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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

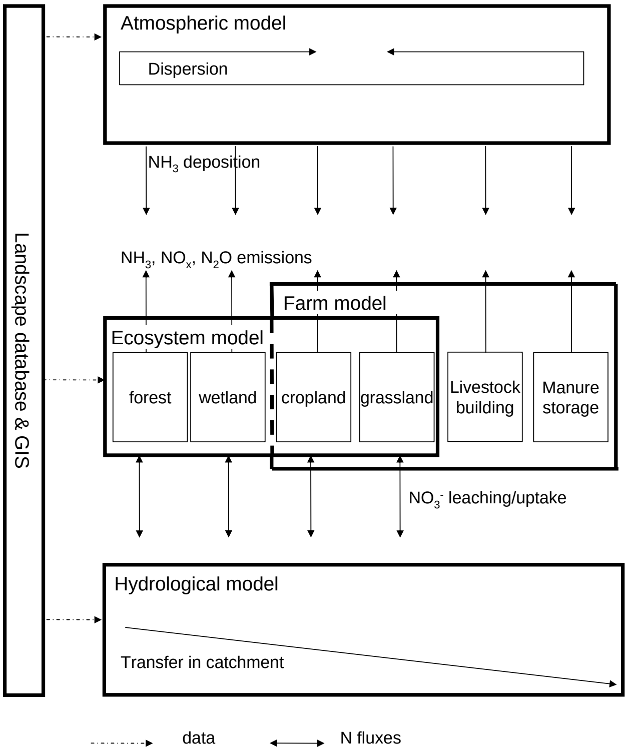

**Fig. 1.** The NitroScape model scheme.

(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 $N_2O$, 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; Ducharne 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 $NH_3$ at the landscape scale (Theobald et al., 2004; Kros et al., 2011) or indirect $N_2O$ 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 (Duretz 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 (Duretz 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, exfiltration, 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 Génermont 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 Michaëlis-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:

$$N_{r,\mathrm{ind,all}} = N_{r,\mathrm{tot,all}} - N_{r,\mathrm{tot,not}} \qquad (1)$$

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

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

$$N_{r,\mathrm{ind,atm}} = N_{r,\mathrm{tot,atm}} - N_{r,\mathrm{tot,not}} \qquad (2)$$

where $N_{r,\mathrm{tot,atm}}$ are the total $N_r$ emissions in the *atm* configuration.

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

$$N_{r,\mathrm{ind,hydro}} = N_{r,\mathrm{tot,hydro}} - N_{r,\mathrm{tot,not}} \qquad (3)$$

where $N_{r,\mathrm{tot,hydro}}$ 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):

$$\mathrm{EF4} = N_2O_{\mathrm{ind,atm}} / \mathrm{capt}_{\mathrm{NH3}} \qquad (4)$$

$$\mathrm{EF5g} = N_2O_{\mathrm{ind,hydro}} / \mathrm{capt}_{\mathrm{NO3}} \qquad (5)$$

$$\mathrm{EF} = N_2O_{\mathrm{ind,all}} / \mathrm{capt}_{\mathrm{N}} \qquad (6)$$

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, $\mathrm{capt}_{\mathrm{NH3}}$ is the total $NH_3$ deposition resulting from atmospheric transfers, $\mathrm{capt}_{\mathrm{NO3}}$ is the total $NO_3^-$ recapture resulting from hydrological transfers and $\mathrm{capt}_{\mathrm{N}}$ 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 $1.75 \times 1.75\,\mathrm{km}^2$ represented by a matrix of $70 \times 70$ pixels of $25 \times 25\,\mathrm{m}^2$ 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

