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Spatialised fate factors for nitrate in catchments: Modelling approach and implication for LCA results

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Abstract

The challenge for environmental assessment tools, such as Life Cycle Assessment (LCA) is to provide a holistic picture of the environmental impacts of a given system, while being relevant both at a global scale, i.e., for global impact categories such as climate change, and at a smaller scale, i.e., for regional impact categories such as aquatic eutrophication. To this end, the environmental mechanisms between emission and impact should be taken into account. For eutrophication in particular, which is one of the main impacts of farming systems, the fate factor of eutrophying pollutants in catchments, and particularly of nitrate, reflects one of these important and complex environmental mechanisms. We define this fate factor as: the ratio of the amount of nitrate at the outlet of the catchment over the nitrate emitted from the catchment's soils. In LCA, this fate factor is most often assumed equal to 1, while the observed fate factor is generally less than 1. A generic approach for estimating the range of variation of nitrate fate factors in a region of intensive agriculture was proposed. This approach was based on the analysis of different catchment scenarios combining different catchment types and different effective rainfalls. The evolution over time of the nitrate fate factor as well as the steady state fate factor for each catchment scenario was obtained using the INCA simulation model. In line with the general LCA model, the implications of the steady state fate factors for nitrate were investigated for the eutrophication impact result in the framework of an LCA of pig production. A sensitivity analysis to the fraction of nitrate lost as N_2O was presented for the climate change impact category. This study highlighted the difference between the observed fate factor at a given time, which aggregates both storage and transformation processes and a "steady state fate factor", specific to the system considered. The range of steady state fate factors obtained for the study region was wide, from 0.44 to 0.86, depending primarily on the catchment type and secondarily on the effective rainfall. The sensitivity of the LCA of pig production to the fate factors was significant concerning eutrophication, but potentially much larger concerning climate change. The potential for producing improved eutrophication results by using spatially differentiated fate factors was demonstrated. Additionally, the urgent need for quantitative studies on the N_2O/N_2 ratio in riparian zones denitrification was highlighted.

Keywords: Eutrophication; Environmental assessment; Life cycle assessment; Climate change; INCA; Denitrification; Riparian zone; Catchment hydrology

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1. Introduction

In regions of intensive agriculture, the contribution of farming systems to the degradation of the environment is increasingly investigated, especially concerning water quality. However, assessing the sustainability of such farming systems considering water pollution only can be misleading, because of possible trade-offs between their different impacts on the environment. More complete environmental assessment tools have been developed, to provide a holistic picture of the environmental impacts of a given system. The challenge of such tools is to be relevant both at a global scale, i.e., for global impact categories such as climate change, and at a smaller scale, i.e., for regional impact categories such as aquatic eutrophication.

Among those, the Life Cycle Assessment (LCA) approach, which considers the whole product life cycle, is recommended by the European Union (Anonymous, 2003) and UNEP (UNEP, 1996). The recent EU communication on Integrated Product Policy (IPP; Anonymous, 2003) states that "All products cause environmental degradation in some way, whether from their manufacturing, use or disposal. Integrated Product Policy (IPP) seeks to minimise these impacts by looking at all phases of a product's life cycle and taking action where it is most effective".

LCA has proved a valuable tool for the environmental evaluation of farming systems (Van der Werf and Petit, 2002). This methodology consists of four stages: the definition of the goal and scope of the study, the inventory analysis, the impact assessment (LCIA) and the interpretation. In the inventory analysis the resources consumed and the emissions to the environment are quantified at all stages of the life cycle of the product studied—from the extraction of resources, through the production of materials, product parts and the product itself, and the use of the product, to its reuse, recycling or final disposal (Guinée et al., 2002). For each environmental impact in the LCIA stage, a characterisation model is used to convert the inventory data contributing to this impact, into impact results. This is done by multiplying the emissions of each substance with a characterisation factor for each impact category to which it may potentially contribute. Characterisation factors are substance-specific, quantitative representations of the additional environmental pressure per unit emission of a substance.

To reach the general objective of taking all the impacts of a given system over time and space into account, in the general LCA model, the impacts are integrated over all the emission/impact locations (world) and over time (infinite horizon) in an assumed steady state (Guinée et al., 2002, p. 413). This situation generally corresponds to ignoring most of the environmental mechanisms between emission and impact as formalised by Heijungs and Wegener Sleeswijk (1999): emission, fate and effect, in each specific context of emission. The *emission* is defined as the output of pollutant from the system studied. The *fate of a pollutant* consists of its transport, its transformation and its accumulation or dilution in a compartment of the environment. The *sensitivity* of the ecosystem describes the way the ecosystem reacts to one dose of pollutant, for example through dose/effect curves.

For regional impact categories such as eutrophication, the simplifications of this generic model are considered excessive (Finnveden et al., 1992; Potting and Blok, 1994; Nichols et al., 1996; Potting and Hauschild, 1997; Finnveden and Potting, 1999; Udo de Haes et al., 1999b; Heijungs et al., 2003) especially when these categories dominate the system studied. SETAC (Society of Environmental Toxicology and Chemistry) now recommends to consider the cause and effect chain between emission and impact and to develop spatially differentiated characterisation factors (Udo de Haes et al., 1999a,b; Potting, 2000). The time differentiation of the characterisation factors is even harder to develop, due to a lack of data and because of the different time scales of each stage of the cause and effect chain.

Concerning the eutrophication characterisation factors, the spatial differentiation has rarely been implemented, or the results obtained still present much uncertainty (Seppälä et al., 2004). Some work has however been done on the fate of eutrophying pollutants in air (NH_3 and NO_x) by using the EMEP model (Potting et al., 1998; Huijbregts et al., 2001; Huijbregts and Seppälä, 2000; Seppälä et al., 2004). Huijbregts and Seppälä (2001) also proposed, on the basis of empirical data from the literature, fate factors for eutrophying pollutants reaching water via the soil. However, their approach is highly questionable for an application of LCA to farming systems. First of all, the system does not include the soil, which leads to defining the emissions as the rates of N and P applied to the soil and the fate factors for these emissions as the fractions of N and P which leave the soil by leaching or runoff. This is not in line with the actual definitions both of the system limits, the emissions and consequently the fate factors, in LCA studies of farming systems (Audsley et al., 1997; Cowell and Clift, 2000; Gosse et al., 2000; Brentrup et al., 2001; Sandars et al., 2003). In these studies, the soil is considered as being part of the studied system at least for the duration of the assessment. The emissions correspond then to the N and P compounds which leave the soil by runoff or leaching.

A lack of specific approaches covering the transfer of diffuse pollution from soil to sensitive ecosystems through the catchment has been diagnosed. Particularly, nitrate excess is problematic both for ecological issues, since nitrogen is the main limiting factor of eutrophication in coastal waters (Ménesguen, 2003), and for drinking water supply. This work focuses on the fate factor of nitrate in catchments.

