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1 **Daily denitrification rates in floodplains under contrasting pedo-**
2 **climatic and anthropogenic contexts: modelling at the watershed**
3 **scale**

4
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9
10 **Abstract**

11 Floodplains play a crucial role in water quality regulation via denitrification. This
12 biogeochemical process reduces nitrate (NO_3^-), with aquifer saturation, organic carbon (OC)
13 and N availability as the main drivers. To accurately describe the denitrification in the
14 floodplain, it is necessary to better understand nitrate fluxes that reach these natural
15 bioreactors and the transformation that occurs in these surface areas at the watershed scale.
16 At this scale, several approaches tried to simulate denitrification contribution to nitrogen
17 dynamics in study sites. However, these studies did not consider OC fluxes influences,
18 hydrological dynamics and temperature variations at a daily time step. This paper focuses on
19 a new model that allows insights on nitrate, OC, discharge and temperature influences on daily
20 denitrification for each water body. We used a process-based deterministic model to estimate
21 daily alluvial denitrification in different watersheds showing various pedo-climatic conditions.
22 To better understand global alluvial denitrification variability, we applied the method to three
23 contrasting catchments: The Amazon for tropical zones, the Garonne as representative of the
24 temperate climate and the Yenisei for cold rivers. The Amazon with a high discharge, frequent

25 flooding and warm temperature, leads to aquifers saturation, and stable OC concentrations.
26 Those conditions favour a significant loss of N by denitrification. In the Garonne River, the low
27 OC delivery limits the denitrification process. While Arctic rivers have high OC exports, the low
28 nitrate concentrations and cold temperature in the Yenisei River hinder denitrification. We
29 found daily alluvial denitrification rates of 119.4 ± 47.5 , 7.6 ± 5.4 and 0.1 ± 0.5 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$
30 during the 2000-2010 period for the Amazon, the Garonne and the Yenisei respectively. This
31 study quantifies the floodplains influence in the water quality regulation service, their
32 contribution to rivers geochemical processes facing global changes and their role on nitrate
33 and OC fluxes to the oceans.

34 **Keywords:** denitrification; nitrate; floodplains; watershed; daily time step; organic carbon

35

36 **1 Introduction**

37 Intensive agriculture brings high amounts of nitrates to rivers by leaching of fertilizers. The
38 nitrate concentrations in free river water are significantly lower than the nitrate concentrations
39 in alluvial aquifers for areas under intense agriculture pressures (Sánchez-Pérez et al., 2003).
40 This difference is explained by the dilution effect when the water flows from aquifers to rivers,
41 together with the N retention capacity of floodplains (Craig et al., 2010). This retention capacity
42 results from plant uptake and denitrification (Pinay et al., 1998; Craig et al., 2010; Ranalli and
43 Macalady, 2010). Denitrification is the process of nitrate reduction (NO_3^-) into nitrous oxide
44 (N_2O) or dinitrogen (N_2). It is the main process that leads to nitrate loss in watersheds (Pinay
45 et al., 1998; Pfeiffer et al., 2006; Baillieux et al., 2014). Denitrifying bacteria are generally
46 facultative aerobic heterotrophs (Zaman et al., 2012). They can switch to anaerobic respiration
47 under low oxygen conditions by completing the denitrification, i.e. by using the oxygen from
48 nitrate. Thus, denitrification is optimized under specific conditions and is limited by three main
49 factors: the availability of nitrate, the availability of organic carbon (OC; Rivett et al., 2008) and
50 the small oxygen availability (Zaman et al., 2012). In this way, denitrification is a microbial

51 process consuming OC (Zaman et al., 2012). The OC used by denitrifying bacteria is taken
52 from soils leaching and in-situ sediments or comes from the river contributions (Gift et al., 2010;
53 Peter et al., 2012). OC in rivers is separated into two classes: particulate organic carbon (POC)
54 and dissolved organic carbon (DOC; Hope et al., 1994). These two forms have two different
55 origins. While POC mostly comes from soil erosion, DOC is a result of soil leaching (Meybeck,
56 1993; Raymond and Bauer, 2001). DOC is the most consumed OC form in denitrification
57 (Peyrard et al., 2011; Zarnetske et al., 2011; Sun et al., 2018).

58 Floodplains are hot spots of denitrification (McClain et al., 2003; Billen et al., 2013). Floodplains
59 are areas connected to the river network and are strongly influenced by the hydrodynamic of
60 the basin, which results in oscillations between aerobic and anaerobic conditions. The location
61 of floodplains intensifies transfers of OC and nitrate by leaching from uplands to the river.
62 These transfers occur at hot moments with a high temporal resolution (Bernard-Jannin et al.,
63 2017). Therefore, daily time step studies should highlight the temporal variability of the
64 denitrification process.

65 Past studies used in situ observations to evaluate large-scale denitrification but they revealed
66 high uncertainties (Groffman et al., 2006). Therefore, modelling appears as an important tool
67 to better assess those processes at large scale (Groffman, 2012). Modelling tools that focus
68 on the exchanges between rivers and floodplains were usually used for hydrology interactions
69 (Yamazaki et al., 2011; Jung et al., 2012). Regarding floodplains biogeochemistry, previous
70 models showed their ability to simulate denitrification (Hattermann et al., 2006; Sun et al.,
71 2018). They can be used to identify nitrate sources and sinks (Boano et al., 2010; Peyrard et
72 al., 2011; Zarnetske et al., 2012) as well as hot spots and hot moments of nutrients cycling
73 (Groffman et al., 2009; Bernard-Jannin et al., 2017). Two options are commonly used to
74 estimate denitrification at large scale: coupling a hydrological with biogeochemical models
75 (Peyrard et al., 2011) or implementing biogeochemical modules in a hydrological model (Sun
76 et al., 2018). Sun et al. (2018) was the first study to show models capacity to simulate daily
77 denitrification variations at the scale of a reach by considering the river-aquifers exchanges of

78 water, nitrate and OC. Denitrification is usually modelled as a nitrate retention rate (Boyer et
79 al., 2006; Ruelland et al., 2007; Peyrard et al., 2011; Sun et al., 2018). Although the integration
80 of the OC availability into floodplains denitrification is a recent effort (Sun et al., 2018), the
81 temporal variations of OC fluxes have not been integrated into models yet. We assume that
82 high temporal resolution of this OC delivery is important to consider in models as a control of
83 the denitrification process. Thus, the accurate modelling approach to better simulate the
84 effective biogeochemical processes with the limiting factors should be done at a daily time
85 step.

86 Past research that uses modelling tools to predict spatial and temporal denitrification variations
87 in floodplains highlighted the potential of these approaches to predict nitrate and OC fluxes at
88 large scale (Peyrard et al., 2011; Bernard-Jannin et al., 2017; Sun et al., 2018). Recent
89 research using new methods tried to estimate alluvial wetlands denitrification with remote
90 sensing data (Guilhen et al., 2020). With a similar approach, this study is the first that aims to
91 simulate denitrification at the scale of several watersheds with contrasting climatic and soil
92 properties. The main objectives of the study are i) to propose a new and easy-to-use
93 methodology to estimate floodplains denitrification at the watershed scale by taking into
94 account spatial and temporal DOC variability, ii) to apply this methodology at the scale of three
95 contrasting watersheds representative of various climatic and soils conditions and iii) to
96 quantify their daily floodplains denitrification.

97

98 **2 Materials and Methods**

99 **2.1 Study cases**

100 To highlight the global denitrification variabilities, we selected three watersheds for their
101 different ranges of nitrate and OC concentrations in the free waters. These three watersheds
102 are the Amazon River, representative of tropical areas, with low nitrate and low OC content,
103 the Yenisei River in Siberia, representative of cold climate, with low nitrate and high OC

104 contents in the free-water, and the Garonne River in France, as a temperate and anthropogenic
105 watershed, with high nitrate and low OC contents (Figure 1). Nitrate contents of the Amazon
106 and the Yenisei rivers are mostly coming from natural sources while the Garonne basin
107 shelters intensive agriculture activities. The Amazon basin is the largest draining area of the
108 world with 6,500,000 km² and displays three large floodplains located in the Northern (the
109 Branco Floodplain) and the Southern (the Madeira Floodplain) part of the basin as well as
110 alongside the mainstream. Based on the GLOBAL-NEWS model results (Mayorga et al.,
111 2010), the Amazon River has a dissolved inorganic nitrogen (DIN) export of 1.6 kgN.ha⁻¹.yr⁻¹
112 and a DOC export of 49.8 kgC.ha⁻¹.yr⁻¹. The DIN export consists mainly of nitrate, which is one
113 of the compounds used in denitrification (Zaman et al., 2012). The basin also has an average
114 soils OC content of 9 kgC.m⁻³ (Batjes, 2009). The Yenisei River is one of the main rivers flowing
115 into the Arctic Ocean with a basin area of 2,500,000 km². The main floodplains of the Yenisei
116 River are in the downstream part of the main channel. The DIN export is around 0.3 kgN.ha⁻¹.yr⁻¹,
117 the DOC export is at 10.6 kgC.ha⁻¹.yr⁻¹ while the average soils OC content is at 34 kgC.m⁻³
118 (Batjes, 2009). Finally, the Garonne River is one of the main French basins with a draining
119 area of 55,000 km². Wide floodplains are mainly located alongside the mainstream in the
120 middle course. The DIN export of this river under high anthropogenic pressures is around 5.6
121 kgN.ha⁻¹.yr⁻¹ (Mayorga et al., 2010) with a DOC export of 14.3 kgC.ha⁻¹.yr⁻¹ and average soils
122 OC content in soils of 9 kgC.m⁻³.

