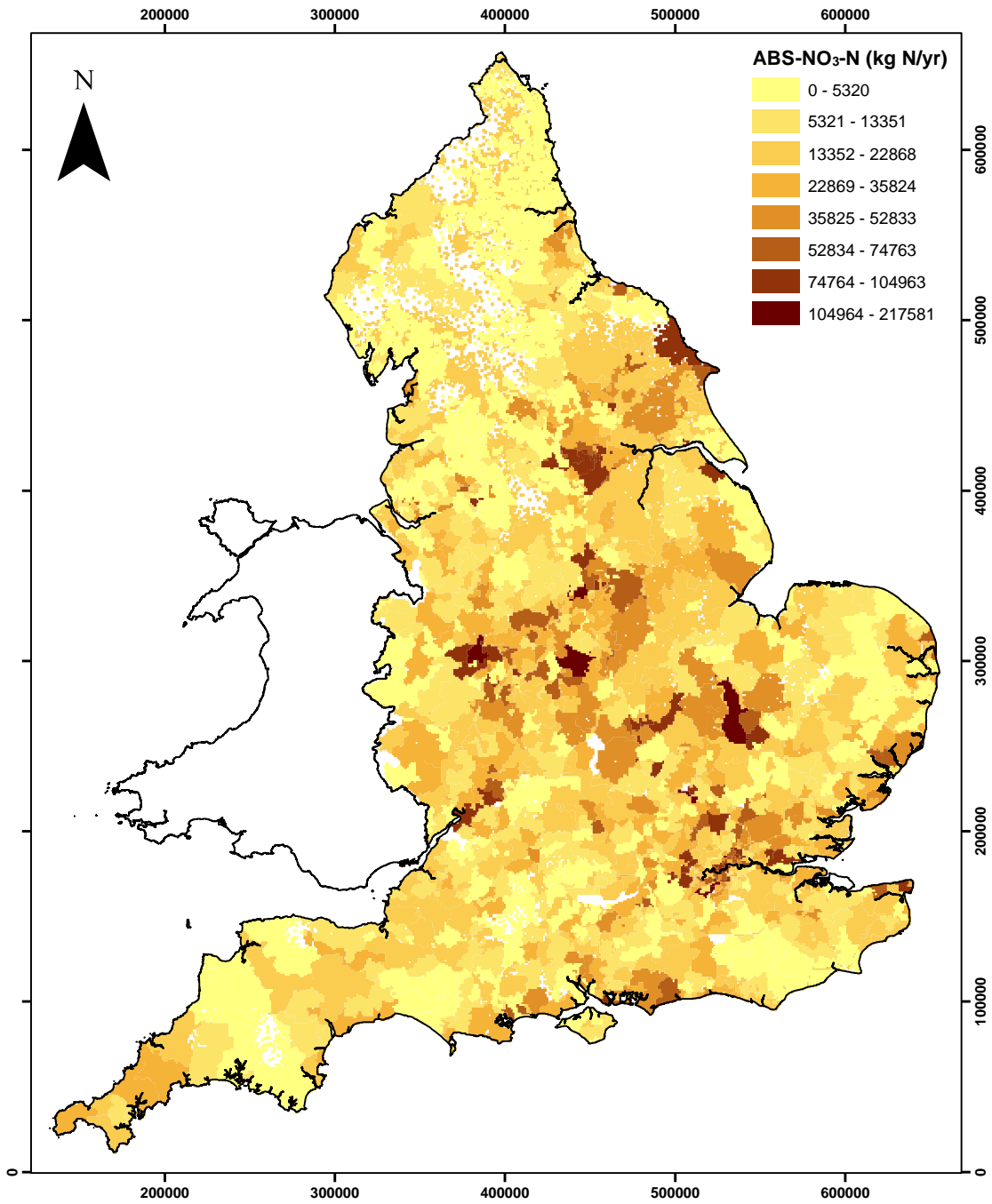
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655 *Figure 1 Water Supply Zones (WSZs) and principal regions in England*