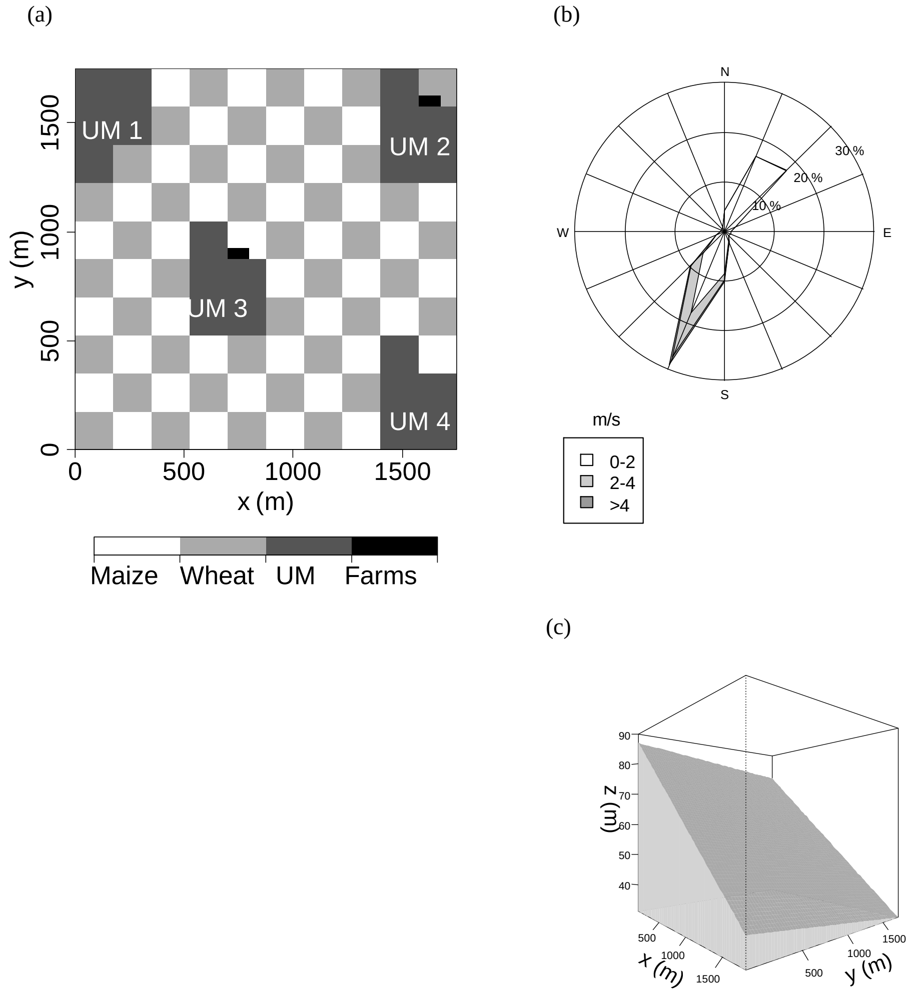

**Fig. 2. (a)** Land use in the test landscape (wheat and maize fields all received 240 kg N $\mathrm{ha}^{-1}$ $\mathrm{yr}^{-1}$ in average, UM: unmanaged ecosystem), **(b)** wind direction and speed and **(c)** topography of the landscape. x- and y-axes in **(c)** correspond to the ones in **(a)**.

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°01′ N, 2°83′ 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 $\mathrm{s}^{-1}$ (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, fishmeal 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 $\mathrm{ha}^{-1}$), March (60 kg N $\mathrm{ha}^{-1}$) and April (120 kg N $\mathrm{ha}^{-1}$). Maize received one manure application in March (120 kg N $\mathrm{ha}^{-1}$) and two mineral fertilizer applications in April (60 kg N $\mathrm{ha}^{-1}$). The unmanaged ecosystems received no fertilizer or manure, but only $\mathrm{N_r}$ from atmospheric transfer and deposition of $\mathrm{NH_3}$ or from hydrological transfer and recapture of $\mathrm{NO_3^-}$.

