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Integrated assessment of agricultural practices on large scale losses of ammonia, greenhouse gases, nutrients and heavy metals to air and water

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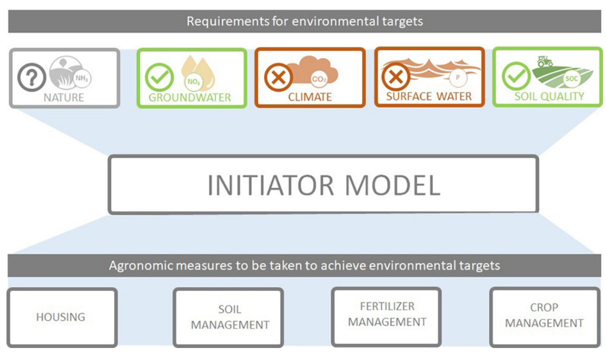
HIGHLIGHTS

- Intensive agricultural production poses a threat to biodiversity, climate, soil and water quality
- Losses of ammonia, GHG, nitrate and phosphorus from Dutch agriculture are predicted with an integrated model.
- Past reductions over 20 years were larger for ammonia and nitrate losses than GHG emissions and P runoff
- Both improved management and livestock reduction is needed to achieve all policy targets
- Long term climate ambitions are more stringent than ammonia emission requirements for biodiversity

GRAPHICAL ABSTRACT



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ABSTRACT

To gain insight in the environmental impacts of crop, soil and nutrient management, an integrated model framework INITIATOR was developed predicting: (i) emissions of ammonia (NH_3) and greenhouse gases (GHG) from agriculture, including animal husbandry and crop production and (ii) accumulation, leaching and runoff of carbon, nutrients (nitrogen, N, phosphorus, P, and base cations) and metals in or from soils to groundwater and surface water in the Netherlands. Key processes in soil are included by linear or non-linear process formulations to maintain transparency and to enable data availability for spatially explicit application from field up to national level. Calculated national trends in nutrient losses over 2000–2020 compared well with independent estimates and showed a reduction in N and P input of 26 to 33 %, whereas the surplus declined by 33 % for N and 86 % for P due to increased crop yields and reduced inputs. This was accompanied by a reduction of 30–35 % in atmospheric emissions of ammonia and nitrous oxide as well a decline in N and P runoff of 35 and 10 %, respectively, whereas the emission of methane increased with 4 %. Model results compared well with (i) large scale observations of ammonia concentrations in air and nitrate concentrations in upper groundwater and ditch water, (ii) with nitrous oxide emissions and phosphorus adsorption in experiments at field scale and (iii) with metal adsorption in large scale soil datasets.

Various mitigation measures were evaluated in view of policy ambitions for climate, soil and environmental quality for 2030, i.e. a reduction of 50 % for NH_3, 11–17 % for GHG, 20 % for N runoff and 40 % for P runoff and an ambition of 50 % GHG emission reduction for 2050. The measures focused on a combination of animal feeding, low emission housing and application technologies, improved crop, soil and nutrient management, all being applied with an effectiveness of 100 % and 50 %, respectively. In addition, we evaluated impacts of 50 % livestock reduction, and combination scenarios of measures and livestock reduction. Full implementation of all measures can reduce NH_3 emission, N leaching and N runoff by approximately 40–50 % and GHG emissions by approximately 30 %, but there is less potential

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to reduce P runoff, being <10 %. The combination of a more likely 50 % implementation/effectiveness of measures with 25 % livestock reduction leads to a comparable reduction. Required reductions from Dutch agriculture seem not possible with improved management only, but also requires livestock reduction, especially when the NH_3 ambitions at the short term (2030) and the climate ambitions for the long term (2050) should be attained.

1. Introduction

The Netherlands is one of the countries with the highest animal densities in the world. The livestock density is 3.8 livestock units per hectare of land, being approximately five times the average European value of 0.8 livestock units per hectare (Eurostat, 2016). The agricultural sector is per unit of product the most productive and efficient sector in the European Union (Van Grinsven et al., 2019). However, this also implies large challenges with respect to soil and nutrient management (recycling) to safeguard soil fertility, air quality, water quality, climate and biodiversity. Most important, high animal densities and high fertilization levels lead to very high reactive nitrogen (N) losses to air and water. Reactive nitrogen stands for all forms of oxidized and reduced nitrogen except for nitrogen gas (N_2), and includes ammonia (NH_3), nitrous oxide (N_2O) and nitrogen oxides (NO_x), being emitted to air, and of nitrate (NO_3) and ammonium (NH_4) lost to ground and surface water. Enhanced levels of reactive nitrogen in air, soil, groundwater and surface water lead to a cascade of effects (Cowling et al., 1998; Erisman et al., 2013). Observed effects in the Netherlands include: (i) negative impacts on human health due to NH_3 and NO_x induced particulate matter formation, and on plants due to ozone (O_3) exposure for which NO_x is a precursor, (ii) decreased biodiversity (especially plant species diversity) and of non-agricultural soils due to NH_3 and NO_x induced eutrophication and acidification, (iii) pollution of groundwater and drinking water due to NO_3 leaching, (iv) eutrophication of surface waters due to high N losses leading to excess algal growth and a declining aquatic biodiversity and (v) global warming due N_2O emissions (Erisman et al., 2001).

The so-called Dutch nitrogen crisis (see e.g. Stokstad, 2019) currently focuses on NH_3 and NO_x emissions to air from agriculture, together with traffic, industries and households in view of biodiversity protection, with agriculture having the dominant share in NH_3 emissions (87 % in 2018; ER, 2020) and a minor share in the NO_x emissions (17 % in 2018; ER, 2020). However, the overall environmental N effects of Dutch agriculture are much larger due to its high contribution to N_2O emissions (74 % in 2018; ER, 2020) and a major contribution to the losses of N to groundwater and surface water (55 % in 2015, Groenendijk et al., 2016) with impacts on groundwater and surface water quality. In addition, livestock farming contributes to methane (CH_4) emissions, only partly compensated by accumulation of soil organic carbon (SOC) in grassland on mineral soils. Changes in soil carbon are relevant in view of soil fertility and the role of soil as a sink or source of CO_2. Approximately 10 % of the total greenhouse gas emissions (GHG) originates from agricultural N_2O and CH_4 emissions (in CO_2-eq) (ER, 2020; Ruyssenaars et al., 2020).

Inorganic and organic fertilization also cause accumulation and/or elevated leaching and runoff of phosphorus (P), base cations (Ca, Mg, K) and metals (Cd, Cu and Zn) from agricultural soils to groundwater and surface water. Phosphorus loading is in most surface waters the key element controlling the ecological biodiversity. The impacts of enhanced P loads on the eutrophication of surface water are a major societal and economical challenge (Van der Zee, 1988; Schoumans and Groenendijk, 2000; Van Gaalen et al., 2016; Van Grinsven et al., 2016). In addition, leaching of base cations implies soil acidification leading to suboptimal soil conditions for crop growth (e.g. Zhu et al., 2020), but in the Netherlands, acidification impacts are counteracted by liming and high application doses of organic manure, but this is not always done effectively. Soil compaction due to intensive machinery use affects a number of environmental parameters like crop yields, effectiveness of fertilizers, greenhouse gas emission and nutrient losses to water (van den Akker, 2004). Finally, leaching and runoff of heavy metals to groundwater and surface water recently gained awareness due to their large impacts on water quality (Bonten et al., 2008; De Vries et al., 2008a). Most relevant metals are copper (Cu) and zinc (Zn), mainly originating from dairy and pig manure, and cadmium (Cd), mainly originating from contaminated P fertilizers. Metal leaching contributes to 20–40 % of the total load to surface waters, exceeding established guidelines for Cu and Zn in many areas (Bonten et al., 2008; De Vries et al., 2008a).

Given the huge impact of agronomic measures on the environment, European and national regulations are designed to increase the sustainability of agriculture. European directives or agreements to reduce the emissions of nutrients, greenhouse gases and pollutants to air and water include (i) the National emission ceilings (NEC) directive (EC, 2001) with NH_3 and NO_x emission targets, (ii) the Birds and Habitats Directive affecting N emissions (EC, 1979, 1992), (iii) the Nitrates Directive (EC, 1991) and the Water Framework Directive (EC, 2000) with critical N and P concentration limits for waterbodies leading to critical N and P doses, and (iv) the Paris Climate Agreement (UN, 2015) and related National Climate Agreement - The Netherlands (NCA; TK, 2019) with emission targets for CO_2, N_2O and CH_4. An overview of the various environmental goals for agriculture in 2030 related to these agreements and the current status (i.e. the year 2018) is given in Table 1.

The national NH_3 emission target is set at 121 kton $NH_3 \cdot yr^{-1}$ (with 106 kton originating from agriculture) for 2030 and onwards according to the NEC directive 2016 (EC, 2016), while the national NO_x emission target is set at 148 kton $NO_x \cdot yr^{-1}$ in 2030 and following years (EC, 2016) being relatively close to the current emission levels. In addition, however, the Birds and Habitats Directive requires that habitats are kept or improved towards in a good ecological condition. Even though the word nitrogen is not mentioned, this implies a reduction in N deposition for all terrestrial ecosystems where the current N deposition exceeds a critical N deposition (critical load) for adverse impacts on biodiversity. This in turn requires a strong reduction in NH_3 and NO_x emissions. The national NH_3 and NO_x emission targets do by far not protect all terrestrial ecosystems across Europe (Hettelingh and Posch, 2019). Consequently, at the beginning of this century, more stringent emission ceilings have already been suggested for the Netherlands ranging from 30 to 55 kton $NH_3 \cdot yr^{-1}$ and 70 to 120 kton $NO_x \cdot yr^{-1}$ for 2030 (Beck et al., 2001; RIVM, 2001). The lower limit refers to sustainable emission levels resulting in 90 % protection of terrestrial ecosystems. Recently, a national advisory board in view of the nitrogen crises (Advisory Board Nitrogen, 2020) advised the government to achieve a 75 % protection of the area with N sensitive habitats in 2030, implying a 50 % reduction of current NH_3 and NO_x emission. In addition, a reduction of 70 % has been advised, based on a calculated 90 % protection of terrestrial ecosystems (Paul, 2021). We used the suggested 50 % and 70 % reduction for 2030 and for 2050, respectively (Table 1).

The aim related to N leaching is such that NO_3 concentrations in upper groundwater stay below 50 $mg \cdot l^{-1}$ (EC, 1991). With respect to N and P runoff, the N and P concentrations in surface waters should stay below the upper limit of good ecological status (EC, 2000), ranging from 1.3 to 4.3 $mg \cdot l^{-1}$ for N (mostly from 2.0 to 2.8 mg $N \cdot l^{-1}$) and from 0.04 to 0.53 $mg \cdot l^{-1}$ for P (mostly from 0.10 to 0.25 mg $P \cdot l^{-1}$). In this study we used a critical limit of 2.4 mg $N \cdot l^{-1}$ and 0.22 mg $P \cdot l^{-1}$ based on good ecological potential of freshwater ditches (STOWA, 2020). The target for the reduction in greenhouse gases, including N_2O, is a 40 % decrease by 2030 compared to 1990 for Europe (Paris Climate Agreement), whereas the national target is a climate neutral agriculture in 2050 including compensation by C sequestration by afforestation at EU scale (European Green Deal, Dutch Climate Agreement). Concentrations of the heavy metals Cd, Pb and Zn in surface water should stay below 0.19 and 11 $\mu g \cdot l^{-1}$ (Staatsblad, 2015) and 15.6 $\mu g \cdot l^{-1}$ (Bonten et al., 2010), respectively. For

Table 1
Environmental goals for agriculture in 2030 related to National Emission Ceilings (NEC), Birds and Habitats Directive (BHD), Nitrates Directive (ND), Water Framework Directive (WFD) and Dutch Climate Agreement (DCA) and the current status (year 2015).