We define the *fate factor* of nitrate in catchments as the ratio of the amount of nitrate at the outlet of the catchment over the nitrate emitted from the catchment's soils. The most common practice in LCA, is to assume this fate factor equal to 1 (Heijungs et al., 1992; Huijbregts and Seppälä, 2001). However, as a rule, the observed fate factor is less than 1, and even less than 0.5 (Aurousseau et al., 1996; Andersen et al., 2001; Haag and Kaupenjohann, 2001; Boyer et al., 2002; Van Breemen et al., 2002; Sebilo et al., 2003). The processes which may explain nitrate fate factors less than 1 are diverse. Nitrate can be stored temporarily in the soil, vadose zone or groundwater (Molénat and Gascuel-Odoux, 2002; Bordenave et al., 1999). It can be taken up by plants or integrated in the organic matter of soil (Mariotti, 1997) especially in buffer zones such as hedges, forests and riparian zones. In riparian zones, it can also be transformed into gas (nitrous oxide and nitrogen gas) by heterotrophic denitrification.

Considering that the general LCA model implies the integration of the impacts over time (Guinée et al., 2002), among these processes, the ones that are important in the estimation of the fate factor are those that affect the long term transformation of the nitrate ion, not its temporary storage. In this respect, the riparian zones are of major importance, especially in shallow groundwater areas. The two major processes responsible for removal of nitrate in riparian buffers are plant uptake and denitrification (Gilliam et al., 1997). Heterotrophic denitrification occurs at the surface (at less than a meter depth) in areas of hydromorphic soils (Curmi et al., 1997; Cey et al., 1999; Dhondt et al., 2002). This process has been studied in controlled conditions (Hwang and Hanaki, 2000), at small scales in situ (little plots along the river) (Curmi et al., 1997; Durand et al., 1998; Cey et al., 1999; Dhondt et al., 2002) and at riparian zone scale (Schipper et al., 1993; Haycock and Burt, 1993; Pinay et al., 1993). In most of these studies, it has been clearly shown to be the main mechanism responsible for nitrogen removal. However, riparian denitrification is still very difficult to quantify at the catchment scale. It is a highly variable process both in spatial (vertical and horizontal) and temporal dimensions (Pinay et al., 1993; Gilliam et al., 1997; Jordan et al., 1998; Rudaz et al., 1999; Clement et al., 2003). The groundwater table variations and the water residence time (Gilliam et al., 1997) are important, but also organic carbon and nitrogen sources, moisture, temperature, oxygen availability (Hénault, 1995; Machefert et al., 2002). Particularly, nitrate availability is often presented as a major controlling factor for denitrification (Jordan et al., 1998; Johnston et al., 2001; Davidsson et al., 2002; Xu et al., 2002). A low nitrate availability can be due to hydrological shortcuts (Durand et al., 1998), the contaminated water from uphill fields avoiding the reactive zones. At the regional scale, a positive relationship between nitrogen input load (estimation of the leachable nitrate at field level by calculating the nitrogen balance) and nitrate removal rate–defined as the ratio of the input load over the nitrate flux at the outlet–in catchments has been calculated by Aurousseau et al. (1996).

The only way to integrate all relevant hydrologic and biochemical processes at the catchment scale consists in conceiving and using modelling tools (Durand et al., 1998; Beaujouan et al., 2001; Machefert et al., 2002). Nitrogen models have been conceived assuming vertical transfer only and aggregated at the catchment scale, while neglecting the spatial interactions in the catchment (e.g. ANSWERS, Bouraoui and Dillaha, 2000; AGNPS, Line et al., 1997). It is suggested that the location of the pollution sources and sinks in the landscape, particularly the distance to the stream and the existence of buffer zones, should have an impact on the water quality. The export coefficient approach integrates this empirically (Johnes, 1996). In classical models of non-point sources pollution, the buffer zone concept (riparian wetlands, vegetated strips or hedgerows) is not explicitly taken into account (Mérot and Durand, 1997). Considering the role of buffer zones and of the spatial location and interaction of landscape elements in the catchment, process-based, relatively complicated and fully distributed models have been conceived (e.g. TNT, Beaujouan et al., 2001). These models require a lot of data, and remain difficult and time consuming to use. One compromise can be found in process-based conceptual models such as INCA (Integrated Nitrogen in Catchments, Whitehead et al., 1998; Wade et al., 2002a). INCA is a semi-distributed model including the major nitrogen processes in the catchment and allowing one to account for riparian denitrification (Durand, 2004).

As a consequence of the high variability of the denitrification process, the proportion of each gas (nitrous oxide or dinitrogen) into which nitrate is transformed remains quite unpredictable and the estimation of this fraction is not integrated in catchment models so far. Measurements of N_2O and N_2 in riparian zones are scarce, and cannot be extrapolated at the catchment scale and for the entire year. Furthermore, they suggest that, depending on the conditions, the fraction of $N–N_2O$ relative to the sum of $N–N_2O$ and $N–N_2$ could be anywhere in the (0–100%) interval: Jordan et al. (1998) obtained values between 10% and 92% in a riparian forest receiving nitrate in drainage from cornfields; in Switzerland, in a fertilised meadow, Rudaz et al. (1999) measured fractions of N_2O between 66% (in spring) and 38% (in autumn) with extreme values ranging from 5.5% to 71%; Speir et al. (1999) found in four nutrient-poor ecosystems extreme values ranging from 28% to 90%. Getting more precise estimates of N_2O is a major challenge for environmental sciences since it is a very potent atmospheric contaminant, both as a greenhouse gas and a destroyer of the ozone layer.

This paper focuses on the application of a generic approach for estimating nitrate fate factors in catchments (spatial fate factors) in a region of intensive agriculture. This approach is based on the analysis of different catchment scenarios using the INCA model (Whitehead et al., 1998; Wade et al., 2002a). In the framework of an LCA of pig production (Basset-Mens and van der Werf, 2005), the implications of these spatial fate factors for nitrate are investigated for the eutrophication impact category. Finally, for the climate change impact category, a sensitivity analysis to the fraction of nitrate transformed to N_2O is presented.

2. Materials and methods

2.1. General approach

The major part of the peninsula of Brittany is dedicated to intensive farming based on dairy production and indoor pig and poultry production. The region has an oceanic temperate climate, with annual precipitation ranging from 1300 mm in the western hills to 650 mm in the eastern low parts. The bedrock is mainly crystalline, comprising granite, gneiss, sandstones and shales. This geology produces a dense network of small rivers, defining relatively small catchments (Fig. 1). In detail, these catchments exhibit contrasting hydrological regimes, geomorphological structures and degrees of agriculture intensification, leading to varying proportions of riparian areas and to very variable nitrogen budgets, both in terms of agriculture input and of stream nitrate output.