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 kg N $ha^{-1}$ $yr^{-1}$ in average within the whole landscape.

| Configuration | $NH_3$ emissions (kg $NH_3$-N $ha^{-1}$ $yr^{-1}$) | $NH_3$ dry deposition (kg $NH_3$-N $ha^{-1}$ $yr^{-1}$) | $NO_3^-$ leaching (kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$) | $NO_3^-$ inputs (kg $NO_3$-N $ha^{-1}$ $yr^{-1}$) | $N_2O$ emissions (kg $N_2O$-N $ha^{-1}$ $yr^{-1}$) | NO emissions (kg NO-N $ha^{-1}$ $yr^{-1}$) |
|---|---|---|---|---|---|---|
| *all* | 39.2 | 9.0 | 65.6 | 7.5 | 5.6 | 0.9 |
| *not* | 38.7 | 0.0 | 45.3 | 0.0 | 4.4 | 0.9 |
| *atm* | 39.4 | 9.1 | 61.5 | 0.0 | 5.1 | 0.9 |
| *hydro* | 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 Duretz 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 kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ in average in the four configurations (Table 1). Total $NH_3$ dry deposition was around 9 kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ 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 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$ in the *not* configuration and 65.6 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$ in the *all* configurations in average, with values around 61.5 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$ 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 kg NO-N $ha^{-1}$ $yr^{-1}$ 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 kg $N_2O$-N $ha^{-1}$ $yr^{-1}$ in average in the four configurations (Table 1). They were 5.6, 4.4, 5.1 and 5.5 kg $N_2O$-N $ha^{-1}$ $yr^{-1}$ 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 kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ in average, but they ranged from 0 to 121 kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ (Fig. 3a). Thus, direct $NH_3$ emissions