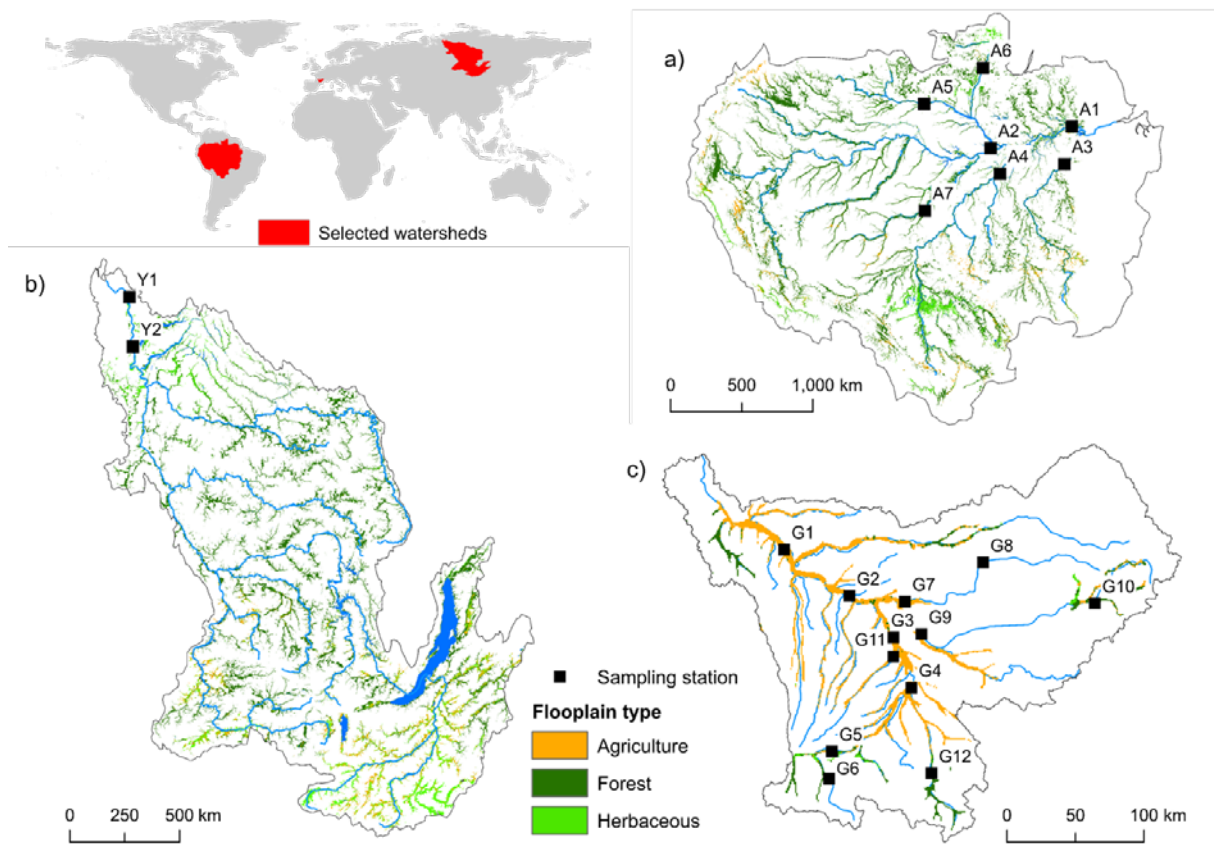
123

124 **2.2 Delineation of the floodplains**

125 An accurate delineation of these areas (Figure 1) was performed to simulate the contribution
126 of the floodplains at the watershed scale spatially. The Amazon and Yenisei floodplains were
127 delineated based on the tools available in the new GIS-interface developed for the SWAT+
128 model (<https://swat.tamu.edu/software/plus/>). This method allows the user to delineate
129 floodplains based on a slope threshold (Rathjens et al., 2015) with a digital elevation model
130 (DEM) from de Ferranti and Hormann (2012). For the Garonne, this method was not able to

131 return consistent delineation. Thus the Garonne floodplain boundaries were based on alluvial
132 soils area (Fluvisols) as proposed by Sun et al. (2018).

133 Floodplains of the three watersheds cover over 660,000 km² (10.2%), 419,000 km² (15.5%),
134 4,000 km² (7.1%) for the Amazon, the Yenisei and the Garonne basins, respectively. Forests
135 and pastures mainly cover Amazon and Yenisei floodplains with 78% and 13% for the Amazon
136 and 66% and 20% for the Yenisei (Figure 1). Nevertheless, some areas are covered by
137 agriculture, especially in the upstream parts of the watersheds. On the contrary, the Garonne
138 floodplains are mostly covered by agriculture, with over 65% of the total area.



139

140 **Figure 1: Study areas: a) The Amazon, b) The Yenisei and c) The Garonne rivers and their respective**
141 **sampling stations used to calibrate the hydrology and the nutrients fluxes. Delineation of the floodplains**
142 **was based on the method of Rathjens et al. (2015) and the Digital Elevation Model of de Ferranti and**
143 **Hormann (2012) for the Amazon and the Yenisei rivers. For the Garonne River, floodplains delineation**
144 **originate from on the soils database of Batjes (2009). Land covers came from the Global Land Cover**
145 **Database 2000 (European Commission, 2003).**

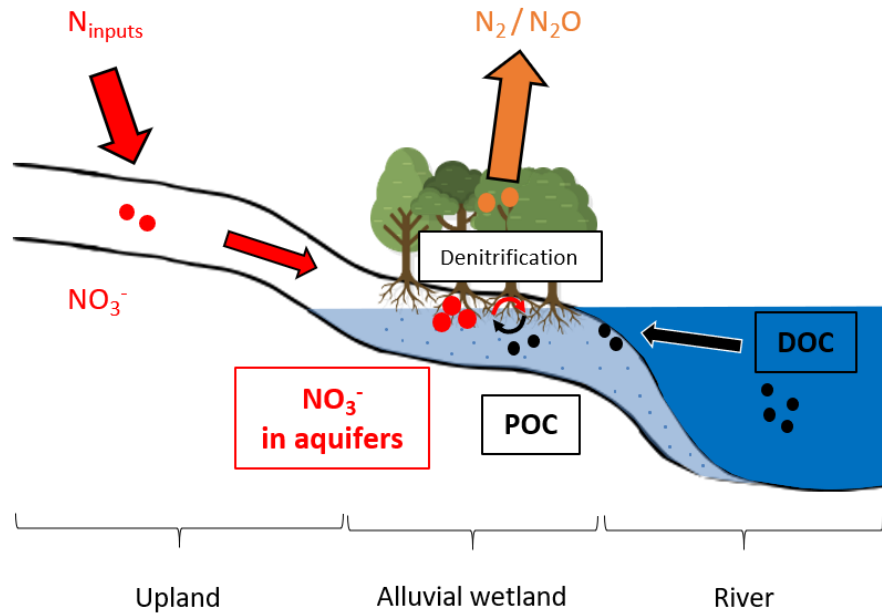
146

147 **2.3 Model implementation for denitrification**

148 The first model applied by Peyrard et al. (2011) estimated the denitrification rate in the
149 hyporheic zone. This rate estimation depends on the availability of POC, DOC and NO_3^- as
150 well as oxygen (O_2) availability and influence of nitrification rate from ammonia (NH_4^+)
151 transformation. Sun et al. (2018) simplified the equation by removing the ammonia term and
152 used surface water-groundwater exchanges to approach the anaerobic conditions. A focus on
153 the soil water content is necessary to assess when anaerobic conditions are occurring to
154 trigger denitrification (Sauvage et al., 2018; Sun et al., 2018).

155 Guilhen et al. (2020) used remote sensing data to assess the extent of water bodies as well
156 as the water saturation in soils where denitrification occurs. Indeed, the denitrification rate in
157 this study depends on the Surface Water Fraction (SWAF) product. Although this product
158 possesses a low spatial resolution (25 km x 25 km for one pixel), its high frequency (3 days to
159 map the whole Amazon Basin; Parrens et al., 2019) makes it possible to record a sudden
160 change in the hydrology. By comparing the brightness temperature of forest and water, a
161 percentage of water cover in a pixel was deduced and used to estimate the anaerobic
162 conditions in the model of Peyrard et al. (2011). Nevertheless, the SWAF data determine the
163 surface water extent in a pixel with a coarse resolution of 25 km x 25 km. However, the SWAF
164 methodology had only been used on the Amazon River so far (Parrens et al., 2017; 2018;
165 2019) and remote sensing data used in this methodology are not available for Arctic zones yet.

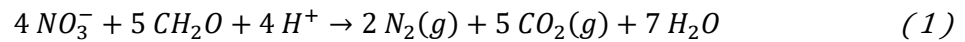
166 In this study, we followed the conceptual schema shown in Figure 2 with denitrification
167 occurring in the floodplain aquifers by using the available nitrate and OC content in aquifers.



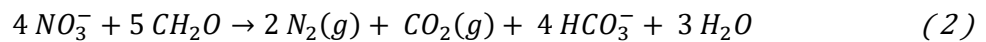
168

169 **Figure 2: Conceptual representation of the denitrification in floodplains based on the previous studies of**
 170 **Sánchez-Pérez and Trémoières (2003), Sauvage et al. (2018) and Sun et al. (2018).**

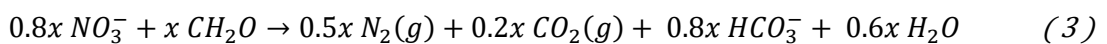
171 The denitrification process studied in past research is as followed:



172 Abril and Frankignoulle (2001) demonstrated an increase in alkalinity due to wetland
 173 denitrification. To take this phenomenon into account, the formation of HCO_3^- from dissolved
 174 CO_2 (eq.2) was coupled to the denitrification (eq.1). Overall, in this study, denitrification was
 175 modelled using the following equation:



176 By using $x = 5$ in (2) to compare the use of organic carbon and the consumption or the
 177 production of the other molecules (Peyrard et al., 2011), we obtain:



178 Sun et al. (2018) showed the capability of Peyrard et al. (2011) model to describe the
 179 denitrification rates in the main floodplains of the Garonne by comparing their simulations with
 180 in-situ denitrification measurements. However, applying this model at the watershed scale or
 181 in other watersheds was not practicable because of its specific design for the middle course

182 Garonne floodplains. To further estimate denitrification in contrasting basins, we investigated
183 a more straightforward method considering OC dynamics and anaerobic conditions.