The approach adopted here aimed at analysing this regional variability by creating combinations of the main factors believed to affect the nitrogen budgets of the catchments, namely, the time series of agriculture inputs, the climate, and the physical environment of the catchment, defined by its hydrological functioning and the extension of riparian wetlands. These catchment scenarios are artificial in the sense of the three types of factors being combined independently, but for each factor, actual observations were used. The N apparent fate factor, defined in practice, as the ratio of the annual stream nitrate flux over the agricultural leachable N for the same year was computed for long term series using the hydrological and nitrogen model INCA (Whitehead

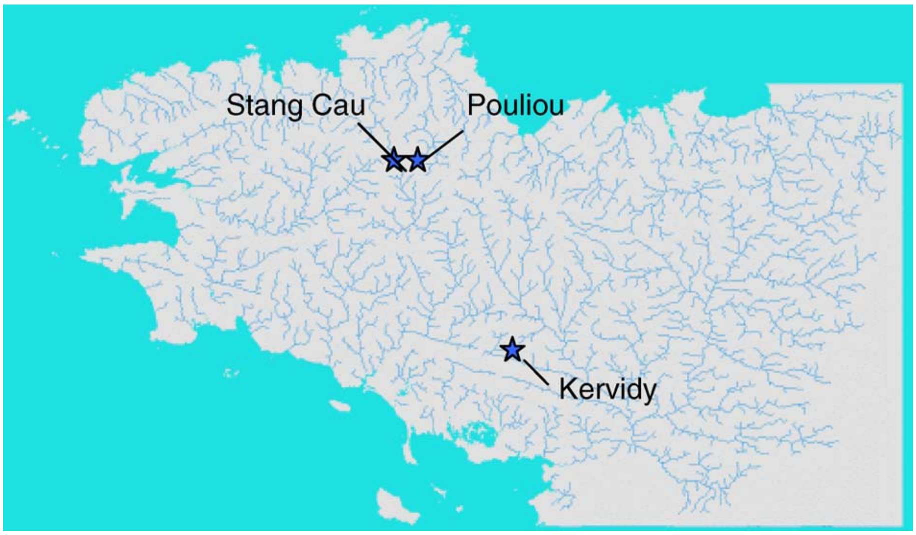

Fig. 1. Network of rivers, defining relatively small catchments in Brittany (France) and location of Stang Cau, Pouliou and Kervidy catchments.

et al., 1998; Wade et al., 2002a). The fate factors determined for the steady state case were used to propose a more precise estimation of the eutrophication result in the frame of an LCA of pig production (Basset-Mens and van der Werf, 2005). Finally, different hypotheses were made for estimating the part of nitrate lost as nitrous oxide (N_2O) in the atmosphere and the sensitivity of the climate change impact to these hypotheses was investigated.

2.2. The INCA model

INCA is a semi-distributed and process-based model simulating water transfer and nitrogen transport and fate through terrestrial systems into rivers. The main components of INCA as described by Whitehead et al. (1998) consist of:

- The hydrological model that calculates the flow of hydrologically effective rainfall in three compartments: the reactive (soil) and groundwater zones of the catchment and the river reach. Surface and subsurface pathways are mixed into the river in the proportions defined by the base flow index (BFI). The BFI represents the proportion of water being transferred to the lower groundwater zone. Each compartment is characterised by its residence time. This component of the model drives N fluxes through the catchment.
- The catchment nitrogen process model that simulates N transformations in the soil and groundwater of the catchment. This component includes plant uptake and microbial processes such as mineralisation, nitrification, denitrification.
- The river nitrogen process model that simulates dilution and in-stream N transformations and losses such as nitrification and denitrification. As shown by Durand (2004), it is possible to consider wetlands as part of this component.

Residence time, BFI and biochemical reaction parameters are defined by calibration. All the equations are of the first-order, but the combined result is non-linear, and the land-phase N-process equations have a dependency on temperature (using a sine function) and soil moisture (with a linear dependency).

2.3. Catchment scenarios

2.3.1. Definition of catchment types

The main features of a catchment assumed here to affect the nitrogen budgets are the proportion of riparian wetland areas and the hydrological regime. The latter can be defined by the proportion of base flow (e.g., base flow index), the difference between the low flow and high flow discharge, etc. (Watremez and Talbo, 1999; Mérot and Buffin, 1996). When available, the seasonal variations of nitrate concentration in the stream give also some hints on the dynamics of the main hydrological stores (Ruiz et al., 2002a).

Among the catchments monitored in Brittany for diffuse pollution studies, we identified three catchments presenting contrasting features: Stang Cau, Pouliou and Kervidy (Fig. 1). Although they are not representing the extremes of the variations observed in Brittany, these catchments exhibit a large range of riparian zone areas (from 5% to 25% of the catchment area), different patterns of flow dynamics and nitrate concentration variations and they have been monitored in detail for at least 5 years. Pouliou and Stang Cau, 0.64 and 0.86 km^2 in area, respectively, have a granitic bedrock overlaid by a sandy weathered material while Kervidy, 5 km^2 in area, has a bedrock made of brioverian shales with a silty–loamy weathered layer. Stang Cau presents the steepest slopes (10% to 25%) followed by Pouliou (2% to 15%) and by Kervidy (mainly less than 5%). The soil depth ranges from 0.4 to 1 m in Stang Cau and Pouliou (Rivière et al., 2002) and from 0.5 to 1.5 m in Kervidy (Durand et al., 2002). The proportion of riparian zones is lowest in Stang Cau (5%), largest in Pouliou (25%) and intermediate in Kervidy (14%). The climate of the three catchments is humid and temperate. The land use is essentially farming, relatively extensive in Stang Cau (sheep and dairy cows), very intensive in Kervidy (dairy cows, pigs and poultry) and intermediate in Pouliou.

The seasonal variations of water discharge are the same for all the catchments, with a minimum in late summer and a maximum in January or February (Fig. 2). The discharge in Kervidy exhibits the largest range, especially due to very low base flow in summer (the stream dries up almost every year). Pouliou and Stang Cau present similar low flow conditions, much higher than Kervidy in summer. This is a common difference between granitic and schistic catchments in the area. The difference between the two granitic catchments is that the quick flow is much more important in Pouliou than in Stang Cau: this is mainly due to the saturated contributive area being much larger in Stang Cau, as shown by the extension of riparian hydromorphic soils (Ruiz et al., 2002b).

The average nitrate concentrations range from 17.8 mg NO_3 l^{-1} for Stang Cau to 66.4 for Kervidy, Pouliou having intermediate concentrations (Fig. 3).

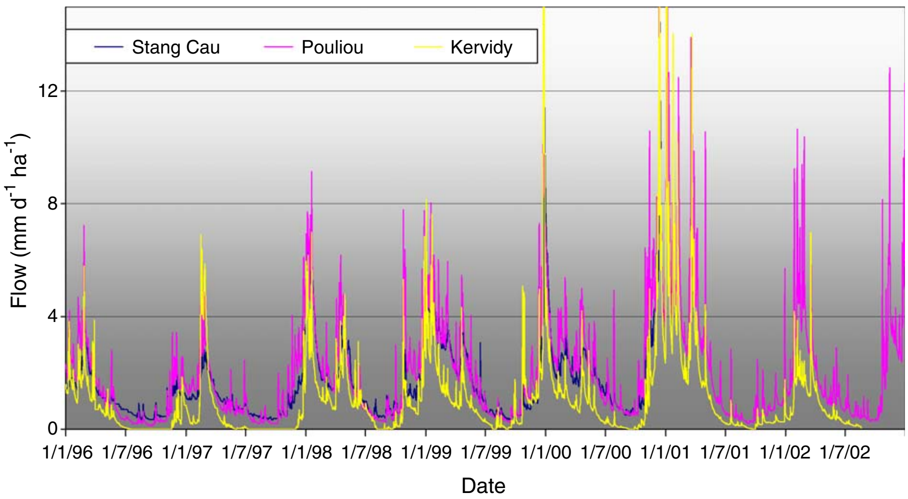

Fig. 2. Temporal variations of the water flow (mm d^{-1} ha^{-1}) for the catchments Stang Cau, Pouliou and Kervidy.