Component	Policy	Entity	Current Status 2015	Reduction goals 2030	Reduction goals 2050
Ammonia (NH_3)	BHD (NEC)	Emission to atmosphere	94 ktonNH$_3$-N[a]	50 %[b]	70 %[c]
Nitrogen oxide (NO_x)	BHD (NEC)	Emission to atmosphere	10 ktonNO$_x$-N[a]	50 %[b]	70 %[c]
Nitrate (NO_3)	ND	Area that exceeds 50 mg NO_3 l^{-1} in groundwater	7.1 %[d]	0 % exceedance[e]	0 % exceedance[e]
Nitrogen(N) and phosphorus (P)	WFD	Leaching and runoff flux to surface water	45 ktonN[f]	20 %[g]	20 %[g]
			3.7 ktonP[f]	40 %[g]	40 %[g]
CO_2, methane (CH_4) and nitrous oxide (N_2O)	DCA	Emission to atmosphere	25 Mton CO_2-eq[h]	11–17 %[i]	50 %[i]

[a] Refers to emissions from Dutch agriculture in 2015 based on NEMA calculations being 114 kton NH_3 and 33 kton NOx (Van Bruggen et al., 2022), which were multiplied by 14/17 and 14/46 for NH3 and 14/46 for NOx.

[b] The reduction percentage of 50 % is based on the advice of the Commission Remkes (Adviescollege Stikstofproblematiek, 2020), assuming that 75 % of the area with N sensitive habitats is then below critical N loads in view of biodiversity impacts. The formal Dutch policy is now to reach a 50 % reduction in 2035. This percentage is based on the assumption that NH_3 and NO_x emissions from other countries reach the NEC ceilings (EC, 2016) by 2030. The Dutch NH_3 and NO_x ceilings for 2030 and onwards are only 121 kton NH_3 and 148 kton NO_x (while 50 % reduction implies 65 kton NH_3 and 118 kton NO_x), but these ceilings are by far not stringent enough to protect all N sensitive habitats.

[c] The reduction of 70 % is based on an advice by Paul (2021), which in turn is bases on a calculated 90 % protection (no exceedance of critical nitrogen loads) of terrestrial ecosystems in view of biodiversity impacts (De Vries et al., 2020; Van den Burg et al., 2021), leading to total emission levels of 39 kton NH_3 and 70 kton NO_x being close to "sustainable emission targets" mentioned 20 years ago, i.e. 30 kton NH_3 and 70 kton NO_x (Beck et al., 2001).

[d] The percentage mentioned is based on calculations with the INITIATOR model.

[e] This is not an official goal, but a target value. The Nitrates Directive only requires to reduce water pollution caused by nitrates from agricultural sources. Therefore measures have to be formulated in national action programs for all Nitrate Vulnerable Zones (i.e. the Netherlands as a whole) where the nitrate concentration in surface and/or groundwater exceeds the target of 50 mg l^{-1}.

[f] The values for the leaching and runoff of N and P are based on calculations with the STONE model (Groenendijk et al., 2016; Groenendijk and van Boekel, 2017). These losses refer, however, to the mean annual load for the period 2010–2013 (https://data.pbl.nl/api/embed/infographic/data/nl/emw17/001s/01/001s_emw17_01_nl.pdf.

[g] Estimates for the reduction of N and P losses to water are based on an evaluation of the Dutch manure law (PBL, 2017) who based the percentage on a detailed modelling study (Groenendijk et al., 2016; Groenendijk and van Boekel, 2017). In this study, the target losses were defined as a reduction of the N and P loads on surface waters from agricultural land required in order to each targets for N and P in surface water that are needed to achieve a "good" ecological state in surface waters.

[h] Refers to emissions of CO_2, N_2O and CH_4 from Dutch agriculture in 2015 based on INITIATOR results. The results for N_2O and CH_4 are in line with NEMA calculations (Van Bruggen et al., 2022), i.e. 18.4 kton N_2O and 482 kton CH_4, using GWP values from IPCC (2006), i.e. 25 CH_4 and 298 for N_2O, being 18 Mton CO_2 eq.

[i] The emission reduction goal for 2030 for agriculture, includes a target of 2.7 Mton CO_2 eq, while the ambition is 4.3 Mton CO_2 eq being a reduction of 11–17 % compared to 2015, as adopted in the National Climate Agreement: (https://www.klimaatakkoord.nl/binaries/klimaatakkoord/documenten/publicaties/2019/06/28/klimaatakkoord/klimaatakkoord.pdf). For 2050, the reduction aim of 50 % is based on the long term strategy of the European Commission (COM, 2018), assuming climate neutrality, with the remaining emissions from Dutch agriculture being compensated by C sequestrations in EU countries with large forested areas (Lesschen et al., 2020).

Cu, we used a value of 50 μg·l^{-1} being a limit for drinking water (Staatsblad, 2015) and also protecting 95 % of aquatic organisms (Bonten et al., 2010).

In summary, agricultural management affects (i) air quality and human health (by emission of NH_3 and to a small extent NO_x), (ii) biodiversity of terrestrial ecosystems due to eutrophication and acidification (by emission and deposition of NH_3), (iii) drinking water quality and eutrophication of aquatic ecosystems (by leaching and runoff of N and P) and (iv) climate (by emission of N_2O, CH_4 and CO_2). Fertilization and soil cultivation also (v) impacts soil fertility by leaching of base cations, due to nitrogen-induced soil acidification and by (vi) soil pollution due to accumulation of Cd, while leaching of Cd, Cu and Zn may further affect water quality. Numerous long-term field experiments have shown that agronomic measures might positively contribute to all these challenges due to optimized soil, crop and nutrient management. Policy regulations however are usually driven by one dimensional target, hampering integrative assessments of best practices and policy measures.

To gain insight in all environmental impacts of soil, crop and nutrient management simultaneously, an Integrated Nutrient ImpacT Assessment Tool On a Regional scale (INITIATOR) was developed. The policy aim of INITIATOR is to present spatially explicit information on the effectiveness of policies, given the desired aims for sustainable agriculture, of all relevant element fluxes (nutrient and contaminants) to atmosphere, groundwater and surface water in the Netherlands. This paper first provides an overview of the integrated model framework. It then demonstrates how INITIATOR can be used to predict temporal and spatial variation in inputs, uptake, soil accumulation and all relevant losses to air and water in response to measures that are currently used for underpinning environmental policies. Results focus on an integrated evaluation of mitigation measures on those losses in view of exceedances of targets or limits for: (i) NH_3 emissions in view of terrestrial biodiversity (ii) NO_3 in groundwater in view of drinking water quality, (iii) N and P concentrations in surface water in view of eutrophication and (iv) greenhouse gas emissions in view of climate change. For various sets of agronomic measures, we identify potential win-wins and trade-offs, to gain insight in the most promising combinations of measures contributing to a sustainable living environment. Insights are relevant for advice to farmers and policy makers in view of the implementation of environmental policies. Details on (i) model descriptions, (ii) input data, (iii) spatial variation of results, (iv) impacts of measures on element accumulation and losses and (v) an assessment of the plausibility of estimated nutrient fluxes and concentrations, in view of observations and independent model assessments, are given in the Supplementary materials S1 to S5, respectively.

2. Modelling approach

2.1. Rationale and overall approach of the INITIATOR model

2.1.1. Model rationale

The model INITIATOR simulates annual fluxes of carbon, nutrients and metals in soil with losses of C and N compounds (CO_2, CH_4, N_2O, NH_3, NO_x and N_2) to air and nutrients (N, P, S, Ca, Mg, K) and metals (Cu, Zn, Cd, Pb) to water for 28,610 unique plots (see also 3.1 for details on the model resolution).

The original emphasis was on nitrogen to: (i) gain insight in the fate of all major N flows in the Netherlands (De Vries et al., 2003b), (ii) calculate regional specific N ceilings defined as the maximum reactive N dose without exceedance of critical limits or targets (De Vries et al., 2001b) and (iii) assess the environmental impacts of agricultural practices and technical measures on N emissions such as changes in animal housing (De Vries et al., 2001a). The model estimates the fate of N fluxes in response to N inputs by fertilizer, manure, biosolids (organic amendments), N fixation and N deposition regarding (i) gaseous emissions of NH_3, NO_x and N_2O from housing and manure storage systems as well as soils and (ii) leaching and runoff of nitrate and ammonium from soil to groundwater and surface water. INITIATOR has been linked to an atmospheric transport model to track atmospheric dispersion of NH_3 and NO_x followed by N deposition in response to changes in agricultural management (De Vries et al., 2011; Kros et al., 2013).

In addition, the model includes: (i) emissions of CH_4 from housing and manure storage systems and CO_2 exchange from soil and (ii) plant uptake, soil accumulation/release (including mineralization/immobilization and adsorption/desorption), leaching and runoff of phosphorus, base cations and heavy metals to groundwater and surface water (De Vries et al., 2011). This integration of carbon, nutrient and metal fluxes on the one hand and the assessment of fluxes due to agronomic measures across soil, water and gaseous environments on the other is unique. It allows an integrative assessment of environmental impacts given a set of site properties and agronomic measures, thereby avoiding pollution swapping and stimulating sustainable management.

2.1.2. General modelling approach

It is imperative that the model approach is appropriate in view of the model objectives and the spatial and temporal scales addressed. The aim of the INITIATOR model is to assess annual element fluxes over a longer time period at high spatial resolution and evaluate the impacts of multiple feed, housing, nutrient, soil and crop management practices on those fluxes. When applying a model on a regional scale, there is a trade-off between model complexity and data availability. When the model complexity increases, the descriptive (model) error decreases but the parameter error increases. The aim is to choose a level of complexity that minimises the total prediction error in large scale environmental applications (see also De Vries et al., 2005).

There are multiple detailed mechanistic models to simulate element fluxes in the field, such as C and N fluxes and GHG emissions, but these models require multiple input data (e.g. initial conditions and parameters) which are often limited or even unknown at the relevant scale. Especially at high spatial resolution, many model parameters cannot be derived in such models and most mechanistic models make us of generic parameter estimates from literature. Mechanistic models are thus very useful to gain insights in processes, in particular for experimental sites that are intensively monitored, but they are mostly not very suitable for large scale applications. This holds even stronger for the implementation of measures, which is generally very hard to parameterize. Considering the aim to predict annual element fluxes, our modelling philosophy was to develop a process-based model at intermediate complexity, including all key processes, well accounting for spatial variability by making use of readily available empirical data based on field measurements. De Vries et al. (1998) showed that model simplification, in terms of less detailed formulations of processes (process aggregation) at low temporal resolution (temporal aggregation) is an adequate step in the upscaling of modelling results from a local to a regional scale when interested in annual average element fluxes. The spatial variability was included by using empirical relationships between model parameters and factors driving the variation, including animal category, land use/crop type, soil type/soil properties (such as pH, clay and organic matter content) and groundwater level to maintain transparency and to increase model applicability across the Netherlands.