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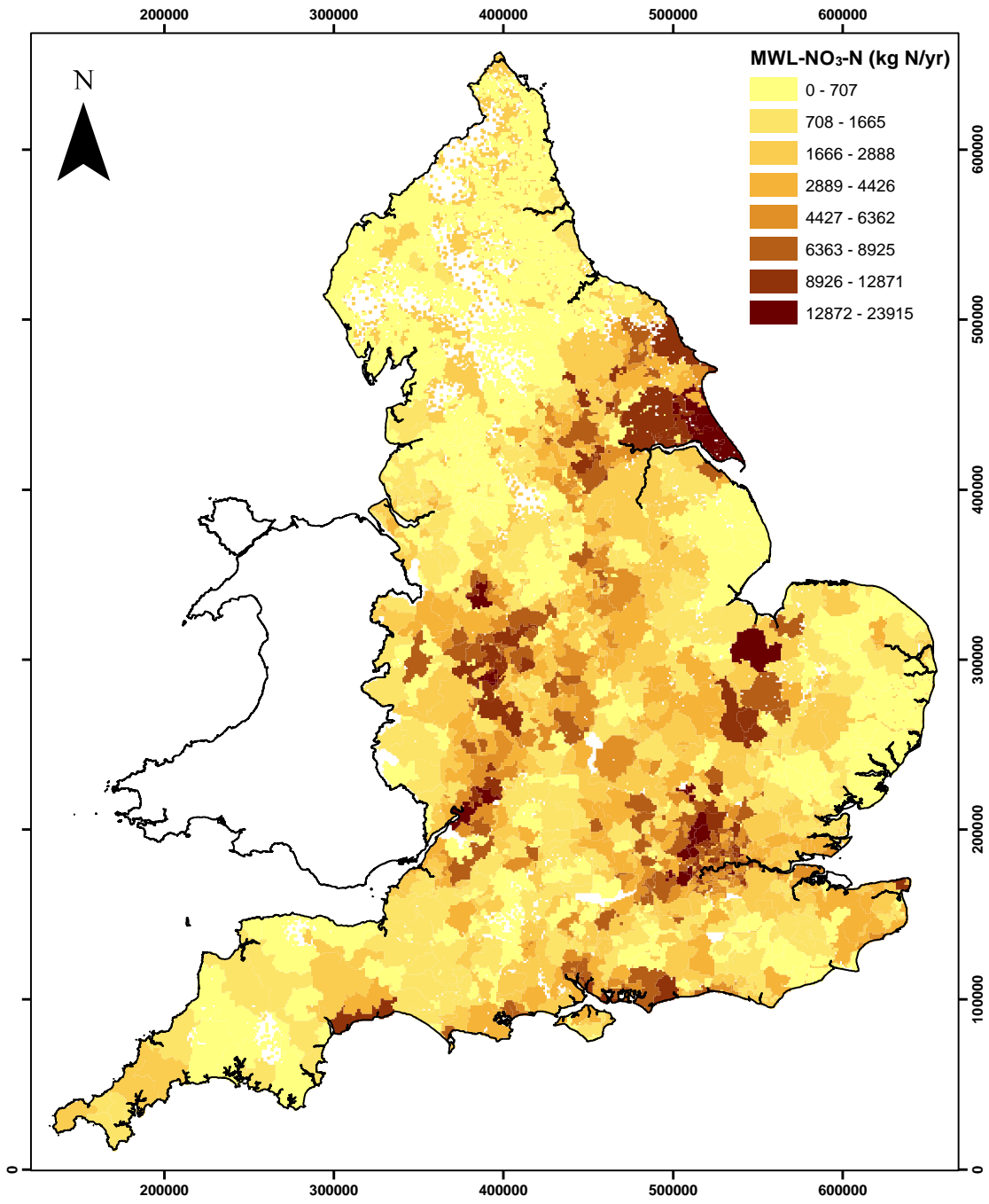


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658 *Figure 2 Distribution of ABS-NO<sub>3</sub>-N in England for 2015*

