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Modelling the contribution of short-range atmospheric and hydrological transfers to nitrogen fluxes, budgets and indirect emissions in rural landscapes

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Abstract. Spatial interactions within a landscape may lead to large inputs of reactive nitrogen (N_r) transferred from cultivated areas and farms to oligotrophic ecosystems and induce environmental threats such as acidification, nitric pollution or eutrophication of protected areas. The paper presents a new methodology to estimate N_r fluxes at the landscape scale by taking into account spatial interactions between landscape elements. This methodology includes estimates of indirect N_r emissions due to short-range atmospheric and hydrological transfers. We used the NitroScape model which integrates processes of N_r transformation and short-range transfer in a dynamic and spatially distributed way to simulate N_r fluxes and budgets at the landscape scale. Four configurations of NitroScape were implemented by taking into account or not the atmospheric, hydrological or both pathways of N_r transfer. We simulated N_r fluxes, especially direct and indirect N_r emissions, within a test landscape including pig farms, croplands and unmanaged ecosystems. Simulation results showed the ability of NitroScape to simulate patterns of N_r emissions and recapture for each landscape element and the whole landscape. NitroScape made it possible to quantify the contribution of both atmospheric and hydrological transfers to N_r fluxes, budgets and indirect N_r emissions. For instance, indirect N_2O emissions were estimated at around 21 % of the total N_2O emissions. They varied within the landscape according to land use, meteorological and soil conditions as well as topography. This first attempt proved that the NitroScape model is a useful tool to estimate the effect of spatial interactions on N_r fluxes and budgets as well as indirect N_r emissions within landscapes. Our approach needs to be

further tested by applying NitroScape to several spatial arrangements of agro-ecosystems within the landscape and to real and larger landscapes.

1 Introduction

Agricultural activities are a major source of emissions of reactive nitrogen (N_r). Two types of emission may be distinguished: (i) direct N_r emissions from areas where nitrogen is applied as mineral fertilizer or manure, (ii) indirect N_r emissions which may occur far from areas of nitrogen application and result from a cascade of transfers and transformations of N_r through the environment (Galloway et al., 2003). Indirect N_r emissions depend on the farming system and the characteristics of the area: variations in meteorological and soil conditions, topography, spatial distribution of N_r sources and sinks which are spatially heterogeneous, in intensity and nature, at a scale of several square kilometres (Beaujouan et al., 2001; Dragosits et al., 2002). Atmospheric NH_3 emitted from an animal house or a field can be deposited to the soil and foliage of nearby ecosystems (Fowler et al., 1998). Similarly, ecosystems at the bottom of slopes can recapture groundwater NO_3^- that originates in N_r applied upstream. Those transfers significantly modify the N_r budget of oligotrophic ecosystems and may lead to indirect N_2O and NO emissions (Skiba et al., 2006; Reay et al., 2009). Indirect N_2O emissions, consecutive to atmospheric deposition of ammonia (NH_3) and recapture of nitrates (NO_3^-), are estimated around 20 % of the total N_2O emissions in Europe

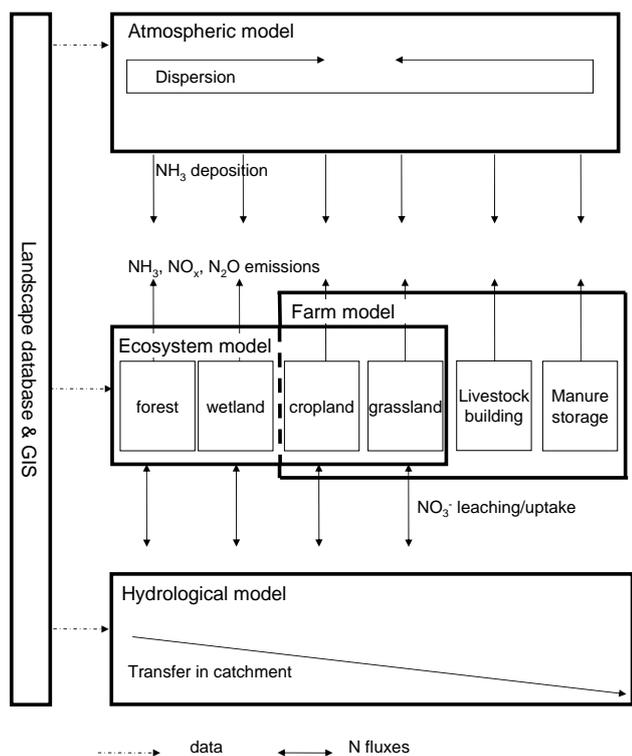


Fig. 1. The NitroScape model scheme.

(IPCC, 2006). A relevant scale to study the fate of N_r is therefore an area, namely the landscape, in which interactions occur between ecosystems and farm management, resulting from atmospheric and hydrological transfers which may be large at short-range, i.e. distances of several square kilometres to several tens of square kilometres. In rural areas, the landscape may include a river or stream catchment, several livestock buildings, agricultural fields and semi-natural ecosystems such as forests and wetlands (Cellier et al., 2011).

Several attempts were carried out to estimate indirect N_r, especially N₂O, emissions from measurements (e.g. Deurer et al., 2008; Reay et al., 2009) or from the IPCC methodology (IPCC, 2006) based on emission factors (e.g. Mosier et al., 1998; Nevison, 2000; Denier van der Gon and Bleeker, 2005). However, those estimates were highly uncertain and rarely account for both atmospheric and hydrological interactions, as well as farm management. Modelling is helpful to study complex dynamic systems such as landscapes, where spatial interactions occur and direct measurements of N_r fluxes are time and cost consuming due to the complexity of the system. Several models were developed to simulate N_r fluxes in rural landscapes. Most of them focused on aquatic ecosystems to describe N_r concentrations and fluxes within and at the outlet of a catchment which may correspond to the landscape scale described above (e.g. Beven, 1997; Whitehead et al., 1998; Beaujouan et al., 2002) or to larger, i.e. regional scales (e.g. Arnold et al., 1998; Billen

and Garnier, 2000). Recent studies attempted to assess the effect of anthropogenic activities on aquatic and terrestrial ecosystems, especially croplands, by coupling hydrological and crop models (e.g. Beaujouan et al., 2001; Ducharme et al., 2007). Other recent modelling studies tried to integrate all compartments of a rural landscape but focusing on one compartment only, the others being described with less detail. A few studies focused on anthropogenic transfers within the terrestrial (croplands, grasslands and farm compartments) and aquatic ecosystems (e.g. Hutchings et al., 2004). Other studies focused on atmospheric transfers between terrestrial ecosystems to assess emission, transfer and deposition of NH₃ at the landscape scale (Theobald et al., 2004; Kros et al., 2011) or indirect N₂O emissions at the regional scale (Denier van der Gon and Bleeker, 2005). However, none of those models dealt with both atmospheric and hydrological N_r transfers in a consistent way regarding temporal and spatial scales. The NitroScape model (Duret et al., 2011) was therefore developed to integrate processes of N_r transfer and transformation with temporal and spatial consistency between various compartments of a rural landscape: the atmosphere, several compartments of the terrestrial ecosystems (livestock buildings, croplands and grasslands) and the aquatic ecosystems (wetlands, streams and groundwater).

In this paper we describe a new approach to estimate direct and especially indirect N_r emissions in relation to spatial interactions by using the NitroScape model. We also estimate the relative contribution of indirect N_r emissions to the N_r fluxes and budgets within a test landscape and the relative contribution of both atmospheric and hydrological pathways to indirect N_r emissions. The test landscape includes livestock buildings, croplands (maize and wheat) and unmanaged ecosystems.

