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HAL Authorization

1 **Long-term ecological observatories needed to understand socioecological systems in the**
2 **Anthropocene**

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26

Abstract

27 Over the last half century, humans have become the dominant force driving many of Earth's cycles.
28 Intensive agriculture and urbanization have simultaneously increased nutrient loading of pastoral landscapes and
29 decreased the capacity of these ecosystems to retain or remove excess nutrients. Widespread degradation of
30 terrestrial and aquatic ecosystems has triggered the establishment of ecological observatories, including the Zone
31 Atelier Armorique (ZAAR) in western France, a part of the International Long-Term Ecological Research network
32 (ILTER). The ZAAR includes a patchwork of land covers and uses, including primary growth forests, intensively
33 cultivated row crops, and ancient *bocage* fields surrounded by hedgerows. In addition to traditional ecological
34 research at ZAAR, which integrated pedology, hydrology, geochemistry, and hydrogeology, the last 8 years have
35 seen the development of multi-proxy and multi-scale approaches to address surface and groundwater quality. Here,
36 we present a global analysis of this 8 year dataset, including biodiversity, vegetation, soil water storage, and
37 stream and groundwater chemistry. Our results highlight a clear relationship between land use and surface water
38 quality, while groundwater quality appeared largely unrelated to land use, suggesting strong differences in
39 nitrogen removal rates. We observed differences among dry and wet years in nutrient fluxes, with multi-year
40 memory effects apparent for some parameters. Given such complex interactions, including emergent dynamics and
41 decadal to centennial time lags, we conclude that multidimensional observations such as those supported by the
42 ZAAR and other ILTER sites, are critical to understanding socioecological systems in the Anthropocene.

43 Keywords: Long-term monitoring, anthropogenic forcing, land use, ecosystem vulnerability, ecosystem
44 resilience, multi-proxy, multi-scale, heterogeneity

45

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50 **1. Introduction**

51 At a global scale, human activity has surpassed geological and biological forces in many dimensions
52 (Steffen et al. 2007, 2011; Berkhout 2014). While there is a controversy over the starting date and the purview of
53 the Anthropocene (Crutzen 2002; Monastersky 2015), widespread anthropogenic effects on aquatic ecosystems
54 and biodiversity expanded exponentially in the 20th century (Steffen et al. 2011; Zalasiewicz et al. 2015). This
55 period was marked by accelerated industrialization, technological advances, and significant economic and
56 demographic growth. Industrial fertilizer production and increases in international trade resulted in a step-change
57 in agricultural output in the decades following the Second World War (Rudel et al. 2009). Combined with large-
58 scale changes in land management (e.g. field consolidation and soil drainage), agricultural production caused
59 widespread degradation of surface water and groundwater quality (Van Meter et al. 2016; Abbott et al. 2018) .
60 Local and global increase of carbon, nitrogen, and phosphorus cycles have been accompanied by changes in the
61 global water cycle and energy balance (Loaiciga 1996; Huntington 2006) threatening biodiversity and human
62 water and food security (Meybeck 2003; Steffen et al. 2015).

63 Despite the degree and extent of human activity, quantifying anthropogenic effects in terrestrial and
64 aquatic ecosystems remains challenging due to lack of appropriate baselines, incomplete time series, and
65 approaches that often focus on individual response variables (Alagona et al. 2012; Worrall et al. 2015; Pinay et al.
66 2018). Additionally, while degradation of ecosystem function and services is proportionally related to human
67 disturbance in some (Howarth 2008) instances, there is growing evidence that ecosystem characteristics strongly
68 modulate resistance to anthropogenic pressures and resilience or recovery trajectory following disturbance (Helton
69 et al. 2018). Because decadal to centennial time lags are often involved in complex interactions in socioecological
70 landscapes (Slavik et al. 2004; Worrall et al. 2015; Abbott et al. 2016; Kolbe et al. 2016), long-term and multi-
71 dimensional observations are needed for sustainable management in the Anthropocene (Burt et al. 2011). Taking
72 water quality as an example, substantial resources have been invested in assessing the impact of agricultural
73 activities on water quality and creating mitigation and adaptation strategies (Singh and Sekhon 1979; Haag and
74 Kaupenjohann 2001; Rodvang and Simpkins 2001).

75 In this paper, we perform a global analysis of such research carried out at a long-term ecological research
76 site in western France. Data were collected by hydrologists, soil scientists, landscape ecologists, hydrogeologists,
77 and sociologists, allowing us to explore ecohydrological linkages and transfers between different catchment
78 components and to test how human activity has influenced a complex landscape. We first provide an overview of
79 the site and the scope of observation, followed by an analysis of ecohydrological functioning at multiple
80 spatiotemporal scales.

81 **2. Zone Atelier Armorique (French LTER)**

82 In Brittany, France, research on surface and subsurface ecology, hydrology, and biogeochemistry has
83 been carried out since 1993 at an international long-term ecological research (ILTER, (Mirtl et al. 2018; Haase et
84 al. 2018) site named the Zone Atelier Armorique (ZAAR). The ZAAR is one of 14 French environmental
85 observatories (<http://www.za-inee.org/fr/reseau>), and is also integrated into the European, (www.lter-europe.net)

86 and international LTER (<https://www.ilternet.edu/>) networks. Located in western France (48° 36' N, 1° 32' W;
87 Fig.1a), the ZAAr was created to conduct interdisciplinary research analysing human activities and their effects on
88 environmental systems including hydrology, ecology, and biodiversity in a socioecological context. The ZAAr
89 tries to promote a multidisciplinary approach to understanding socioecological problems, which do not respect
90 traditional boundaries between disciplines and environments (e.g. terrestrial-aquatic, surface-subsurface). Such
91 networks aim to integrate complexity of environmental systems with the interactions between natural systems and
92 society. Specifically, the ZAAr aims to understand the linked dynamics and co-evolution of human and water
93 systems (Thomas et al. 2016b; Abbott et al. 2017; Marcais et al. 2018). A long-term monitoring strategy was
94 developed in the ZAAr to investigate interactions between agricultural practices, landscape structure (e.g.
95 hedgerows, topography, and river networks), water quality, and catchment hydrology. Variable spatial and
96 temporal characteristics in the ZAAr have provided a high-resolution picture of biogeochemical and hydrological
97 drivers of soil, subsurface, and surface water quality.

