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# Long-term ecological observatories needed to understand ecohydrological systems in the Anthropocene: a catchment-scale case study in Brittany, France

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 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.

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

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- 46 Length of manuscript: 5302 words
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# 50 **1. Introduction**

At a global scale, human activity has surpassed geological and biological forces in many dimensions 51 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 approaches that often focus on individual response variables (Alagona et al. 2012; Worrall et al. 2015; Pinay et al. 65 2018). Additionally, while degradation of ecosystem function and services is proportionally related to human 66 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).

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

#### 81

# 2. Zone Atelier Armorique (French LTER)

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

and international LTER (https://www.ilternet.edu/) networks. Located in western France (48° 36' N, 1° 32' W; 86 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 km<sup>2</sup>, (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**

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

The long legacy of land use and excess nutrients have degraded aquatic ecosystems in this area, including 124 125 lakes, rivers, groundwater, and estuaries (Ben Maamar et al. 2015; Kolbe et al. 2016; Abbott et al. 2018). Average nitrate concentration increased from 9 mg L<sup>-1</sup> in 1976 to 65 mg L<sup>-1</sup> in 1989 (Cheverry 1998), exceeding numerous 126 regulatory limits (European Nitrate directive 1991), and causing widespread harmful algal blooms along Brittany 127 128 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 wetlands, aquifers, and hyporheic zones (Gilliam 1994; Vidon and Hill 2004; Trauth et al. 2015). Areas of 140 141 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 hedges, embankments, and ditches (Bernhardt et al. 2003; Hall et al. 2009; Worrall et al. 2012; Schelker et al. 144 145 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**

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### 3.1 Land use and meteorological data

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

Meteorological data were available from the Meteo France weather station located in *Louvigné du desert*, 37 km from the outlet of the studied area. A weather station was installed in 2009 inside the ZAAr to record air 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 the ZAAr. The time step of our measurements ranged between 10 minutes for groundwater level and discharge, 2 166 167 weeks for soil moisture, and monthly for water quality. Seventeen agricultural wells from across the ZAAr were selected for water quality analyses (Fig. 1b) and thirteen piezometers were installed on a highly instrumented 168 169 hillslope crossed by 3 hedgerows near the boundary of a wetland. Groundwater level was measured for each piezometer using pressure head sensors from OTT (Orpheus mini). Table S1 summarises the characteristics of the 170 171 groundwater wells and shallower piezometers.

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

177 
$$\Delta S_{year} = \int_{0}^{z} (\theta_{max} - \theta_{min}) . dz$$
 Equation 2

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

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

Monthly to bimonthly stream and groundwater samples were collected from December 2012 to June 2016. Stream water from 6 nested catchment outlets were sampled at each date (Fig. 1b) and 2 additional catchments (G1 and S1) from Thomas et al. (2016) were also integrated in our global analysis. Water samples from surface sampling points and groundwater wells were filtered to 0.45 µm through polyethersulfone filters and major anions were measured using ion chromatography as described in (Thomas et al. 2016). Table S2 summarises the frequency of sampling and the list of parameters analysed. All the time series analyses (i.e. precipitation, discharge, potential evaporation, groundwater level, water quality, and soil moisture) were calculated for each hydrological year from 1 of September to 31 of August of the next year.

#### **3.5 Temperature monitoring**

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

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

#### **4. Results**

#### 208 **4.1 ZAAr land use evolution**

Land use changed substantially from 1993 to 2013 (Fig. S1). In 1993 to 1998, pasture occupied 15-30 % 209 210 of the landscape, increasing to 35-45% in 1999 to 2013, with a maximum of around 60 % in 2009. Maize and cereal cultures covered 15-25 % of the area from 2000 to 2009, increasing to 40% in 2010 to 2013. Initially, 15-211 28% of the landscape was not used for agricultural practices, decreasing to ~10% in 2010. The forested area 212 213 (~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-Vilaine). 216

#### 217 **4.3 Hydrological compartments characterization**

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#### 4.3.1 Soil moisture and groundwater variation under a contrasted climatic context

Historical data from Meteo-France over a 30 years period revealed substantial variability in precipitation and evapotranspiration (Fig. 2). The hydrological years 1988-1993, 1995-1998, 2001-2006, 2009-2012, and 2014-2016 were relatively dry with net precipitation (P-PET) lower than 200 mm. 2009-2010 and 2016-2017 were the driest years with negative net precipitation of -10 mm and -137 mm, respectively. The wettest hydrological years were 1998-2000, 2006-2007, and 2013-2014 with net precipitation between 450 and 600 mm (Fig. S2). The cumulative rainfall was lowest in 2009-2010 (736 mm) and highest in 2013-2014 (1180 mm; Fig.2 and Fig. S2). Soil moisture measurements at the recharge zone of the hillslope within and beyond the rooting zone were used to calculate water storage ( $\Delta S_{year}$ ) (Fig. 3). Soil water storage in both zones followed net precipitation except for the beginning of the observed time series. Soil moisture within the rooting zone was systematically drier in 6 of the 8 observed years, independent of other hydrological changes. Water storage was highest beyond the rooting zone in the middle of a meadow far away from the root system of hedgerows.