A flow chart of all element inputs and transformation processes is given in Fig. 1. INITIATOR contains three main modules describing the element excretion and emissions on farm scale (housing systems), the spatial explicit distribution of manure and fertilizer, and the fate of those elements in soil and atmosphere. In addition it is coupled to an atmospheric transport model that calculates the N deposition to agricultural soils.

Relevant data from animal numbers, housing types, grazing time and geographic location for each farm for each year were derived from census data in the GIAB database (Van Os et al., 2016). The emissions of NH_3, NO_x, N_2O and CH_4 from housing and manure storage systems are quantified by multiplication of animal numbers with either N excretion factors and emission fractions (for NH_3, NO_x, N_2O) or with emission factors (for CH_4) per animal category and housing type (for NH_3, N_2O). The model accounts for >60 categories affecting both N excretion and NH_3 emission. The excretion of C, N and P in manure is calculated for each farm by a multiplication of the animal numbers with the excretion per animal for the particular year. The excretion of metals and base cations is calculated from generic ratios to P for the included manure types. This excretion, corrected for gaseous N emissions, is input to a manure and fertilizer distribution model that predicts the inputs of C, N, P, base cations and metals to the soil, both by manure and inorganic fertilizers.

The soil module calculates emissions of NH_3, NO_x, N_2O, CH_4 and CO_2 from terrestrial systems and accumulation, leaching and runoff of carbon, nutrients (nitrogen, phosphorus and base cations) and metals to groundwater and surface water. The NH_3 emissions by field application of manure or

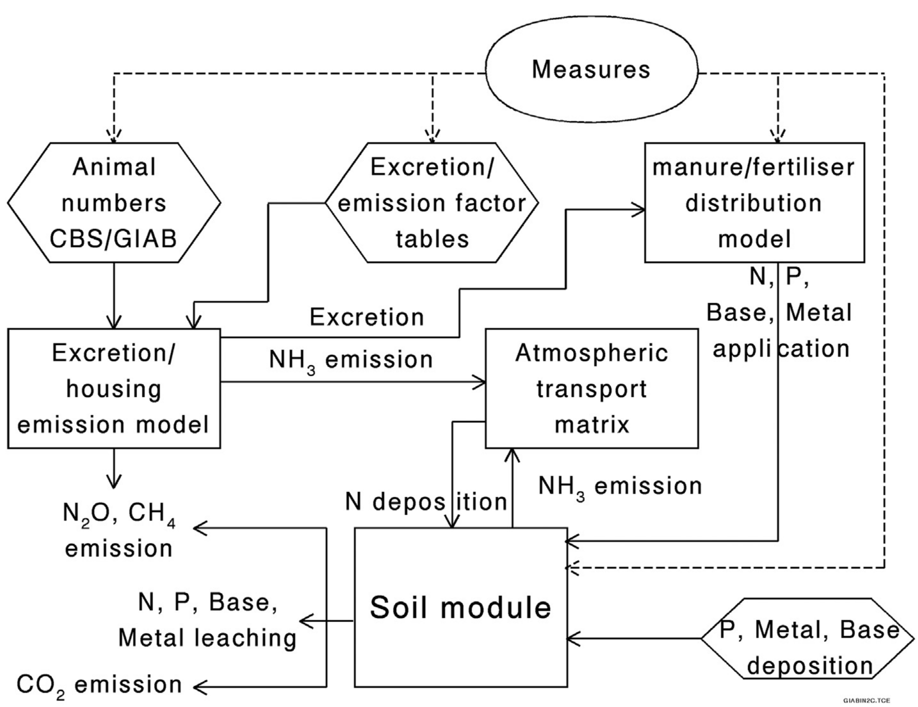

Fig. 1. Coupling of modules and model outputs in INITIATOR.

Table 2

List of symbols used in the process descriptions in INITIATOR.

Symbol	Explanation	Unit
Excretion of carbon, nutrients and metals		
acat	Animal category	–
n_{acat}	The number of animals in each animal category	–
$M_{ex,acat}$	Excreted mass of manure	$kg \cdot animal^{-1}$
$E_{c,acat}$	Element concentration in manure	$g \cdot kg^{-1}$
E_{ex}	Excreted element via animal manure	$kg \cdot ha^{-1} \cdot yr^{-1}$
Gaseous emissions from housing systems		
$N_{in,pr}$	Total excretion of N in animal manure	$kg \cdot ha^{-1} \cdot yr^{-1}$
N_{em_h}	Total N (NH_3, NO_x, N_2O and N_2) loss from housing and storage systems, divided in emissions for ammonia, $NH_{3,em,h}$, nitrous oxide, $N_2O_{em,h}$, nitrogen oxide, $NO_{x,em,h}$, and di-nitrogen, $N_{2,em,h}$	$kg \cdot yr^{-1}$
frN_{em_h}	N emission fractions from manure in housing and manure storage systems, divided in ammonia, $frNH_{3,em,h}$, nitrous oxide, $frN_2O_{em,h}$, nitrogen oxide, $frNO_{x,em,h}$, and di-nitrogen, $frN_{2,em,h}$	–
$frNH_{3,em,s}$	Ammonia emission fractions from manure in storage systems	–
$CH4_{em\_h}$	Total CH_4 emission from manure in housing and manure storage systems	$kg \cdot yr^{-1}$
$frCH4_{acat}$	CH_4 emission factor per animal in an animal category	$kg \cdot animal^{-1} \cdot yr^{-1}$
$frCH4_{em\_h}$	CH_4 emission per volume manure produced	$kg\ CH_4 \cdot m^{-3}$
D_{acat}	Manure density per animal category	$kg \cdot m^{-3}$
Manure and fertilizer redistribution		
E_{ex_farm}	The excreted amount of elements (N and P) for a farm	$kg \cdot yr^{-1}$
E_{em_hs}	The element fraction lost via gaseous emissions from a farm	–
E_{exp_farm}	The amount of elements (N and P) exported to farms outside the NL	$kg \cdot yr^{-1}$
Espace_farm	The amount of manure elements (N and P) that can be applied on farm scale	$kg \cdot yr^{-1}$
$Pdose_{field}$	The maximum dose of manure P that one is allowed to apply	$kg \cdot ha^{-1} \cdot yr^{-1}$
$Ndose_{field}$	The maximum dose of manure N that one is allowed to apply	$kg \cdot ha^{-1} \cdot yr^{-1}$
Soil related emissions of carbon		
C_{soil}	The Carbon pool in soil	$kg \cdot ha^{-1}$
k	The decomposition constant for organic carbon	yr^{-1}
hc	The humification constant for organic products, different for manure type, organic residues and compost	–
C_{in}	The amount of C applied to the soil	$kg \cdot ha^{-1}$
s_{mv}	The annual lowering of peat soils due to drainage	$m \cdot yr^{-1}$
ρ_{om}	The density of unripened peat	$kg \cdot m^{-3}$
fr_c and fr_{om}	The organic matter fraction of peat, and the organic C fraction of the organic matter	–
D_{peat}	The depth of the peat layer	Cm
C_{lime}	The net C release due to liming	$kg \cdot ha^{-1} \cdot yr^{-1}$
$CO_{3,lime}$	The amount of added carbonates via liming products	$kg \cdot ha^{-1} \cdot yr^{-1}$
$HCO_{3,uit}$	The amount of C leached in the form of HCO_3	$kg \cdot ha^{-1} \cdot yr^{-1}$
$CH4_{em\_s}$	The emitted CH_4 from the soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
gwl	Groundwater level below surface	Cm
Soil related emissions of ammonia		
$NH_{3,em,a}$	Ammonia emission due to manure and fertilizer application and grazing	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{in,am}$	Nitrogen input to the soil via animal manure	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{in,g}$	Nitrogen input to the soil via dung and urine from grazing animals	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{in,f}$	Nitrogen input to the soil via fertilizer	$kg \cdot ha^{-1} \cdot yr^{-1}$
$frNH_{3,em,a}$	Ammonia emission fraction from manure applied to land	–
$frNH_{3,em,g}$	Ammonia emission fraction from dung and urine from grazing animals	–
$frNH_{3,em,f}$	Ammonia emission fraction from fertilizer	–
Nitrogen uptake		
N_{up}	Net nitrogen uptake in crops removed from the field	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{up,min}$	Net nitrogen uptake at zero input of nitrogen	$kg \cdot ha^{-1} \cdot yr^{-1}$
N_{dep}	Nitrogen deposition	$kg \cdot ha^{-1} \cdot yr^{-1}$
N_{fix}	Biological nitrogen fixation	$kg \cdot ha^{-1} \cdot yr^{-1}$
fr_{up}	Nitrogen uptake fraction	–
$f_{a,am}$	Factor describing the availability of animal manure relative to fertilizers	–
$f_{a,g}$	Factor describing the availability of excrements from grazing animals relative to fertilizers	–
Nitrogen immobilization, mineralization, nitrification and denitrification		
$N_{im,s}$	Net nitrogen immobilization in soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
N_{in}	Total N input to the soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
$fr_{im,s}$	Immobilization fraction for the soil	–
$N_{ni,s}$	Nitrification in the soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
$fr_{ni,s}$	Nitrification fraction for the soil	–
$N_{de,s}$	N losses via denitrification in the soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
$fr_{de,s}$	Denitrification fraction for the soil	–
DA, CNM	Dissimilation-to-Assimilation ratio as well as the C:N ratio of microorganisms involved in mineralization of organic matter	–
CN_{soil}	The C:N ratio of the soil	–
$rf_{mi,cn}$	Correction factor to convert C:N ratio given the C:N ratio of peat	–
Leaching of nitrogen to and denitrification in upper groundwater		
$NH_{4,ex}$	Excess ammonium input to the soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
$NO_{3,ex}$	Excess nitrate input to the soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
N_{ex}	Excess nitrogen input to the soil	$kg \cdot ha^{-1} \cdot yr^{-1}$
$NH_{4,le}$	Ammonium leaching from the unsaturated zone	$kg \cdot ha^{-1} \cdot yr^{-1}$
$NO_{3,le}$	Nitrate leaching from the unsaturated zone to groundwater	$kg \cdot ha^{-1} \cdot yr^{-1}$
N_{le}	Nitrogen leaching from the unsaturated zone to groundwater	$kg \cdot ha^{-1} \cdot yr^{-1}$
fr_{ro}	Runoff (lateral flow) fraction	–
$N_{de,gw}$	N losses via denitrification in upper groundwater	$kg \cdot ha^{-1} \cdot yr^{-1}$
$fr_{de,gw}$	Denitrification fraction for upper groundwater	–
$NH_{4,if,gw}$	Ammonium inflow to upper groundwater	$kg \cdot ha^{-1} \cdot yr^{-1}$
$NO_{3,if,gw}$	Nitrate inflow to upper groundwater	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{if,gw}$	Nitrogen inflow to upper groundwater	$kg \cdot ha^{-1} \cdot yr^{-1}$
Inflow and denitrification in ditches and outflow to surface waters		
$NH_{4,if,di}$	Ammonium inflow to ditches by runoff from terrestrial systems	$kg \cdot ha^{-1} \cdot yr^{-1}$
$NO_{3,if,di}$	Nitrate inflow to ditches by runoff from terrestrial systems	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{if,di}$	Nitrogen inflow to ditches by runoff from terrestrial systems	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{ni,di}$	Nitrification flux in ditches	$kg \cdot ha^{-1} \cdot yr^{-1}$
$N_{de,di}$	N loss via denitrification in ditches	$kg \cdot ha^{-1} \cdot yr^{-1}$
$fr_{de,di}$	Denitrification fraction for ditches	–
$N_{of,di}$	Nitrogen outflow from ditch to large surface waters	$kg \cdot ha^{-1} \cdot yr^{-1}$
Phosphorus fluxes		
$P_{ox,t}$	Reactive P pool size at time step t	$mmol \cdot kg^{-1}$
$P_{acc,t}$	P accumulation in soil at time step t	$mmol \cdot kg^{-1}.d^{-1}$
$P_{surplus}$	The difference between P inputs and outputs	$mmol \cdot kg^{-1} \cdot d^{-1}$
Δt	Time step for P balance calculations	d^{-1}
P_{in}	The soil input of P from manure, fertilizer, deposition and seepage	$kg \cdot ha^{-1} \cdot yr^{-1}$
P_{up}	The net P uptake by the crop	$kg \cdot ha^{-1} \cdot yr^{-1}$
$P_{le,t}$	The total loss of P by leaching and runoff	$kg \cdot ha^{-1} \cdot yr^{-1}$
ΔP_{org}	the change in organic P due to net mineralization of peat soils	$kg \cdot ha^{-1} \cdot yr^{-1}$
z, ρ, M, γ	Parameters to convert $kg \cdot ha^{-1} \cdot yr^{-1}$ to $mmol \cdot kg^{-1} \cdot d^{-1}$, z is the thickness of the soil layer (m), ρ is the bulk density ($kg \cdot m^{-3}$), M the molar weight of P ($mg \cdot mmol^{-1}$) and γ is a time conversion factor (1/365)	–
C_{pt}, C_{pi}	The total P (Pt) and ortho P (Pi) concentration of soil solution	$mg \cdot l^{-1}$
L, L_m	Amount of adsorbed (labile) P as well as the adsorption maximum (L_m)	$mmol \cdot kg^{-1}$
K_L	The Langmuir affinity constant	$m^3 \cdot kg^{-1}$
$P_{diss,L}$	The P flux from soil solution to the stable adsorbed P pool	$mmol \cdot kg^{-1} \cdot d^{-1}$
P_{loss}	The loss of P by leaching or runoff	$mmol \cdot kg^{-1} \cdot d^{-1}$
μ_{DisS}, μ_{SDis}	The rate constant for the transfer from soil solution to a stable pool (μ_{DisS}) and vica versa (μ_{SDis})	d^{-1}
K_F, n	Freundlich parameters: the Freundlich constant and exponent of the stable pool	$mmol \cdot kg(soil)^{-1} (mg \cdot l(water)^{-1})^{-n}$, –
S	The size of the stable adsorbed P pool	$mmol \cdot kg^{-1}$