2 Materials and methods

2.1 The NitroScape model

The NitroScape model integrates in a spatially distributed and dynamic way four types of models representing processes of N_r transfer and transformation within the four corresponding compartments of a rural landscape: the atmosphere, the hydrological network, the agro-ecosystems and the farm buildings (Duret et al., 2011, Fig. 1). For each compartment of NitroScape, models were selected according to their ability to simulate N_r processes at the landscape scale and their consistency within the NitroScape model regarding temporal and spatial scales:

- the atmospheric model OPS (van Jaarsveld, 2004) describes processes of dispersion, transfer and deposition of N_r pollutants over a domain where surface characteristics may vary in space. It works at various spatial scales by combining long-range (Lagrangian) and short-range (Gaussian) modelling of pollutant transfer.

We used the grid-based version of OPS describing NH_3 processes only and working at a time step of 12 h (one day- and one night-time calculation per 24 h). OPS was validated for NH_3 concentrations simulated on a landscape of 3 km by 3 km (van Pul et al., 2008);

- the hydrological model TNT (Beaujouan et al., 2002) represents water and NO_3^- transfer in the hydrological network of a catchment. It accounts for runoff, ex-filtration, leaching, deep flows and uptake from deep soils (below 180 cm). It is mainly based on the assumptions of the hydrological model TOPMODEL (Beven, 1997). It is a distributed model that takes into account dual porosity (retention and drainage porosity). Computations are performed at a daily time step, following a mono-directional (a pixel flows into only one pixel) or multi-directional (one pixel can flow into several pixels) scheme. This scheme directly depends on the surface topography and is calculated from a digital elevation model at the beginning of the simulation;
- the agro-ecosystem model CERES-EGC (Gabrielle et al., 2006) is a process-based model which simulates water, carbon and nitrogen cycles in agro-ecosystems at a daily time step and at the field scale. It models vegetation growth and development, energy balance, evapotranspiration, heat and water transfer in soil above 180 cm. It accounts for mineral and organic N inputs from the farmer and simulates NO_3^- leaching and gaseous emissions of NH_3 , N_2O and NO . Water and NO_3^- fluxes are modelled by using a semi-empirical Darcy's law. Simulation of NH_3 emissions uses the approach of the process-based model from Générumont and Cellier (1997). CERES-EGC uses the semi-empirical model NOE (Hénault et al., 2005) to simulate N_2O emissions from both nitrification and denitrification processes. It also uses the module developed by Laville et al. (2005) to simulate NO emissions from nitrification processes. Total denitrification in soil is expressed as the product of a potential denitrification rate with three factors related to soil water content, NO_3^- content, and temperature. The fraction of denitrified NO_3^- that evolves as N_2O is considered as constant. Similarly, nitrification is expressed as the product of a potential nitrification rate with three factors related to soil water content, temperature and NH_4^+ as substrate of a Michaelis-Menten reaction. As for denitrification, a soil-specific proportion of total nitrification evolves as N_2O ;
- the farm model FASSET (Berntsen et al., 2003) simulates N_r species in a dynamic way and accounts for N_r transfer at the farm scale and exchanges with the outside of the landscape. It was adapted by (i) including production of animal manure either in the livestock housing or in the field and manure storage and (ii) removing the

agro-ecosystem component of FASSET. The updated version of FASSET, namely FASSET-farm, runs at a daily time step and deals with a range of livestock systems, livestock housing types and manure store types. NH_3 losses from manure in animal housing and manure storage are modelled according to Hutchings et al. (1996). N_2O emissions by farms (livestock housings and manure storage) are calculated using the IPCC methodology (IPCC, 2006).

Since all those processes occur simultaneously, the four models were integrated into a common modelling framework using the PALM dynamic coupler (Buis et al., 2006). They are called modules hereafter. An additional module, namely the linker, was developed and integrated into PALM to specify the exchange of data between the other four modules. It received and sorted fluxes and made it possible to calculate N_r budgets.

For simulating spatial interactions, a raster approach was used in NitroScape, in which the landscape was divided into pixels. TNT and OPS, which perform simulations on a grid, were directly integrated in this framework, using a one-to-one relationship between pixels. Individual runs of CERES-EGC and FASSET-farm were performed as many times as there were pixels occupied by an agro-ecosystem or a livestock building. Exchange of data between modules was performed at the pixel scale: each agro-ecosystem pixel received and sent data which were different from those of its neighbouring pixels.

The modules of NitroScape exchanged data in a dynamic way at a daily time step (i.e. the shortest time step common to all modules) during the simulation. Each module provided information to the other modules for the next daily time step.

2.2 The methodology to estimate indirect and direct emissions of N_r

The concept of indirect emissions was generally considered for N_2O emissions, less often for other N_r species (NH_3 , NO_3^- , NO). It represents the fraction of local emissions which result from N_r transfer by the atmospheric and hydrological pathways, and not from direct anthropogenic application of N_r .

The NitroScape model made it possible to account for and simulate the indirect emissions of several N_r species (NH_3 , NO_3^- , N_2O , NO) and the interactions between them. The relative contribution of short-range transfers of N_r on N_r fluxes and budgets was estimated by implementing four configurations of NitroScape in which atmospheric or hydrological or both pathways of N_r transfer were accounted for or not. The indirect emissions were estimated as the difference between emissions simulated by accounting for all types of N_r transfer and emissions simulated by cutting one or two pathways of transfer:

- the “all transfers” (*all*) configuration corresponded to the reference configuration in which (i) NH_3 was emitted, laterally transferred through the atmospheric pathway and deposited in areas which could be far from areas of N_r application, and (ii) NO_3^- was lost by leaching in interaction with the groundwater, laterally transferred through the hydrological pathway (runoff and lateral transfer in the saturated zone) and recaptured in areas which could be far from areas of N_r application;
- the “atmospheric transfers” (*atm*) configuration corresponded to the case in which only short-range lateral transfers by the atmospheric pathway were calculated. This was implemented by (i) sending null wet deposition and null NO_3^- concentration to the hydrological module to prevent leaching and lateral transfers of NO_3^- and (ii) cutting recapture of groundwater NO_3^- by croplands and unmanaged ecosystems. NO_3^- recapture resulted from two types of interaction between the soil unsaturated zone and the groundwater: (i) capillary rise and (ii) groundwater uprising when the water table rose and brought groundwater and dissolved NO_3^- into the soil unsaturated zone. Daily NO_3^- leaching was stored to be taken into account in the final budget of each pixel;
- the “hydrological transfers” (*hydro*) configuration corresponded to the case in which only short-range lateral transfers by the hydrological pathway were calculated. This was implemented by sending null NH_3 emissions from agro-ecosystems and farm buildings to the atmospheric module, which prevented lateral transfers and deposition of NH_3 . Daily NH_3 emissions were stored to be included in the final budget of each pixel;
- the “no short-range transfer” (*not*) configuration corresponded to the case in which lateral transfers by both atmospheric and hydrological pathways were cut. This was implemented by (i) sending null NH_3 emissions from agro-ecosystems and farm buildings to the atmospheric module to prevent emissions, lateral transfers and deposition of NH_3 , (ii) sending null wet deposition and null NO_3^- concentration to the hydrological module to prevent leaching, lateral transfers and recapture of groundwater NO_3^- by croplands and unmanaged ecosystems. Daily NH_3 emissions and daily NO_3^- leaching were stored to be taken into account in the final budget of each pixel.