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

#### 236 **4.3.3 Stream discharge**

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

#### **4.4 Biogeochemical reactivity in catchment compartments**

#### 244 **4.4.1 Spatial analysis of nitrate concentrations**

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

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

Figure 4a shows the mean nitrate concentration in stream water measured over the periods indicated in Table S2. Overall, concentration was much less variable in surface waters than in groundwater. For the headwater catchments located in the granitic part of ZAAr, nitrate concentration was lower than 25 mg  $L^{-1}$  (Fig. 4a). Nitrate concentration increased with drainage area and intensive agricultural land contribution but the mean value remained below 40 mg  $L^{-1}$  except for the catchments S1 and BV8, and *Couesnon* station.

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

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

We used the ratio of nitrate to chloride (NO<sub>3</sub>:Cl) to identify potential processes controlling nitrate concentration (Fig. 8). Three processes can be identified from the NO<sub>3</sub>:Cl ratio: (i) denitrification is characterized by a NO<sub>3</sub>:Cl ratio less than 1 and low chloride concentration, (ii) agricultural inputs are characterized by a NO<sub>3</sub>:Cl ratio much higher than 1 and low chloride concentration, (iii) and sewage inputs are characterized by a NO<sub>3</sub>:Cl ratio much smaller than 1 and high chloride concentration. Deep wells fell into two groups, with F3, F8, F14, and F15 expressing a high NO3:Cl ratio and low chloride concentration (Fig. 8a), and the other deep wells showing a NO3:Cl ratio <1 and low chloride concentration.

In the shallow GW (P1-P11), the NO<sub>3</sub>:Cl ratio as well as the chloride concentration were highly variable and changed over time (Fig. 10b). In the near stream and wetland area NO<sub>3</sub>:Cl ratio was lower than one and chloride concentration was low (Fig. 10c). The stream water ratio (Fig. 8a) varied in time except for the catchment located in the forested zone, which had a very low NO<sub>3</sub>:Cl.

**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 NO<sub>3</sub>:Cl 297 ratio characteristic of agricultural inputs while others showed a NO<sub>3</sub>: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. 2015). For the shallow groundwater in the hillslope, there was high temporal and spatial variation in nitrate 299 300 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  $\sim 40 \text{ mg l}^{-1}$ .

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).

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

323

## 5.3 Using long-term studies to promote socio-hydrological research

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

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

- (ii) Learning disciplinary language and perspectives, which often use the same terms or interpret the
   same findings differently
- (iii) Obtaining relevant metrics and measurements to quantify vulnerability, resilience, and
   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 that they are increasingly dominated by anthropogenic drivers (i.e. direct disturbance and climate change 338 339 (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 production and slowed ecological degradation in many areas, achieving ecological and societal good 361 362 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 are needed to address both the ecological and social aspects of this formidable challenge. The relationship between 365 366 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.

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#### 394 Bibliography

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

- Aquilina L, Vergnaud-Ayraud, V., Labasque, T., Bour, O., Molenat, J., Ruiz, L., de Montety,
  V., De Ridder, J., Roques, C., Longuevergne, L. (2012) Nitrate dynamics in agricultural
  catchments deduced from groundwater dating and long-term nitrate monitoring in
  surface- and groundwaters Science of the Total Environment 435:167-178
  doi:10.1016/j.scitotenv.2012.06.028
- Ben Maamar S, Aquilina L, Quaiser A, Pauwels H, Michon-Coudouel S, Vergnaud-Ayraud V,
  Labasque T, Roques C, Abbott BW, Dufresne A (2015) Groundwater Isolation Governs
  Chemistry and Microbial Community Structure along Hydrologic Flowpaths Frontiers
  in microbiology 6:1457 doi:10.3389/fmicb.2015.01457
- Bouraoui F, Grizzetti B (2011) Long term change of nutrient concentrations of rivers
  discharging in European seas The Science of the total environment 409:4899-4916
  doi:10.1016/j.scitotenv.2011.08.015
- Burt TP, Howden NJK, Worrall F, McDonnell JJ (2011) On the value of long-term, lowfrequency water quality sampling: avoiding throwing the baby out with the bathwater
  Hydrological Processes 25:828-830 doi:10.1002/hyp.7961
- Burt TP, Pinay, G, (2005) Linking hydrology and biogeochemistry in complex landscapes.
  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,
- Forman RTT, Baudry J (1984) Hedgerows and hedgerow networks in landscape ecology
   Environmental management 8:495-510 doi:10.1007/bf01871575
- Gao HK, Hrachowitz M, Sriwongsitanon N, Fenicia F, Gharari S, Savenije HHG (2016)
  Accounting for the influence of vegetation and landscape improves model
  transferability in a tropical savannah region Water Resources Research 52:7999-8022
  doi:10.1002/2016wr019574
- Grimaldi C, Fossey M, Thomas Z, Fauvel Y, Merot P (2012) Nitrate attenuation in soil and
  shallow groundwater under a bottomland hedgerow in a European farming landscape
  Hydrological Processes 26:3570-3578 doi:10.1002/hyp.8441
- Guo ZL, Zhong C, Cai CF, Ding SW, Wang ZM (2007) Nitrogen competition in contour
  hedgerow systems in subtropical China Nutrient Cycling in Agroecosystems 81:71-83
  doi:10.1007/s10705-007-9153-z
- Haase P, Tonkin JD, Stoll S, Burkhard B, Frenzel M, Geijzendorffer IR, Hauser C, Klotz S,
  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