Table 2 (*continued*)

Symbol	Explanation	Unit
Metal fluxes		
ΔMe	Change in total metal (Cu, Zn, Cd, and Pb) pool	$g \cdot kg^{-1} \cdot yr^{-1}$
Me_{in}, Me_{up}, Me_{le}	Total input, uptake and leaching of metals	$g \cdot kg^{-1} \cdot yr^{-1}$
K_{sp}, n	Two soil plant transfer coefficients for metal uptake	$mg \cdot kg^{1-n}$, –
Me_{crop}	The metal concentration in the harvested crop	$mg \cdot kg^{-1}$
$\alpha_1 .. \alpha_3$, β_0 … β_3	Regression coefficients to estimate K_{sp}	–
Water fluxes		
PE	Precipitation excess	$mm \cdot yr^{-1}$
fr_{int}	Interception fraction for precipitation	–
P	Precipitation	$mm \cdot yr^{-1}$
E_s	Soil evaporation	$mm \cdot yr^{-1}$
$E_{t,ref}$	Transpiration rate for a reference situation	$mm \cdot yr^{-1}$
fr_{tr}	Transpiration fraction, crop specific	–

fertilizers and those from housing systems are subsequently uses as input of the atmospheric transport model OPS (Sauter et al., 2015; Wichink Kruit et al., 2017) to assess N deposition on (non-)agricultural systems. Measures can affect animal numbers, excretion factors or emission fractions or soil properties affecting the element fate in soil and emissions to air and water systems (Fig. 1).

2.2. Calculation of element flows

We outline the main element flows here while details of the calculation procedures are given in the supplementary material part S1 (further denoted as S1, same holds for S2-S5). To give an impression of the input data needed for the model, the list of symbols used in the process descriptions in INITIATOR is given in Table 2.

2.2.1. Excretion and housing emissions

Excretion of N and P is described by a multiplication of: (i) an excretion factor (kg N or kg P per animal per year) for >60 animal categories with (ii) the number of animals in each category, based on a geographically explicit census database (GIAB; Van Os et al., 2016) with data for each farm. Similarly, for base cations and heavy metals, the excretion is calculated by a multiplication of: (i) a manure production factor (kg manure per animal per year) for various animal categories (22 for base cations and 10 for heavy metals) with (ii) the number of animals in each category, and (iii) the estimated base cation and heavy metal content in each type of manure. More details on the animal categories and element contents are given in S2.

The NH_3, NO_x and N_2O emissions from housing and manure storage are calculated by multiplication of the N excretion with an emission factor (kg NH_3, NO_x or N_2O per kg N excretion) for either major animal manure types (NO_x and N_2O) or >200 housing systems (NH_3), following the procedure as used for the national emission reporting, NEMA (Velthof et al., 2012). In case of NH_3, a distinction is also made in emission factors for housing systems and for manure storage systems. The CH_4 emission from agriculture (enteric fermentation and manure management) is calculated according to NEMA (Lagerwerf et al., 2019). The CH_4 emission from enteric fermentation in animals is derived from an emission factor (kg CH_4 per animal per year), depending on the gross energy intake per animal category, multiplied by the corresponding livestock numbers. The CH_4 emission from manure management is calculated by a multiplication of an emission factor CH_4 per volume manure produced (kg $CH_4 \cdot m^{-3}$) with the manure volume in the storage system. The manure volume is calculated by a multiplication of a manure excretion factor (kg per animal per year) per animal category with the number of animals and the reciprocal of the bulk density of the manure ($m^3\ kg^{-1}$); (see S1 for details of the calculation procedure).

2.2.2. Production and distribution of nutrients and heavy metals in manure, biosolids and fertilizers

The production of carbon, nutrients and metals in manure is calculated at farm level by a multiplication of the animal numbers with the excretion per animal, using animal numbers and grazing hours from GIAB and annual excretion rates from WUM/CBS (CBS, 2019). The excreted manure in housing and manure storage systems was corrected for gaseous N losses (see above) and for manure export/processing before application. Manure export/processing are on national data until 2010 and thereafter on farm based census data aggregated to agricultural regions. Leaching from housing and manure storage systems was assumed negligible. At farm level the excreted manure during grazing was applied first on grassland. Next, housing manure is applied to the fields per farm, where the manure dose is not allowed to exceed the maximum doses given by the manure legislation. We distinguished cattle, pig and poultry manure. If farms produce more manure than legally can be applied on their fields, the excess is distributed over farms within a nearby region (we distinguish 239 agricultural regions of about 7000 ha each) that have the capacity to utilise more manure. If an excess exists in a region, the excess is distributed over surrounding regions with a shortage of manure while accounting for distance and the degree of acceptance of a given manure type. For the situation that there is more manure than legally can be applied over all regions together, implying a violation of the legal national N or P application limits, the excess manure is distributed over maize fields in areas with a manure excess in proportion to the excretion rates per region. The application of base cations and metals by manure is derived by using the weighted average P/base cation and P/metal ratios in the manure for the corresponding region (see S1 for details of the calculation procedure).

Next to the application of animal manure, biosolids such as compost and sewage sludge, are applied. Biosolids amounts are based on national data until 2015 and thereafter on farm statistics on produced and sold amounts of biosolids according to census data and distributed equally over all arable and maize fields of particular farms that accept biosolids.

The inorganic N and P fertilizer dose per field is subsequently calculated by the difference between the legal standards (crop and soil specific) for a particular year and the calculated effective nutrient dose of applied animal manure and biosolids. This was done by assuming balanced fertilization as the common agricultural practice in the Netherlands, which is an appropriate assumption considering that the calculated total N fertilizer amount fitted well with national statistics of N and P fertilizer inputs (see Section 5.1). The input of base cations and metals by fertilizers is calculated from the mix of applied fertilizer types for a particular year (according to national statistics on sold amounts) and generic base cations and metals contents per fertilizer type.

2.2.3. Net uptake of nutrients and heavy metals

The yearly net uptake of nutrients and cations is calculated on the basis of yearly crop yields per crop type (23 main crop types) and region (province) for the period 2000–2020 and yearly element contents per crop type, except for N, Cd and Zn. The yearly N content is calculated as a maximum value at optimal N supply corrected for the effective N input via fertilization, grazing animal, fixation and deposition. Element uptake by residues is ignored since these largely remain on the field after harvest. Element contents for other main nutrients, Na, Cl, Pb and Cu are fixed for each crop. The contents of Cd and Zn are derived from soil contents, using soil-plant relationships (De Vries et al., 2008a), accounting for differences in soil texture, organic matter content and pH, discussed in more detail in S1.

2.2.3.1. Soil emissions of CH_4 and CO_2. The methane (CH_4) emission from soils is set at a constant value with the exception of natural grasslands that emit CH_4 at a rate depending on the groundwater level (Van den Pol-van Dasselaar et al., 1999). The CO_2 emission from soils is calculated as the sum of the net C pool change in mineral soils, C release due to peat oxidation and C release due to liming to counteract acidification of agricultural soils.

The net C pool change in mineral soils is calculated as the difference in net annual C input by animal manure, compost and crop residues, being a fraction of total C input remaining after 1 year (each with its own humification coefficient) and the annual C release due to organic matter decomposition represented by a first order decay process with one C pool. The net C release due to oxidation of peat is calculated by a multiplication of: the annual lowering of the peat soil as a function of the groundwater level with the bulk density of the peat and the fraction of organic carbon in the peat. The net C release due to liming is calculated by predicting the liming requirement from the fate of added nitrogen and base cations in manure and fertilizer. Details for all these processes are given in S1.