Total indirect N_r (N_r being NH_3 or NO_3^- or NO or N_2O) emissions in the *all* configuration were calculated as:

$$\text{N}_{r,\text{ind,all}} = \text{N}_{r,\text{tot,all}} - \text{N}_{r,\text{tot,not}} \quad (1)$$

where $\text{N}_{r,\text{tot,all}}$ are the total N_r emissions in the *all* configuration and $\text{N}_{r,\text{tot,not}}$ are the total N_r emissions in the *not* configuration.

The indirect N_r emissions due to atmospheric transfers were calculated as:

$$\text{N}_{r,\text{ind,atm}} = \text{N}_{r,\text{tot,atm}} - \text{N}_{r,\text{tot,not}} \quad (2)$$

where $\text{N}_{r,\text{tot,atm}}$ are the total N_r emissions in the *atm* configuration.

The indirect N_r emissions due to hydrological transfers were calculated as:

$$\text{N}_{r,\text{ind,hydro}} = \text{N}_{r,\text{tot,hydro}} - \text{N}_{r,\text{tot,not}} \quad (3)$$

where $\text{N}_{r,\text{tot,hydro}}$ are the total N_r emissions in the *hydro* configuration.

Direct N_r emissions corresponded to N_r emissions in the *not* configuration.

In contrast with most previous approaches on indirect N_r emissions, which could only account for a landscape or a catchment as a whole, this approach made it possible to give an estimate of indirect emissions for each pixel and thus each agro-ecosystem type. However, for a given pixel, indirect N_r emissions were related to N_r applied in other agro-ecosystem pixels and not, as generally made, to N_r applied in this pixel.

The model outputs were also used to estimate the indirect emission factors as defined by the IPCC guidebook (IPCC, 2006). Indirect emission factors were calculated for the whole landscape and for croplands and unmanaged ecosystems by accounting for pixels related to each type of agro-ecosystem. They were calculated from the following equations derived from Mosier et al. (1998):

$$\text{EF4} = \text{N}_2\text{O}_{\text{ind,atm}} / \text{capt}_{\text{NH}_3} \quad (4)$$

$$\text{EF5g} = \text{N}_2\text{O}_{\text{ind,hydro}} / \text{capt}_{\text{NO}_3} \quad (5)$$

$$\text{EF} = \text{N}_2\text{O}_{\text{ind,all}} / \text{capt}_{\text{N}} \quad (6)$$

where EF4 is the emission factor due to atmospheric NH_3 deposition, EF5g is the emission factor due to hydrological NO_3^- recapture, EF is the emission factor due to both atmospheric deposition and hydrological recapture, $\text{capt}_{\text{NH}_3}$ is the total NH_3 deposition resulting from atmospheric transfers, $\text{capt}_{\text{NO}_3}$ is the total NO_3^- recapture resulting from hydrological transfers and capt_{N} is the total N_r recapture corresponding to atmospheric deposition and hydrological recapture.

2.3 The test landscape

NitroScape was applied to a simplified landscape with a size of $1.75 \times 1.75 \text{ km}^2$ represented by a matrix of 70×70 pixels of $25 \times 25 \text{ m}^2$ each. That corresponded to the minimal landscape size at which hydrological transfers occur and to the number and size of pixels needed to simulate atmospheric

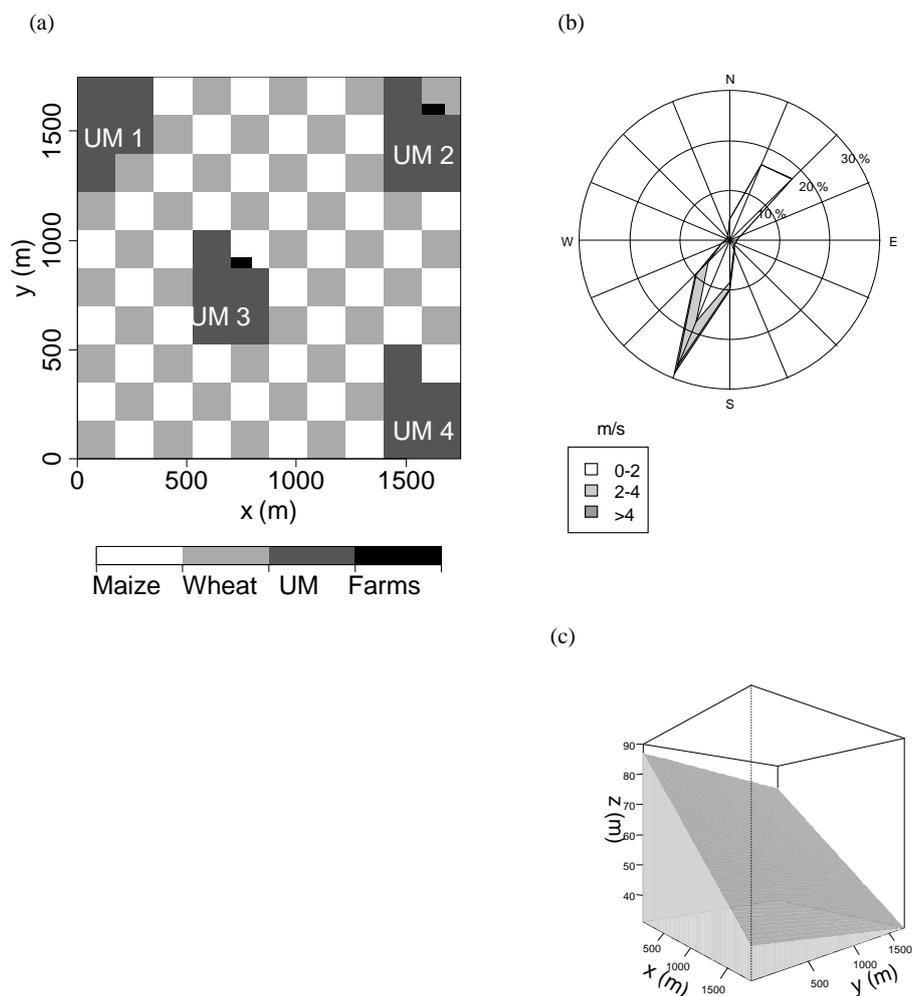


Fig. 2. (a) Land use in the test landscape (wheat and maize fields all received $240 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in average, UM: unmanaged ecosystem), (b) wind direction and speed and (c) topography of the landscape. x- and y-axes in (c) correspond to the ones in (a).

deposition. The agricultural activity on this landscape corresponded to farm management in intensive rural areas with mixed crops and pig farming (Fig. 2a). From a topographical point of view, the landscape was characterized by a linear slope with a gradient of 50 m between the highest and the lowest parts of the landscape (Fig. 2c). Meteorological data used for the simulation were taken from a meteorological station located on the Kervidy-Naizin catchment ($48^{\circ}01' \text{ N}$, $2^{\circ}83' \text{ O}$, Brittany, France). This catchment was characterized by humid climatic conditions (total rainfall: 1968 mm, average relative humidity: 90 %) and a relatively small range of temperature (average temperature: 10° C , standard deviation: 5° C). The prevailing winds were from the north-east and the south-west, with an average wind speed of about 1.8 m s^{-1} (Fig. 2b). The soil type was a uniform silty loamy soil. Farms were mixed crop-pig farms characterized by indoor pigs (200 sows, 2000 piglets and 2000 baconers). Pig feed was mainly based on imported feed such as wheat, soybean, barley, fish-meal and fat. Baconers were also fed with barley, pea, rye and

rapeseed. Croplands cultivated on the farm were wheat and maize (Fig. 2a). Wheat received three applications of mineral fertilizer in February (60 kg N ha^{-1}), March (60 kg N ha^{-1}) and April (120 kg N ha^{-1}). Maize received one manure application in March (120 kg N ha^{-1}) and two mineral fertilizer applications in April (60 kg N ha^{-1}). The unmanaged ecosystems received no fertilizer or manure, but only N_r from atmospheric transfer and deposition of NH_3 or from hydrological transfer and recapture of NO_3^- .