The seasonal variations are also quite different, Stang Cau and Kervidy showing cycles with a winter maximum and a summer minimum, whereas in Pouliou, the lowest concentrations are observed in winter and highest ones in summer. The variations are more marked for Kervidy, Stang Cau showing the most stable concentrations.

All these features are summarised by the calibrated values of the INCA parameters for the three catchments (Table 1). The base flow index and the residence times of the soil and groundwater stores were adjusted to get the best fit between the observed and simulated daily discharge. The initial concentrations and the capacity of the stores were then calibrated using the concentrations variations.

2.3.2. Climate

Three levels of annual hydrologically effective rainfall (resulting runoff of the rain falling on a catchment) were selected: 300 (Le Rheu, Eastern Brittany), 435 (Kervidy, Center Brittany) and 700 mm (Kerbernez, Western Brittany). These three levels span the rainfall gradient of the region and correspond to three distinctive agro-climatic subregions. In each case,

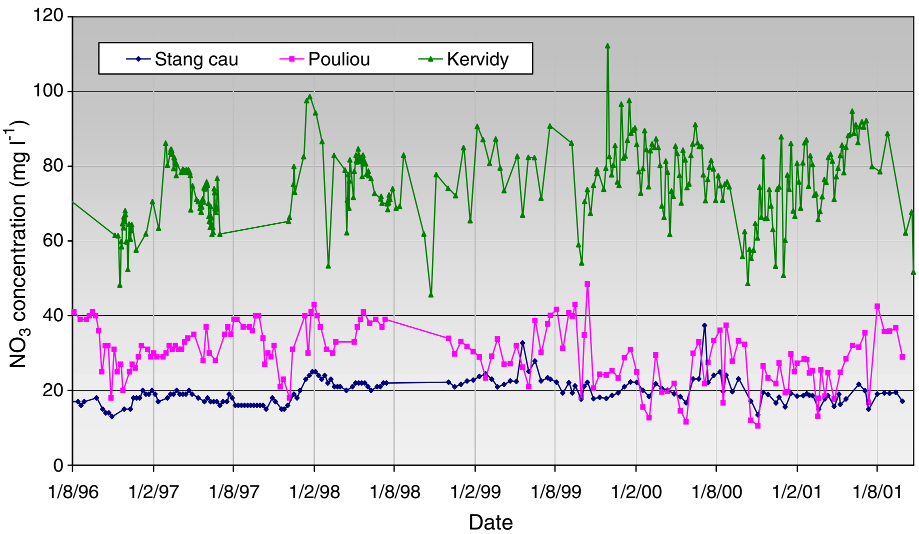

Fig. 3. Temporal variations of nitrate concentration (mg NO_3 l^{-1}) for the catchments Stang Cau, Pouliou and Kervidy.

Table 1
Main calibrated parameters and goodness of fit of the INCA model for the three catchments

	Kervidy	Pouliou	Stang Cau
Base flow index	0.8	0.5	0.7
Soil water store residence time (day)	1	1	3
Ground water store residence time (day)	20	60	50
Soil water store retention volume (m^3 m^{-2})	1	0.3	0.3
Soil water store drainage volume (m^3 m^{-2})	0.206	0.156	0.814
Ground water store drainage volume (m^3 m^{-2})	2.06	15.6	0.582
Mean wetland water volume (m^3 m^{-2})[a]	0.013	0.106	0.009
Initial stream NO_3 concentration (mg N l^{-1})	15	12	4
Goodness of fit (R^2) of simulated discharge	0.80	0.86	0.82
Goodness of fit (R^2) of simulated N flux	0.82	0.80	0.81

[a]The wetland water volume is calculated from the a and b parameters of the reach module using the formula: $V = L \cdot \frac{Q^{(1-b)}}{a}$ where Q is the mean daily discharge (m) and A the total area of the catchment (m^2) (see Durand, 2004).

a climatic record of eight years (from August to August) was used, which encompassed the major year to year variations observed in the past decades (Durand, 2004).

2.3.3. Historical trajectory

Three historical trajectories of the nitrogen input to the catchments were obtained by combining over time three contrasting agricultural situations, to represent the diversity of agricultural intensification in Brittany (Fig. 4). The first situation of agriculture in Brittany corresponded to extensive dairy production, mainly based on pastures (80% meadow/0% maize/20% cereal). The second situation corresponded to intensive dairy production based both on pastures and maize (40% meadow/40% maize/20% cereal). The third situation corresponded to mixed intensive animal (dairy, pig, chicken) and crop productions mainly based on maize and cereals (20%/40%/40%). The leachable nitrate from each land use in each agricultural situation was estimated according to Simon and Le Corre (1998) for maize, Simon et al. (1997) for meadows and expert advice and measurements for cereals. These three situations corresponded respectively to low, medium and high N input. It can be noted that the variability of the nitrate finally leached to the groundwater was simulated by the model when applying the eight climatic years to the leachable nitrate estimates, for each catchment scenario.

The first trajectory reached the most current intensive situation (as in Kervidy catchment), with an increase of leachable nitrate from 3.7 kg ha^{-1} of N–NO_3 (first agricultural situation), considered as the pre-intensification reference (1965), to more than 100 kg in the 1990 decade (third agricultural situation) followed by a slight decrease and then a stabilisation at 93 kg ha^{-1} of N–NO_3. The second trajectory was the same as the first one until the top of the curve but a more important decrease, up to 66 kg ha^{-1} (second agricultural situation), was assumed (induced for example by an extensification of the production per surface unit). Finally, the third trajectory reached a less intensified situation (first agricultural situation), with an increase from 3.7 up to 20 kg ha^{-1} of N–NO_3 and no decrease. In each case, the final load was maintained constant as long as necessary for the system to reach a steady state. For the low input scenario, the land use was kept constant during all the historical evolution (first agricultural situation, 80% meadow/0% maize/20% cereal). For the high input

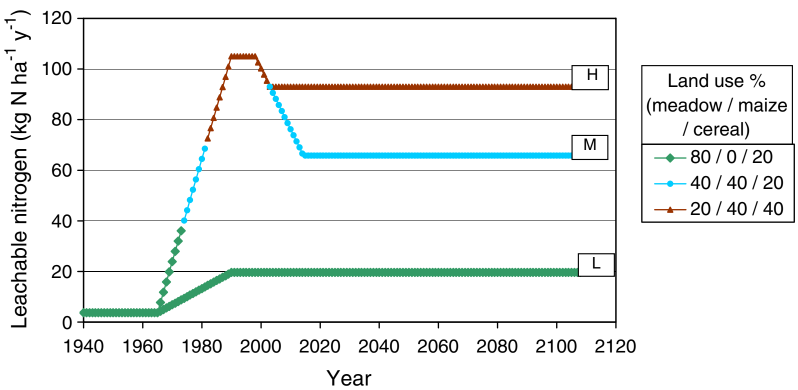

Fig. 4. Scenarios of the historical evolution of nitrogen inputs (as leachable nitrogen) in Brittany's catchments: the nitrogen inputs are stabilised at high (H), medium (M) and low (L) levels. The evolution of the land use as percentage of meadow/maize/cereal is given for each input scenario.