2.2.3.2. Soil emissions of NH_3, N_2O, N leaching and runoff. The N fluxes from and in soils are calculated with a consistent set of simple linear equations (De Vries et al., 2003b). First the total N input to the soil is calculated as the sum of inputs by animal manure, fertilizer, atmospheric deposition and biological N fixation. The fate of N in soils is calculated as a sequence of occurrences in the order ammonia emission, followed by uptake, mineralization/immobilization, nitrification and denitrification in the soil. The mineralization/immobilization is related to the net C accumulation or release of carbon, multiplied by the N/C ratio of the soil. All other N transformation processes are calculated as a linear response to N inputs. Emissions of NH_3 are calculated by a multiplication of the N inputs by manuring, fertilization and grazing with specific N emission fractions for these inputs derived from the national ammonium based NH_3-inventory model NEMA (Velthof et al., 2012). As mentioned, the N uptake is calculated as a function of effective N input, land use, soil type and hydrological regime. The N surplus (all N inputs minus N uptake minus net N immobilization) determines the loss to the environment. The leaching loss is partitioned to surface water and to groundwater by multiplying the total leaching loss with a runoff fraction (including all pathways like runoff, shallow leaching and drainage) and a leaching fraction (the remaining fraction). The N_2O and NO_x emission are calculated as fixed fractions of the nitrification and denitrification flux respectively. Nitrogen losses occurring in surface and groundwater bodies are also calculated (see S1).

2.2.3.3. Soil accumulation and leaching and runoff of P. The accumulation or release of P is calculated using a mass balance approach, subtracting P uptake, leaching to groundwater and runoff to surface water from the P input by fertilizer, manure and deposition. The root zone is divided in three layers of respectively 0–5 cm, 5–20 cm and 20–50 cm depth. Below 50 cm, an average background dissolved P concentration per unique combination of soil type, land use and geohydrology (called Hydrological Response Unit, HRU is used, based on about 1060 measurements at depths between 5 and 2 m between 1970 and 2010 available in the DINO repository (Data and Information on the Dutch Subsurface, www.dinoloket.nl/en). Data were upscaled by kriging of log transformed concentrations in combination of bias adjusting of the kriging variance and final back transformation (Brus et al., 2010).

The P mineralization rate is derived by multiplying the C mineralization rate with an average P/C ratio. P uptake is calculated by multiplying the yield with a constant crop P content, as described before. P leaching is described by multiplying the water flux with a total dissolved (inorganic and organic) P concentration. The change in the inorganic P concentration in soil solution is determined by an adsorption/desorption process on a labile Pool (L) using a Langmuir equation as well as a rate-limited P transfer to and from a stable pool (S) using a Freundlich equation (Van der Zee, 1988). The maximum amounts of P in the labile and stable pool were set equal to 1/6 and 1/3 of the amount of oxalate extractable Al and Fe respectively (Van der Zee, 1988; Schoumans and Groenendijk, 2000). Details on the process descriptions are given in Van der Salm et al. (2016) and in S1.

Soil acidification: the change in soil pH due to acidification is ignored assuming that agricultural soils are limed to counteract acidification. Lime requirements are assessed from the calculated loss of base cations by subtracting base cation crop uptake and losses to water from the input by fertilizer, manure and deposition. The leaching of base cations was set equal to the leaching of major anions, including sulphate (SO_4), nitrate (NO_3), chloride (Cl) and bicarbonate (HCO_3). The leaching of SO_4 and Cl was set equal to the input minus crop removal (SO_4 only), assuming no soil interaction, whereas HCO_3 leaching was calculated by multiplying a HCO_3 concentration (dependent on soil pH) with the precipitation surplus.

Soil accumulation and leaching and runoff of heavy metals: similar to P, metal accumulation is calculated by using a mass balance approach, subtracting metal uptake and metal leaching and runoff to water from the metal input by fertilizer, animal manure, other organic sources and atmospheric deposition. Metals included are copper (Cu), Zinc (Zn), lead (Pb) and cadmium (Cd). Copper and Zn are mainly supplied by animal manure, Pb input is mainly due to deposition whereas inorganic fertilization is the largest source of Cd. The possible impact of soil erosion was neglected since most sites are located in flat areas (De Vries et al., 2004). The net crop uptake rate was derived by multiplying crop yield by the metal content in that crop, being either fixed (Pb and Cu) or depending on the metal concentration in the soil and soil properties, i.e. organic matter content, clay content and pH (Cd and Zn). The leaching rate of metals from the topsoil was derived by multiplying the precipitation surplus with a dissolved metal concentration in soil solution, derived from the reactive metal content, using a Freundlich equation, with the Freundlich adsorption constant depending on organic matter content, clay content and pH (De Vries et al., 2008a). Details on the process descriptions are given in S1.

3. Model application

3.1. Model inputs, model parameters and model evaluation

3.1.1. Schematization of the study area and data derivation approach

INITIATOR was applied to all agricultural land of the Netherlands to estimate the fate of organic matter, nutrients and metals of the rooting zone. Parcels from one farm with similar crop type and soil type (sand, loess, clay, peat) and located in the same agricultural region were combined to one spatial unit. At this level the manure and fertilizer distribution and the ammonia emissions from the fields were calculated. Soil related entities the (aggregated) parcels were assigned to 28,610 unique so-called Hydrological Response Units (HRU) (Van der Bolt et al., 2020). The HRUs consist of multiple spatial grid cells with a resolution of 250 m × 250 m with the same land-use (grass, maize and arable land), soil type, and geohydrological conditions for which soil properties, geohydrological properties and water balances are available. In brief, the inputs by fertilizer and manure and related emissions of NH_3 were calculated at parcel level (BRP, 2017) and then aggregated to HRUs at which levels the soil fluxes. Emissions of NH_3 and CH_4 from housing systems were calculated for all individual farms, based on georeferenced information of the housing systems (GIAB; Van Os et al., 2016). All fluxes in and from soil were both calculated and presented at HRU level.

This section gives an overall description of the derivation of and ranges on input data used (details in S2). The databases used to derive inputs for these plots are listed in Table 3.

Model inputs and model parameters vary as a function of land use (grassland, maize, arable), soil type (sand, loess, clay and peat) and wetness class (see also Tables 3 and 4) and are allocated over the HRU plots using a geostatistical interpolation where needed. Clayey and sandy soils are subdivided in calcareous and non-calcareous soils since pH largely affects the uptake and leaching of metals. The wetness classes were derived from the mean highest groundwater (MHW) table of the 1:50,000 soil map with the classes (i) wet and poorly drained (MHW < 40 cm), (ii) moist and moderately drained (MHW 40 to 80 cm) and (iii) dry and well drained (MHW > 80 cm).

3.1.2. Model inputs

Overall ranges in input data, used to estimate inputs, uptake, accumulation and leaching of elements (C, N, P, Ca, Mg, K, Na, Cl, Cu, Zn, Pb) in agricultural soils are given in Table 4, for manure inputs and in Table 5 for all other input data, with their dependence on hydrological response unit (combination of soil type, land use and groundwater level) and crop type. In addition, data on emissions of NH_3, N_2O, NO_x, N_2 and CH_4 from housing systems, animals and animal manure are also given in Table 4.

Table 3
Geographic databases used with their temporal and spatial resolution.

Aspect	Database	Temporal resolution	Spatial resolution	Availability/description
Animal numbers and location per housing system	RVO/CBS/GIAB, census data	Yearly (2000–2019)	Point level, in total ranging from 97,000 farms in 2010 to 54,000 in 2018, with their location (from 2015 at stable level)	Private, only available for research institutes involved. (GIABplus; Van Os et al., 2016) (GIAB; Gies et al., 2015).
Manure application technique and grazing time	RVO/CBS/GIAB, census data	Yearly (2000, 2005, 2015, 2016) (Used for 2001–2004: 2000 2006–2007:2005 2008–2014: 2015)	Farm level	Private
Fertilizer inputs	CBS/NEMA	Yearly (2000–2019)	National level, downscaled to BRP level	Public at national scale
Land use	BRP (2017)	Yearly: 2002, 2004, 2006–2019 (Used for 2000, 2001: 2002 2003: 2004 2005: 2006)	Parcel level ranging from 0.1 – to 7 ha (90 %), with a mean of ca. 2.3 ha and in total ca. 800,000 parcels.	Partly public[a] (only the location and crop type, but not the linkage with farms) but not open for each year
Crop yields	CBS, census data	Yearly (2000–2019)	Provincial yields for all (ca. 25) CBS crop types, in total 12 provinces.	Public (CBS, 2003, 2020)[c]
Soil types and groundwater levels; soil physical and chemical status	Dutch Soil Information System (SIS)	Fixed	Polygons; original scale1:50,000	Public (De Vries, 1999; Kroon et al., 2001; De Vries et al., 2003a; Van der Bolt et al., 2016)
Soil types in view of Manure Act	Digital soil map	Fixed (2016)	Parcel level	Public[b]
P-AL and Pw status	RVO census data	Yearly (2013–2019)	Parcel level	Private
Derogation status	RVO census data	Yearly (2006–2019)	Farm level	Private (cf. Hooijboer et al., 2017)
Hydrological fluxes	LWKM model results	Fixed (30 years mean balance; 1980–2010)	Hydrological response Unit (in total 28,610).	Private, only available for research institutes involved.

[a] https://www.pdok.nl/introductie/-/article/basisregistratie-gewaspercelen-brp-.
[b] http://www2.hetlnvloket.nl/mijndossier/grondsoortenkaart/grondsoorten15.html.
[c] https://opendata.cbs.nl/statline/#/CBS/nl/dataset/7100oogs/table?fromstatweb.

3.1.2.1. Element inputs. Geo-referenced data for the inputs of C, N, P, base cations and metals via animal manure were based on yearly CBS/GIAB data of animal numbers at farm level for the years 2000–2018 combined with average excretions of N and P for major animal categories according to WUM/CBS (see above), average concentrations of base cations (Ca, Mg, K, Na and Cl) in manure (for sows, solid poultry manure and horsed form Rinsema (1985) and Mooij (1996) for all other manure types) and of metals (Cu, Zn, Pb and Cd) in manure (Römkens and Rietra, 2008; Deltares, 2018). Ranges for the various input data are given in Table 4.

Nitrogen, sulphur, base cation and metal deposition data for the period 2000–2020 were derived from results of the atmospheric transport models OPS at a 1 km × 1 km grid for N and S (RIVM, 2020), a 5 km × 5 km grid for the base cations Ca, Mg, K and Na (Van Jaarsveld et al., 2010), and a 10 km × 10 km grid for metals (Bleeker, 2004). Ranges in Table 5 refer to the whole period. Phosphorus deposition was set at 0.5 kg P ha^{-1} yr^{-1} (De Vries et al., 2019). Cl deposition was derived from the Na deposition using a fixed Na to Cl ratio in sea water of 0.858 (Van Jaarsveld et al., 2010).