98 Research at the ZAAr has been carried out on a long-term basis in partnership with local landowners and
99 decision makers (e.g. local governments) to develop and quantify effectiveness of adaptation and mitigation
100 strategies for sustaining water quality, biodiversity, and human quality of life. The ZAAr focuses on agricultural
101 and urban landscapes and employs both observational and experimental scientific approaches. The ZAAr is
102 composed of three contrasted landscape units: (i) a landscape intersected by a hedgerow network in the *Pleine-*
103 *Fougères* area of 93 km², (ii) an alluvial plain of the *Couesnon river* which is a Natura 2000 site, and (iii) an urban
104 area in the city of Rennes. These units have been monitored since 1993, 2006, and 2011, respectively. In this
105 paper, we will focus on the *Pleine-Fougères* ZAAr site (which we refer to simply as the ZAAr), because it is the
106 site with the longest land-use, ecology, and biodiversity time series (i.e. 25 years), as well as high frequency
107 hydrological data since June of 2009.

108 **2.1 Land-use history and effects on water quality**

109 Farming in north-western France started millennia ago, consisting of small cultivated fields interspersed
110 with nut and apple trees (Forman and Baudry 1984). Intensive farming appeared in Brittany after the Second
111 World War, when many agricultural fields were consolidated and many hedgerows were cut down (Thomas and
112 Abbott 2018). The hedgerow network covers the ancient landscape of the *Massif Armoricain*. Dairy farming is the
113 dominant agricultural system. Cows are mostly fed from maize and grass (rotational and permanent grassland) and
114 imported proteins (soybean). Large farms also grow wheat.

115 Current land use of the ZAAr is dominated by corn, wheat, and pastureland, which follow topographic
116 and geological gradients in the ZAAr (Thomas et al. 2016a). There is a higher density of hedgerows and more
117 pastures for dairy cows in the southern half of the ZAAr, which is granitic and hilly, while there are larger, open
118 fields in the schist lowlands to the north (Fig. 1b). Deeper, loamy soils of 80 to 85 cm have developed on the
119 granite bedrock in the south part of the ZAAr and shallower soils of 45 to 70 cm have developed on the schist
120 bedrock in the northern part (*UMR 1069 SAS INRA - AGROCAMPUS OUEST*). The difference in elevation
121 between the south and the north of the catchment is about 110m (Fig. 1b). The estimated input of nitrogen,
122 including manure and synthetic fertilizer, is around 164 kg N per ha per year, exceeding agricultural nutrient
123 removal by ~30% (van Grinsven et al. 2012; Poisvert et al. 2016).

124 The long legacy of land use and excess nutrients have degraded aquatic ecosystems in this area, including
125 lakes, rivers, groundwater, and estuaries (Ben Maamar et al. 2015; Kolbe et al. 2016; Abbott et al. 2018). Average
126 nitrate concentration increased from 9 mg L⁻¹ in 1976 to 65 mg L⁻¹ in 1989 (Cheverry 1998), exceeding numerous
127 regulatory limits (European Nitrate directive 1991), and causing widespread harmful algal blooms along Brittany
128 coasts (Perrot et al. 2014). National and international political actions and legal frameworks were set up to reduce
129 fluvial nutrient loads at European, national, and local scales (PMPOA; Action programs for the nitrate directive;
130 European Nitrate directive; Brittany Pure Water *Bretagne Eau Pure – BEP*; the governmental plan against green
131 algae). The efficiency of these actions is unclear, with some areas where water quality parameters show
132 improvement, and others areas appearing to remain stable (Bouraoui and Grizzetti 2011; Abbott et al. 2018). For
133 example, nitrate concentration in surface water shows a slight improvement, and to a lesser extent, in shallow
134 groundwater (Aquilina 2012). As elsewhere, recovery trajectories (ecosystem resilience) depends not only on
135 nutrient inputs, but also on retention and removal capacity in different catchment components (i.e. stream,
136 groundwater, wetland, riparian vegetation, hedgerows, etc. (Pinay et al. 2018). Specifically, nitrate retention and
137 removal in a catchment depend on physical and biological conditions in soils, groundwater, and streams, which
138 determine assimilatory uptake and dissimilatory use in denitrification (Thomas and Abbott 2018). Denitrification
139 requires nitrate, an electron donor (e.g. carbon or pyrite), and anoxic conditions that co-occur in areas such as
140 wetlands, aquifers, and hyporheic zones (Gilliam 1994; Vidon and Hill 2004; Trauth et al. 2015). Areas of
141 preferential denitrification change in space and time (McClain et al. 2003), due to variation in hydrological and
142 biological conditions (Moatar et al. 2017), making the assessment of the removal capacity at catchment-scale
143 difficult. The distribution of nutrient sources and sinks is strongly influenced by landscape structure, including
144 hedges, embankments, and ditches (Bernhardt et al. 2003; Hall et al. 2009; Worrall et al. 2012; Schelker et al.
145 2016), though these relationships do not always hold at small spatial scales (Burt 2005; Abbott et al. 2017). At the
146 catchment scale, hedgerows play an important role in the regulation of floods (Merot 1999), and at the local scale,
147 they can alter water-table depth and nutrient transfer (Caubel-Forget and Grimaldi 2001; Guo et al. 2007; Thomas
148 et al. 2016a).

149 **3. Methods**

150 **3.1 Land use and meteorological data**

151 From 1993 until 2013, land use mapping was carried out annually in the ZAAR. Digital maps were
152 produced from summer aerial photography by geographers at the University of Rennes 2 (COSTEL, UMR LETG).
153 This detailed classification was used to evaluate agricultural practices, in combination with regional accounting
154 (farmers reporting their land use directly as required by law into a geographic information system). These two
155 types of data provided information on how crops were distributed among farms and farm types. Finally, detailed
156 ground surveys have been carried out since 1995 four times a year from the same 300 field margins distributed
157 across areas of the ZAAR with different field margin densities.

158 Meteorological data were available from the Météo France weather station located in *Louigné du desert*,
159 37 km from the outlet of the studied area. A weather station was installed in 2009 inside the ZAAR to record air
160 temperature, humidity, wind speed and direction, global and reflected radiation, and soil temperature at 3 depths

161 (50, 100, and 180 cm) at a 5 minute interval. An automatic rain gauge recorded instantaneous pulses of rainfall
162 with a 0.24mm resolution. A manual rain gauge was also installed to verify cumulative rainfall.