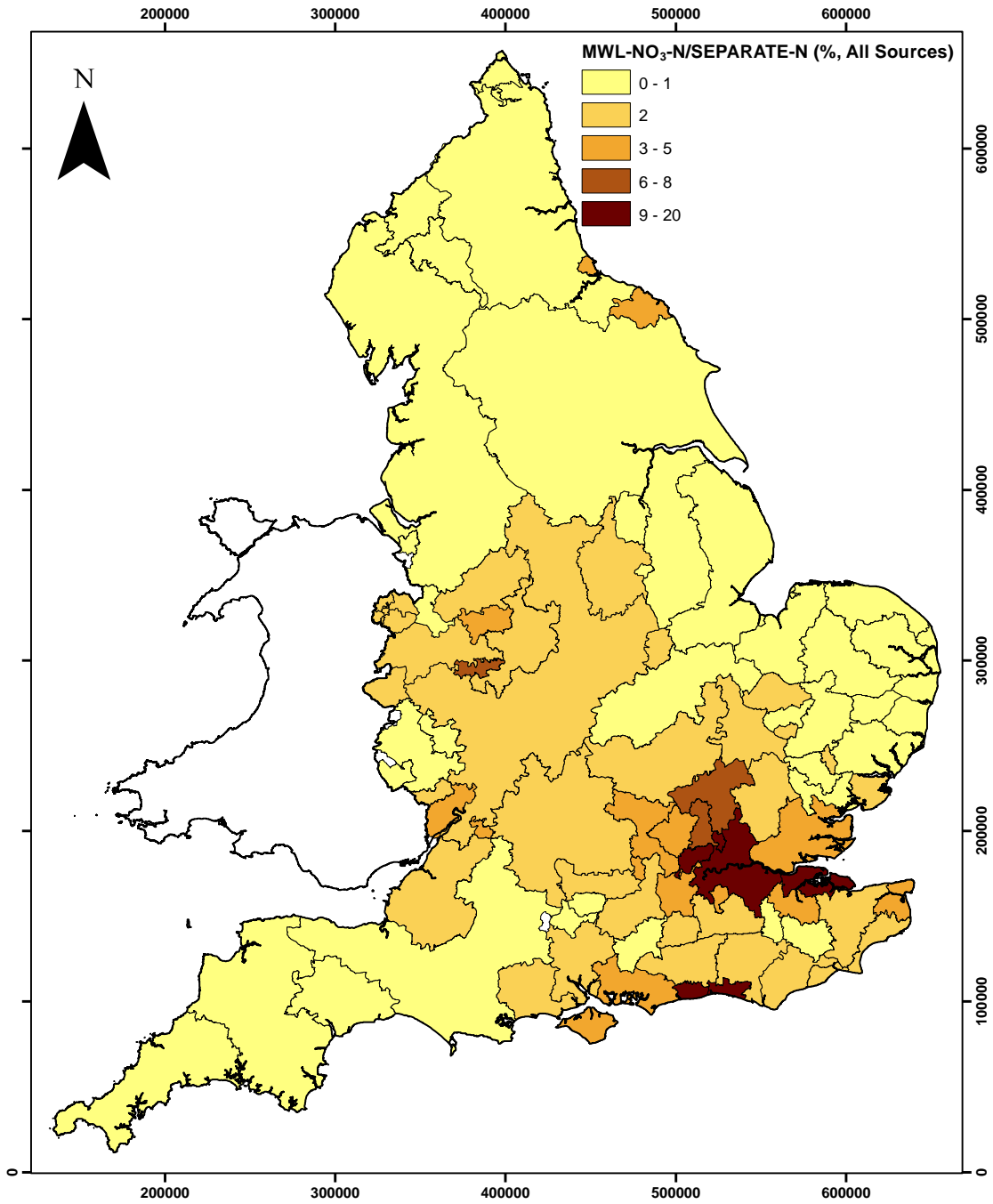
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662 *Figure 3 MWL- $\text{NO}_3\text{-N}$  fluxes for WSZs in England for 2015*