Table 2
Definition of contrasting catchment scenarios by combining catchment types, effective rainfall and historical nitrogen inputs in catchments

Catchment type	Low nitrogen inputs (L)			Medium nitrogen inputs (M)			High nitrogen inputs (H)		
	Rainfall			Rainfall			Rainfall		
	Low (1)	Middle (2)	High (3)	Low (1)	Middle (2)	High (3)	Low (1)	Middle (2)	High (3)
Stang Cau[a]	SL1	SL2	SL3	SM1	SM2	SM3	SH1	SH2	SH3
Pouliou[b]	PL1	PL2	PL3	PM1	PM2	PM3	PH1	PH2	PH3
Kervidy[c]	KL1	KL2	KL3	KM1	KM2	KM3	KH1	KH2	KH3

[a]Granitic bedrock, 5% wetland, nitrate concentrations maximum in winter; [b]granitic bedrock, 25% wetland, nitrate concentrations maximum in summer; [c]schistic bedrock, 14% wetland, nitrate concentrations maximum in winter.

scenario, the land use was assumed to change step by step from the less intensified (first agricultural situation, 80%/0%/20%) to the most intensified (third agricultural situation, 20%/40%/40%) and finally, for the medium input scenario, the same evolution was assumed as for the high input scenario except that the final extensification leads to an intermediate land use (second agricultural situation, 40%/40%/20%) (Fig. 4). The effect of the land use on actual evapotranspiration was taken into account according to the evolution over time of the land use defined in these three trajectories.

2.3.4. Combined scenarios

27 scenarios were obtained by combining the three catchment types, the three levels of effective rainfall and the three nitrogen input scenarios in catchments (Table 2).

2.4. Simulation over time of the nitrogen output and fate factor calculation

The first step was to calibrate the model against flow and chemistry data from the three selected catchments (Fig. 5). Since the input of nitrogen in this approach is defined as the leachable nitrogen, the land-phase nitrogen process model was by-passed (i.e., all the rate parameters were set to zero) and the leachable nitrate, as defined in the historical trajectories, was directly injected into the soil. This means that, for the land-phase part of the model, the hydrological processes only were considered (i.e., transfer and storage in the soil water and in the groundwater), because the soil transformation processes were already included in the calculation of the leachable nitrogen. The in-stream process model was used to estimate the denitrification process in the stream and the wetland zone.

Then the initial conditions (initial concentrations in soil water, ground water and stream water) were set to obtain a steady state of the catchments in equilibrium with the estimated nitrogen load in 1930 (3.7 kg ha^{-1}), and the historical load time series were applied as N input over time. The evolution of the nitrogen output in the river for each scenario was then simulated. The nitrate fate factor was defined as the ratio of annual fluxes of N export from the catchment in the river over annual fluxes of N input in the catchment, namely leachable nitrate. The nitrate fate factor was calculated over the entire simulation period for each catchment

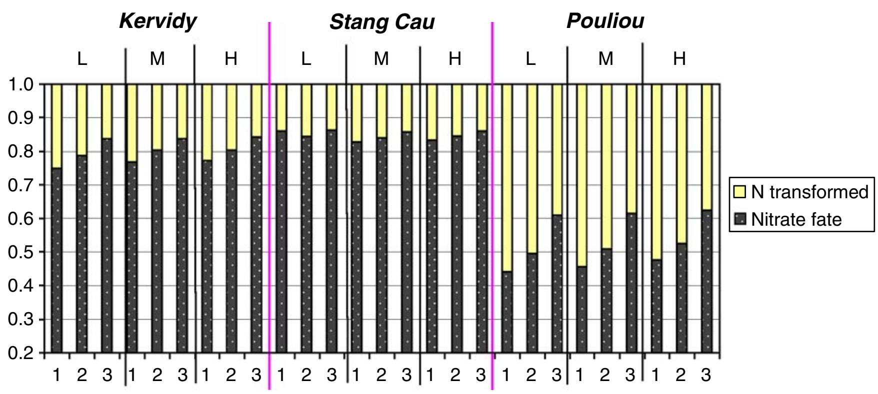

Fig. 5. Nitrate fate factors versus nitrate reduction after catchment stabilisation for 27 scenarios of catchment combining three hydrological catchment types (Kervidy, Stang Cau, Pouliou), three effective rainfall levels (1, 2 and 3 refer to low, medium and high effective rainfall, respectively) and three nitrogen input scenarios (L=low input, M=medium input, H=high input).

scenario. The fate factor used in the LCA was obtained after stabilisation at the final steady state (as a consequence of the stabilisation of the nitrogen load inputs—see Section 2.3.3.).

2.5. Consequences for LCA results

An LCA of pig production (Basset-Mens and van der Werf, 2005) was used to analyse the sensitivity of LCA results to the nitrate fate factors and to the indirect emission factors for N_2O. In this LCA study, an environmental inventory of all emissions and resources consumed at all stages of the life cycle of pig production had been performed and converted to seven impact categories. The impacts were expressed per kilogram of pig. Eutrophication potential (EP) was calculated using the generic EP factors in kilogram PO_4-eq. as recommended by Guinée et al. (2002): NH_3: 0.35, NO_3: 0.1, NO_2: 0.13, NO_x: 0.13, PO_4: 1. Global Warming Potential (GWP) was calculated, as recommended by Guinée et al. (2002), for a 100-year time horizon (GWP_{100}) according to the GWP_{100} factors by IPCC in kg CO_2-eq.: CO_2: 1, N_2O: 310, CH_4: 21.

In the general LCA model, the integration of the impacts over time is considered (Guinée et al., 2002). That is why we analysed the sensitivity of the eutrophication potential results to the steady state fate factors obtained in this study. The generic EP characterisation factor for nitrate (0.1) was multiplied by the range of values obtained for this steady state factor. Very few data exist concerning the fraction of nitrate lost as N_2O during denitrification processes in catchments, and it was concluded from the literature review that this fraction can be anywhere between 0 and 1. Consequently, to analyse the sensitivity of the global warming potential to the fraction of nitrate lost as N_2O, three hypotheses where tested, namely 30%, 50% and 70% of the denitrified nitrate lost as N_2O. The emission thus calculated replaces the general reference of Mosier et al. (1998) for indirect emission of N_2O after nitrate leaching (2.5% of nitrogen leached transformed to N_2O).