Within that landscape 39 fields of 49 pixels each and one field of 41 pixels were dedicated to maize, 39 fields of 49 pixels each and one field of 41 pixels were dedicated to wheat, four fields of 245 pixels each were dedicated to unmanaged ecosystems and 16 pixels were dedicated to pig buildings (Fig. 2a). One of the four unmanaged ecosystems (UM 1) was located in the north-west, i.e. the highest part of the landscape; another unmanaged ecosystem was located in the centre of the landscape (UM 3), close to one of the livestock building. The other two unmanaged ecosystems were located

Table 1. Average N_r fluxes within the whole landscape. N_r inputs were $191 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in average within the whole landscape.

Configuration	NH_3 emissions ($\text{kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$)	NH_3 dry deposition ($\text{kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$)	NO_3^- leaching ($\text{kg NO}_3^-\text{-N ha}^{-1} \text{ yr}^{-1}$)	NO_3^- inputs ($\text{kg NO}_3\text{-N ha}^{-1} \text{ yr}^{-1}$)	N_2O emissions ($\text{kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$)	NO emissions ($\text{kg NO-N ha}^{-1} \text{ yr}^{-1}$)
<i>all</i>	39.2	9.0	65.6	7.5	5.6	0.9
<i>not</i>	38.7	0.0	45.3	0.0	4.4	0.9
<i>atm</i>	39.4	9.1	61.5	0.0	5.1	0.9
<i>hydro</i>	38.7	0.0	61.5	8.3	5.5	0.9

in the lowest part of the landscape: one in the north-east close to the other livestock building (UM 2) and the other in the south-east of the landscape (UM 4) far from the livestock buildings (Fig. 2a).

NitroScape simulations integrated a whole year from 1 January to 31 December.

3 Results – discussion

NitroScape made it possible to simulate N_r fluxes and budgets at the whole landscape scale, their spatial distribution within the landscape as well as the interactions between the different types of N_r transfer, especially by the atmospheric and hydrological pathways. This first attempt was made on a theoretical landscape which was constructed in such a way that the spatial arrangement of croplands and unmanaged ecosystems made it possible to simulate a range of N_r transfers from croplands and livestock housings to unmanaged ecosystems. Only indirect N_r emissions occur in unmanaged ecosystems, induced by both atmospheric and hydrological pathways (see Duret et al., 2011, for more details). This would not have been possible by using a real landscape characterized by such a diversity of agro-ecosystems within a small area of around 3 km^2 . Further work is needed to evaluate the ability of the NitroScape model to simulate N_r fluxes and budgets, including short-range transfers and indirect N_r emissions, within real and larger landscapes.

3.1 Total N_r fluxes (N_r being NH_3 or NO_3^- or NO or N_2O) at the landscape scale

Total (i.e. direct and indirect) NH_3 emissions, NO_3^- losses, NO and N_2O emissions were calculated from the *all* configuration. Their comparison with fluxes calculated from the other three configurations (i.e. *not*, *atm* and *hydro*) provided a first overview of the relative weight of direct and indirect N_r emissions at the whole landscape scale.

Total NH_3 emissions by the whole landscape, including farms (livestock housings and manure storage), croplands and unmanaged ecosystems, were around $39 \text{ kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ in average in the four configurations (Table 1). Total NH_3 dry deposition was around $9 \text{ kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ in the *all* and *atm* configurations, while it was around zero in

the *not* and *hydro* configurations. These differences between the *all* and *atm* configurations in the one hand and the *not* and *hydro* configurations in the other hand show an effect of NH_3 transfer and deposition by the atmospheric pathway on NH_3 emissions by agro-ecosystems at the landscape scale. Total NO_3^- losses to the groundwater from both leaching and dilution varied at the landscape scale between $45.3 \text{ kg NO}_3^-\text{-N ha}^{-1} \text{ yr}^{-1}$ in the *not* configuration and $65.6 \text{ kg NO}_3^-\text{-N ha}^{-1} \text{ yr}^{-1}$ in the *all* configurations in average, with values around $61.5 \text{ kg NO}_3^-\text{-N ha}^{-1} \text{ yr}^{-1}$ in the *hydro* and *atm* configurations (Table 1). That result indicates that both hydrological and atmospheric transfers of N_r play a role in NO_3^- losses at the landscape scale.

Total NO emissions by the whole landscape were around $0.9 \text{ kg NO-N ha}^{-1} \text{ yr}^{-1}$ in average within the whole landscape in the four configurations (Table 1). There were no indirect NO emissions due to atmospheric or hydrological or both transfers. Since the atmospheric model OPS did not account for NO transfer, discrepancies in NO emissions between the four configurations, if they had occurred, might have only resulted from processes of nitrification and denitrification in emitting areas. Total N_2O emissions by the whole landscape were estimated around $5 \text{ kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$ in average in the four configurations (Table 1). They were 5.6, 4.4, 5.1 and $5.5 \text{ kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$ in the *all*, *not*, *atm* and *hydro* configurations, respectively. That result indicates that, as for NO_3^- losses, both hydrological and atmospheric transfers play a role in N_2O emissions at the landscape scale. The soil was a silty loamy soil, but its texture had no direct effect on processes of NO and N_2O production in CERES-EGC, and consequently in NitroScape, since those processes in CERES-EGC only depended on soil NO_3^- or NH_4^+ content, soil water content and temperature, and constants.

3.2 Direct NH_3 emissions, NO_3^- losses and N_2O emissions within the landscape

Direct NH_3 emissions, NO_3^- losses and N_2O emissions were calculated from the *not* configuration.

Direct NH_3 emissions by croplands and unmanaged ecosystems were estimated at around $38.7 \text{ kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ in average, but they ranged from 0 to $121 \text{ kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ (Fig. 3a). Thus, direct NH_3 emissions