Element inputs by the application of biosolids were based on yearly national amounts of the included biosolid types for the years 2000–2018 (CBS, 2020) combined with average concentrations of base cations and Cl and metals (pers. comm. Aterro; attero.nl for VGF compost, pers. comm. BVOR; bvor.nl, for green compost and CBS (https://www.cbs.nl/nl-nl/cijfers/detail/83400NED for sewage sludge) in these biosolids. Biological N fixation was estimated as a function of land use, i.e. 25 kg ha^{-1} yr^{-1} for grassland, 15 kg ha^{-1} yr^{-1} for arable land and 8 kg ha^{-1} yr^{-1} for maize land. The estimate for maize is an average value for the fixation by free living N fixing bacteria in all land use types. The additional N fixation in grassland is due to N fixation by clovers, whereas the additional input in arable land is due to fixation by legumes (De Vries et al., 2003b).

3.1.2.2. Element uptake. Annual crop yields, used to estimate crop nutrient and metal uptake were derived at province level from census data, as derived from extensive sample surveys of farms per crop and region for the period 2000–2020 (CBS, 2020). Element contents per crop were derived from national crop content databases, based on analyses done by agricultural laboratories (N and P), on literature data (Ca, Mg, K, Na, Cl; Houba and Uittenbogaard (1994) Pb and Cu) or derived from soil metal contents (Cd and Zn) as described before. The Pb and Cu contents of crops are median values in two large datasets as described in De Vries et al. (2008b).

3.1.2.3. Soil contents. Data for soil C, N, P and metal (Cd, Cu, Pb and Zn) contents and of Fe and Al hydroxides for major soil types in each HRU plot were derived from the Dutch Soil Information System including >6000 soil profiles corresponding to soil mapping units. Data on soil Cd and Zn contents were derived from approximately 3000 individual soil samples in provincial and national monitoring networks (Finke et al., 2001). Soil properties (De Vries et al., 2008a) such as organic matter content, clay content and pH, affecting relationships between metals in soil and plant, were all derived from the national soil database (Dutch Soil Information System).

3.1.2.4. Hydrological fluxes. Each HRU plot has a detailed hydrological schematization with water fluxes in different soil layers down to 5 m below the soil surface, based on calculations with the SWAP model (Kroes et al., 2000). For this study we only focus on the water and nutrient fluxes entering and leaving the topsoil. Water fluxes (including precipitation, leaching, runoff, evaporation, seepage) represent the 30-year averaged (1981–2010) hydrology and are regionally normalized.

3.1.3. Model parameters

Overall ranges in major parameters used to estimate emissions, transformations and soil retention or release of elements in agricultural soils, their dependency on animal category, land use (crop category), soil type and groundwater level are given in Table 6.

3.1.4. Housing emissions

Housing emission fractions for N_2O, NO_x, N_2 and CH_4 were based on the yearly reported emission inventory based on the NEMA model per animal manure type, while emissions fractions of NH_3 were derived for >200 housing types (Van Bruggen et al., 2022) Ranges in emission fractions of NH_3, N_2O, NO_x, N_2 and CH_4 from housing systems or in the field for all included animal categories vary widely (Tables 5, S2).

3.1.4.1. Carbon transformation parameters. Data on humification constants for organic inputs were based on Velthof et al. (1999) and vary between 0.3 and 0.5 for animal manure and 0.01–0.8 for biosolids. Humification constants for crop residues were based on Velthof and Kuikman (2000) and vary between 0.21 and 0.33 (Table 6). Decomposition rate constants for the organic carbon in the soil were calibrated on the basis of reported SOC trends in topsoil of grassland and of arable land in the Netherlands during the period 1984–2004 (Reijneveld et al., 2009). The trends are based on

Table 4
Ranges (5 %–95 %) in parameters affecting inputs of elements (C,N, P, Ca, Mg, K, Na, Cl, Cu, Zn, Pb) by animal manure and biosolids for major animal categories and biosolid categories and emissions of NH_3, N_2O, N_2 and CH_4 from housing systems for the year 2015. Details of the data per category for the years 2000–2019 are given in S2.

Parameter	Explanation	Unit	Ranges		
			Cattle[a]	Pigs	Granivores[b]
Livestock related parameters					
n_{acat}	Animal number[a]	n·category^{-1}	3–359	11–4481	60–150,000
$M_{ex,acat}$	Produced fresh manure[a]	kg·animal^{-1}·yr^{-1}	2416–27,000	1020–4500	9–268
D_{acat}	Manure density[a]	kg·m^{-3}	700–1005	600–1040	570–1020
N_{ex}	N excretion[a]	kg N·animal^{-1}·yr^{-1}	16.4–141.6	13.9–29.5	0.4–6.0
P_{ex}	P excretion[a]	kg P·animal^{-1}·yr^{-1}	5.4–47	5.8–14	0.2–3.1
NH_4	TAN (NH_4) content in manure[a]	%	51–73	64–71	64–77
$CH_{4,acat}$	CH_4 animal emission rate[b]	kg CH_4·animal^{-1}·yr^{-1}	7–131	1.5–1.5	0
$frCH_{4,em\_h}$	CH_4 loss from housing systems	kg CH_4·m^{-3} manure	0.181–36	8.5–18	0.015–0.44
hc_{man}	Humification constants for manure		0.35–0.5	0.3–0.3	0.44–0.5
Housing emission fractions[a]					
$frNH_{3,em\_hs}$	NH_3 emission fraction housing	–	0.059–0.44	0.084–0.49	0.003–0.58
$frNH_{3,em\_st}$	NH_3 emission fraction storage	–	0.003–0.05	0.003–0.03	0.003–0.08
$frN2O_{em\_hs}$	N_2O emission fraction housing	–	0.002–0.011	0.011–0.002	0.002–0.005
$frNO_{x,em\_hs}$	NO_x emission fraction housing	–	0.002–0.005	0.005–0.002	0.002–0.005
$frN_{2,em,h}$	N_2 emission fraction housing	–	0.020–0.025	0.025–0.02	0.020–0.025
Element contents in manure[c]					
OM_{man}	OM content in manure	g·kg^{-1}	71–164	27–138	93–424
Ca_{man}	Ca content in manure	g·kg^{-1}	1.5–2.9	2.7–3.7	5.6–21.8
Mg_{man}	Mg content in manure	g·kg^{-1}	0.7–2.5	0.7–1.4	1.3–3.9
K_{man}	K content in manure	g·kg^{-1}	4.4–11.1	3.6–6.8	4.8–16
Na_{man}	Na content in manure	g·kg^{-1}	0.6–1.4	0.5–0.8	0.7–5.6
Cl_{man}	Cl content in manure	g·kg^{-1}	2–3	1.5–1.9	1.7–8
S_{man}	S content in manure	g·kg^{-1}	0.2–0.6	0.3–0.5	0.4–3.2
Cu_{man}	Cu content in manure	mg·kg^{-1}	50–130	397–398	50–138
Zn_{man}	Zn content in manure	mg·kg^{-1}	190–300	644–966	300–307
Pb_{man}	Pb content in manure	mg·kg^{-1}	3.5–11.8	3.9–18.4	4.4–11.8
Cd_{man}	Cd content in manure	mg·kg^{-1}	0.2–0.2	0.3–0.56	0.15–0.2

Amounts biosolids OMin		kton	Spent lime	VGF compost	Green compost	Champost	Sewage sludge
			18	58	137	119	7
Element contents in biosolids[c]							
N_{bs}	N content in biosolids	g·kg^{-1}	4.2	3.5	2.9	3.4	8.2
P_{bs}	P content in biosolids	g·kg^{-1}	31	4.4	3.3	4.8	23
Ca_{bs}	Ca content in biosolids	g·kg^{-1}	490	9.5	18	29	–
Mg_{bs}	Mg content in biosolids	g·kg^{-1}	22	2.7	2.2	1.9	–
K_{bs}	K content in biosolids	g·kg^{-1}	2.0	3.5	3.2	3.2	–
Na_{bs}	Na content in biosolids	g·kg^{-1}	0	–	–	0.6	–
Cl_{bs}	Cl content in biosolids	g·kg^{-1}	–	0.85	0.54	1.2	0.24
S_{bs}	S content in biosolids	g·kg^{-1}	25	2.0	1.8	17	–
Cu_{bs}	Cu content in biosolids	mg·kg^{-1}	133	118	85	100	535
Zn_{bs}	Zn content in biosolids	mg·kg^{-1}	558	518	438	223	1395
Pb_{bs}	Pb content in biosolids	mg·kg^{-1}	31	147	127	15	116
Cd_{bs}	Cd content in biosolids	mg·kg^{-1}	5.2	1.1	1.5	0.52	1.4

[a] This includes cows, horses, sheep and goats.
[b] This includes poultry, turkeys, ducks, mink and rabbits.
[c] All data are time dependent, except for element contents in manure and biosolids which are assumed to be time invariant.

a data base with ~2 million SOC data from farmers' fields and are divided in changes per land use type (grassland, maize land and arable land) and soil type (sand, clay, loess and peat). The rate constants were derived by first assuming a steady state between the input of effective organic matter (C_{in}) in the year 2000 and the current (period around 2000) carbon pool in the top soil (Cpool), i.e. $k = C_{in}/C_{pool}$, and then correcting the values such that the organic C sequestration rate in the year 2000 equals the changes as published by (Reijneveld et al., 2009) for the various land use types and soil types. Rate constants thus derived ranged between 0.008 and 0.05 per year (Table 6).

3.1.4.2. Nitrogen transformation data. Model parameters for NH_3 emission due to grazing, animal manure application and the application of biosolids were set equal to those used in the NEMA model, used for the yearly reported emission inventory (see above). The NH_3 emission fractions for animal manure application (and also for housing) were related to the content of total ammonia nitrogen (TAN). Fractions of the TAN in excreted manure during housing and grazing were also based on NEMA (see above).

The NH_3 emission fractions due to animal manure application depend on the used application technique (ranging from injection to above ground application) and crop type (grass or arable). The NH_3 emission fractions due to fertilizer application depend on the used mix of fertilizer types at the national scale (one value per year for the country as a whole).

Nitrogen uptake fractions, nitrification fraction in soils, denitrification fractions in soil, upper groundwater and ditches and fraction relating total nitrification and denitrification to N_2O emissions are all based on literature, as summarized in (De Vries et al., 2003b).

3.1.4.3. Phosphorus and metal sorption constants. Langmuir adsorption constants (K_P, in $m^3\ g^{-1}$) for various soils have been derived from field data from Koopmans and van der Salm (2011), showing increasing values from peat (2400 $m^3\ kg^{-1}$) up to sandy soils (6530 m^3 kg), as further described in as described in Van der Salm et al. (2016). Metal sorption constants, i.e. the soil plant transfer constant for metals and the Freundlich coefficient for metals were related to soil properties (clay and organic matter content and soil pH).

Table 5
Ranges (5 %–95 %) in model inputs (year 2015), soil data, initial values of soil concentrations (state variables) and hydrological fluxes used to estimate inputs, uptake, accumulation and leaching of elements (C,N, P, Ca, Mg, K, Na, Cl, Cu, Zn, Pb) in agricultural soils. The dependency of each input on location, indicated by HRU, crop category and time is also indicated. Details of the data are given in S2.