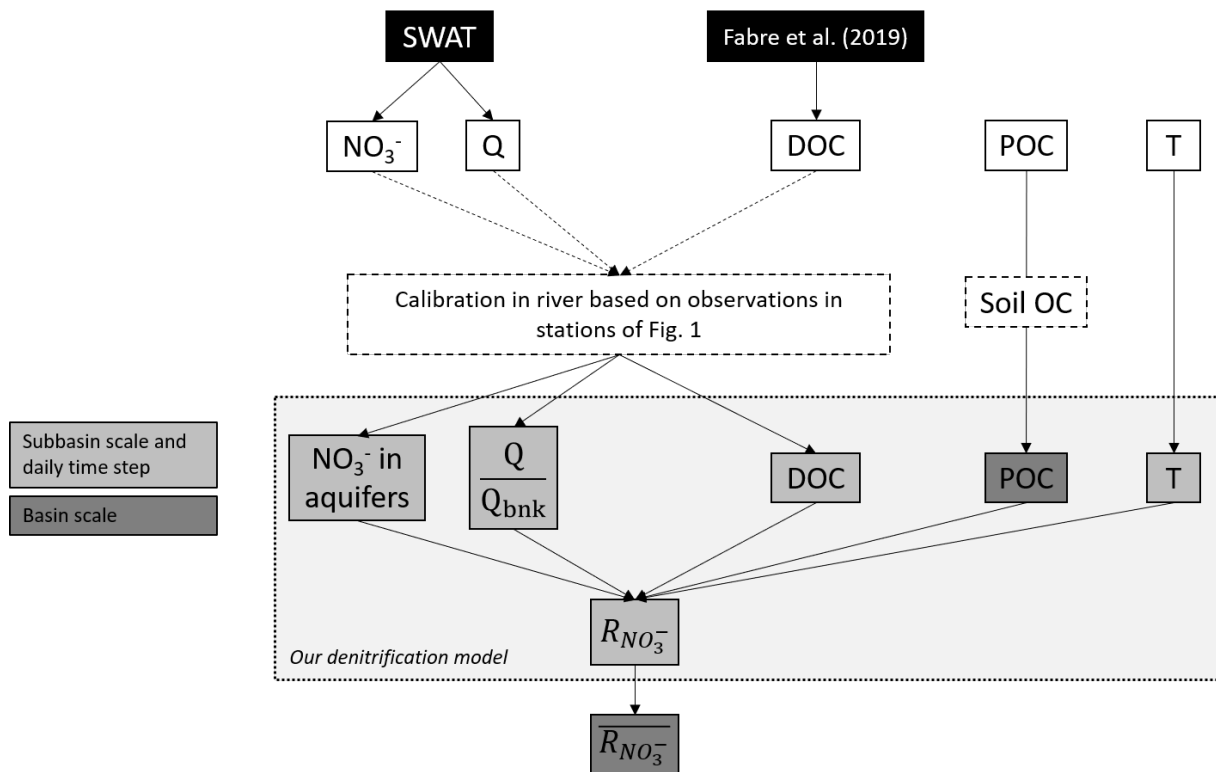
184 Therefore, we applied a new version of the model allowing an estimation of the denitrification
185 rate based on easy-to-obtain variables as followed:

$$R_{NO_3,i} = -0.8x \left(\rho \cdot \frac{1-\varphi}{\varphi} \cdot k_{POC} [POC_i] \cdot \frac{10^6}{M_c} + k_{DOC} [DOC_i] \right) \cdot \frac{[NO_{3,i}]}{K_{NO_3} + [NO_{3,i}]} \cdot \frac{Q_i}{Q_{bnk}} \cdot e^{-\frac{(T_i - T_{opt})^2}{100}} \quad (4)$$

186 where $R_{NO_3,i}$ is the denitrification rate in $\mu\text{mol.L}^{-1}$ on day i , $0.8x$ is the stoichiometric proportion
187 of nitrate consumed in denitrification compared to the organic matter used with $x = 5$, ρ is the
188 dry sediment density in kg.dm^{-3} , φ corresponds to the sediment porosity, k_{POC} and k_{DOC} are
189 the mineralization rate constants of POC and DOC (day^{-1}), $[POC_i]$ and $[DOC_i]$ are the
190 concentrations on day i ($\mu\text{mol.L}^{-1}$) of POC in alluvial soils and DOC in the river, M_c is the carbon
191 molar mass (g.mol^{-1}), $[NO_{3,i}]$ is the nitrate concentration in the aquifer on day i ($\mu\text{mol.L}^{-1}$), K_{NO_3}
192 is the half-saturation constant for nitrate limitation ($\mu\text{mol.L}^{-1}$), Q_i and Q_{bnk} are the discharge on
193 day i and the discharge at bank full depth, T_i and T_{opt} are the temperature in the subbasin on
194 day i and the optimal temperature for denitrification. T_{opt} was fixed to 27°C (Saad and Conrad,
195 1993; Canion et al., 2014; Brin et al., 2017). The stoichiometric ratio between the consumption
196 of nitrate and OC in the denitrification is $0.8x$ as in (3). More details on the conceptualization
197 of the model could be found in Peyrard et al. (2011).

198 Our global modelling strategy consists in the application of the former model (equation 4) with
199 the help of N and C entry data coming from two different sources (Figure 3). Firstly, a generic
200 model calculates the DOC concentrations in rivers, and secondly, the SWAT model estimates
201 nitrate concentrations in aquifers. We correlated the daily DOC concentrations to the daily
202 discharge with the relation proposed by Fabre et al. (2019) for the study case of the Yenisei
203 River. We assumed that POC concentrations in soils are not profoundly affected in time. POC
204 concentrations were considered much larger than the other nutrients involved in the
205 denitrification model. Thus, we fixed the values of average POC content in soils for each
206 watershed based on Batjes (2009).

207 DOC concentrations in the river and NO_3^- content in aquifers were extracted or calculated in
 208 each subbasin, as explained in the following paragraphs. Then, our model estimates the
 209 denitrification rate at a daily time step for each water body. Finally, these calculations helped
 210 to determine an average annual denitrification rate. Figure 3 summarizes our approach used
 211 to estimate the daily denitrification rate in the floodplains of the three watersheds.



212
 213 **Figure 3: Details of the different steps of the denitrification model setup. First, the Soil and Water**
 214 **Assessment Tool (SWAT) and the Fabre et al. (2019) model are calibrated to estimate discharge and riverine**
 215 **nutrients concentrations. Then, these results are used to follow nutrients contents in floodplains aquifers**
 216 **and to calculate the denitrification rate.**

217
 218 Denitrifying bacteria are more efficient at an optimal temperature of around 25-30°C (Saad and
 219 Conrad, 1993; Canion et al., 2014; Brin et al., 2017). Therefore a temperature term following
 220 a Gaussian function with an optimum was added into the model of Sun et al. (2018) to better
 221 describe the denitrification variability according to the watersheds with various climates.

222 We fixed the half-saturation constant for nitrate limitation based on Peyrard et al. (2011)
 223 estimations in the hyporheic zone from in-field measurements. The two OC mineralization rate
 224 constants were calculated by Sun et al. (2018) based on in-situ observations on the Garonne

225 River. These two parameters integrate the temperature effect on the microbial ability to
 226 degrade the organic matter. New k_{POC} and k_{DOC} values independent from the temperature
 227 allow exporting this calibration to the two other watersheds. These new values were obtained
 228 by dividing k_{POC} and k_{DOC} of Sun et al. (2018) by the temperature term of (4) filled with the
 229 average temperature in the Garonne watershed ($\bar{T}_{Garonne}$) as followed:

$$\left\{ \begin{array}{l} k_{POC} = \frac{k_{POC, Sun\ et\ al.(2018)}}{e^{\frac{-(\bar{T}_{Garonne} - T_{opt})^2}{100}}} \\ k_{DOC} = \frac{k_{DOC, Sun\ et\ al.(2018)}}{e^{\frac{-(\bar{T}_{Garonne} - T_{opt})^2}{100}}} \end{array} \right. \quad (5)$$

230 These new k_{POC} and k_{DOC} values were assumed valid to be used for the two other watersheds
 231 since the daily temperatures control the denitrification rates variations.