3. Results

3.1. Fate factors at steady state

The nitrate fate factors at steady state for the 27 catchment scenarios are shown in Fig. 5. For the Kervidy catchment type, the nitrate fate factor ranges from 0.75 to 0.84 with an average value of 0.80. The Stang Cau catchment type shows a higher average value of 0.85 with a lower variability, from 0.83 to 0.86. For the Pouliou catchment type, the fate factor is much lower, with an average value of 0.53 and a higher variability, from 0.44 to 0.62. The catchment type (essentially through the proportion of riparian zones) is the major parameter affecting the nitrate fate factor at steady state: the larger the riparian zone, the lower the fate factor. It can also be observed from these results that for a given catchment type, the fate factor value decreases when the effective rainfall decreases. High rainfall decreases the average residence time of water in the riparian zone and thus decreases the proportion of nitrate denitrified. This effect is more marked for the Pouliou catchment, with the highest proportion of riparian zone.

The nitrogen input scenario has a very minor effect on the fate factor value at the steady state. This is logical, since INCA uses first order equations to model the denitrification, i.e., the N denitrified and the nitrate concentration are proportional, and the fate factor is constant whatever the N concentration. As compared to the constant value of 1 most commonly used in LCA, these results suggest that the fate factor may vary from 0.44 to 0.86 depending on catchment type and amount of rainfall.

3.2. Evolution of the nitrate fate factor over time

When applying "historical" nitrogen load scenarios in the catchments, important variations of the nitrate fate factor over time are obtained between the initial and final steady states. The curves of the nitrate fate factor and the nitrogen load over the simulation period are compared in Fig. 6 for the scenario KM3. When the N input starts to increase in 1972 (A), the fate factor value falls to reach its minimum value (B). As the N input goes on increasing, the fate factor value increases progressively, and this increase continues for several years after the input has began to decrease. The highest value of the fate factor is then obtained (C), and when the N input stabilises, the fate factor gradually decreases to its initial value (D). This graph illustrates the importance of the build up and decrease of the N stock in the catchment. This N stock is represented in the model as the amount of N–NO_3 dissolved in the soil water and groundwater of the whole catchment. The building up of the N stock (i.e., through the increase of N concentrations in soil and ground water) acts actually as a sink at the catchment scale, resulting in a decrease of the fate factor. Conversely, the decrease of the N stock (following the decrease of the N input) acts as a nitrate source that increases the fate factor. As a result of these

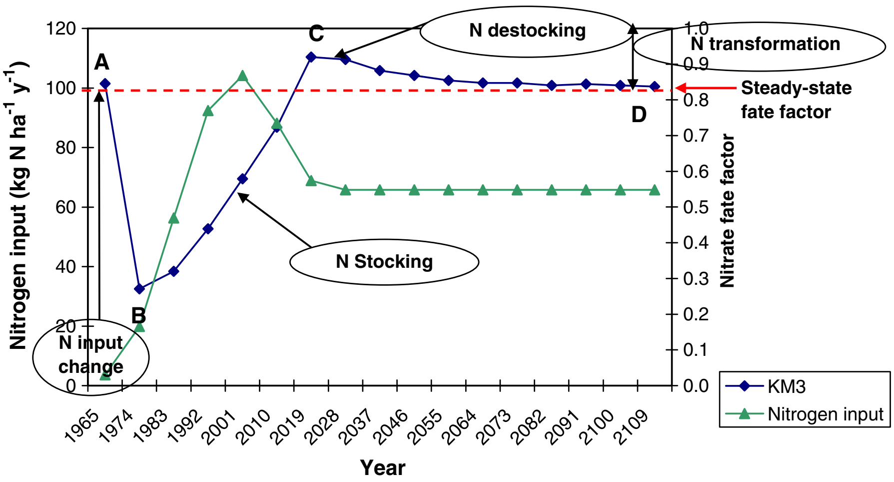

Fig. 6. Temporal variations of the nitrate fate factor and of the nitrogen input scenario (each point corresponds to a 9 year average) for the scenario KM3 combining the Kervidy catchment type, the medium input scenario and the third level of effective rainfall—illustration of the stocking, destocking and nitrogen transformation in riparian areas.

processes the annual fate factor calculated during unsteady conditions is variable and different from the fate factor sensu stricto, due to N transformation only. In other words, it shows that in the short term, the trends of the nitrate flux in the stream can be independent, and even opposite to the trends of the agricultural input due to the time-lags in the system.

Considering the 27 scenarios, and despite a similar shape, the curves of the fate factor over time present important differences, mainly between catchment types (Fig. 7a,b,c). Pouliou curves exhibit smoother variations in time and more marked sensitivity to rainfall than the curves for the other two catchments. Stang Cau curves show the quickest response to N load variations and reach the steady state earlier. Kervidy curves present the widest range of variation in time (difference between points B and C as defined in Fig. 6). Relative to the fate factor value at steady state (points A and D) the decrease in B is about 70% and the increase at point C is 10%, while for Stang Cau and Pouliou, the decrease at B is 40% and the increase at C is 10% and 0%, respectively. This means that the variations of the nitrogen stock are larger in Kervidy than in Stang Cau and Pouliou. The main reason is the higher base flow index in this catchment (0.8), the groundwater acting as the major nitrate stock. In P, the overall volume of groundwater is higher than in Kervidy, but the base flow index is 0.5. Another interesting feature suggested by these figures is the responsiveness of the catchments to the changes in N load, particularly when considering the evolution of the fate factor in the medium input scenario, after the drastic decrease of N load. In Pouliou, the C–D decreasing phase is not observed. Conversely, for Stang Cau and Kervidy, this rapid change in N load makes it possible to illustrate the contribution of the decrease of the N stock to the fate factor (i.e., point C higher than A and D), for the three rainfall scenarios in S (SM1, SM2, SM3) and for the wettest only in Kervidy (KM3). This phenomenon is not visible for the high input scenario, where the decrease of N load is not as marked as in the medium input scenario, and never visible in Pouliou. These results show that the capacity to dampen a change of the N load input is the highest for Pouliou followed by Kervidy and lowest for Stang Cau.

All these differences are primarily linked with the catchment type, i.e., the importance of hydrological stocks and the hydrological reactivity of the catchment. If the catchment type consists in important hydrological stocks and riparian zones (like Pouliou), the effective rainfall will have a major influence on the nitrate fate factor curve. Conversely, if the catchment is less dampened (like Stang Cau), the N load scenario will play a major role on the evolution of the fate factor value over time.