163 **3.3 Hydrological monitoring**

164 Since 2009, high-frequency monitoring of soil moisture, groundwater, discharge, and water quality has
165 been performed with a particular focus on the role of landscape structures in the variability of nitrogen fluxes in
166 the ZAAr. The time step of our measurements ranged between 10 minutes for groundwater level and discharge, 2
167 weeks for soil moisture, and monthly for water quality. Seventeen agricultural wells from across the ZAAr were
168 selected for water quality analyses (Fig. 1b) and thirteen piezometers were installed on a highly instrumented
169 hillslope crossed by 3 hedgerows near the boundary of a wetland. Groundwater level was measured for each
170 piezometer using pressure head sensors from OTT (Orpheus mini). Table S1 summarises the characteristics of the
171 groundwater wells and shallower piezometers.

172 Soil moisture at the instrumented hillslope was measured at 25 locations using a capacitance sensor
173 (Diviner 2000, Sentek) (Paltineanu and Starr 1997; Green et al. 2006). Measurements were performed along a soil
174 profile of 140 cm. Water storage (ΔS_{year}) was calculated from monthly or bimonthly soil moisture measurements
175 to a depth of 60 cm for sensors located within and beyond the rooting zone. For each year and each location, the
176 maximum (θ_{max}) and minimum (θ_{min}) values were used to assess water storage (ΔS_{year}) using Equation 2.

$$177 \quad \Delta S_{year} = \int_0^z (\theta_{max} - \theta_{min}) \cdot dz \quad \text{Equation 2}$$

178 Stream discharge and water temperature were monitored for 13 small catchments within the ZAAr,
179 ranging from 2 to 35 km² (Fig. 1b). Data from one of the 13 catchments and 2 additional catchments were
180 collected from 1996 to 1999 and from 2012 until 2017 (catchments G1, S1 and GS1; Thomas et al 2016). Table S2
181 summarises the characteristics of the stream water data of these 15 catchments.

182 Additional discharge and nitrate concentration data were available from the public institution DREAL
183 (*Directions Régionales de l'Environnement, de l'Aménagement et du Logement*) at two hydrometric stations
184 adjacent to the ZAAr (*Le Guyoult* and the *Couesnon Rivers*; Fig. 1b). Maps of weathered layer thickness, aquifer
185 thickness, and bedrock are available through the French geological survey (BRGM), and hydrological and
186 geological information are available through openGIS web map services (WMS; Kolbe et al., 2016).

187 Monthly to bimonthly stream and groundwater samples were collected from December 2012 to June
188 2016. Stream water from 6 nested catchment outlets were sampled at each date (Fig. 1b) and 2 additional
189 catchments (G1 and S1) from Thomas et al. (2016) were also integrated in our global analysis. Water samples
190 from surface sampling points and groundwater wells were filtered to 0.45 μm through polyethersulfone filters and
191 major anions were measured using ion chromatography as described in (Thomas et al. 2016). Table S2 summarises
192 the frequency of sampling and the list of parameters analysed.

193 All the time series analyses (i.e. precipitation, discharge, potential evaporation, groundwater level, water
194 quality, and soil moisture) were calculated for each hydrological year from 1 of September to 31 of August of the
195 next year.

196 **3.5 Temperature monitoring**

197 In addition to air temperature measurements collected at the weather station, individual temperature
198 probes were installed to characterise the thermal signature in stream water T_{sw} , groundwater T_{gw} , multiple depths
199 of the hyporheic zone T_{hypo} (3 or 6 depths for each site), and 3 key-locations in the soil along the hillslope:
200 recharge zone; $T_{soi_{rz}}$, for the mid and bottom locations; T_{soi_m} , and T_{soi_b} , respectively. Some of the temperature
201 sensors were installed at the beginning of the monitoring period (September 2009), allowing quantification of inter
202 and intra-annual variation as well as long-term trends.

203 We also deployed fibre optic distributed temperature sensors (T_{fo} ; FO-DTS) along a 600 m reach in one
204 of the nested catchments. Continuous FO-DTS measurements allowed characterization of the location and time of
205 stream-groundwater exchange. Spatial resolution of the FO-DTS was about 0.5m with a sampling frequency of 20
206 minutes from July 2016 to July 2017.

207 **4. Results**

208 **4.1 ZAAr land use evolution**

209 Land use changed substantially from 1993 to 2013 (Fig. S1). In 1993 to 1998, pasture occupied 15-30 %
210 of the landscape, increasing to 35-45% in 1999 to 2013, with a maximum of around 60 % in 2009. Maize and
211 cereal cultures covered 15-25 % of the area from 2000 to 2009, increasing to 40% in 2010 to 2013. Initially, 15-
212 28% of the landscape was not used for agricultural practices, decreasing to ~10% in 2010. The forested area
213 (~10% of the landscape) was stable over the investigated time period. Wild field margins represented less than 3 %
214 of the ZAAr area initially, decreasing to less than 1% in 2010. These trends towards less permanent pasture and
215 more cultivated land matched larger regional changes from 2006 to 2014 (Public Sata Source : RPG Ille-et-
216 Vilaine).

217 **4.3 Hydrological compartments characterization**

218 **4.3.1 Soil moisture and groundwater variation under a contrasted climatic context**

219 Historical data from Meteo-France over a 30 years period revealed substantial variability in precipitation
220 and evapotranspiration (Fig. 2). The hydrological years 1988-1993, 1995-1998, 2001-2006, 2009-2012, and 2014-
221 2016 were relatively dry with net precipitation (P-PET) lower than 200 mm. 2009-2010 and 2016-2017 were the
222 driest years with negative net precipitation of -10 mm and -137 mm, respectively. The wettest hydrological years
223 were 1998-2000, 2006-2007, and 2013-2014 with net precipitation between 450 and 600 mm (Fig. S2). The
224 cumulative rainfall was lowest in 2009-2010 (736 mm) and highest in 2013-2014 (1180 mm; Fig.2 and Fig. S2).

225 Soil moisture measurements at the recharge zone of the hillslope within and beyond the rooting zone
226 were used to calculate water storage (ΔS_{year}) (Fig. 3). Soil water storage in both zones followed net precipitation
227 except for the beginning of the observed time series. Soil moisture within the rooting zone was systematically drier
228 in 6 of the 8 observed years, independent of other hydrological changes. Water storage was highest beyond the
229 rooting zone in the middle of a meadow far away from the root system of hedgerows.