- Helton AM, Hall RO, Bertuzzo E (2018) How network structure can affect nitrogen removal by
  streams Freshwater Biology 63:128-140 doi:10.1111/fwb.12990
- Helton AM, Poole GC, Payn RA, Izurieta C, Stanford JA (2012) Scaling flow path processes to
  fluvial landscapes: An integrated field and model assessment of temperature and
  dissolved oxygen dynamics in a river-floodplain-aquifer system Journal of Geophysical
  Research: Biogeosciences 117:n/a-n/a doi:10.1029/2012jg002025
- Howarth RW (2008) Coastal nitrogen pollution: A review of sources and trends globally and
   regionally Harmful Algae 8:14-20 doi:10.1016/j.hal.2008.08.015
- Huntington TG (2006) Evidence for intensification of the global water cycle: Review and
   synthesis Journal of Hydrology 319:83-95 doi:10.1016/j.jhydrol.2005.07.003
- Jasechko S, Debra Perrone, Befus KM, M. Bayani Cardenas, Grant Ferguson, Tom Gleeson,
   Elco Luijendijk, McDonnell JJ, Richard G. Taylor, Wada Y, Kirchner JW (2017)
   Global aquifers dominated by fossil groundwaters but wells vulnerable to modern
   contamination Nature Geoscience doi:DOI: 10.1038/NGEO2943
- Kolbe T, Marçais J, Thomas Z, Abbott BW, de Dreuzy J-R, Rousseau-Gueutin P, Aquilina L,
  Labasque T, Pinay G (2016) Coupling 3D groundwater modeling with CFC-based age
  dating to classify local groundwater circulation in an unconfined crystalline aquifer
  Journal of Hydrology 543, Part A:31-46 doi:doi.org/10.1016/j.jhydrol.2016.05.020
- Loaiciga HA, Valdes, J. B., Vogel, R., Garvey, J., Schwarz, H. (1996) Global warming and the
  hydrologic cycle Journal of Hydrology 174:83-127
- Malone ET, Abbott BW, Klaar MJ, Kidd C, Sebilo M, Milner AM, Pinay G (2018) Decline in
   Ecosystem δ13C and Mid-Successional Nitrogen Loss in a Two-Century Postglacial
   Chronosequence Ecosystems doi:10.1007/s10021-018-0245-1
- Marçais J, de Dreuzy JR, Ginn TR, Rousseau-Gueutin P, Leray S (2015) Inferring transit time
  distributions from atmospheric tracer data: Assessment of the predictive capacities of
  Lumped Parameter Models on a 3D crystalline aquifer model Journal of Hydrology
  525:619-631 doi:10.1016/j.jhydrol.2015.03.055
- Marcais J, Gauvain A, Labasque T, Abbott BW, Pinay G, Aquilina L, Chabaux F, Viville D, de
   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
- Meybeck M (2003) Global analysis of river systems: from Earth system controls to
   Anthropocene syndromes Philosophical transactions of the Royal Society of London
   Series B, Biological sciences 358:1935-1955 doi:10.1098/rstb.2003.1379
- Mirtl M, E TB, Djukic I, Forsius M, Haubold H, Hugo W, Jourdan J, Lindenmayer D,
  McDowell WH, Muraoka H, Orenstein DE, Pauw JC, Peterseil J, Shibata H, Wohner C,
  Yu X, Haase P (2018) Genesis, goals and achievements of Long-Term Ecological
  Research at the global scale: A critical review of ILTER and future directions The
  Science of the total environment 626:1439-1462 doi:10.1016/j.scitotenv.2017.12.001
- Moatar F, Abbott BW, Minaudo C, Curie F, Pinay G (2017) Elemental properties, hydrology,
  and biology interact to shape concentration-discharge curves for carbon, nutrients,
  sediment, andmajor ions water Resources Research doi:10.1002/2016WR019635
- Perrot T, Rossi N, Ménesguen A, Dumas F (2014) Modelling green macroalgal blooms on the
   coasts of Brittany, France to enhance water quality management Journal of Marine
   Systems 132:38-53 doi:10.1016/j.jmarsys.2013.12.010
- Pinay G, Bernal S, Abbott BW, Lupon A, Marti E, Sabater F, Krause S (2018) Riparian
   Corridors: A New Conceptual Framework for Assessing Nitrogen Buffering Across
   Biomes Frontiers in Environmental Science 6 doi:10.3389/fenvs.2018.00047
- Poisvert C, Curie F, Moatar F (2016) Annual agricultural N surplus in France over a 70-year
   period Nutrient Cycling in Agroecosystems 107:63-78 doi:10.1007/s10705-016-9814-x
- Rudel TK, Schneider L, Uriarte M, Turner BL, 2nd, DeFries R, Lawrence D, Geoghegan J,
  Hecht S, Ickowitz A, Lambin EF, Birkenholtz T, Baptista S, Grau R (2009) Agricultural
  intensification and changes in cultivated areas, 1970-2005 Proceedings of the National
  Academy of Sciences of the United States of America 106:20675-20680
  doi:10.1073/pnas.0812540106
- Savenije HHG, Hoekstra AY, van der Zaag P (2014) Evolving water science in the
   Anthropocene Hydrology and Earth System Sciences 18:319-332 doi:10.5194/hess-18 319-2014
- Steffen W, Richardson K, Rockstrom J, Cornell SE, Fetzer I, Bennett EM, Biggs R, Carpenter
  SR, de Vries W, de Wit CA, Folke C, Gerten D, Heinke J, Mace GM, Persson LM,
  Ramanathan V, Reyers B, Sorlin S (2015) Sustainability. Planetary boundaries: guiding