3.3. Implication for eutrophication and climate change LCA results

With the standard LCA methodology, the nitrate fate factor is assumed to be equal to 1 and the indirect emission factor from nitrate leaching for N_2O is assumed to be 2.5%, as proposed by Mosier et al. (1998). With these factors, the eutrophication and climate change impacts obtained for 1 kg of pig produced in a conventional system are respectively 0.0208 kg PO_4-eq. and 2.3 kg CO_2-eq. (Basset-Mens

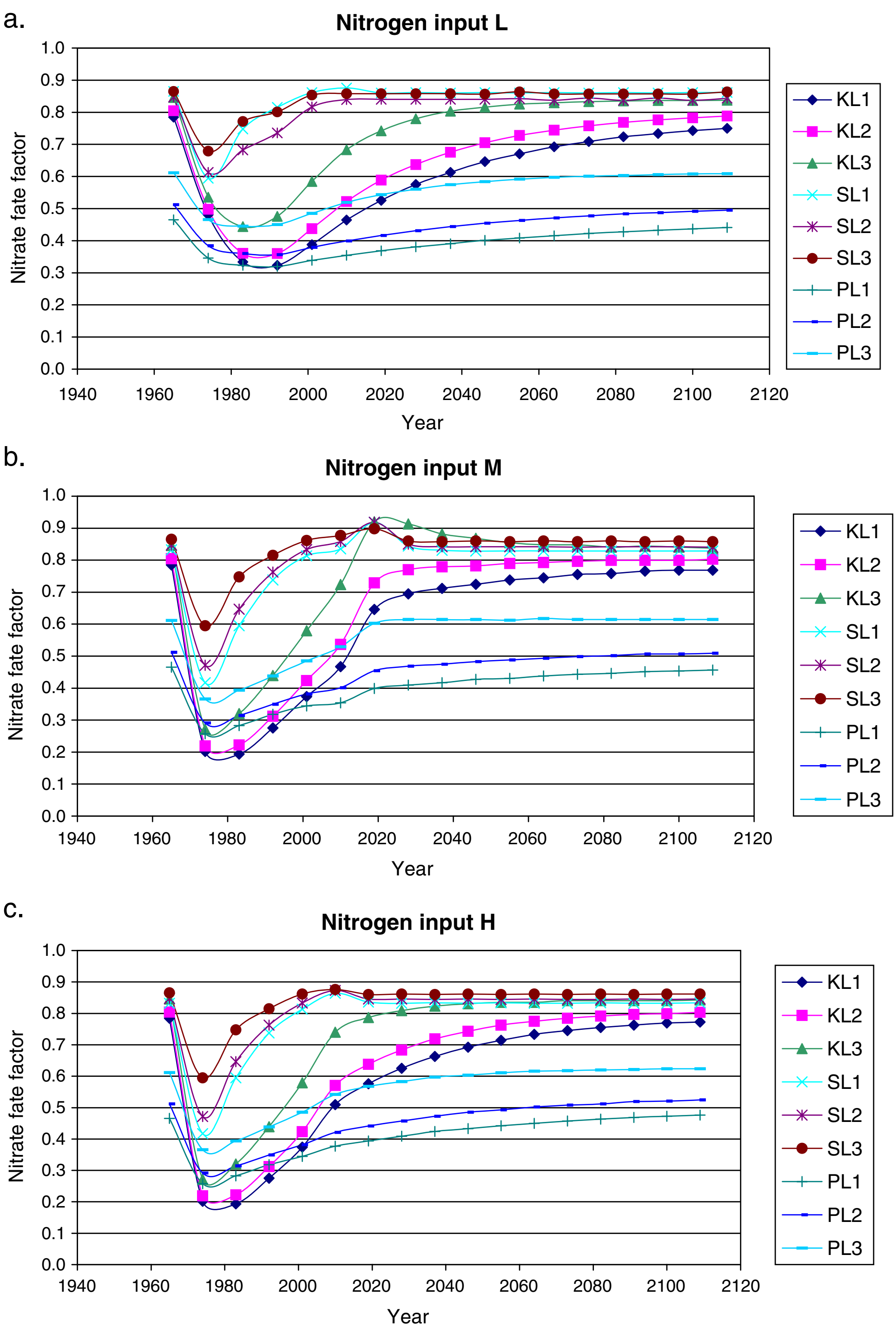

Fig. 7. Temporal variations of the nitrate fate factor for the nine scenarios of the low input scenario (L) (a), the medium input scenario (M) (b) and the high input scenario (H) (c), combining the three hydrological types (Kervidy, Stang Cau, Pouliou) and three levels of effective rainfall (1, 2 and 3 refer to low, medium and high effective rainfall, respectively).

and van der Werf, 2005). When applying the nitrate fate factors calculated in this study, the eutrophication result is reduced by 5% (nitrate fate factor of 0.9) to 32% (nitrate fate factor of 0.4) (Fig. 8). Conversely, the climate change impact remains constant (nitrate fate factor of 0.9 and fraction of nitrate lost as N_2O of 0.3) or

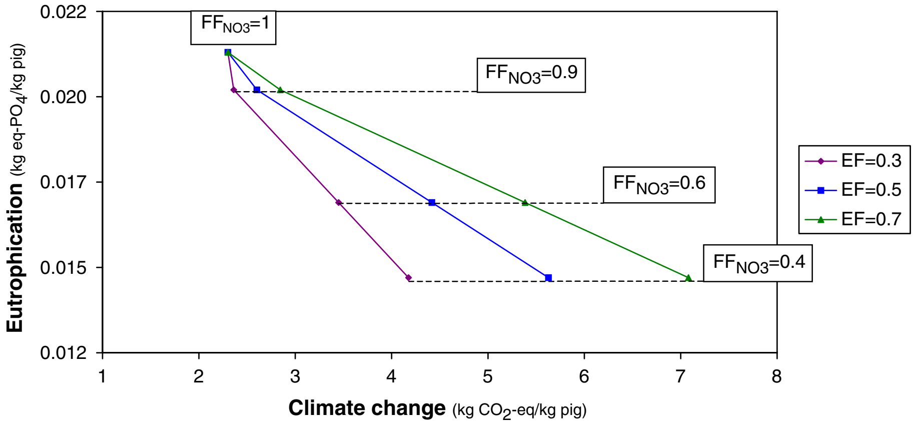

Fig. 8. Influence of the nitrate fate factor (FF_{NO3}) and different hypotheses of the fraction of nitrate lost as N_2O (EF) on LCA results (for 1 kg of pig) for eutrophication and climate change impact categories.

increases by more than 200% (for nitrate fate factor of 0.4 and fraction of nitrate lost as N_2O of 0.7). The scenario combining a nitrate fate factor of 0.9 and a fraction lost as N_2O of 0.3 is close to the hypothesis of Mosier et al. (1998) as it corresponds to an overall indirect emission factor of $(1-0.9)*0.3=3\%$ for N_2O after nitrate leaching whereas a fate factor of 0.4 and a fraction lost as N_2O of 0.7 corresponds to an indirect emission factor of 42%.

These results suggest that the outcomes of the LCA regarding the aquatic eutrophication impact can be significantly improved by taking into account the spatial variability of the nitrate fate factor. They also make clear that there is potentially a trade-off between eutrophication and climate change impacts due to the emission of N_2O during denitrification. In this framework, the necessity to work at the catchment scale on denitrification processes and on a reliable quantification of the nitrate lost as N_2O is paramount.