232

233 **2.4 Model choice to estimate Nitrate and DOC dynamics**

234 This study uses the Soil and Water Assessment Tool (SWAT) model to assess and quantify
 235 nitrate and OC dynamics based on discharge simulations of the three selected watersheds.
 236 SWAT is a hydro-agro-climatological model developed by USDA Agricultural Research Service
 237 (USDA-ARS; Temple, TX, USA) and Texas A&M AgriLife Research (College Station, TX, USA;
 238 Arnold et al., 1998). Its performance has already been tested at multiple catchment scales in
 239 various climatic and soil conditions on water, sediment and water chemistry especially nitrogen
 240 (Fu et al., 2019) and organic carbon (Oeurng et al., 2011) exports. Theory and details of
 241 hydrological and water quality processes integrated into SWAT are available online
 242 (<http://swatmodel.tamu.edu/>). For the Garonne River, we integrated most of the anthropogenic
 243 pressures in the basin to represent the watershed dynamics. Irrigation and dam management
 244 were implemented into the modelling based on national surveys from CACG
 245 (<https://www.cacg.fr/fr/>) and Electricité de France (REGARD-RTRA/STAE program). In the
 246 same way, city effluents were calibrated based on European databases of UWWTP – EUDB

247 (EEA Report, 2013; <https://ec.europa.eu/>). Finally, land-use databases were updated to better
248 simulate the fertilizers supply in the basin and to better match with national crop yields, as
249 demonstrated in Cakir et al. (2020). The SWAT model integrates the nitrogen cycle. SWAT
250 calculates the denitrification in soils but does not consider the denitrification occurring in
251 aquifers.

252 The nitrogen cycle in SWAT was calibrated with observed in-stream nitrate concentrations
253 available at the different gauging stations shown in Figure 1. Based on the correlation between
254 observed and simulated concentrations during low flow periods, we assumed that simulated
255 nitrate concentrations in aquifers are representative of real conditions. Thus, the nitrate
256 concentrations in aquifers, as new denitrification model inputs (equation 4), were extracted
257 from the SWAT model at the subbasin scale and at a daily time step. Concerning the anaerobic
258 conditions, as it was demonstrated in Sun (2015), the denitrification rate is linked to the water
259 volume stored in floodplains aquifers. The latter is linked to the water level in the channel
260 (Helton et al., 2014; Sun et al., 2018). Therefore, we considered a ratio between the daily
261 discharge in the stream extracted from SWAT and the discharge at bank full depth. The ratio
262 is limited to 1 and depicts the gap between the current discharge and the discharge needed to
263 produce a flooding. It is linked to the aquifers filling and trigger denitrification when it is close
264 to 1. SWAT accurately simulates the discharges at different time steps and at small or large
265 scales (Ferrant et al., 2011; Lu et al., 2019). However, the SWAT model encounters difficulties
266 to estimate discharges at bank full depth with accuracy due to the different resolutions of the
267 Digital Elevation Models (DEMs) used. Based on rating curves in gauging stations of the three
268 watersheds, we adjusted the value of the discharge at bank full depth (Q_{bnk}) to allow better
269 variations of the Q/Q_{bnk} ratio in time and space. We used ratios of 7/8, 1/5 and 1/4 to refine
270 bank full depth discharges for the Amazon, the Garonne and the Yenisei, respectively.
271 Consequently, bank full depth discharges were changed from 262,000 $m^3.s^{-1}$ to around
272 200,000 $m^3.s^{-1}$ at Obidos for the Amazon River, from 7,700 $m^3.s^{-1}$ to 640 $m^3.s^{-1}$ at Verdun for

273 the Garonne River and from above 1,400,000 m³.s⁻¹ to 140,000 m³.s⁻¹ at Dudinka for the
274 Yenisei River.

275

276 **2.5 Hydrology calibration**

277 Hydrology was first manually calibrated. Then an automatic calibration with three loops of 500
278 calibrations was done on the Yenisei and the Garonne basins as evoked in Fabre et al. (2017)
279 and Cakir et al. (2020) with the SWAT-CUP software. For the Amazon River, the hydrology
280 was calibrated manually as for the OC and the nitrate dynamics. The calibration was performed
281 with available observations in rivers extracted from the Observation Service SO HYBAM
282 (<https://hybam.obs-mip.fr/>), the French Water Agency of the Garonne River ([http://www.eau-](http://www.eau-adour-garonne.fr/)
283 [adour-garonne.fr/](http://www.eau-adour-garonne.fr/)) and the Arctic Great Rivers Observatory (Holmes et al., 2018) datasets for
284 the Amazon, the Garonne and the Yenisei, respectively. For the Amazon, we calibrated and
285 validated the model manually over the 2000-2009 period and the 2010-2016 period,
286 respectively. For the Garonne, the model was calibrated from 2000 to 2005 and was validated
287 from 2006 to 2010 based on Cakir et al. (2020). For the Yenisei, the model was calibrated over
288 2003 to 2010 and validated over the 2011-2016 period based on Fabre et al. (2019).

289

290 **2.6 Validity of simulated Nitrate and DOC dynamics**

291 We used two indices to validate our simulated riverine nitrate and DOC concentrations with
292 observed data: the coefficient of determination (R²) and the percentage of bias (PBIAS). These
293 indices are detailed in Moriasi et al. (2007). R² ranges from 0 to 1, with higher values indicating
294 less error variance. R² higher than 0.3 could be considered acceptable for daily biogeochemical
295 modelling (Moriasi et al., 2015). PBIAS expresses the percentage of deviation between
296 simulations and observations. Thus, the optimal value is 0. PBIAS can be positive or negative,
297 which reveals a model underestimation or overestimation bias, respectively (Moriasi et al.,
298 2007).

299 **2.7 Water quality efficiency ratio**

300 We used an efficiency ratio R based on the exported fluxes out of the basin (F_{outlet}) to test the
301 denitrification efficiency in the watershed. This ratio compares the nutrients flux consumed by
302 denitrification (F_{denit}) to the total fluxes exported, e.g. exported at the watershed outlet and
303 removed by denitrification:

$$R = \frac{F_{denit}}{F_{denit} + F_{outlet}} \quad (6)$$

304

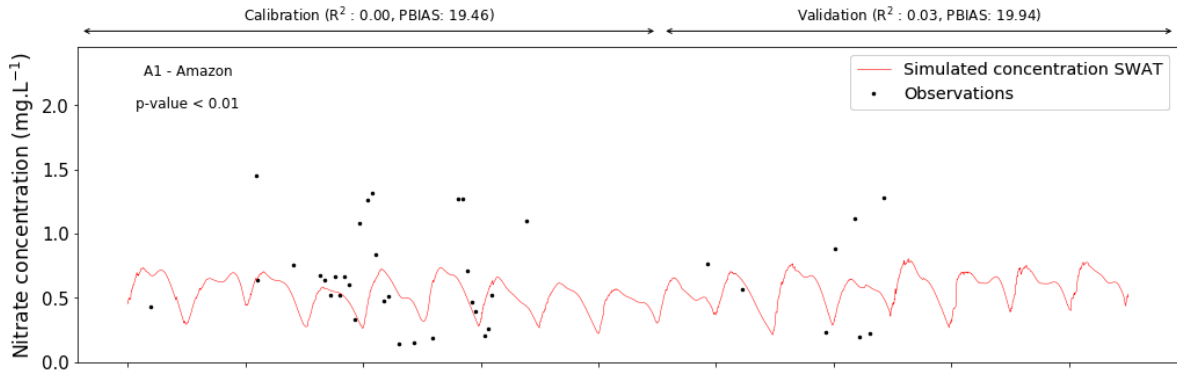
305 **3 Results**

306 **3.1 Nitrate and DOC simulations from SWAT in the three watersheds**

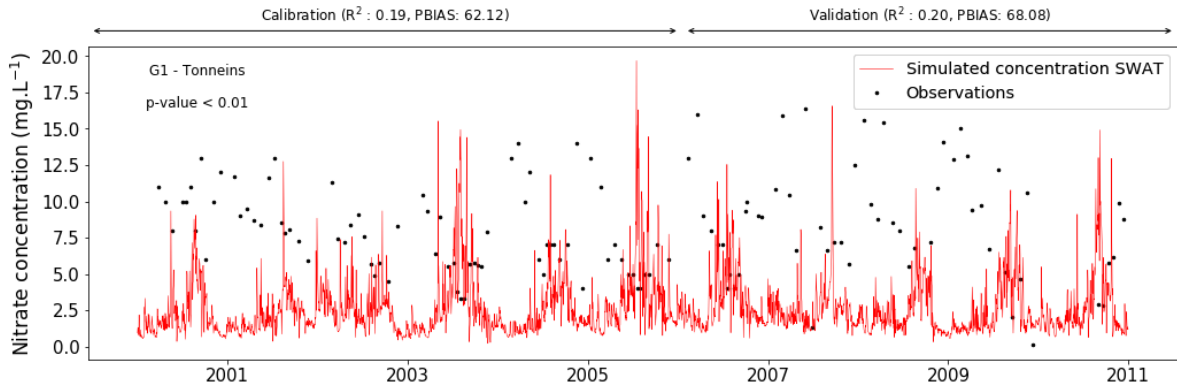
307 The results of nitrate and DOC dynamics at the Amazon outlet are in the range of in-situ
308 observations regarding the PBIAS index. Still, they show discrepancies with the temporal
309 variations (Figure 4a and Figure 5a). On the Garonne River, the simulated nitrate
310 concentrations are in the range of observations during high flow periods but display
311 underestimations during low flow periods (Figure 4b). Concerning DOC concentrations, the
312 simulations are in agreement with the observations ranges on the three watersheds. They do
313 not simulate the dynamics of observed data in the Garonne and Amazon rivers accurately
314 (Figure 5). However, these simulations are conserved because the PBIAS index and the p-
315 values show that they are in the range of the observations with regards to the low number of
316 observed data (Moriasi et al., 2015). Based on this assessment, the simulated DOC fluxes
317 (Appendix 1) are assumed to describe the observed data adequately. In the same way, the
318 good representation of low-water nitrate concentrations upstream to the floodplains indicates
319 that the simulated nitrate content in the floodplains aquifers should be close to reality. Table 1
320 shows the fitted parameters used to obtain these theoretical C and N concentrations.