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

- Tesoriero AJ, Liebscher H, Cox SE (2000) Mechanism and rate of denitrification in an
   agricultural watershed: Electron and mass balance along groundwater flow paths Water
   Resources Research 36:1545-1559 doi:10.1029/2000wr900035
- Thomas Z, Abbott B (2018) Hedgerows reduce nitrate flux at hillslope and catchment scales
  via root uptake and secondary effects Journal of contaminant hydrology July 5, 2018
  doi:https://doi.org/10.1016/j.jconhyd.2018.07.002.
- Thomas Z, Abbott B, Troccaz O, Baudry J, Pinay G (2016) Proximate and ultimate controls on
   carbon and nutrient dynamics of small agricultural catchments Biogeosciences 13:1863 1875 doi:10.5194/bg-13-1863-2016
- Thomas Z, Rousseau-Gueutin P, Kolbe T, Abbott BW, Marçais J, Peiffer S, Frei S, Bishop K,
  Pichelin P, Pinay G, de Dreuzy JR (2016) Constitution of a catchment virtual
  observatory for sharing flow and transport models outputs Journal of Hydrology 543,
  Part A:59-66 doi:doi.org/10.1016/j.jhydrol.2016.04.067
- van Grinsven HJM, ten Berge HFM, Dalgaard T, Fraters B, Durand P, Hart A, Hofman G,
  Jacobsen BH, Lalor STJ, Lesschen JP, Osterburg B, Richards KG, Techen AK, Vertès
  F, Webb J, Willems WJ (2012) Management, regulation and environmental impacts of
  nitrogen fertilization in northwestern Europe under the Nitrates Directive; a benchmark
  study Biogeosciences 9:5143-5160 doi:10.5194/bg-9-5143-2012
- Van Meter KJ, Basu NB, Veenstra JJ, Burras CL (2016) The nitrogen legacy: emerging
   evidence of nitrogen accumulation in anthropogenic landscapes Environmental
   Research Letters 11:035014 doi:10.1088/1748-9326/11/3/035014
- Worrall F, Howden NJK, Burt TP (2015) Evidence for nitrogen accumulation: the total
   nitrogen budget of the terrestrial biosphere of a lowland agricultural catchment
   Biogeochemistry 123:411-428 doi:10.1007/s10533-015-0074-7
- 544
- 545 **Figure caption**

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

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

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

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

**Fig. 5** Nitrate concentration in shallow groundwater along the highly monitored hillslope (P1-P10), in the forested bottomland (P11), in the wetland (P12-P13), and in the stream (a). Mean nitrate concentration (± SD) among points and through time along the hillslope (b). Vertical black lines indicate piezometers position and depth. Discontinuous and continuous blue lines indicate maximum (GWL max) and minimum groundwater (GWL min) 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)

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

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