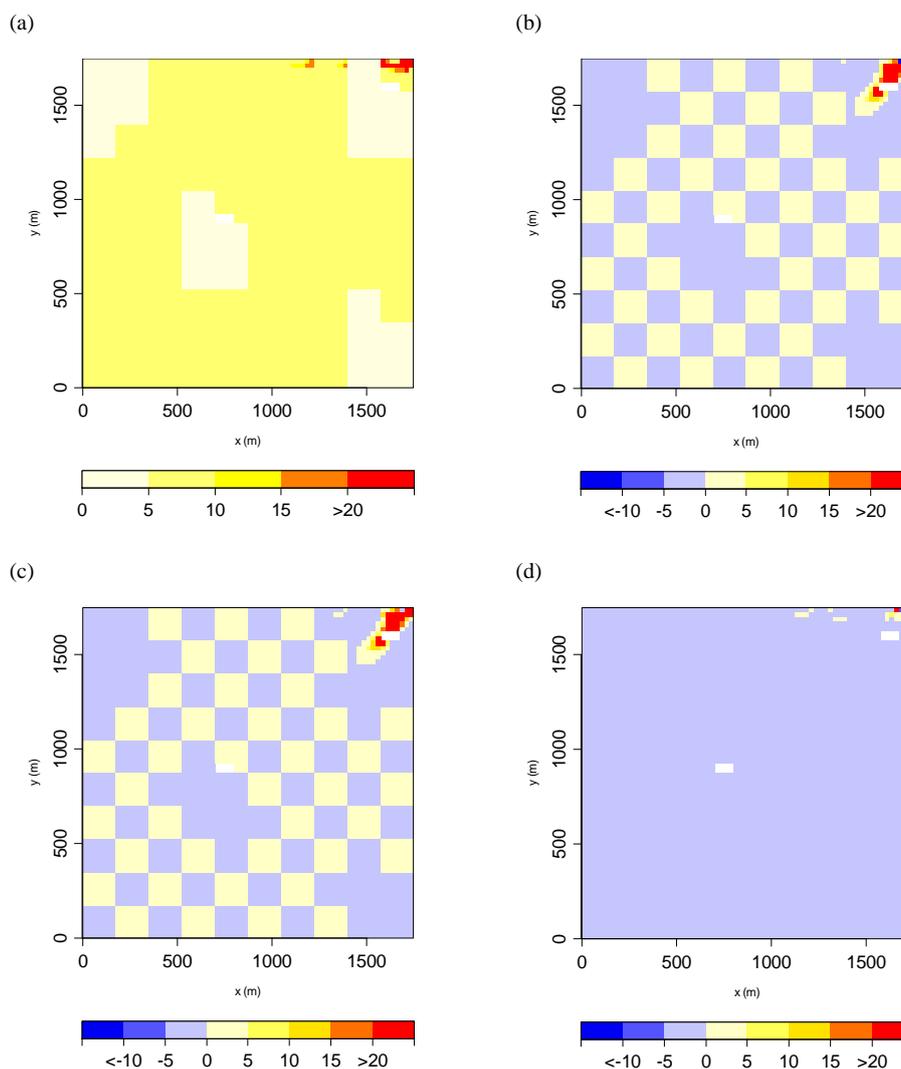


Fig. 3. (a) Direct NH_3 emissions by agro-ecosystems in the *not* configuration ($\text{kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$). Indirect NH_3 emissions in the (b) *all*, (c) *atm* and (d) *hydro* configurations ($\text{kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$). Negative values in (b), (c) and (d) mean that NH_3 emissions are lower in each of the *all*, *atm* and *hydro* configurations than in the *not* configuration.

represented 20 % of the N_r inputs in average at the landscape scale. This value was close to the maximal value of 19 % derived from EEA/EMEP Guidebook (2009) giving a maximal NH_3 emission factor of 68 % for pig slurry and 1 % for mineral fertilizer. That value is higher than the value of 9 % derived from the results presented by Leip et al. (2011) for EU27 and taking into account mineral fertilizers and manure for N_r inputs. NH_3 emissions varied within the landscape according to the N_r input patterns. The highest NH_3 emissions were simulated for croplands located in the north-east of the landscape which was the lowest part of the landscape and where soil was highly saturated. Nitrification was therefore limited, leading to high NH_4^+ content (Hénault et al., 2005) and therefore high NH_3 emissions (Génermont and Cellier, 1997). The lowest NH_3 emissions were simulated for the un-

managed ecosystems located in the north-west of the landscape where soil was not saturated.

Direct NO_3^- losses to the groundwater by leaching were estimated at around $45.3 \text{ kg NO}_3^-\text{-N ha}^{-1} \text{yr}^{-1}$, but they ranged from 0 to $150 \text{ kg NO}_3^-\text{-N ha}^{-1} \text{yr}^{-1}$ (Fig. 4a). Direct NO_3^- losses corresponded to 23 % of the N_r inputs in average at the landscape scale. The highest leaching values were simulated for the wheat fields located in the west and the centre of the landscape, while the lowest ones were simulated for the unmanaged ecosystems and the croplands located in the east of the landscape where soil was saturated. The leaching rates mainly varied according to the land use and the N_r inputs.

Direct N_2O emissions by croplands and unmanaged ecosystems were around $4.4 \text{ kg N}_2\text{O-N ha}^{-1} \text{yr}^{-1}$ in average,

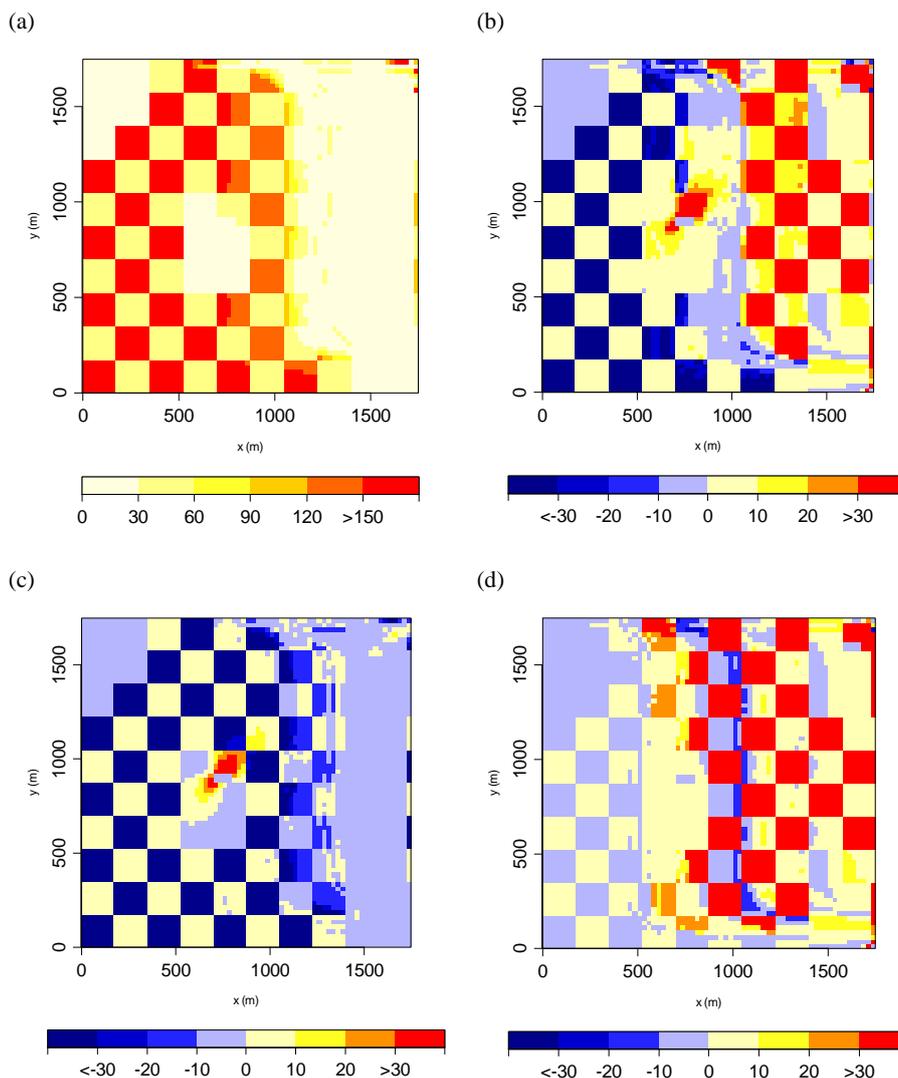


Fig. 4. (a) Direct NO_3^- losses to the groundwater in the *not* configuration and indirect NO_3^- losses in the (b) *all*, (c) *atm* and (d) *hydro* configurations ($\text{kg NO}_3^- \text{-N ha}^{-1} \text{yr}^{-1}$). Negative values in (b), (c) and (d) mean that NO_3^- losses are lower in each of the *all*, *atm* and *hydro* configurations than in the *not* configuration.

ranging from 0 to more than $60 \text{ kg N}_2\text{O-N ha}^{-1} \text{yr}^{-1}$ (Fig. 5a). The value of the average direct N_2O emission factor was then estimated at 2.3 % of the N_r inputs. This value was higher than the expected value of 1 % given by the IPCC methodology (IPCC, 2006) and the value of 0.6 % reported by Reay et al. (2009). The highest direct N_2O emissions were simulated for pixels located in the north and the east of the landscape. These areas were the lowest parts of the landscape where soil was highly saturated, leading to high denitrification rates (Hénault et al., 2005). Direct N_2O emissions also varied according to the land use and the N_r inputs, with the highest direct N_2O emissions simulated for the wheat fields receiving more N_r inputs. The lowest direct N_2O emissions were simulated for the unmanaged ecosystems receiving no direct N_r inputs.