321

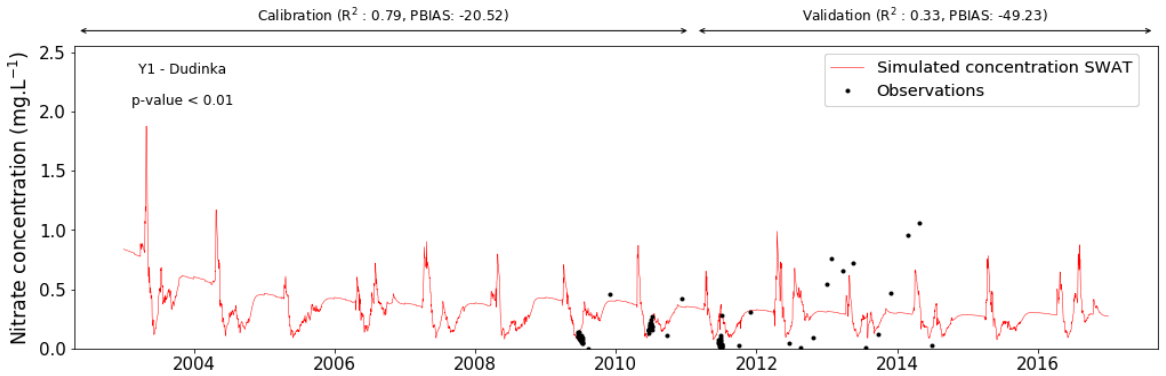
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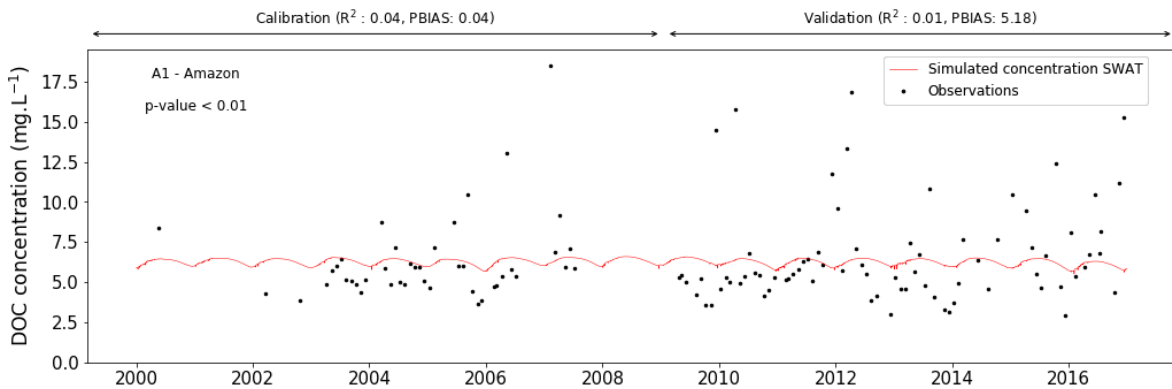


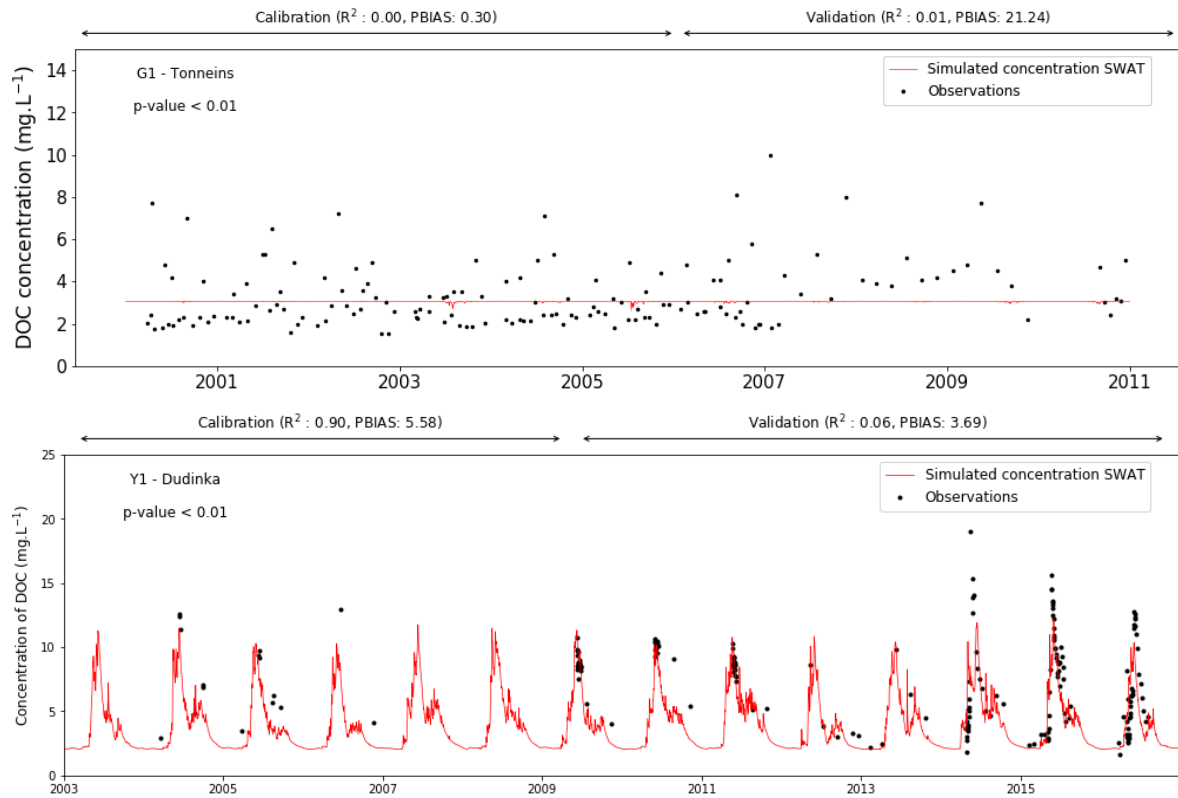
325

Figure 4: Daily observed and simulated nitrate concentrations (mg.L⁻¹) at the outlet of a) the Amazon, b) the Garonne and c) the Yenisei rivers. Locations of the sampling stations are found in Figure 1.

326

327





330 **Figure 5: Daily observed and simulated DOC concentrations at the outlet of a) the Amazon, b) the Garonne**
 331 **and c) the Yenisei rivers. The Yenisei graph is adapted from Fabre et al. (2019). Locations of the sampling**
 332 **stations are found in Figure 1.**

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Table 1: Fitted values of the SWAT parameters for the three watersheds for nitrate and organic carbon. The parameters for the Amazon River were calibrated manually based on riverine observations from the Observation Service SO HYBAM. The settings for the Garonne and Yenisei Rivers were adapted from Cakir et al. (2020) and Fabre et al. (2019), respectively. The SWAT parameters linked to denitrification refer to the one in soils. SWAT does not integrate the denitrification occurring in floodplains aquifers.

File	Parameter	Definition	Default	Value for:		
				Amazon	Garonne	Yenisei
<u>Nitrate parameters:</u>						
*.bsn	CDN	Denitrification exponential rate coefficient	1.0	0.5	1.0	3.0
	CH_ONCO_BSN	Channel organic nitrogen concentration in channel (ppm)	0	0	25	0
	CMN	Rate factor for humus mineralization of active organic nitrogen	0.0003	0.06	0.001	0.06
	IWQ	In-stream water quality (QUAL2E module)	1	0	1	0
	N_PERCO	Nitrogen percolation coefficient	0.2	3	0.58	0
	N_UPDIS	Nitrogen uptake distribution parameter	20	20	40	20
	RSDCO	Residue decomposition coefficient	0.05	0.1	0.1	0.01
	SDNCO	Denitrification threshold water content in soils	1.1	1.1	1.5	0.8
*.chm	SOL_NO3	Initial nitrate concentration in the soil layer	0	0	19	0
	SOL_ORGN	Initial organic N concentration in the soil layer	0	0	30	0
*.swq	BC1	Rate constant for biological oxidation of NH3 in the reach at 20°C (1/day)	0.550	0.550	1	0.550
	BC2	Rate constant for biological oxidation of NO2 to NO3 in the reach at 20°C (1/day)	1.100	1.100	2	1.100
	BC3	Rate constant for biological oxidation of NO2 to NO3 in the reach at 20°C (1/day)	0.210	0.210	0.21	0.210
	RS4	Rate coefficient of organic N settling in the reach at 20°C (1/day)	0.050	0.050	0.001	0.050
*.wwq	Al1	Fraction of algal biomass that is nitrogen (mg N/mg alg)	0.08	0.08	0.09	0.08
<u>Organic carbon:</u>						
*.bsn *.sub	α	Potential maximum DOC concentration in the river (mg.L ⁻¹)		5.72 – 12.43	2.10 – 3.38	15.0
	β	Discharge at which the DOC concentration equals half of α (mm.day ⁻¹)		0.001 – 0.74	0.001 – 0.03	1.22

339

340 **3.2 Simulated average denitrification rates in contrasting watersheds**

341 With the help of the new denitrification model (exposed in Equation 4), the parameters detailed
342 in Table 2 and the previous works on DOC exports, we were able to assess the floodplains
343 denitrification rates for the three considered watersheds. The average annual rates of the

344 floodplain denitrification are at $73.0 \pm 6.2 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ for the Amazon, $4.5 \pm 1.4 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$
 345 for the Garonne and $0.7 \pm 0.2 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ for the Yenisei.