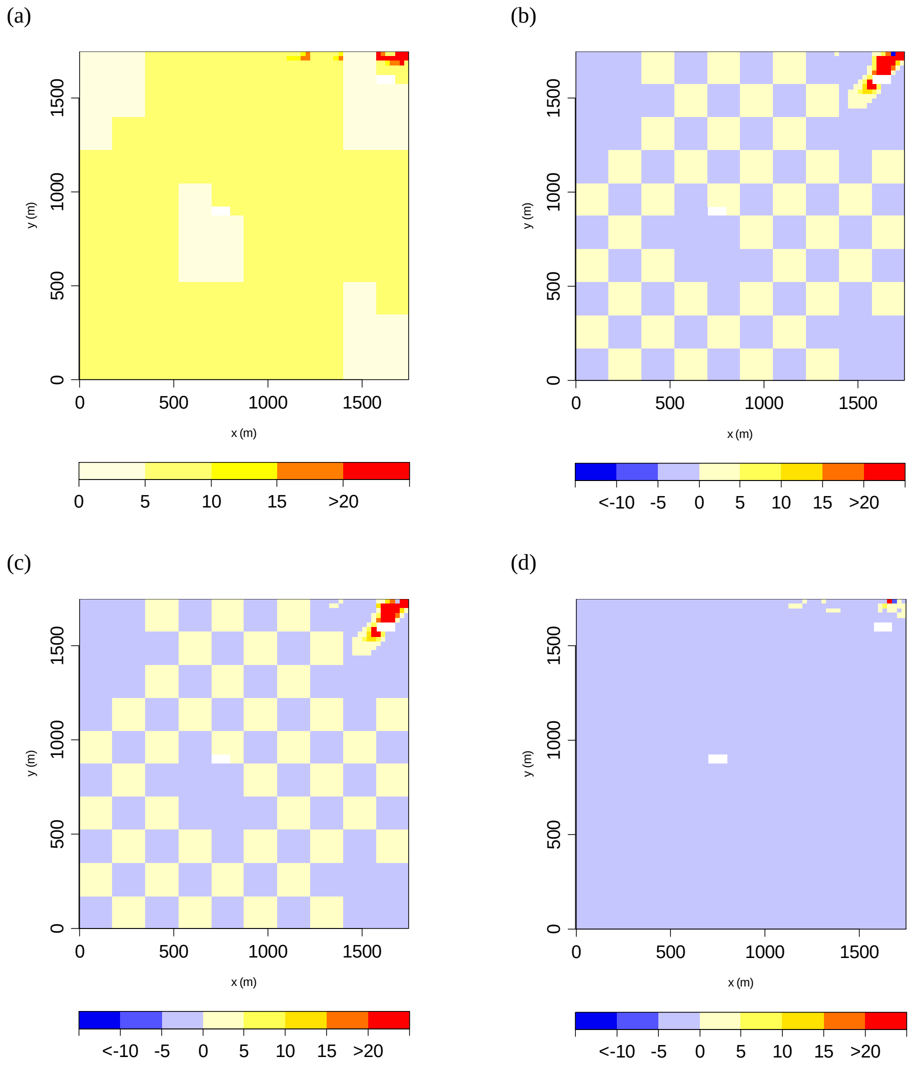

**Fig. 3. (a)** Direct $NH_3$ emissions by agro-ecosystems in the *not* configuration (kg $NH_3$-N $ha^{-1}$ $yr^{-1}$). Indirect $NH_3$ emissions in the **(b)** *all*, **(c)** *atm* and **(d)** *hydro* configurations (kg $NH_3$-N $ha^{-1}$ $yr^{-1}$). Negative values in **(b)**, **(c)** and **(d)** 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 unmanaged 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 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$, but they ranged from 0 to 150 kg $NO_3$-N $ha^{-1}$ $yr^{-1}$ (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 kg $N_2O$-N $ha^{-1}$ $yr^{-1}$ in average,