3.3 NH_3 deposition and NO_3^- recapture within the landscape

NH_3 dry deposition on croplands and unmanaged ecosystems was around $9 \text{ kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$ in average in the *all* and *atm* (Table 1) configurations, but they ranged from 0.2 to $360 \text{ kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$ (Fig. 6a and b). The highest NH_3 deposition rates were simulated close to the farm buildings. The lowest NH_3 deposition rates were simulated in the north-west and the south-east of the landscape for pixels not located in the lee of the farm buildings. NH_3 deposition was theoretically zero in the *not* and *hydro* configurations. There was no interaction between the atmospheric and hydrological pathways for NH_3 deposition. The simulated NH_3 deposition rates ranged within the same values as those observed

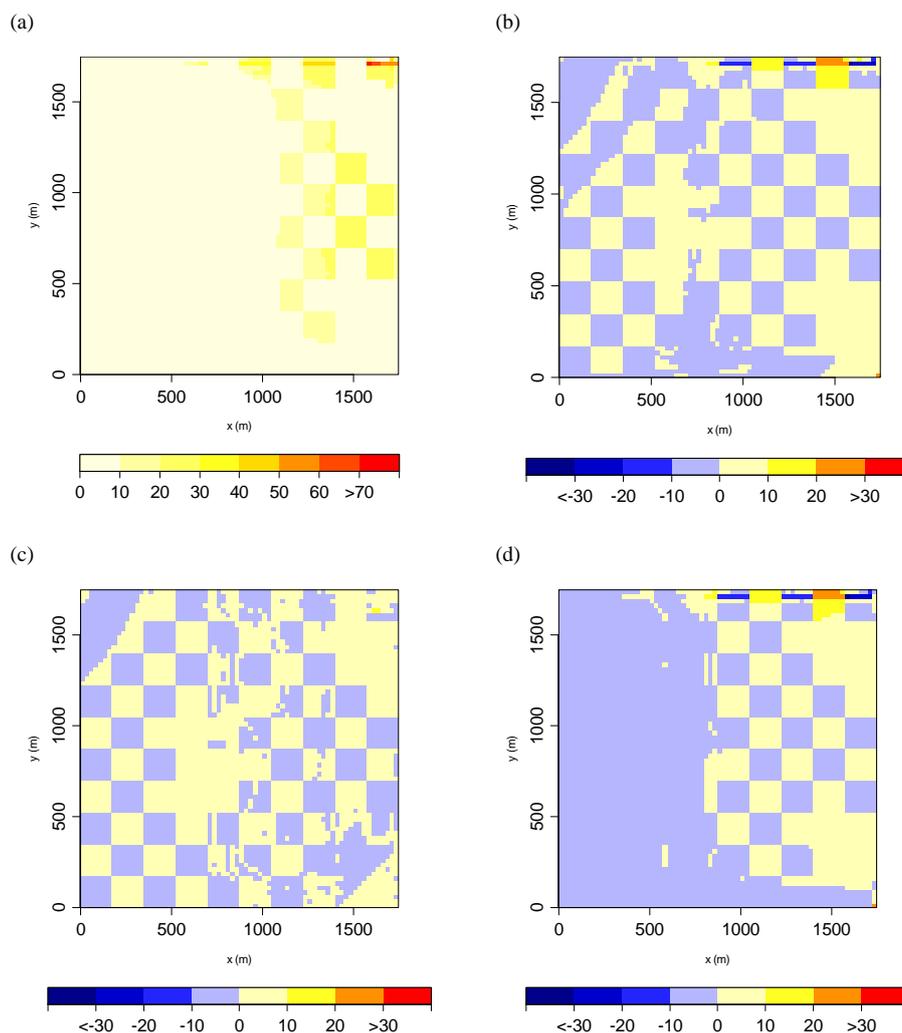


Fig. 5. (a) Direct N_2O emissions in the *not* configuration and indirect N_2O emissions in the (b) *all*, (c) *atm* and (d) *hydro* configurations ($\text{kg N}_2\text{O-N ha}^{-1} \text{yr}^{-1}$). Negative values in (b), (c) and (d) mean that N_2O emissions are lower in each of the *all*, *atm* and *hydro* configurations than in the *not* configuration.

by Fowler et al. (1998), with high deposition rates close to the downwind of the farm buildings (Loubet et al., 2009). The lowest NH_3 deposition rates were found in the north-west and the south-east of the landscape for pixels located far from the farm buildings and receiving low N_r inputs from them due to wind direction distribution.

NO_3^- inputs, including direct inputs and recapture, resulting from capillary rise and groundwater uprising was around 7.5 (resp. 8.3) $\text{kg NO}_3^- \text{-N ha}^{-1} \text{yr}^{-1}$ in average in the *all* (resp. *hydro*) configuration, ranging from 0 to 130 $\text{kg NO}_3^- \text{-N ha}^{-1} \text{yr}^{-1}$ (Fig. 7a resp. 7b). NO_3^- recapture was theoretically zero in the *not* and *atm* configurations. However, there was interaction between the atmospheric and hydrological pathways for NO_3^- recapture: NO_3^- inputs were higher when accounting for hydrological transfers only than accounting for both hydrological and atmospheric transfers. A hypothesis to explain that discrepancy of 0.8 $\text{kg NO}_3^- \text{-N ha}^{-1} \text{yr}^{-1}$

might be that the atmospheric compartment was a sink for NH_3 emitted by agro-ecosystems and, consequently, agro-ecosystems were sinks for NO_3^- . Those sinks were not accounted for in the *hydro* configuration which led to a higher accumulation of NO_3^- in soils, then higher NO_3^- recapture in the *hydro* configuration than in the *all* one. The highest values of NO_3^- inputs by both capillary rise and groundwater uprising were found for the unmanaged ecosystems in the east of the landscape, especially for groundwater uprising (around 60 $\text{kg NO}_3^- \text{-N ha}^{-1} \text{yr}^{-1}$). That means that groundwater reached the soil surface in the east of the landscape with a higher NO_3^- content than the soil NO_3^- content of the unmanaged ecosystems. On the contrary, the highest values of NO_3^- inputs by capillary rise were simulated for the maize fields located in the west of the landscape (around 5 $\text{kg NO}_3^- \text{-N ha}^{-1} \text{yr}^{-1}$). Soil water content was lower in the west than in the east of the landscape resulting in a higher capillary rise.