230 Groundwater level in the recharge zone (P02 in Fig. 5) was most variable for the 8-year period. 2016-
231 2017 showed the lowest variability in groundwater level, with only 3 m of change observed during the
232 hydrological year, contrasted with 8 m of change in 2012-2013 and 2013-2014, when net precipitation was high
233 (Fig. S3). Annual minimum groundwater level was surprisingly stable across the 8-year time series, varying only
234 ~70 cm. The highest groundwater level observed during the low water period was ~12 m A.S.L in August 2014
235 and the lowest was 11.3 m A.S.L. in early November 2016.

236 **4.3.3 Stream discharge**

237 Long-term discharge over 30 years at the *Couesnon* gauging station showed highest discharge in 2000-
238 2001, the wettest year of the period of record, and the lowest discharge in 1991-1992 (Fig.2). 2009-2010 was the
239 driest year, but fall and winter precipitation was high, preventing extreme low flows in the late fall. Discharge
240 remained high from September 2013 to September 2016 despite a small decrease in annual rainfall over these
241 years (Fig. 5, Fig. S2). The hydrological year 2016-2017 was extremely dry, surpassing the 2 previous dry years
242 with low cumulative rainfall, high cumulative PET, and extremely low maximum discharge (Fig.2).

243 **4.4 Biogeochemical reactivity in catchment compartments**

244 **4.4.1 Spatial analysis of nitrate concentrations**

245 Mean nitrate concentration in deep groundwater (i.e. 28 to 98m) showed high spatial variability over the
246 ZAAr area (Fig. 4b). No clear relationship between well depth and nitrate concentration was found. The wells
247 located on granite bedrock tended to have nitrate concentration lower than 35 mg L⁻¹ (wells F1 to F6), the well
248 F16, at the transition between granite and schist, had a very high concentration of nitrate (75 mg L⁻¹), and 5 of the
249 10 wells in schist bedrock had no nitrate (F7, F9, F10, F11 and F12) but the other half had concentration between
250 33 and 64 mg L⁻¹ (F8, F13, F14, F15 and F17).

251 Nitrate concentration measured in the shallow groundwater (i.e. <10 m from soil surface (P1 to P13))
252 showed a gradient between the recharge zone and wetland (Fig. 5a and B). In the recharge zone, high nitrate
253 concentration (40-60 mg L⁻¹) was observed, approximately the same concentration as the deep groundwater (Fig.
254 5b). Nitrate concentration was highest (~70 mg L⁻¹) at the mid-slope where an artesian upwelling zone occurred
255 (Clement et al. 2003). Nitrate concentration decreased abruptly from 65 to 0 mg L⁻¹ in the wetland.

256 Figure 4a shows the mean nitrate concentration in stream water measured over the periods indicated in
257 Table S2. Overall, concentration was much less variable in surface waters than in groundwater. For the headwater
258 catchments located in the granitic part of ZAAr, nitrate concentration was lower than 25 mg L⁻¹ (Fig. 4a). Nitrate

259 concentration increased with drainage area and intensive agricultural land contribution but the mean value
260 remained below 40 mg L^{-1} except for the catchments S1 and BV8, and *Couesnon* station.

261 At the catchment scale, there was generally a good agreement between land use type and surface-water
262 exports, with nitrate concentration increasing with intensive row-crop land use (Fig. 4a). These results highlight a
263 direct relationship between land use and nitrate concentration in stream water. However, there was little or no
264 relationship between land use and groundwater nitrate concentration, suggesting that ecosystem removal processes
265 control water quality in this catchment component.

266 **4.4.2 Temporal analyses of nitrate concentration and sources**

267 We tested for interannual trends in nitrate concentration using mean nitrate concentration for each year
268 across all sites within each of the different hydrological compartments (i.e. deep groundwater, shallow
269 groundwater, wetland, near stream-groundwater, and stream water). Nitrate concentration was high for deep and
270 shallow groundwater and showed more temporal variability in shallow than in deep groundwater. The
271 concentration in the wetland was very low or below detection through the years. Temporal variability (as indicated
272 by the standard deviation) decreased over the time series, though part of this is likely due to changes in sampling
273 frequency (Fig. 7). Mean annual nitrate concentration and discharge were correlated, though this linkage differed
274 across timescales. At a finer time scale (Fig. 6), stream nitrate concentration varied between groundwater
275 concentration and near stream groundwater. However, starting in the winter of 2014, we observed an increase in
276 nitrate concentration at the beginning of the recharge period and a decrease until the end of recharge period.

277 Stream discharge over the 29 years period showed high variability, ranging from $2.5 \text{ m}^3 \text{ s}^{-1}$ to $19 \text{ m}^3 \text{ s}^{-1}$,
278 but nitrate concentration was quite stable in time, ranging less than a factor of 2 from 32 mg L^{-1} to 50 mg L^{-1} (Fig.
279 6). Seasonal analysis of mean nitrate concentration and discharge showed no correlation except during the winter
280 where dilution is observed as nitrate concentration decreases when discharge increases (Fig. S4).

281 We used the ratio of nitrate to chloride ($\text{NO}_3:\text{Cl}$) to identify potential processes controlling nitrate
282 concentration (Fig. 8). Three processes can be identified from the $\text{NO}_3:\text{Cl}$ ratio: (i) denitrification is characterized
283 by a $\text{NO}_3:\text{Cl}$ ratio less than 1 and low chloride concentration, (ii) agricultural inputs are characterized by a $\text{NO}_3:\text{Cl}$
284 ratio much higher than 1 and low chloride concentration, (iii) and sewage inputs are characterized by a $\text{NO}_3:\text{Cl}$
285 ratio much smaller than 1 and high chloride concentration. Deep wells fell into two groups, with F3, F8, F14, and
286 F15 expressing a high $\text{NO}_3:\text{Cl}$ ratio and low chloride concentration (Fig. 8a), and the other deep wells showing a
287 $\text{NO}_3:\text{Cl}$ ratio <1 and low chloride concentration.