Parameter	Explanation	Unit	HRU[a]	Crop type	Time	Range
Fertilizer input						
N_{fert}	N fertilizer input	kg ha^{-1} yr^{-1}	x		x	0–273
P_{fert}	P fertilizer input	kg ha^{-1} yr^{-1}	x		x	0–8.8
Ca_{fert}	Ca fertilizer input	kg ha^{-1} yr^{-1}	x		x	0.0007–46
Mg_{fert}	Mg fertilizer input	kg ha^{-1} yr^{-1}	x		x	0–25
K_{fert}	K fertilizer input	kg ha^{-1} yr^{-1}	x		x	0–19
S_{fert}	S fertilizer input	kg ha^{-1} yr^{-1}	x		x	0–2.7
Na_{fert}	Na fertilizer input	kg ha^{-1} yr^{-1}	x		x	0–0.5
Cl_{fert}	Cl fertilizer input	kg ha^{-1} yr^{-1}	x		x	0–12
Cu_{fert}	Cu fertilizer input	g ha^{-1} yr^{-1}	x		x	0.001–3.5
Zn_{fert}	Zn fertilizer input	g ha^{-1} yr^{-1}	x		x	7.2–82
Pb_{fert}	Pb fertilizer input	g ha^{-1} yr^{-1}	x		x	0.01–25
Cd_{fert}	Cd fertilizer input	g ha^{-1} yr^{-1}	x		x	0.0002–1.4
Deposition						
N_{dep}	N deposition	kg ha^{-1} yr^{-1}	x		x	17–27
P_{dep}	P deposition	kg ha^{-1} yr^{-1}				0.5–0.5
Ca_{dep}	Ca deposition	kg ha^{-1} yr^{-1}	x			1.8–4.1
Mg_{dep}	Mg deposition	kg ha^{-1} yr^{-1}	x			0.46–4.8
K_{dep}	K deposition	kg ha^{-1} yr^{-1}	x			1.5–4
S_{dep}	S deposition	kg ha^{-1} yr^{-1}	x			3.9–6.7
Na_{dep}	Na deposition	kg ha^{-1} yr^{-1}	x			6.6–79
Cl_{dep}	Cl deposition	kg ha^{-1} yr^{-1}	x			12–143
Cu_{dep}	Cu deposition	g ha^{-1} yr^{-1}	x			26–55
Zn_{dep}	Zn deposition	g ha^{-1} yr^{-1}	x			43–102
Pb_{dep}	Pb deposition	g ha^{-1} yr^{-1}	x			3.5–7.2
Cd_{dep}	Cd deposition	g ha^{-1} yr^{-1}	x			0.39–0.89
Fixation						
N_{fix}	N fixation	kg N ha^{-1} yr^{-1}		x		2–9.8
Crop yield and element contents in crops				x		
Y_{crcat}	Crop yield	ton ha^{-1} yr^{-1}	x	x	x	7.6–20
$N_{ct,crcat}$	N content in crop	g kg^{-1}		x	x	6–29.8
$P_{ct,crcat}$	P content in crop	g kg^{-1}		x	x	0.95–4.3
$S_{ct,crcat}$	S content in crop	g kg^{-1}		x		1.1–2.9
$Ca_{ct,crcat}$	Ca content in crop	g kg^{-1}		x		0.6–4.8
$Mg_{ct,crcat}$	Mg content in crop	g kg^{-1}		x		0.9–2.9
$K_{ct,crcat}$	K content in crop	g kg^{-1}		x		4.7–33
$Cu_{ct,crcat}$	Cu content in crop	mg kg^{-1}		x		3.9–12
$Zn_{ct,crcat}$	Zn content in crop	mg kg^{-1}		x	x[c]	0.2–129
$Pb_{ct,crcat}$	Pb content in crop	mg kg^{-1}		x		0.1–2.3
$Cd_{ct,crcat}$	Cd content in crop	mg kg^{-1}		x	x[c]	0.01–0.3
Soil data peat[b]						
ρ_{om}	Density of unripened peat	kg·m^{-3}	x			158–966
fr_c	Organic matter fraction of peat	–				0.55–0.55
fr_{om}	Organic C fraction of peat-OM	–	x			0.031–0.85
Soil properties[b]						
Clay	Clay content	%	x			2.9–52
SOC	Soil organic carbon content	%	x			1.7–32
pH	Soil pH	–	x			4.4–7.4
Al + Feox	Oxalate extractable Al and Fe content in soil	mmol kg^{-1}	x			45–361
Element concentrations in soils[b]						
P_{re}	Reversibly adsorbed soil P content	mmol kg^{-1}	x			14–40
Cu_{soil}	Total Cu content in soil	mg·kg^{-1}	x			12–59
Zn_{soil}	Total Zn content in soil	mg·kg^{-1}	x			15–116
Pb_{soil}	Total Pb content in soil	mg·kg^{-1}	x			6–32
Cd_{soil}	Total Cd content in soil	mg·kg^{-1}	x			0.15–0.58
Hydrological data						
P	Precipitation	mm·yr^{-1}	x			808–891
E_s	Soil evaporation	mm·yr^{-1}	x			196–240
E_t	Transpiration	mm·yr^{-1}	x			212–324
fr_{int}	Interception fraction	–	x			278–427
fr_{ro}	Runoff (lateral flow) fraction	–	x			0.001–0.41
GHG	Groundwater level	cm	x			16–256
GLG		cm	x			77–338

[a] HRU is the hydrological response unit, being a unique combination of soil type, major land use (grass, maize and arable) and groundwater level (GHG and GLG).
[b] Soil data were derived from a geo-referenced soil database (Dutch Soil Information System).
[c] Concentrations of Cd and Zn in crop were based on soil-plant relationships with soil Cd and Zn concentrations varying in time due to simulations.

Table 6
Ranges (5%–95%) in model parameters (year 2015) used to estimate emissions, transformations and soil retention or release of elements in agricultural soils. The dependency of parameters on animal category, land use, soil type, groundwater level and time is also indicated. Details of the data are given in S2.

Parameter	Explanation	Unit	Animal category	Land use	Soil type	Groundwater level	Time	Range
Carbon transformation parameters								
k	Decomposition constant	yr^{-1}		x[a]	x[a]			0.008–0.05
hc_{cr}	Humification constants for crop residues	–		x[b]				0.21–0.33
Nitrogen transformation data								
$frNH_{3,em,a}$	NH_3 emission fraction from manure	–	x			–	x	0.0018–0.303
	NH_3 emission fraction from solid manure							0.68
$frNH_{3,em,g}$	NH_3 emission fraction from dung and urine from grazing animals	–		–	–	–		0.04
$frNH_{3,em,f}$	NH_3 emission fraction from fertilizer	–		–	–	–	x	0.037
fr_{up}	Nitrogen uptake fraction	–		x	–	–		0.26–0.48
$fr_{ni,s}$	Nitrification fraction in soil	–		x	x	x		0.89–0.99
$fr_{de,s}$	Denitrification fraction in soil	–		x	x	x		0.35–0.89
$f_{a,am}$	N availability of animal manure-N	–	x[c]		–	–	–	0.011–0.49
$f_{a,g}$	N availability of excreted N during grazing	–	–	–	–	–	–	0.15
CN	C/N ratio from C to N mineralisation				x			16–32
$fr_{de,gw}$	Denitrification fraction upper groundwater	–			x	x		0.12–0.88
$fr_{de,di}$	Denitrification fraction ditches	–			x	x		0–0.79
frN_2O_{ni}	Fraction relating total nitrification to N_2O emissions	–		–	x	x		0.014–0.022
frN_2O_{de}	Fraction relating total denitrification to N_2O emissions	–		–	x	x		0.035–0.07
Phosphorus and metal sorption constants								
K_L	Langmuir adsorption constant for P	$m^3\ kg^{-1}$		–	x	–		500–2000[d]
K_F	Freundlich Constant for irreversible P adsorption	$m^3\ g^{-1}$		–	x	–		4.5–40[e]
μ_{DisS}	Rate constant for the transfer of P from soil solution to a stable pool	(d^{-1})			x			1.4×10^{-3}
μ_{SDis}	The rate constant for the transfer from a stable pool to soil solution	(d^{-1})			x			$2–44 \times 10^{-6}$
K_{sp}	Soil plant transfer constant for metals	$mg^{1-n} \cdot kg^{n-1}$		–	x	–		Variable
K_f	Freundlich coefficient for metals	$mol^{1-n} \cdot l^n \cdot kg^{-1}$		–	x	–		Variable

[a] Decomposition rate constants are calibrated on trends in SOC constants, being land use and soil type dependent.
[b] Here the land use category actually refers to the type of crops with respect to the residues.
[c] Here the animal category actually refers to the used application technique.
[d] Values vary with soil type, i.e. 500 for peat, 1000 for sand and loess and 2000 for clay, based on in-situ field based measurements in the Netherlands (Koopmans and van der Salm, 2011).
[e] Value vary with soil type and with soil depth and the range is 5.2–38 for the depth of 0–5 cm, 4.5–40 for 5–20 cm and 4.6–34 for the depth of 20–50 cm.

3.1.5. Model evaluation

The plausibility of crucial model inputs and outputs was assessed by a comparison with independent estimates on national trends in: (i) N and P inputs, uptake and surpluses, derived from national statistics (CBS, 2020) and (ii) NH_3 emissions and N_2O emissions based on the NEMA model (Van Bruggen et al., 2022), making use of the national CBS data. In addition, predicted ammonia emissions and nitrate and phosphorus concentrations in groundwater and surface water were compared with measured data from national monitoring networks.

3.2. Integrated evaluation of agronomic measures

The agricultural sector faces the challenge of combining current efficient production with environmental goals for water, soil, biodiversity, climate and air quality as formulated in national and European directives and regulations. Numerous studies have shown that agronomic measures positively contribute to one or multiple of these goals (Young et al., 2021). We here assessed the advantages and trade-offs of five sets of measures, each designed and focused on a specific part of the nutrient cycle in farm management using the year 2015 as the base year (Table 7). Predictions were made for the year 2050. The first action taken, before evaluating the measures was to reduce the N and P input to acceptable agronomic input levels at each spatial calculation unit, since that is not everywhere the case in 2015. This was done by enhancing the manure export although it could have also been accomplished by reducing the number of livestock.