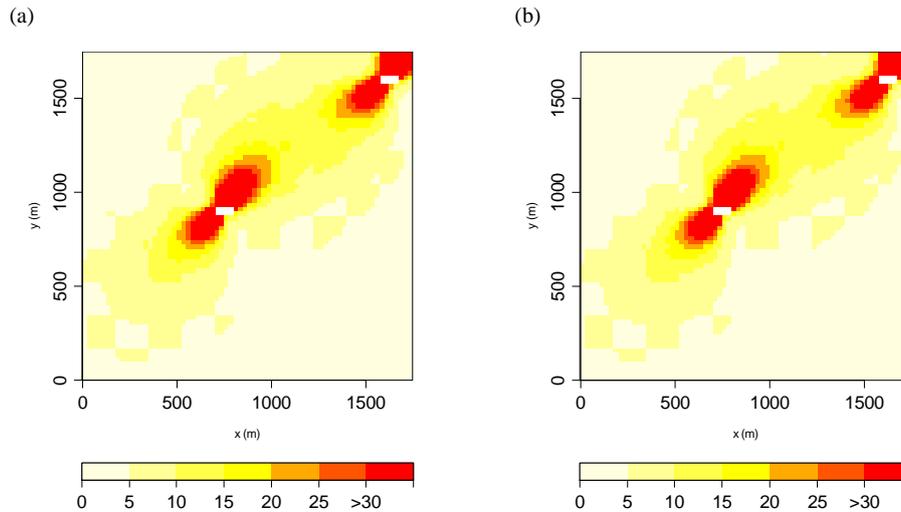


Fig. 6. NH_3 deposition in the (a) *all* and (b) *atm* configurations ($\text{kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$).

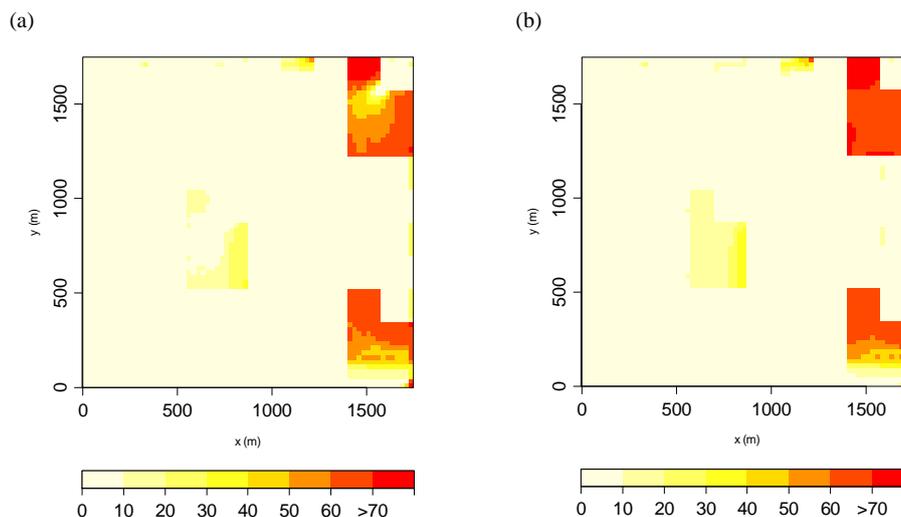


Fig. 7. NO_3^- inputs by capillary rise and groundwater uprising in the (a) *all* and (b) *hydro* configurations ($\text{kg NO}_3^- \text{-N ha}^{-1} \text{yr}^{-1}$).

That might be explained by the fact that the east of the landscape received water by rainfall and lateral transfers from the upper part of the landscape, while the west of the landscape mainly received water by rainfall. The highest NO_3^- inputs to soils by lateral transfers were simulated in the north-east of the landscape where water accumulated.

3.4 Indirect emissions of NH_3 , NO_3^- and N_2O within the landscape

Indirect N_r emissions were calculated by using the methodology described in Sect. 2.2.

Indirect NH_3 emissions due to both atmospheric and hydrological transfers were $0.5 \text{ kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$ in average, ranging from -7 to $108 \text{ kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$ (Table 1, Fig. 3b). Indirect NH_3 emissions resulting from atmospheric

(resp. hydrological) transfers and deposition (resp. recapture) were 0.7 (resp. 0) $\text{kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$ in average, ranging from 0 to 108 (resp. -14 to 7) $\text{kg NH}_3\text{-N ha}^{-1} \text{yr}^{-1}$ (Table 1, Fig. 3c resp. 3d). Hydrological transfers and recapture did not lead to indirect NH_3 emissions, while atmospheric transfers and deposition led to indirect NH_3 emissions. Moreover, indirect NH_3 emissions due to both atmospheric and hydrological transfers were not the sum of indirect NH_3 emissions due to atmospheric transfers and those due to hydrological transfers. That result indicates interactions between the atmospheric and hydrological pathways of N_r transfer. In the *all*, *atm* and *hydro* configurations the highest indirect NH_3 emissions were simulated close to the farm buildings located in the north-east of the landscape where soil saturation led to low nitrification.

Indirect NO_3^- losses to the groundwater due to both atmospheric and hydrological transfers were $20.3 \text{ kg NO}_3^- \text{-N ha}^{-1} \text{ yr}^{-1}$ in average, ranging from -20 to $283 \text{ kg NO}_3^- \text{-N ha}^{-1} \text{ yr}^{-1}$ (Table 1, Fig. 4b). Indirect NO_3^- losses to the groundwater due to atmospheric (resp. hydrological) transfers were 16.2 (resp. 16.2) $\text{kg NO}_3^- \text{-N ha}^{-1} \text{ yr}^{-1}$ in average, ranging from -9 to 151 (resp. -24 to 121) $\text{kg NO}_3^- \text{-N ha}^{-1} \text{ yr}^{-1}$ (Table 1, Fig. 4c resp. 4d). Thus, indirect NO_3^- leaching due to both hydrological and atmospheric transfers were not the sum of indirect NO_3^- leaching due to hydrological transfers and those due to atmospheric transfers. Like for NH_3 emissions, there were interactions between the hydrological and atmospheric pathways of N_r transfer. Like for NO_3^- recapture described above, those interactions might be related to exchange of the different species of N_r between the soil-groundwater compartment, the agro-ecosystems and the atmospheric compartment. In the *all* and *hydro* configurations the highest indirect NO_3^- losses were simulated for the wheat fields located in the east of the landscape while the lowest ones were for the unmanaged ecosystems. Hydrological lateral transfers therefore led to accumulation of NO_3^- at the bottom of the slope. High losses might be explained by the conditions of soil saturation which might lead to strong interaction with the groundwater and potentially high dilution of NO_3^- in the groundwater. In the *atm* configuration the highest indirect NO_3^- losses were simulated close to the farm buildings located in the centre of the landscape, especially for the wheat fields. In this part of the landscape high N_r inputs from fertilizer application and NH_3 deposition as well as favourable conditions for nitrification led to high soil NO_3^- content and consequently high NO_3^- leaching.