288 In the shallow GW (P1-P11), the $\text{NO}_3:\text{Cl}$ ratio as well as the chloride concentration were highly variable
289 and changed over time (Fig. 10b). In the near stream and wetland area $\text{NO}_3:\text{Cl}$ ratio was lower than one and
290 chloride concentration was low (Fig. 10c). The stream water ratio (Fig. 8a) varied in time except for the catchment
291 located in the forested zone, which had a very low $\text{NO}_3:\text{Cl}$.

292 **5. Discussion**

293 **5.2 Vulnerability of water bodies to nitrate contamination**

294 Nitrate concentration at the ZAAr seems to be controlled by two main processes: agricultural inputs and
295 denitrification. Wetland and stream NO₃:Cl ratios suggest that denitrification is the main process controlling
296 nitrate concentration in these hydrological compartments. For the deep groundwater, some wells showed a NO₃:Cl
297 ratio characteristic of agricultural inputs while others showed a NO₃:Cl ratio characteristic of denitrification. This
298 agrees with observations that pyrite and anoxic conditions in aquifers can be very localised (Ben Maamar et al.
299 2015). For the shallow groundwater in the hillslope, there was high temporal and spatial variation in nitrate
300 concentration (Fig. 6). The nitrate signature in piezometers located at the crest to the mid-slope suggests that
301 agricultural inputs are the main source of nitrate to the system. The nitrate signature of the wells at the mid-slope
302 hedgerow suggests that denitrification occurs at this location, probably stimulated by the presence of the hedgerow
303 (Caubel-Forget and Grimaldi 2001; Grimaldi et al. 2012; Thomas and Abbott 2018). This is the case as well for
304 the piezometer P11 which is localised in a more forested area of the catchment and nevertheless has a nitrate
305 concentration of ~40 mg l⁻¹.

306 These results suggest that for surface-water compartments such as streams, riparian areas, wetlands, and
307 near-surface groundwater are quite vulnerable to nitrate contamination from agricultural activity. On the other
308 hand, deep groundwater was resistant to excess nutrient inputs, with ecosystem characteristics, in this case
309 conditions for denitrification (Kolbe et al. 2016; Abbott et al. 2016; Marcais et al. 2018), determining water
310 chemistry. However, on longer time-scales those favourable conditions for denitrification could change,
311 potentially due to depletion of electron donors or changes in groundwater recharge caused by climate change
312 (Tesoriero et al. 2000; Jasechko et al. 2017). For both surface and groundwater, best management practices such as
313 cover crops, riparian buffers, and hedgerows could increase ecosystem resistance and resilience to excess
314 nutrients, while also enhancing other ecosystem functions and services (Caubel-Forget et al. 2001; Grimaldi et al.,
315 2012; Thomas and Abbott, 2018; Pinay et al. 2018).

316 As for many complex landscapes, the ZAAr is experiencing multiple changes in pressures and internal
317 structure simultaneously. Variability of nitrate concentration and flux in time was mainly driven by meteorological
318 conditions and associated changes in discharge (Moatar et al. 2017; Thomas et al. 2016a). Spatial variability, on
319 the other hand, depended on landscape structure, including subsurface denitrification capacity, the presence of
320 wetland and hedgerows, and land use. Climate is a second forcing to explain nitrate concentrations in the stream
321 water. Consequently, the overall ecosystem state of this landscape in the future will depend on the interaction
322 between direct human disturbance and climate change.

323 **5.3 Using long-term studies to promote socio-hydrological research**

324 Understanding processes at the catchment scale requires multidisciplinary approaches capable of
325 characterizing human actions such as land use, physical characteristics such as geology and hydrology, and
326 ecological processes such as denitrification, succession, and migration (Alvarez-Cabria et al. 2016; Malone et al.
327 2018). While involving multiple perspectives is laudable and in many cases essential, it also poses several
328 challenges, including:

- 329 (i) Identifying processes and parameters of concern in the socioecological domain, which are not
330 easily comparable

- 331 (ii) Learning disciplinary language and perspectives, which often use the same terms or interpret the
332 same findings differently
- 333 (iii) Obtaining relevant metrics and measurements to quantify vulnerability, resilience, and
334 adaptation capacities
- 335 (iv) Generalizing these metrics and measurements to predict ecosystem functioning in areas with less
336 data or without data

337 Two characteristics of socio-hydrological systems is that they scale by stream order and connectivity and
338 that they are increasingly dominated by anthropogenic drivers (i.e. direct disturbance and climate change
339 (Meybeck 2003; Helton et al. 2012; Savenije et al. 2014). This is particularly apparent in the ZAAr, where
340 connectivity between surface water and groundwater varies spatially from small headwater subcatchments to large
341 lowland catchments, integrating multiple landscape types and disturbance modes. The contribution of nutrients
342 from each tributary depends on multiple overlapping temporal signals stemming from weather, climate, land use,
343 topography, soil thickness, and geological bedrock properties (Gao et al. 2016; Abbott et al. 2017). Characteristic
344 response times of water or elemental flow depend on the distribution of transit times, which itself depends on
345 system size and properties fixing the balance between vulnerable young water systems and resilient old water
346 systems (Marçais et al. 2015; Abbott et al. 2016; Jasechko et al. 2017).

347 Planning and decision making for socio-hydrological systems that do not integrate the interactions
348 between multiple scales and dimensions are bound to fail, as they do not consider how upstream and downstream
349 effects in space and time can alter adaptation and mitigation or increase vulnerability. It will likely always be
350 difficult to determine multi-dimensional ecosystem resistance and resilience, but long-term and diverse
351 observations give us the best chance to apply precautionary principles and protect ecosystems in the
352 Anthropocene. A spatial approach can help map vulnerable patches, and repeat sampling can quantify how these
353 patches change in time, allowing identification of both spatial and temporal drivers of ecological functioning
354 (Abbott et al. 2017).

355 **5.4 How to promote sustainable management**

356 Landscape management and land-use distribution are both strongly contingent on the past, including
357 cultural, technological, and ecological legacies. In an engineering perspective, achieving sustainable management
358 simply entails controlling human activities so they do not exceed the capacity of natural environments. However,
359 neither collective human behaviour nor ecological dynamics can be handled on engineering principles. Over the
360 past 50 years, changes in agricultural techniques and regional management have significantly increased food
361 production and slowed ecological degradation in many areas, achieving ecological and societal good
362 simultaneously. In the same time period, anthropogenic climate change is increasing environmental systems'
363 variability, potentially undermining the mutually beneficial shifts in management. The current challenge is to
364 throttle human footprint adaptively to match dynamic regional carrying capacities. Multidisciplinary approaches
365 are needed to address both the ecological and social aspects of this formidable challenge. The relationship between
366 pressure from agricultural sources and changes in ecosystems and water quality is inherently difficult because
367 several physical parameters of the environment and human actions are acting in the same direction at the same
368 time. This collinearity complicates identifying causality and reliably establishing best practices.