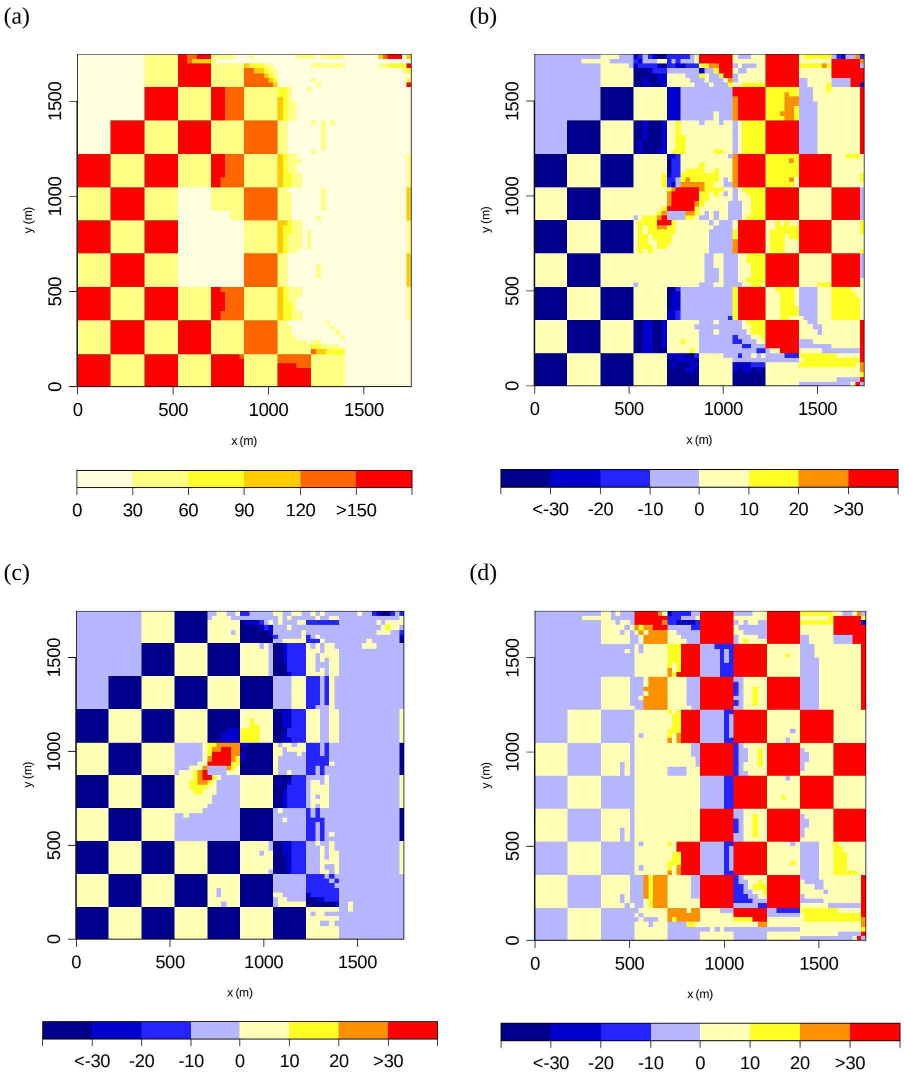

**Fig. 4. (a)** Direct $NO_3^-$ losses to the groundwater in the *not* configuration and indirect $NO_3^-$ losses in the **(b)** *all*, **(c)** *atm* and **(d)** *hydro* configurations (kg $NO_3^-$-N ha$^{-1}$ yr$^{-1}$). Negative values in **(b)**, **(c)** and **(d)** mean that $NO_3^-$ losses are lower in each of the *all*, *atm* and *hydro* configurations than in the *not* configuration.

ranging from 0 to more than 60 kg $N_2O$-N ha$^{-1}$ 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 kg $NH_3$-N ha$^{-1}$ yr$^{-1}$ in average in the *all* and *atm* (Table 1) configurations, but they ranged from 0.2 to 360 kg $NH_3$-N ha$^{-1}$ 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