4. Discussion

4.1. Comparison with experimental observations

Given the continuous changes in agriculture during the past decades, no experimental data can validate the nitrate fate factors at steady state estimated in this study. However, for the Kervidy catchment, an annual input/output budget reveals a fate factor of 0.47 (70 kg N ha^{-1} y^{-1} in the river against 150 leachable at the soil level) (Bordenave and Merceron, 1999; Abrassart, 1999). This value, which is low compared to our average result of 0.80, can be explained by the catchment not being at steady state, the average streamwater concentration increasing continuously. The N stored annually in the Kervidy catchment is probably still important, given these results. Another way to validate the results is to estimate the denitrification rate of the riparian zones. Durand et al. (1998) have estimated for Kervidy the denitrification rate of the riparian zones at around 200 kg N ha^{-1} y^{-1}. Considering a riparian surface for this catchment of 14%, and a nitrogen input by leaching at soil level of 150 N ha^{-1} y^{-1}, the resulting fate factor for this catchment should be around 0.81. This figure is very similar to the value obtained around 0.80 for the scenario combining the Kervidy catchment and its real effective rainfall (intermediate).

For the other two catchments: Pouliou and Stang Cau, precise measurements of the real nitrogen leaching and of the nitrogen measured in the river have been performed for two years (Ruiz et al., 2002b). The ratio between the river nitrogen and the nitrogen leached was much higher for Stang Cau (98%) than for Pouliou (56%). These results should be taken with caution, given the short period of monitoring, but they roughly confirm the results of our simulation study.

Some studies suggest (not only in Brittany) a relationship between the relative surface of buffer zones and the nitrate fate factor at the catchment scale (Curmi et al., 1997; Gilliam et al., 1997; Ruiz et al., 2002b). By construction, the nitrate fate factor is strongly correlated to the riparian zone area in the model, which is coherent with these findings. It must be noted that other authors highlight the major effect of the shape and continuity of the riparian zones, rather than their area, on their overall efficiency (Gilliam et al., 1997; Pinay and Trémolières, 2000; Beaujouan et al., 2002; Clement et al., 2003). This could only be investigated with a distributed model, which is beyond the scope of this paper.

Aurousseau et al. (1996) found a strong relationship, at the regional scale, between the nitrogen input load and the fate factor, which is contradictory with the results on the fate factor at steady state presented here, which show a major effect of catchment type and climate and almost no influence of the nitrogen input. Two main reasons can explain this contradiction. First, the change in nitrogen input in the less intensified catchments having been less important and slower than in the more intensified ones, the former could be nearer the steady state than the latter, and therefore present a larger fate factor. Second, it is possible that the INCA model structure is not adapted, or too simple, to simulate adequately the relationship between riparian zone denitrification and nitrogen input. In particular, it is conceivable that the volume of soil and water where denitrification is active within the catchment (i.e., in reduced conditions and with sufficient active carbon and nitrate) increases when surface receiving fertiliser in excess increases, while in the model this volume is constant.

4.2. Broader considerations

This study highlights the difference between the observed fate factor at a given time, which aggregates both storage and transformation processes and a “steady state fate factor”, specific to the system considered. This steady state fate factor is theoretical, since human-impacted systems are not likely to reach a real equilibrium, as the rate of change of human activities is faster than the rate of change of the state variables of the system. However, this steady state factor is the one that must be considered to assess the environmental sustainability of the human activity considered. This implies that long term simulations with biophysical models are very useful to provide reliable inputs for environmental evaluation methods.

The results also show that the eutrophication result of an LCA can be sensitive to spatial fate factors for nitrate. On the basis of this first sensitivity analysis of LCA results to nitrate fate factors, some more practical aspects for implementing the approach proposed in this study should be discussed. The spatialisation of fate factors would require the spatialisation of emissions. However, this spatialisation of eutrophying emissions and fate factors would be rather laborious for assessing systems with multiple sites of eutrophying emissions. The spatialisation of nitrate fate factors would be important and feasible for systems with eutrophying emissions mainly located in a given area, ideally in a given catchment. In practice, this condition will most often be satisfied for farming systems. Developed at the regional scale, the proposed approach would allow assigning for each catchment type and its corresponding emissions, a nitrate fate factor. Finally, similar work for the fate of all the major substances responsible for eutrophication, namely phosphate in addition to nitrate, would be worthwhile to improve eutrophication result. The Phosphorus version of the INCA model (Wade et al., 2002b) could be used for this purpose.

5. Conclusions and perspectives

The improvement of the eutrophication impact assessment in a complete evaluation methodology such as LCA implies a better integration of the environmental mechanisms between emission and impact. In a context of intensive agriculture and serious coastal eutrophication, the fate of nitrate in the catchment represents a crucial stage of the cause and effect chain. To produce spatially differentiated fate factors for nitrate, the approach developed here is adaptable in any other similar context. This approach consists of:

- The identification of contrasted types of catchments in the region relative to their nitrate transfer capacity.
- The conception of contrasting scenarios of catchments crossing key-parameters for nitrate fate (such as : effective rainfall, nitrogen input, hydrological type).
- The simulation over a long period of the nitrogen input (nitrate leaching at soil level) and output (recovered in the river) taking into account the historical context (increase, decrease and stabilisation).
- At steady state, the calculation of the fate factor for each scenario.
- The synthesis of these data in a table for LCA practitioners, allocating to each scenario combining a catchment type and a rainfall level the corresponding fate factor.

The adaptation of the approach to other regions must take into account important features such as hydrological settings (bedrock conductivity and depth to water table, variability of base flow index, size of coastal catchments…), climatic gradient, major soil types, etc.

By using the INCA model, the relative contributions of both storage and transformation processes at the catchment scale were distinguished and quantified for different catchment scenarios. The range of fate factors obtained for the study region was wide, from 0.44 to 0.86, depending primarily on the hydrological catchment type and secondarily on the effective rainfall.

Although further work is needed to check that the results for the test catchments used here really span the whole range of fate factor variability in this region, it is already possible to draw useful conclusions on how this would affect LCA results. The sensitivity of the eutrophication result in an LCA to the spatial fate factors is significant, but the sensitivity of the climate change result is potentially much larger.

Concerning the improvement of the eutrophication impact assessment in complete evaluation methodologies, important and complementary research should be planned:

- a similar work for integrating phosphate fate,
- the integration of ecosystem sensitivity,
- the conception of a coherent and exhaustive procedure for eutrophication, consisting of the identification of the limiting factor in the study context, the integration of the fate of this substance particularly and of the ecosystem sensitivity.

This study highlights the potential for producing improved eutrophication results by using spatially differentiated fate factors. Additionally, it reveals the urgent need for quantitative studies of denitrification at the catchment scale, and particularly on the N_2O/N_2 ratio in riparian zones. This lack of knowledge hinders the improvement of the estimation of the climate change impact of agriculture, and casts a doubt on the overall environmental benefit of riparian buffer zones. The potential transfer of pollution between eutrophication and climate change constitutes a good illustration of the general problem of pollution transfer, and underlines once more the relevance of holistic approaches such as LCA.

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