369 In Brittany and other regions of Europe, high nitrate concentration triggered multi-level regulation and
370 reform. Management practices such as field margins, cover crops, and grass buffer strips were implemented.
371 While no single intervention has been a panacea, ecologists from the ZAAr are still finding evidence that many of
372 these measures effectively preserve biodiversity and serve society (Alignier and Baudry 2015; Baudry et al. 2017).
373 The regulatory framework may highlight a single ecosystem dimension (in this case nitrate concentration) but
374 secondary and tertiary costs and benefits must be considered as well (PMPOA 1993 to 2002, Action programs for
375 the nitrate directive, since 1996 , European Nitrate directive, Brittany Pure Water (*Bretagne Eau Pure - BEP*)
376 from 1994 to 2006, The governmental plan against green algae – since 2009). The efficacy of these actions remains
377 uncertain, with some areas and water quality parameters showing improvement, and others appearing to remain
378 stable (Bouraoui and Grizzetti 2011). In the ZAAr it is still difficult to draw firm conclusions about the efficacy of
379 interventions at catchment scales, but it is clear that the positive or null results will only become clear with
380 deliberate and long-term monitoring of holistic socioecological functioning. Collaborative research that involves
381 the community moves us towards the goal of helping each actor of the system to be involved and informed about
382 the risks and profits of each action.

383

384

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Bibliography

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- 396 Abbott BW, Baranov V, Mendoza-Lera C, Nikolakopoulou M, Harjung A, Kolbe T,
397 Balasubramanian MN, Vaessen TN, Ciocca F, Campeau A, Wallin MB, Romeijn P,
398 Antonelli M, Gonçalves J, Datry T, Laverman AM, de Dreuzy J-R, Hannah DM,
399 Krause S, Oldham C, Pinay G (2016) Using multi-tracer inference to move beyond
400 single-catchment ecohydrology *Earth-Science Reviews* 160:19-42
401 doi:10.1016/j.earscirev.2016.06.014
- 402 Abbott BW, Gruau G, Zarnetske JP, Moatar F, Barbe L, Thomas Z, Fovet O, Kolbe T, Gu S,
403 Pierson-Wickmann AC, Davy P, Pinay G (2017) Unexpected spatial stability of water
404 chemistry in headwater stream networks *Ecology letters* 21:296-308
405 doi:10.1111/ele.12897
- 406 Abbott BW, Moatar F, Gauthier O, Fovet O, Antoine V, Ragueneau O (2018) Trends and
407 seasonality of river nutrients in agricultural catchments: 18years of weekly citizen
408 science in France *The Science of the total environment* 624:845-858
409 doi:10.1016/j.scitotenv.2017.12.176
- 410 Alagona P, Sandlos J, Wiersma Y (2012) Past imperfect: using historical ecology and baseline
411 data for conservation and restoration projects in North America *Environ Philos* 9:49-70
412 doi:doi: 10.5840/envirophil2012914
- 413 Alvarez-Cabria M, Barquin J, Penas FJ (2016) Modelling the spatial and seasonal variability of
414 water quality for entire river networks: Relationships with natural and anthropogenic
415 factors *The Science of the total environment* 545-546:152-162
416 doi:10.1016/j.scitotenv.2015.12.109

- 417 Aquilina L, Vergnaud-Ayraud, V., Labasque, T., Bour, O., Molenat, J., Ruiz, L., de Montety,
418 V., De Ridder, J., Roques, C., Longuevergne, L. (2012) Nitrate dynamics in agricultural
419 catchments deduced from groundwater dating and long-term nitrate monitoring in
420 surface- and groundwaters *Science of the Total Environment* 435:167-178
421 doi:10.1016/j.scitotenv.2012.06.028
- 422 Ben Maamar S, Aquilina L, Quaiser A, Pauwels H, Michon-Coudouel S, Vergnaud-Ayraud V,
423 Labasque T, Roques C, Abbott BW, Dufresne A (2015) Groundwater Isolation Governs
424 Chemistry and Microbial Community Structure along Hydrologic Flowpaths *Frontiers*
425 *in microbiology* 6:1457 doi:10.3389/fmicb.2015.01457
- 426 Bouraoui F, Grizzetti B (2011) Long term change of nutrient concentrations of rivers
427 discharging in European seas *The Science of the total environment* 409:4899-4916
428 doi:10.1016/j.scitotenv.2011.08.015
- 429 Burt TP, Howden NJK, Worrall F, McDonnell JJ (2011) On the value of long-term, low-
430 frequency water quality sampling: avoiding throwing the baby out with the bathwater
431 *Hydrological Processes* 25:828-830 doi:10.1002/hyp.7961
- 432 Burt TP, Pinay, G, (2005) Linking hydrology and biogeochemistry in complex landscapes.
433 *Progress in Physical Geography* 29:297–316
- 434 Caubel-Forget V, Grimaldi C, Rouault, F., (2001) Contrasted dynamics of nitrate and chloride
435 in groundwater submitted to the influence of a hedge *C R Acad Sci Paris* 332:107-113
- 436 Cheverry C (1998) *Agriculture intensive et qualité des eaux*. Editions Quae. INRA,
- 437 Forman RTT, Baudry J (1984) Hedgerows and hedgerow networks in landscape ecology
438 *Environmental management* 8:495-510 doi:10.1007/bf01871575
- 439 Gao HK, Hrachowitz M, Sriwongsitanon N, Fenicia F, Gharari S, Savenije HHG (2016)
440 Accounting for the influence of vegetation and landscape improves model
441 transferability in a tropical savannah region *Water Resources Research* 52:7999-8022
442 doi:10.1002/2016wr019574
- 443 Grimaldi C, Fossey M, Thomas Z, Fauvel Y, Merot P (2012) Nitrate attenuation in soil and
444 shallow groundwater under a bottomland hedgerow in a European farming landscape
445 *Hydrological Processes* 26:3570-3578 doi:10.1002/hyp.8441
- 446 Guo ZL, Zhong C, Cai CF, Ding SW, Wang ZM (2007) Nitrogen competition in contour
447 hedgerow systems in subtropical China *Nutrient Cycling in Agroecosystems* 81:71-83
448 doi:10.1007/s10705-007-9153-z
- 449 Haase P, Tonkin JD, Stoll S, Burkhard B, Frenzel M, Geijzendorffer IR, Hauser C, Klotz S,
450 Kuhn I, McDowell WH, Mirtl M, Muller F, Musche M, Penner J, Zacharias S,