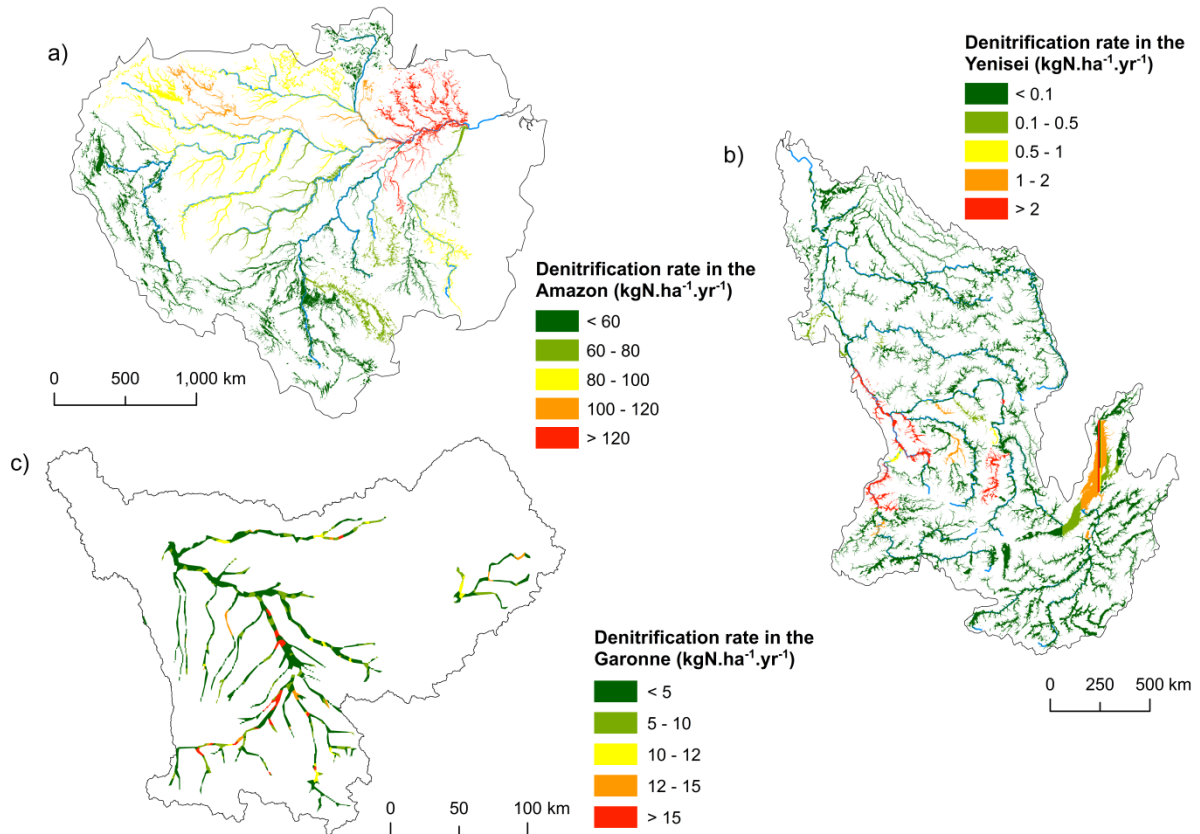
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347 **Table 2: Calibrated values of the different parameters used in the floodplains denitrification rates**
 348 **calculations based on the work of Peyrard et al. (2011) and Sun et al. (2018) on the Garonne River.**

Basin	Amazon	Garonne	Yenisei
$\rho \text{ (kg}\cdot\text{dm}^{-3}\text{)}$	0.25	0.15	0.1
φ	1.03	1.3	1.03
$k_{DOC} \text{ (day}^{-1}\text{)}$	$1.88\cdot 10^{-3}$	$3.63\cdot 10^{-3}$	$1.88\cdot 10^{-3}$
$k_{POC} \text{ (day}^{-1}\text{)}$	$2.75\cdot 10^{-4}$		
$[POC] \text{ (}\mu\text{mol}\cdot\text{L}^{-1}\text{)}$	1	1.5	33
$K_{NO_3} \text{ (}\mu\text{mol}\cdot\text{L}^{-1}\text{)}$	30		
$T_{opt} \text{ (}^\circ\text{C)}$	27		

349

350 Figure 6 shows the annual average denitrification fluxes in floodplains found in this study. It
 351 highlights the hot spots of denitrification for each of the three watersheds. The hot spots for
 352 the Amazon basin are located in the Northern part of the watershed. At the same time, the
 353 denitrification in the Garonne basin is usually higher in the primary active floodplains between
 354 the stations G3 and G4 but also in the upstream parts near G5 (see Figure 1 for stations
 355 locations). For the Yenisei watershed, the hotspots are located in the unfrozen parts of the
 356 basin and in the Lake Baikal.



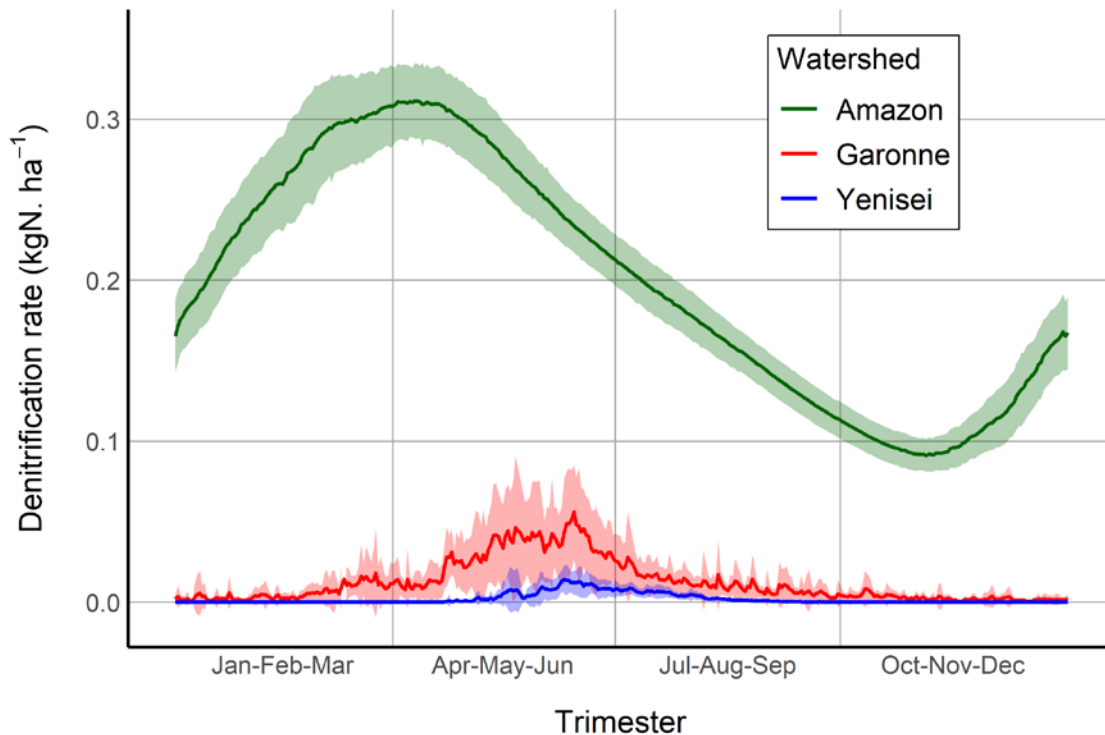
357

358 **Figure 6: Representation of the mean annual average DOC consumption ($\text{kgC}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$) in denitrification in**
 359 **floodplains of the three selected watersheds on the 2000-2010 period.**

360

361 **3.3 Temporal variability of the denitrification**

362 Figure 7 shows the average daily denitrification rates (R_{NO_3}) on the 2000-2010 period for the
 363 three watersheds. For the Amazon, R_{NO_3} is maximal in April with a removal around $0.31 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{day}^{-1}$.
 364 The lowest values are around $0.09 \text{ kgN}\cdot\text{ha}^{-1}\cdot\text{day}^{-1}$ in October. R_{NO_3} reaches 0.06
 365 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{day}^{-1}$ in May on the Garonne and is lower during the cold season between October
 366 and February. With the same pattern, the Yenisei shows higher rates during the unfreezing
 367 period around May, but these rates are still low compared to the two other basins.



368
 369 *Figure 7: Average daily variations of the denitrification rates in the floodplains of the three selected*
 370 *watersheds on the period 2000-2010.*

371

372 4 Discussion

373 4.1 Methodologies used

374 This paper exposed the capability of a simple model to describe daily denitrification rates in
 375 floodplains of contrasting watersheds. It is the first attempt to simulate, understand and
 376 compare daily denitrification rates in three different basins by applying a dynamic model.
 377 Previous large-scale denitrification models provided either estimation of interannual fluxes or
 378 assessed the denitrification contribution to the nitrogen budget (Birgand et al., 2007; Boyer et
 379 al., 2006; Groffman, 2012; Thouvenot-Korpoo et al., 2009). Others studies used models to
 380 estimate the denitrification at the global scale (Seitzinger et al., 2006). However, none have
 381 supplied daily denitrification rates yet. The need for a daily time step is important, particularly
 382 for basins subjected to sudden changes in the hydrological dynamic (as flash-flood in the
 383 Garonne River).

384 Compared to previous research, the model used in this paper was modified to integrate a new
385 temperature dependence of R_{NO_3} with an optimal set at 27°C. This term allowed the comparison
386 of the denitrification rates between watersheds with different climates. This dependence is
387 essential, especially for the Yenisei River, where the solutes are available, but the cold climate
388 inhibits the microbial activity. Other studies mentioned an optimal temperature around 45°C
389 for this process in soils (Benoit et al., 2015; Billen et al., 2018). More research is needed to
390 better understand and consider this temperature effect in the proposed method.