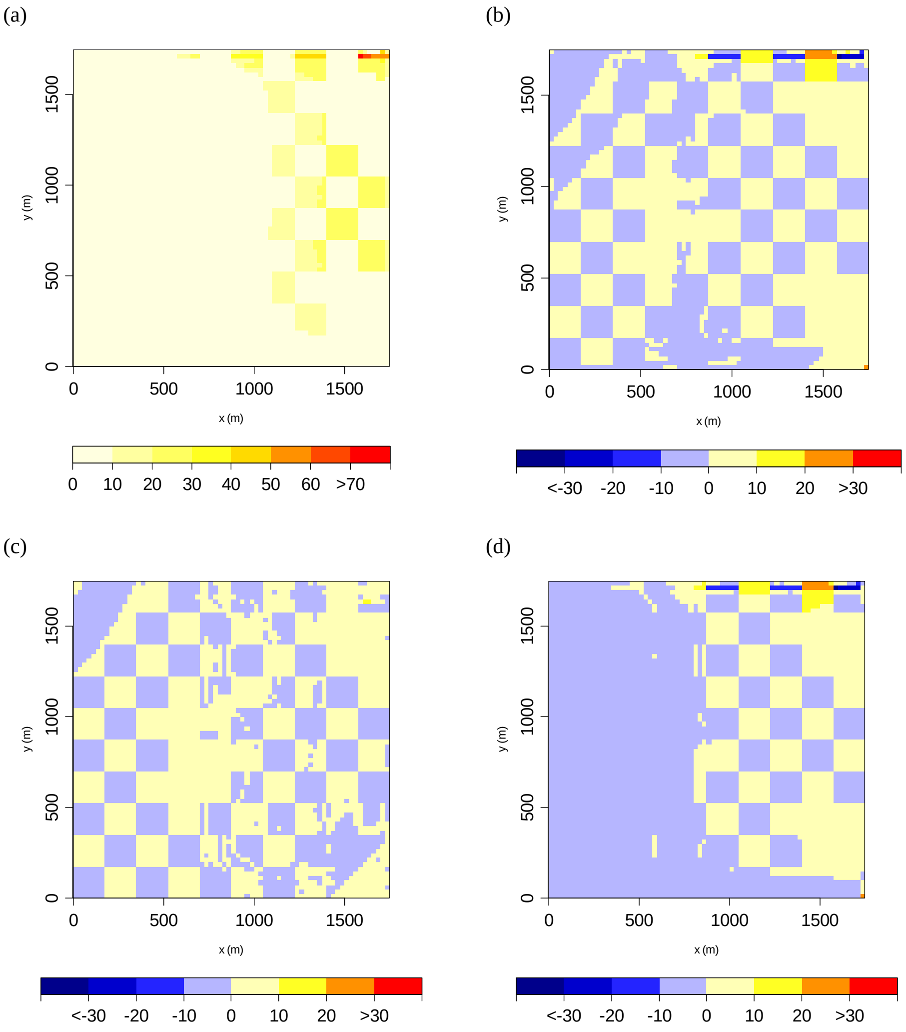

**Fig. 5. (a)** Direct $N_2O$ emissions in the *not* configuration and indirect $N_2O$ emissions in the **(b)** *all*, **(c)** *atm* and (d) *hydro* configurations (kg $N_2O$-N $ha^{-1}$ $yr^{-1}$). Negative values in **(b)**, **(c)** and **(d)** mean that $N_2O$ emissions are lower in each of the *all*, *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) kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$ in average in the *all* (resp. *hydro*) configuration, ranging from 0 to 130 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$ (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 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$ 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 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$). 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 kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$). Soil water content was lower in the west than in the east of the landscape resulting in a higher capillary rise.