- 451 Schmeller DS (2018) The next generation of site-based long-term ecological
452 monitoring: Linking essential biodiversity variables and ecosystem integrity *The*
453 *Science of the total environment* 613-614:1376-1384
454 doi:10.1016/j.scitotenv.2017.08.111
- 455 Helton AM, Hall RO, Bertuzzo E (2018) How network structure can affect nitrogen removal by
456 streams *Freshwater Biology* 63:128-140 doi:10.1111/fwb.12990
- 457 Helton AM, Poole GC, Payn RA, Izurieta C, Stanford JA (2012) Scaling flow path processes to
458 fluvial landscapes: An integrated field and model assessment of temperature and
459 dissolved oxygen dynamics in a river-floodplain-aquifer system *Journal of Geophysical*
460 *Research: Biogeosciences* 117:n/a-n/a doi:10.1029/2012jg002025
- 461 Howarth RW (2008) Coastal nitrogen pollution: A review of sources and trends globally and
462 regionally *Harmful Algae* 8:14-20 doi:10.1016/j.hal.2008.08.015
- 463 Huntington TG (2006) Evidence for intensification of the global water cycle: Review and
464 synthesis *Journal of Hydrology* 319:83-95 doi:10.1016/j.jhydrol.2005.07.003
- 465 Jasechko S, Debra Perrone, Befus KM, M. Bayani Cardenas, Grant Ferguson, Tom Gleeson,
466 Elco Luijendijk, McDonnell JJ, Richard G. Taylor, Wada Y, Kirchner JW (2017)
467 Global aquifers dominated by fossil groundwaters but wells vulnerable to modern
468 contamination *Nature Geoscience* doi:DOI: 10.1038/NGEO2943
- 469 Kolbe T, Marçais J, Thomas Z, Abbott BW, de Dreuzy J-R, Rousseau-Gueutin P, Aquilina L,
470 Labasque T, Pinay G (2016) Coupling 3D groundwater modeling with CFC-based age
471 dating to classify local groundwater circulation in an unconfined crystalline aquifer
472 *Journal of Hydrology* 543, Part A:31-46 doi:doi.org/10.1016/j.jhydrol.2016.05.020
- 473 Loaiciga HA, Valdes, J. B. , Vogel, R., Garvey, J., Schwarz, H. (1996) Global warming and the
474 hydrologic cycle *Journal of Hydrology* 174:83-127
- 475 Malone ET, Abbott BW, Klaar MJ, Kidd C, Sebilo M, Milner AM, Pinay G (2018) Decline in
476 Ecosystem $\delta^{13}\text{C}$ and Mid-Successional Nitrogen Loss in a Two-Century Postglacial
477 Chronosequence Ecosystems doi:10.1007/s10021-018-0245-1
- 478 Marçais J, de Dreuzy JR, Ginn TR, Rousseau-Gueutin P, Leray S (2015) Inferring transit time
479 distributions from atmospheric tracer data: Assessment of the predictive capacities of
480 Lumped Parameter Models on a 3D crystalline aquifer model *Journal of Hydrology*
481 525:619-631 doi:10.1016/j.jhydrol.2015.03.055
- 482 Marçais J, Gauvain A, Labasque T, Abbott BW, Pinay G, Aquilina L, Chabaux F, Viville D, de
483 Dreuzy JR (2018) Dating groundwater with dissolved silica and CFC concentrations in

- 484 crystalline aquifers *The Science of the total environment* 636:260-272
485 doi:10.1016/j.scitotenv.2018.04.196
- 486 Merot P (1999) The influence of hedgerow systems on the hydrology of agricultural
487 catchments in a temperate climate. *Agronomie* 19:655-669
- 488 Meybeck M (2003) Global analysis of river systems: from Earth system controls to
489 Anthropocene syndromes *Philosophical transactions of the Royal Society of London*
490 *Series B, Biological sciences* 358:1935-1955 doi:10.1098/rstb.2003.1379
- 491 Mirtl M, E TB, Djukic I, Forsius M, Haubold H, Hugo W, Jourdan J, Lindenmayer D,
492 McDowell WH, Muraoka H, Orenstein DE, Pauw JC, Peterseil J, Shibata H, Wohner C,
493 Yu X, Haase P (2018) Genesis, goals and achievements of Long-Term Ecological
494 Research at the global scale: A critical review ofILTER and future directions *The*
495 *Science of the total environment* 626:1439-1462 doi:10.1016/j.scitotenv.2017.12.001
- 496 Moatar F, Abbott BW, Minaudo C, Curie F, Pinay G (2017) Elemental properties, hydrology,
497 and biology interact to shape concentration-discharge curves for carbon, nutrients,
498 sediment, and major ions water *Resources Research* doi:10.1002/2016WR019635
- 499 Perrot T, Rossi N, Ménesguen A, Dumas F (2014) Modelling green macroalgal blooms on the
500 coasts of Brittany, France to enhance water quality management *Journal of Marine*
501 *Systems* 132:38-53 doi:10.1016/j.jmarsys.2013.12.010
- 502 Pinay G, Bernal S, Abbott BW, Lupon A, Marti E, Sabater F, Krause S (2018) Riparian
503 Corridors: A New Conceptual Framework for Assessing Nitrogen Buffering Across
504 Biomes *Frontiers in Environmental Science* 6 doi:10.3389/fenvs.2018.00047
- 505 Poisvert C, Curie F, Moatar F (2016) Annual agricultural N surplus in France over a 70-year
506 period *Nutrient Cycling in Agroecosystems* 107:63-78 doi:10.1007/s10705-016-9814-x
- 507 Rudel TK, Schneider L, Uriarte M, Turner BL, 2nd, DeFries R, Lawrence D, Geoghegan J,
508 Hecht S, Ickowitz A, Lambin EF, Birkenholtz T, Baptista S, Grau R (2009) Agricultural
509 intensification and changes in cultivated areas, 1970-2005 *Proceedings of the National*
510 *Academy of Sciences of the United States of America* 106:20675-20680
511 doi:10.1073/pnas.0812540106
- 512 Savenije HHG, Hoekstra AY, van der Zaag P (2014) Evolving water science in the
513 Anthropocene *Hydrology and Earth System Sciences* 18:319-332 doi:10.5194/hess-18-
514 319-2014
- 515 Steffen W, Richardson K, Rockstrom J, Cornell SE, Fetzer I, Bennett EM, Biggs R, Carpenter
516 SR, de Vries W, de Wit CA, Folke C, Gerten D, Heinke J, Mace GM, Persson LM,
517 Ramanathan V, Reyers B, Sorlin S (2015) Sustainability. Planetary boundaries: guiding