391 The significant improvement of this model comes from the integration of the different carbon
392 sources, together with nitrates as substrates, so that the stoichiometric ratio controls the
393 denitrification rates. This operation was made possible with the help of C & N data sources
394 with accurate temporal and spatial scales. The integration of the model of Fabre et al. (2019)
395 to estimate the daily variations of DOC concentrations in the river as a source of C data to use
396 for control of stoichiometric ratio the new model makes part of the new aspect. DOC plays a
397 predominant role in the denitrification process. Therefore, the integration of the simulated DOC
398 concentrations at a daily time step in the river is a notable improvement to refine denitrification
399 estimates at the watershed scale.

400 The other part of the model concerned by the organic carbon integrates the role of the POC.
401 POC was set up in the model depending on the average soils OC content of the three
402 watersheds floodplains. A first improvement would be to spatialize more accurately the POC
403 content at the subbasin scale. Therefore, more research should be conducted to validate global
404 datasets of soil OC. Plus, the POC content used in this study does not consider the POC
405 renewal by deposition during flooding events. The soil OC turnover may boost floodplain
406 denitrification but was not studied yet.

407 Nonetheless, this model does not consider OC lability, which is essential in the estimation of
408 denitrification rates (Zarnetske et al., 2011). Around 20% of the DOC is labile in freshwater
409 ecosystems (Søndergaard and Middelboe, 1995; Guillemette and del Giorgio, 2011;
410 McLaughlin and Kaplan, 2013). Integrating the lability in the denitrification model may improve

411 C and N dynamics in floodplains. Moreover, DOC is the most consumed form in denitrification
412 (Peyrard et al., 2011; Zarnetske et al., 2011). Yet, the model does not integrate the dominant
413 use of the DOC compared to the POC.

414 The delineation system from Rathjens et al. (2015) showed its capability to visualise the
415 floodplains as a functional and active area. This tool could be further compared to remote
416 sensing data from SWAF on the Amazon and other systems to see if easy-to-obtain data such
417 as the DEM are sufficient to estimate floodplains coverage at the watershed scale.

418 Concerning the ratio between daily discharge and discharge at bank full depth, correction
419 parameters were applied on the discharges at bank full depth based on known parts of the
420 three watersheds. Uncertainties could remain in some other parts of the catchments, which
421 would have a significant impact on the denitrification variations in surrounding areas. A better
422 definition of discharge at bank full depth in the different parts of the basins may improve
423 floodplains denitrification estimates at the watershed scale.

424 Moreover, we defined the mineralization rate constants for DOC and POC based on scarce in-
425 situ measurements (Sun et al., 2018) which are non-representative of the entire watershed.
426 Indeed, k_{POC} and k_{DOC} vary under the influence of multiple drivers such as soils characteristics,
427 temperature and microorganism's activity (geophysical and biological characteristics). An
428 improvement in the calculations of denitrification rates could be to measure these coefficients
429 in different areas and determine their temporal and spatial variability in the floodplains.

430 Concerning the half-saturation constant for nitrate limitation, this variable was based on in-situ
431 observations in the Garonne hyporheic zone (Peyrard et al., 2011). Again, other
432 measurements are needed to refine the value of K_{NO_3} in floodplains of various watersheds.

433 Lastly, this study outlines some weaknesses in the estimation of denitrification rates. Alluvial
434 wetlands show higher denitrification than other areas in floodplains (McClain et al., 2003;
435 Harrison et al., 2011). However, the model proposed here does not distinguish alluvial
436 wetlands from the rest of the floodplains. Accurate mapping of alluvial wetlands at the

437 watershed scale would help the scientific community to better estimate the specific
438 denitrification rates in these highly reactive areas and consequently would improve the
439 estimates in other areas of the floodplains.

440 **4.2 Calibration of the inputs for denitrification**

441 The concentrations of nutrients in floodplains were extracted from the SWAT model. This
442 model, as shown in Table 1, already integrates denitrification processes. However, the
443 denitrification represents the one occurring in uplands soils and stream but does not integrate
444 the predominant role of floodplains aquifers (McClain et al., 2003). Therefore, our model
445 proposed in this study could fill the gap and help to approach the floodplains denitrification
446 contribution to N and C dynamics at the watershed scale.

447 This paper shows that the N and C inputs were calibrated successfully in different areas of the
448 three watersheds. Nevertheless, these calibrations may be improved by better representing
449 in-stream and uplands processes to improve the calibration of nitrate and organic carbon
450 concentrations in floodplains aquifers. In the same way, nitrate concentrations in the Garonne
451 River are underestimated and could induce lower denitrification.

452 Concerning OC, uncertainties remain in the simulation of DOC concentrations in the three
453 watersheds on the one hand (Figure 5). Different processes and conditions are not considered
454 in the model of Fabre et al. (2019) yet. Anthropogenic pressures in the Garonne River, as well
455 as consumption, deposition, or floodplain deliveries for the Amazon River, are conditions that
456 could explain the observed DOC variations. Process-based models could help to improve the
457 DOC simulations by considering various in-stream processes such as in-stream assimilation
458 or production (Du et al., 2020). However, DOC and nitrate concentrations are in the range of
459 observations. Thus, the other components of the model regulate the denitrification rates on the
460 Amazon River and the Garonne River.

461 On the other hand, the variations of observed DOC concentrations are so intense that the data
462 quality could be discussed. Sampling nitrate and DOC in streams is difficult in large watersheds

463 due to in-situ conditions. Thus, an improvement in the quality of data could be required to refine
464 the parameters of the DOC model and to improve the modelling efforts for the nitrate
465 concentrations or to confirm that the DOC model of Fabre et al. (2019) is adapted to various
466 climatic and soils conditions.

467 **4.3 Temporal and spatial validity of the resulting floodplains denitrification** 468 **rates**

469 We showed that even if the dynamics of nitrate and DOC concentrations in rivers are hard to
470 obtain, these concentrations are in the range of observed data. Nevertheless, the
471 concentrations at the outlet already integrate the complex processes occurring in the
472 watershed. Consequently, we were able to extract from the SWAT model the average nitrate
473 concentrations in the floodplains aquifers and compared it with the literature. In the Amazon
474 watershed, the simulated nitrate concentrations in aquifers are around $0.9 \pm 0.6 \text{ mgN-NO}_3\text{.L}^{-1}$.
475 These values are in the range of the observations made in previous works ($0.04\text{-}2.8 \text{ mgN-}$
476 $\text{NO}_3\text{.L}^{-1}$; McClain et al., 1994; Leite et al., 2011). Concerning the Garonne basin, the SWAT
477 model simulated average nitrate concentrations of $8.6 \pm 5.8 \text{ mgN-NO}_3\text{.L}^{-1}$ in aquifers while past
478 research measured concentrations between 3.86 and 17.95 $\text{mgN-NO}_3\text{.L}^{-1}$ (Jégo et al., 2012;
479 Sun et al., 2018). The average nitrate concentrations in the Yenisei aquifers were far lower
480 than the other basins with values around $0.01 \pm 0.14 \text{ mgN-NO}_3\text{.L}^{-1}$. As no literature is available
481 to validate these values in the Yenisei, we assumed that they are representative and could be
482 used to estimate denitrification rates.

483 To validate our simulated denitrification rates, we compared our outputs with results from other
484 studies in the same watersheds. Sánchez-Pérez et al. (2003) and Sun et al. (2018), based on
485 in-situ observations, found that a highly reactive ecological corridor including efficient alluvial
486 wetlands in the floodplains of the Garonne watershed provides a denitrification rate of 21-25
487 $\text{kgN-NO}_3\text{.ha}^{-1}\text{.yr}^{-1}$. On the same part of the watershed, our study gives a nitrate removal of 19.9
488 $\text{kgN-NO}_3\text{.ha}^{-1}\text{.yr}^{-1}$. Our rates are in the same order of magnitude, which could allow validating
489 the method used in this paper.

490 We compared our results from the Amazon watershed with the estimation of Guilhen et al.
491 (2020). This work focused on the three main floodplains of the Amazon: one alongside the
492 mainstream near Obidos (station A1), one alongside the Branco and Negro rivers and one in
493 the upstream Bolivian parts of the Madeira Basin. They found denitrification rates of 142.5
494 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the mainstream floodplain, 38.8 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the Branco floodplain and 60.4
495 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the Madeira floodplain. In our study, we found denitrification rates of 165.7
496 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the mainstream floodplain, 144.3 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the Branco system and 67.6
497 $\text{kgN}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the Madeira upstream part. Only the Branco floodplain shows different results.
498 This offset could be due to different drivers influence. Guilhen et al. (2020) estimated the DOC
499 concentrations at a monthly time step with high variations. In our study, the daily DOC
500 concentrations are relatively constant but are still closer to the real concentrations. Their
501 denitrification rates depend on the presence of water in the soil surface with a binary approach.
502 In our study, the integration of the ratio between discharge and discharge at bank full depth
503 improves the understanding of the denitrification dynamic. This improvement is not obvious in
504 tropical systems such as the Amazon River because denitrification is occurring during the
505 frequent and long-lasting flooding events. Therefore, our approach may be more relevant for
506 basins where flooding events occur at a high temporal frequency, such as the Garonne River.
507 The temporal resolution of this study highlighted preferential periods of denitrification. For the
508 three watersheds, the periods of high-water flows show a higher denitrification rate as implied
509 by the model.