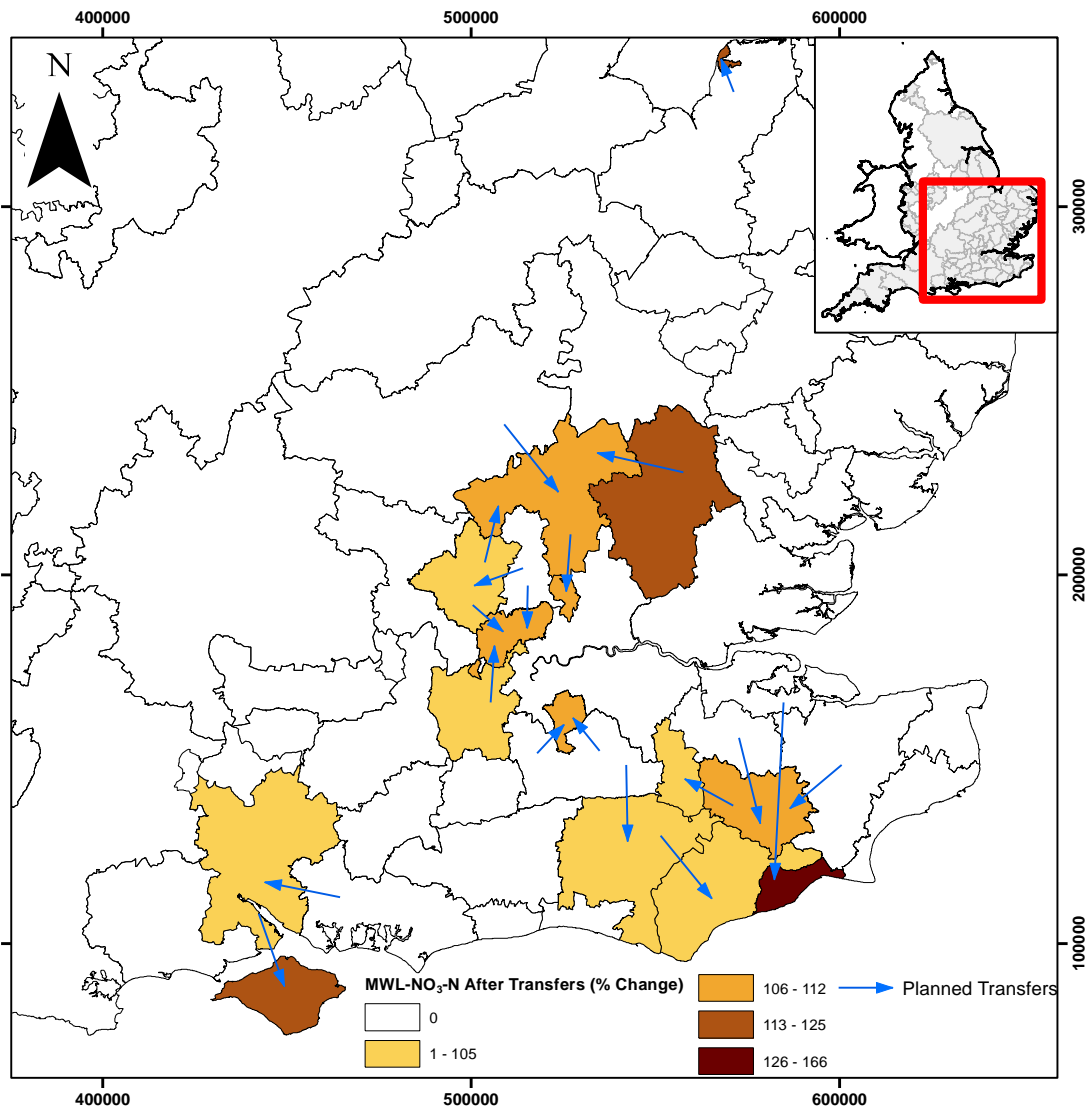
663

664 *Figure 4 Percentage of MWL-NO<sub>3</sub>-N to all N sources (as derived from SEPARATE<sup>20</sup>) at the WRZ scale in England for 2015*

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669 *Figure 5 Percentage change in MWL-NO<sub>3</sub>-N flux compared to 2015 baseline in receiving WRZs following implementation of*  
 670 *treated drinking water transfers by 2020*

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678 **8 Tables**

679

680 *Table 1 Total flux of nitrate removed from the aquatic environment by abstraction (ABS-NO<sub>3</sub>-N) in England for 2015 in*  
 681 *comparison to previous estimates of aquatic N retention (organic N retention and storage) and removal processes*  
 682 *(denitrification)*

<b>Flux Name</b>	<b>Reference</b>	<b>Flux Value (kt N/yr)</b>	<b>ABS-NO<sub>3</sub>-N/Flux Value (-)</b>
ABS-NO <sub>3</sub> -N	This study	24.2	-
Organic N retention by abstraction	Finlay et al. (2016)	3.9	6.21
Organic N storage in floodplains	Finlay et al. (2016)	0.8	30.03
Organic N storage in channel	Finlay et al. (2016)	1.6	15.02
Denitrification in the hydrosphere (rivers, groundwater and coastal marine environments)	Leip et al. (2011); Worrall et al. (2012)	62 -716	0.39 – 0.03

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685

686 *Table 2 Total flux of nitrate entering the environment from mains water leakage (MWL-NO<sub>3</sub>-N) in England for 2015 in*  
 687 *comparison to previous estimates of N sources (MWL-NO<sub>3</sub>-N, N from leaking sewers, all N sources, urban diffuse N sources).*  
 688 *MWL-NO<sub>3</sub>-N/Flux Value refers to the MWL-NO<sub>3</sub>-N estimate made in this study.*

<b>Flux Name</b>	<b>Reference</b>	<b>Flux Value (kt N/yr)</b>	<b>MWL-NO<sub>3</sub>-N/Flux Value (-)</b>
MWL-NO <sub>3</sub> -N	This study	3.62	-
MWL-NO <sub>3</sub> -N	Environment Agency (2013)	20.31	0.178
SEWER-N	Environment Agency (2013)	4.06	0.891
All N sources	Zhang et al. (2014)	277	0.013
Urban diffuse N sources	Zhang et al. (2014)	2.31	1.567

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