518 human development on a changing planet *Science* 347:1259855
519 doi:10.1126/science.1259855

520 Tesoriero AJ, Liebscher H, Cox SE (2000) Mechanism and rate of denitrification in an
521 agricultural watershed: Electron and mass balance along groundwater flow paths *Water*
522 *Resources Research* 36:1545-1559 doi:10.1029/2000wr900035

523 Thomas Z, Abbott B (2018) Hedgerows reduce nitrate flux at hillslope and catchment scales
524 via root uptake and secondary effects *Journal of contaminant hydrology* July 5, 2018
525 doi:<https://doi.org/10.1016/j.jconhyd.2018.07.002>.

526 Thomas Z, Abbott B, Troccaz O, Baudry J, Pinay G (2016) Proximate and ultimate controls on
527 carbon and nutrient dynamics of small agricultural catchments *Biogeosciences* 13:1863-
528 1875 doi:10.5194/bg-13-1863-2016

529 Thomas Z, Rousseau-Gueutin P, Kolbe T, Abbott BW, Marçais J, Peiffer S, Frei S, Bishop K,
530 Pichelin P, Pinay G, de Dreuzy JR (2016) Constitution of a catchment virtual
531 observatory for sharing flow and transport models outputs *Journal of Hydrology* 543,
532 Part A:59-66 doi:doi.org/10.1016/j.jhydrol.2016.04.067

533 van Grinsven HJM, ten Berge HFM, Dalgaard T, Fraters B, Durand P, Hart A, Hofman G,
534 Jacobsen BH, Lalor STJ, Lesschen JP, Osterburg B, Richards KG, Techen AK, Vertès
535 F, Webb J, Willems WJ (2012) Management, regulation and environmental impacts of
536 nitrogen fertilization in northwestern Europe under the Nitrates Directive; a benchmark
537 study *Biogeosciences* 9:5143-5160 doi:10.5194/bg-9-5143-2012

538 Van Meter KJ, Basu NB, Veenstra JJ, Burras CL (2016) The nitrogen legacy: emerging
539 evidence of nitrogen accumulation in anthropogenic landscapes *Environmental*
540 *Research Letters* 11:035014 doi:10.1088/1748-9326/11/3/035014

541 Worrall F, Howden NJK, Burt TP (2015) Evidence for nitrogen accumulation: the total
542 nitrogen budget of the terrestrial biosphere of a lowland agricultural catchment
543 *Biogeochemistry* 123:411-428 doi:10.1007/s10533-015-0074-7

544

545 **Figure caption**

546 **Fig. 1** The Location of the Zone Atelier Armorique (ZAAr), an international long-term
547 ecological research site in north-western France (a); ZAAr boundaries, topography, stream
548 network, and location of sampling points (wells and outlets), as well as the location of the
549 highly monitored hillslope (b). Hedgerows density is highest in the hilly southern half of the
550 ZAAr while open fields are dominant in the schist plateau

551 **Fig. 2** Daily discharge, precipitation (P), and potential evapotranspiration (PET)
552 measured or modeled for the past 30 years. Discharge until 2012 is from the hydrometric
553 station of the *Couesnon River* (Fig. 1), which is adjacent to the ZAAr, and discharge from 2012
554 to 2017 (red line) was obtained by transposing the discharge measured at the outlet of the BV6
555 subcatchment (Fig. 1). Net precipitation was calculated starting in 1993, when PET data
556 became available. Positive and negative values of P-PET (net precipitation) were separated to
557 highlight times of water excess and deficit. Over the period of record, mean annual
558 precipitation (\bar{P}) is 914 mm and mean annual PET is 687 mm

559 **Fig. 3** Water storage (ΔS_{year}) evolution over 8 years since 2009. Water storage
560 calculated for each hydrological year from mean soil moisture measurements along 60 cm soil
561 profiles in the highly monitored hillslope within (In) and beyond (Out) the rooting zone of
562 hedgerows (Fig. 1). P is the annual precipitation and P-PET is net precipitation

563 **Fig. 4** Spatial variability of nitrate concentration in stream water (a) and groundwater
564 (b). Each point is the mean value of the time series (see Table S1 and Table S2)

565 **Fig. 5** Nitrate concentration in shallow groundwater along the highly monitored
566 hillslope (P1-P10), in the forested bottomland (P11), in the wetland (P12-P13), and in the
567 stream (a). Mean nitrate concentration (\pm SD) among points and through time along the
568 hillslope (b). Vertical black lines indicate piezometers position and depth. Discontinuous and
569 continuous blue lines indicate maximum (GWL max) and minimum groundwater (GWL min)
570 level, respectively. The black continuous line shows the soil surface

571 **Fig. 6** Nitrate concentration variation since 2012 in the main hydrological
572 compartments, i.e. streams, deep groundwater (deep GW), shallow groundwater (shallow GW),
573 wetland, and near-stream groundwater (NS-GW)

574 **Fig. 7** Annual mean and standard deviation of stream water nitrate concentration (red
575 line) and discharge (black line) over the past 29 years. The number of nitrate samples per year
576 was about 4 at the beginning of the sampling period, it was about 24 samples per year since
577 2001-2002

578 **Fig. 8** Relationship between nitrate:chloride ratio and chloride for hydrological
579 compartments: deep groundwater (a), shallow groundwater (b), wetland (c), and streams (d)

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