Indirect N_2O emissions due to both atmospheric and hydrological transfers were $1.2 \text{ kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$ in average, ranging from -33 to $29 \text{ kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$ (Table 1, Fig. 5b). They therefore represented 21 % of the total N_2O emissions in average at the landscape scale, which is in accordance with the value of 20 % proposed by IPCC (2006) and the value of 25 % reported by Reay et al. (2009). Indirect N_2O emissions due to atmospheric (resp. hydrological) transfers were around 0.7 (resp. 1.1) $\text{kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$, ranging from -1 to 26 (resp. -34 to 29) $\text{kg N}_2\text{O-N ha}^{-1} \text{ yr}^{-1}$ (Table 1, Fig. 5c resp. 5d). Like for NH_3 and NO_3^- fluxes, indirect N_2O emissions due to both atmospheric and hydrological transfers were not the sum of indirect N_2O emissions due to atmospheric transfers and those due to hydrological transfers. There were interactions between the hydrological and atmospheric pathways of N_r transfer, especially N_r exchanges between the soil, the vegetation and the atmosphere. The highest indirect N_2O emissions were simulated at different locations within the landscape according to the type of transfer. Indirect N_2O emissions due to atmospheric transfers were located close to the farm buildings, especially those located in the north-east of the landscape. The highest indirect N_2O emissions due to hydrological transfers were simulated

for the maize fields and the unmanaged ecosystems located in the north-east of the landscape which received high amounts of NO_3^- from recapture and where conditions of soil saturation were favourable to denitrification and then N_2O emissions. Negative indirect N_2O emissions due to hydrological transfers were simulated for the wheat fields located in the north-east of the landscape, which might be explained by dilution of NO_3^- in the groundwater. The highest indirect N_2O emissions due to atmospheric transfers were simulated close to the farm buildings, especially those located in the north-east of the landscape where NH_3 deposition led to higher soil NO_3^- content. In the other parts of the landscape, NH_3 deposition led to lower N_2O emissions, which might be explained by the fact that NH_3 deposition led to an increase of NO_3^- uptake by plants.

3.5 Indirect N_2O emission factor

The values of the indirect N_2O emission factors (i.e. EF4, EF5g and EF) simulated for the whole landscape were around 8, 9 and 6 % in the *atm*, *hydro* and *all* configurations, respectively (Fig. 8). For croplands only, those values were around 10, 9 and 4 % in the *atm*, *hydro* and *all* configurations, respectively. For the unmanaged ecosystems only, they were around 3, 10 and 6 % in the *atm*, *hydro* and *all* configurations, respectively. The values of the indirect N_2O emission factor were therefore higher for the unmanaged ecosystems than for croplands. The unmanaged ecosystems emitted more N_2O than croplands for the same amount of recaptured NO_3^- . That might be explained by competition for NO_3^- uptake by croplands than by unmanaged ecosystems. Moreover, the high productivity of croplands might also be linked to high water uptake by croplands, leading to reducing soil saturation, then reducing denitrification and consequently reducing indirect N_2O emissions by croplands in comparison with unmanaged ecosystems. Another hypothesis to explain patterns of indirect N_2O emissions might be that atmospheric NH_3 deposition was more limiting than hydrological NO_3^- recapture: deposited NH_3 needed to be first nitrified before being denitrified, while NO_3^- might be directly denitrified. This hypothesis might also explain the discrepancy between EF4 and EF5g for unmanaged ecosystems. This result supports the idea of the land-use receptor approach proposed by Denier van der Gon and Bleeker (2005) to estimate atmospheric indirect emissions.

The values of the indirect N_2O emission factor due to hydrological transfers might be compared with the ones of the EF5g emission factor derived from the IPCC methodology: 1.5 % ranging between 0.3 and 6 % (Mosier et al., 1998), 0.1 % ranging between 0.01 and 1 % (Nevison, 2000), 0.75 % ranging between 0.05 and 2.5 % (IPCC, 2006). They were higher than the maximum values proposed by those authors. The values of the indirect N_2O emission factor due to atmospheric transfers were higher than the ones of the

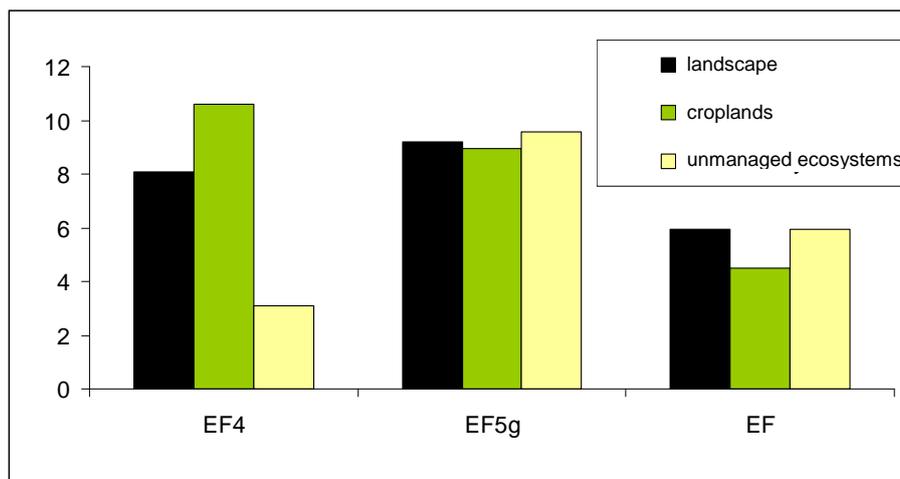


Fig. 8. Indirect N_2O emission factor in the *atm* (EF4), *hydro* (EF5g) and *all* (EF) configurations for the whole landscape, all croplands and the four unmanaged ecosystems. Values are in percentage of $kg N_2O-N$ emitted by $kg N$ captured.

EF4 emission factor derived from the IPCC methodology: 1 % ranging between 0.2 and 2 % (Mosier et al., 1998), 2.5 % ranging between 0.5 and 4 % (Denier van der Gon and Bleeker, 2005), 1 % ranging between 0.2 and 5 % (IPCC, 2006). The values of EF4 proposed by those authors included both short-range and long-range transfers and they were related to NH_3 , NH_4^+ and NO_x emissions, while values calculated from NitroScape only included short-range transfers and NH_3 emissions. Moreover, there were large uncertainties on values derived from the IPCC methodology and IPCC (2006) revised the EF4 values since emissions from some unmanaged ecosystems were higher than those previously reported (e.g. Denier van der Gon and Bleeker, 2005). The high values of EF4 simulated with NitroScape might result from high denitrification rates leading to both high direct and indirect N_2O emissions.

NitroScape simulations were carried out on a small test landscape and showed the role of short-range transfers in N_r fluxes, but further simulations on real landscapes are required to estimate N_r fluxes, budgets and indirect N_r emission factors on real conditions.

4 Conclusions

The NitroScape model integrates processes of N_r (N_r being NH_3 , NO_3^- , N_2O or NO) transformation and short-range transfer in a dynamic and spatially distributed way to simulate N_r fluxes and budgets at the landscape scale. By using four configurations of NitroScape taking into account or not the atmospheric, hydrological or both pathways of N_r transfer, we showed the ability of NitroScape to simulate patterns of N_r losses and recapture, and their large variability, for each landscape element (i.e. pixel with a size of $25 m \times 25 m$) within a test landscape. Moreover, NitroScape

made it possible to estimate the relative contribution of indirect N_r emissions to the N_r fluxes and budgets within the landscape, the relative contribution of the atmospheric and hydrological pathways to indirect N_r emissions as well as interaction between both pathways. The need of an integrated, spatially distributed and dynamic model is emphasized by the high variability of N_r losses and gains which were simulated within the landscape, and the effect of landscape topography and short-range processes on N_r fluxes. We also showed that N_2O emissions by unmanaged ecosystems were affected by both atmospheric deposition of NH_3 and hydrological recapture of NO_3^- , which emphasized the need to model dynamically both atmospheric and hydrological transfers of N_r . Taking into account both pathways of N_r transfers led to simulate high values of indirect N_2O emissions, estimated at around 21 % of the total N_2O emissions. Indirect N_2O emissions were affected by both the location of NH_3 deposition and NO_3^- recapture within the landscape and the land use of receptors. Thus, the spatial arrangement of agro-ecosystems, especially those located in areas of N_r recapture, may affect N_2O emissions. This hypothesis needs to be further tested by applying NitroScape to various scenarios of spatial arrangements of agro-ecosystems within the landscape and to real and larger landscapes.

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