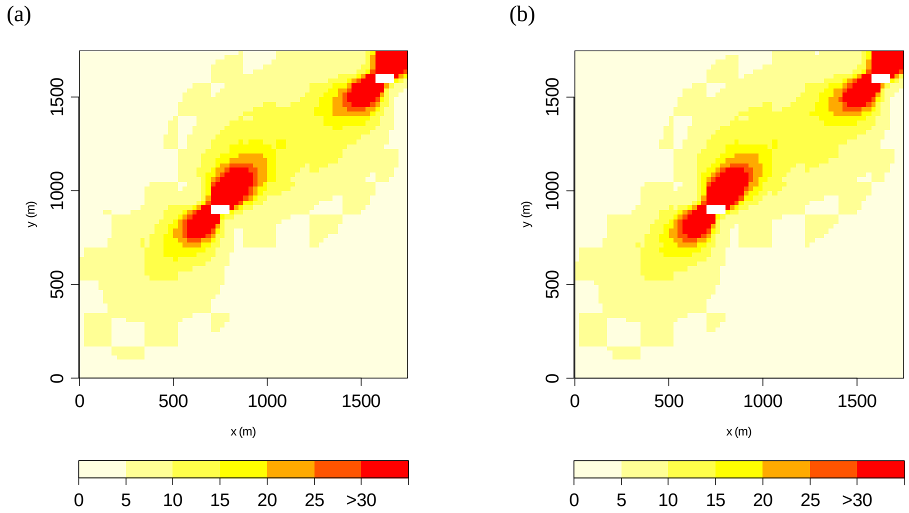

**Fig. 6.** $NH_3$ deposition in the **(a)** *all* and **(b)** *atm* configurations (kg $NH_3$-N $ha^{-1}$ $yr^{-1}$).

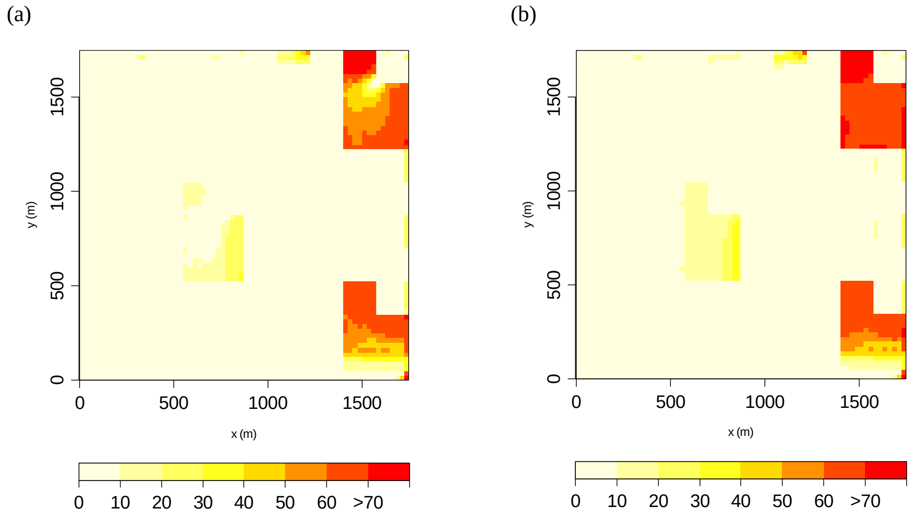

**Fig. 7.** $NO_3^-$ inputs by capillary rise and groundwater uprising in the **(a)** *all* and **(b)** *hydro* configurations (kg $NO_3^-$-N $ha^{-1}$ $yr^{-1}$).

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 kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ in average, ranging from −7 to 108 kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ (Table 1, Fig. 3b). Indirect $NH_3$ emissions resulting from atmospheric (resp. hydrological) transfers and deposition (resp. recapture) were 0.7 (resp. 0) kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ in average, ranging from 0 to 108 (resp. −14 to 7) kg $NH_3$-N $ha^{-1}$ $yr^{-1}$ (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 kg $NO_3^-$-N ha$^{-1}$ yr$^{-1}$ in average, ranging from −20 to 283 kg $NO_3^-$-N ha$^{-1}$ 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) kg $NO_3^-$-N ha$^{-1}$ yr$^{-1}$ in average, ranging from −9 to 151 (resp. −24 to 121) kg $NO_3$-N ha$^{-1}$ 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 kg $N_2O$-N ha$^{-1}$ yr$^{-1}$ in average, ranging from −33 to 29 kg $N_2O$-N ha$^{-1}$ 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) kg $N_2O$-N ha$^{-1}$ yr$^{-1}$, ranging from −1 to 26 (resp. −34 to 29) kg $N_2O$-N ha$^{-1}$ 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 $NO_3^-$ between plant uptake and denitrification with higher $NO_3^-$ 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

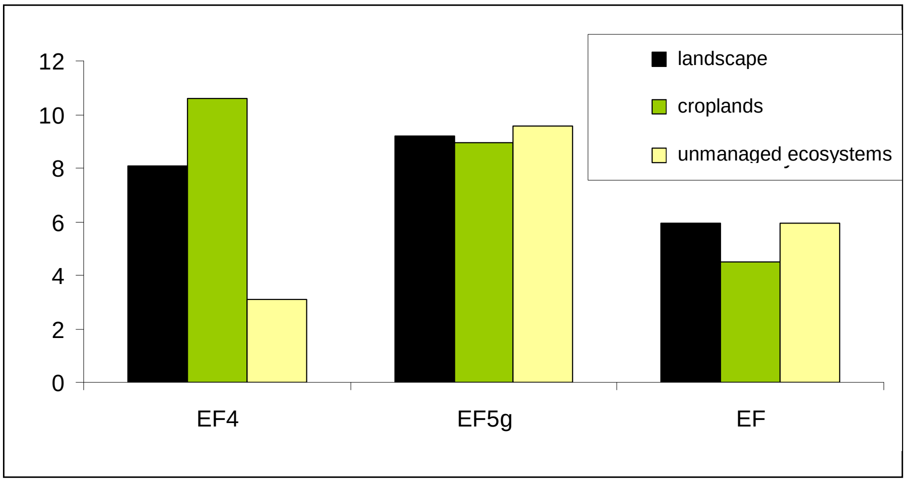

**Fig. 8.** Indirect $N_2O$ emission factor in the *atm* (EF4), *hydro* (EF5g) and *all* (EF) configurations for the whole landscape, all croplands and the four unmanaged ecosystems. Values are in percentage of kg $N_2O$-N emitted by kg N captured.

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 des 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 × 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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