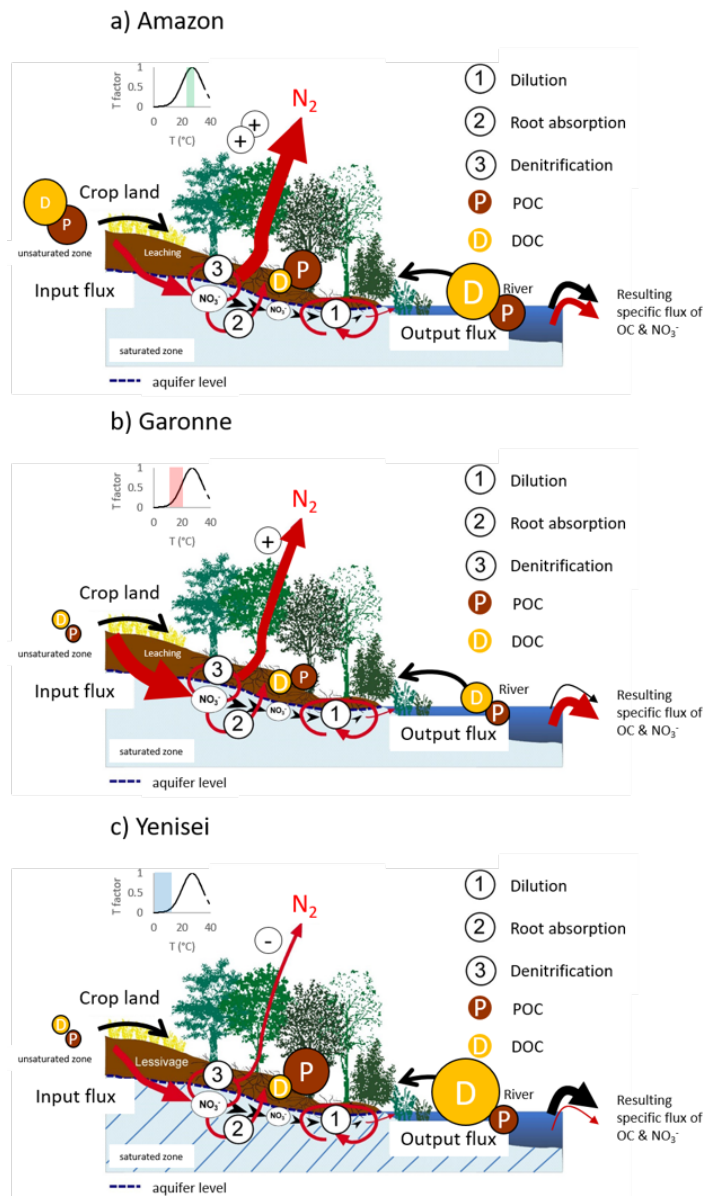
510 **4.4 Efficiency of the different floodplains**

511 By including contrasting watersheds, this paper brings to light a comparison of the efficiency
512 of different types of floodplains with various anthropogenic and climatic contexts. Denitrification
513 dynamics follow the hydrological cycles. Denitrification rates peak when and where both nitrate
514 and DOC are not limiting factors like in the Amazon basin. On the contrary, the Garonne River
515 has high exports of nitrate due to the anthropogenic pressures within the watershed. The DOC
516 concentrations are always low except for some upstream parts of the watershed, which lead

517 to higher denitrification rates. The Yenisei River has high DOC concentrations during the
518 unfreezing period, but the low nitrate concentrations and the cold temperatures limit the
519 denitrification. The suggested conceptualization integrates all of these contrasts between
520 watersheds for the denitrification (Figure 8).

521 By comparing the average exports of nitrate and OC at the outlets of the three watersheds with
522 the denitrification rates, we were able to evaluate the floodplains contribution to the regulating
523 services of surface waters. The denitrification occurring in uplands and streams is already
524 integrated into the flux exported to the oceans and is negligible compared to the one in
525 floodplains. The DOC used for denitrification accounts for 10.4% of the total DOC flux exiting
526 the Amazon basin (exported to the ocean or consumed by denitrification). This ratio reaches
527 3.0% in the Garonne and amounts to 0.9% for the Yenisei basin. Concerning nitrates, those
528 processed in the denitrification represents 85% of the total nitrate flux exiting the Amazon basin
529 and 34% in the Garonne watershed. For the Yenisei watershed, only 13% of the total nitrate
530 flux exiting the basin is used for denitrification.

531 Concerning the Amazon and the Yenisei River, as the DOC concentrations are generally
532 higher than nitrate, only a few of the total DOC yield is needed for the denitrification. The
533 Garonne River, which is under high anthropogenic pressures, is characterized by soils with
534 low organic matter contents and high exports of nitrates. The resulting concentrations in the
535 river are quite in the same range, and a large part of the DOC export is needed to consume a
536 small amount of the aquifers nitrate content.



537

538 **Figure 8: Conceptualization of the denitrification model for the selected watersheds. In each case of study,**
 539 **the different variables favouring the process shows various intensities. Adapted from Sánchez-Pérez and**
 540 **Trémolières (2003) and Bernard-Jannin et al. (2017). The red arrows represent nitrogen dynamics, and black**
 541 **arrows represent organic carbon pathways. The top-left graph shows the variation of the temperature index**
 542 **in the denitrification model. The coloured zones are the temperature intervals for each watershed.**

543

544 5 Conclusion

545 This paper demonstrated the possibility of a simple model to simulate the floodplains
 546 denitrification rates in contrasting watersheds. We showed that tropical catchments that
 547 combine an average temperature around the optimal temperature for denitrification and large
 548 C availability show the highest amounts for the process. On the other hand, we confirmed that

549 C and N availability, as well as average temperature, could be limiting factors for floodplains
550 denitrification on both cold and temperate watersheds. This study also highlighted the role of
551 floodplains on water quality and their contribution to the stability and the resilience of the basins
552 subjected to future climate and land use changes.

553

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556 Electricité De France (project REGARD-RTRA/STAE), Compagnie d'Aménagement des
557 Coteaux de Gascogne, Banque Hydro, the Arctic Great Rivers Observatory and the TOMCAR-
558 Permafrost Project for the sharing of their data on DOC, nitrate and daily discharge in the
559 studied watersheds. This work is part of the governmental PhD program of Clément Fabre.

560

561 **7 Authors contributions**

562 C.F., J.G., S.S. and J.M.S.P. designed and developed the model with the help of R.C. C.F.
563 performed and analyzed the modelling. C.F. wrote the paper with considerable contributions
564 from J.M.S.P., S.S.; J.G., M.G. and R.C.

565

566 **8 Additional information**

567 This research did not receive any specific grant from funding agencies in the public,
568 commercial, or not-for-profit sectors.

569

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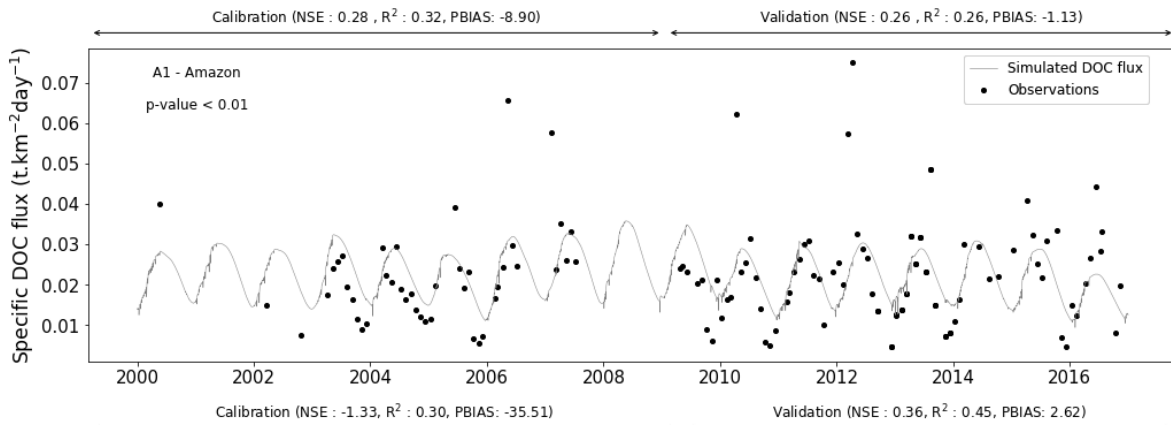
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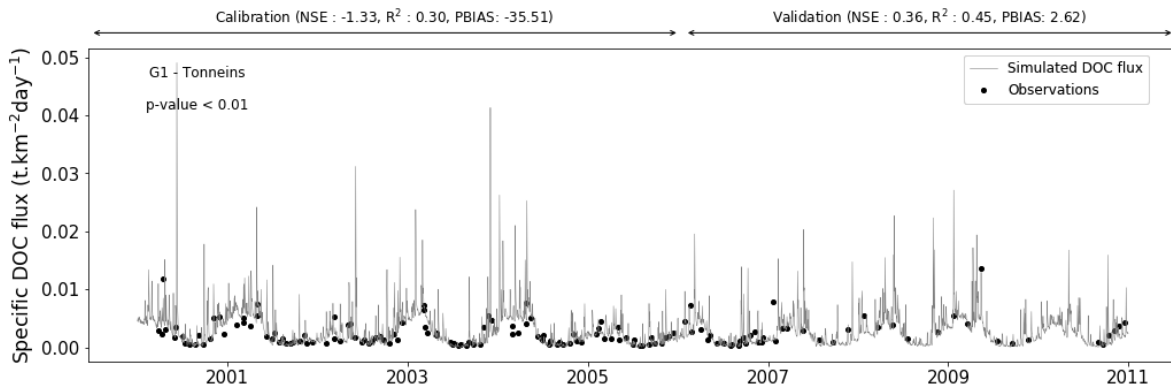
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831 **10 Appendices**

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835 **Appendix 1: Daily DOC fluxes exported at the outlet of a) the Amazon River, b) the Garonne River and c)**
836 **the Yenisei River. The Yenisei graph is adapted from Fabre et al. (2019). Locations of the sampling stations**
837 **are found in Figure 1.**