The first set of measures (Animal feeding and housing period) aims to increase the share of nutrients that contribute to the production of animal products and reduce the proportion of nutrients excreted or used for maintenance. The second set (Low emission housing and application) contains measures to reduce gaseous emissions from housing and storage facilities, grazing and field application of manure, whereas the third set (Optimized fertilization) focuses on technologies and strategies to increase the effectiveness of manure and fertilizers applied. The fourth set (Improved soil management) contains measures to reduce N and P losses to air and water, to improve crop yields and to increase carbon sequestration, whereas the fifth set (Improved water protection) focuses on specific measures reducing nutrient runoff to surface

Table 7
Overview of the evaluated measures for emission reductions (relative to the year 2015).

nr	Measure Description	NH_3	NO_3	P	N_2O	CH_4	CO_2
0	**Enhanced manure export up to acceptable agronomic input levels**	x	x	x	x	x	x
1	**Animal feeding**						
1.1	Reduced protein content	x	x	x	x		
1.2	Increased grazing time	x	x				
2	**Low emission housing and application**						
2.1	Innovative manure separation techniques; closed manure storage with thermal oxidation; improved animal breed; feed additives	x	x			x	
2.2	Improved application techniques; separate application of solid and liquid manure[a]	x	x	x	x		
3	**Improved nutrient management**						
3.1	Stringent N and P application targets[b]	x	x	x	x		
3.2	Precision fertilization (right place, time)	x	x		x		
3.3	Urease and nitrification inhibitors while using NH_4 based fertilizers	x			x		
4	**Improved soil management**						
4.1	Reduced tillage		x				x
4.2	Buffer strips grassland and arable land		x	x			
4.3	Submerged infiltration drains	x	x	x	x	x	x
5	**Improved crop management**						
5.1	Increased use of cover crops		x	x			x
5.2	Use of efficient high yield crop varieties	x	x		x		
5.3	More leguminous (N fixing) crops		x	x	x		

[a] Note that manure separation also requires an adequate housing management of the farmer.
[b] Here the animal numbers will also be affected when manure export is not increased.

waters. The last set of measures (optimized crop management) aims for further diversification by using more leguminous (N fixing) crops and optimized crop rotation schemes. The identification and quantification of the effect of the included measures on model parameters were based on Lesschen et al. (2020).

Animal feeding (set 1) contains measures that reduce the protein content in the feed ration for dairy cows from 145 $g \cdot kg^{-1}$ to 120 $g \cdot kg^{-1}$ (Vellinga et al., 2013) and the P content from 3.3 to 3.2 $g \cdot kg^{-1}$, whereas the productivity slightly increased (1.5 %) in 2050. For pigs and poultry no ration adaptions were assumed, except that benzoic acid was added for pigs in order to mitigate the NH_3 emission. As a consequence of these changes for dairy cows, the excreted nutrients decline with 9 % for N and with 4 % for P. For pigs and poultry the N and P excretion remain unchanged. Excretion changes due to ration and productivity changes where based on GLEAM calculations (MacLeod et al., 2013). In addition, the measures assume that all dairy cows will graze outside the stable for 3600 h a year whereas all poultry and pigs will stay outside for 25 % and 5 % of their time respectively. Due to these extra grazing hours, about 7 % of the excreted N by cows ends up in the field, whereas for pigs and poultry 25 % and 5 % of the excreted N ends up outside the stable respectively. This requires new or adapted housing systems with free ranging facilities. Increased grazing times reduces NH_3 proportional to the change in grazing hours.

Optimizing housing systems (set 2) includes the use of innovative manure separation techniques, optimum storage facilities, ammonia scrubbers and ventilation inside the stable, leading to a reduction in NH_3 emission. It is assumed that the combined effect of all these measures results in a decline of the NH_3 emission factors with 65 to 73 % for dairy and pig livestock farms and with 3 to 50 % for poultry farms, depending on the animal category. For dairy cows a housing systems with separate urine collection in combination with chemical air scrubber with about 70 % lower NH_3 emission compared to a conventional dairy stable was recently provisory certified in the Netherlands (Rav = A1.39; https://zoek.officielebekendmakingen.nl/stcrt-2021-40346.html). We did not include application of urease inhibitors in housing systems in view of potential impacts on animal health.

The newly built stables with separated collection of excreted urine and faeces, were combined with closed manure storages with thermal oxidation, resulting in CH_4 emission reduction from housing manure, which is estimated at 63 % for manure from dairy cows and 87 % from pigs (Lesschen et al., 2020). Considering the very limited contribution of poultry to CH_4 emissions (approximately 0.6 %; Van Bruggen et al., 2021), reductions in this sector have not been included, also since there has hardly been any research. Furthermore, a 40 % reduction in enteric CH_4 emission was applied, based on an improved breed of lower methane-emitting animals, and use of synthetic feed additives (Lesschen et al., 2020). All these measures do not affect N_2O emission, except for pig stables where an increase in N_2O emission with 10 % is assumed due to the use of straw and free ranging. In addition, improved application techniques and acidification of manure are assumed to reduce NH_3 emissions by 35 % for cattle and pig manure (Groenestein et al., 2017) with an additional reduction of 15 % (so a total reduction of 50 %) of cattle manure by adding of water to manure and the separate application of solid manure and urine (Bussink and van Rotterdam-Los, 2011; Huijsmans et al., 2015).

Optimized fertilizer strategies (set 3) focusing on the right type, dose, timing and location have potential for lowering the effective nutrient surpluses in agriculture (Velthof and Mosquera, 2011). Use of innovative precision farming technologies enables increased efficiency of the fertilizers, and better timing of fertilizers reduces the risk for NH_3 and runoff losses. These measures are implemented by an increase in NUE near 5 % while applying balanced fertilization, implying a decrease in N fertilizer doses of 10 kg N ha^{-1} for grass and 25 kg N ha^{-1} for arable land, and a reduction in P fertilizer dose of 1 kg P ha^{-1}, a 10 % reduction of N_2O emission and no NH_3 emission from artificial fertilizers through a shift to nitrate based fertilizers alone (Bouwman et al., 2002). We parameterized the impact of nitrification inhibitors by a 50 % reduction in N_2O emissions (Akiyama et al., 2010; Ruser and Schulz, 2015; Velthof and Rietra, 2018), while assuming a change in fertilizer type from calcium ammonium nitrate (CAN), being the dominant fertilizer in the Netherlands, to ammonium sulphate (Velthof and Rietra, 2018). The effect of nitrification inhibitors was combined with urease inhibitors since it strongly enhances NH_3 emissions from manure (Wu et al., 2021) being a trade-off that we like to avoid. We assumed that by applying combining urease and nitrification inhibitors, there is no enhanced NH_3 emission from manure, while we assumed a 5 % (non-calcareous soil) and 10 % (calcareous soils) increase in NH_3 emission from ammonium sulphate (Wu et al., 2021). In addition, we assumed a reduction in N leaching and N runoff of 10 %, considering a similar enhancement in N use efficiency by nitrification inhibitors (Abalos et al., 2014).

Improved soil management (set 4) focuses on: (i) improvement of soil fertility by reduced tillage, (ii) reduced runoff of N and P by the application of buffer strips and (iii) reduction in the emissions of CO_2 and N_2O by use of submerged infiltration drains in peat soils. Reduced soil tillage aims to counteract the current subsoil compaction, and enhance the carbon input via solid manure, compost and the use of cover crops during harvest and winter on arable land. Soil structure improvement was assumed to enhanced crop yields by 10 % for arable crops and by 5 % for grassland and maize (Groenendijk et al., 2016) and an average reduction in runoff of 12 % and 19 % for N and of 1 % and 0 % for P on arable land and grassland, respectively (Groenendijk et al., 2016). Buffer strips across arable and grassland fields, attached to water courses, aim to reduce nutrient losses due to runoff. The application of these strips, which often include the growth of wild flowers to also enhance biodiversity across the landscape, was implemented by assuming that the runoff fractions of N and P are reduced with 10 % (Groenendijk et al., 2021). The use of "Water Infiltration Systems with submerged infiltration drains" (WIS) implies that the summer groundwater level, which is deeper than 40 cm below surface for all peat soils in the Netherlands, is adapted to a summer groundwater level that is higher than 40 cm below surface. In the model, the impact on CO_2 and N_2O emissions and on runoff of N and P was applied by adapting the current GLG (mean lowest groundwater level) of all peat soils to a GLG of 40 cm, thereby causing a reduction in CO_2 emission and in N and P mineralization with a related reduction in N_2O emissions and N and P runoff. We excluded, however, peat soils with a peat layer of <80 cm to allow proper installation of a submerged infiltration drain.

The last set of measures (set 5) on improved crop management includes the use of cover crops, more efficient high yield crop varieties and enhanced use of N fixing crops. The use of cover crops was assumed to increase crop N uptake with 5 kg N ha^{-1} given the same fertilization levels. The use of high yield crop varieties was included by enhancing crop yields by 3–17 %, depending on crop type (implying 3–17 %) more uptake of N and P (Schröder et al., 2016). The increased use of N fixing crops was included by decreasing the N fertilization dose with 20 kg N ha^{-1} for grassland and with 15 kg N ha^{-1} on arable land, assuming that this amount is compensated by enhanced N fixation (Shantharam and Mattoo, 1997; Vitousek et al., 2013).

We combined the results of all sets of measures in a scenario called S1M. Since, we expected that the needed reductions for N, P and GHG losses to air and water could not be reached with technical innovations and management measures only, we also included a scenario in which we reduced livestock by 50 % (Scenario S2R) and a scenario in which we combined all measures and livestock reduction (Scenario S3MR). The reduction in livestock was related to the policy idea that 50 % livestock reduction implies 50 % reduction in ammonia emissions. It is slightly higher than the 42 % reduction in the so-called Productivity stricter scenario presented by Lesschen et al. (2020), aiming to achieve all national and international agreements in 2050 in combination with measures (Gonzalez-Martinez et al., 2021; Kros et al., 2021). In the scenario with livestock reduction, we did not reduce the area of agricultural land, implying a lower livestock density on the current land area. Since full application of measures with the assumed attainable reduction percentage is highly unlikely, and the effectiveness is in practice lower than in theory we also evaluated scenarios in which the effectiveness of the measures was set at 50 % (scenario S1MH and S3MRH). We assumed that the agricultural area stays constant over the next decades, thus implying a lower livestock density per hectare.

For all the scenarios, including the separate sets of measures, we evaluated their impact in view of the targets defined for NH_3 emissions (NH_3 emission ceilings and the Habitats Directive), for NO_3 leaching to groundwater (Nitrates Directive), for N and P runoff to surface water (Water Framework Directive) and for CO_2, CH_4 and N_2O (greenhouse gas) emission from agriculture (National Climate Agreement) as given in Table 1. To make results comparable to formally reported national scale results for the year 2015 (see Table 1, indicated as 2015*), the INITIATOR model results for the emissions of NH_3, NO_x, N_2O and CH_4 were scaled to the values by NEMA (results were within 5–15 %) and for the calculated losses of N and P to water to those of the STONE model (results were within 10–25 %). In addition, the effect of measures was compared to the scaled results for 2015, while not taking excess manure application into account (indicated as 2015 Figs. 5 and 6).

4. Results

4.1. Trends in inputs, uptake and losses of elements at national scale

4.1.1. Trends in element inputs, uptake and surpluses

National trends in inputs, uptake and surpluses of carbon, nitrogen, phosphorus, base cations (Ca, Mg and K) and metals (Cu, Zu and Cd) for the years 2000–2020 are presented in Fig. 2.

Inputs of carbon from manure and compost remain quite stable over time, ranging from 1.1 to 1.2 Mton C yr^{-1}, with a 10 % carbon increase via compost and a comparable decline of crop residue inputs. As a consequence, total organic matter levels in mineral soil stabilize. The national observed decline in soil C is largely due to the oxidation of peat soils. The nitrogen and phosphorus input declined with 26 % and 33 %, respectively up to the year 2010, as a result of more stringent fertilizer regulations and lower protein content of the forages and ration, and remained relatively

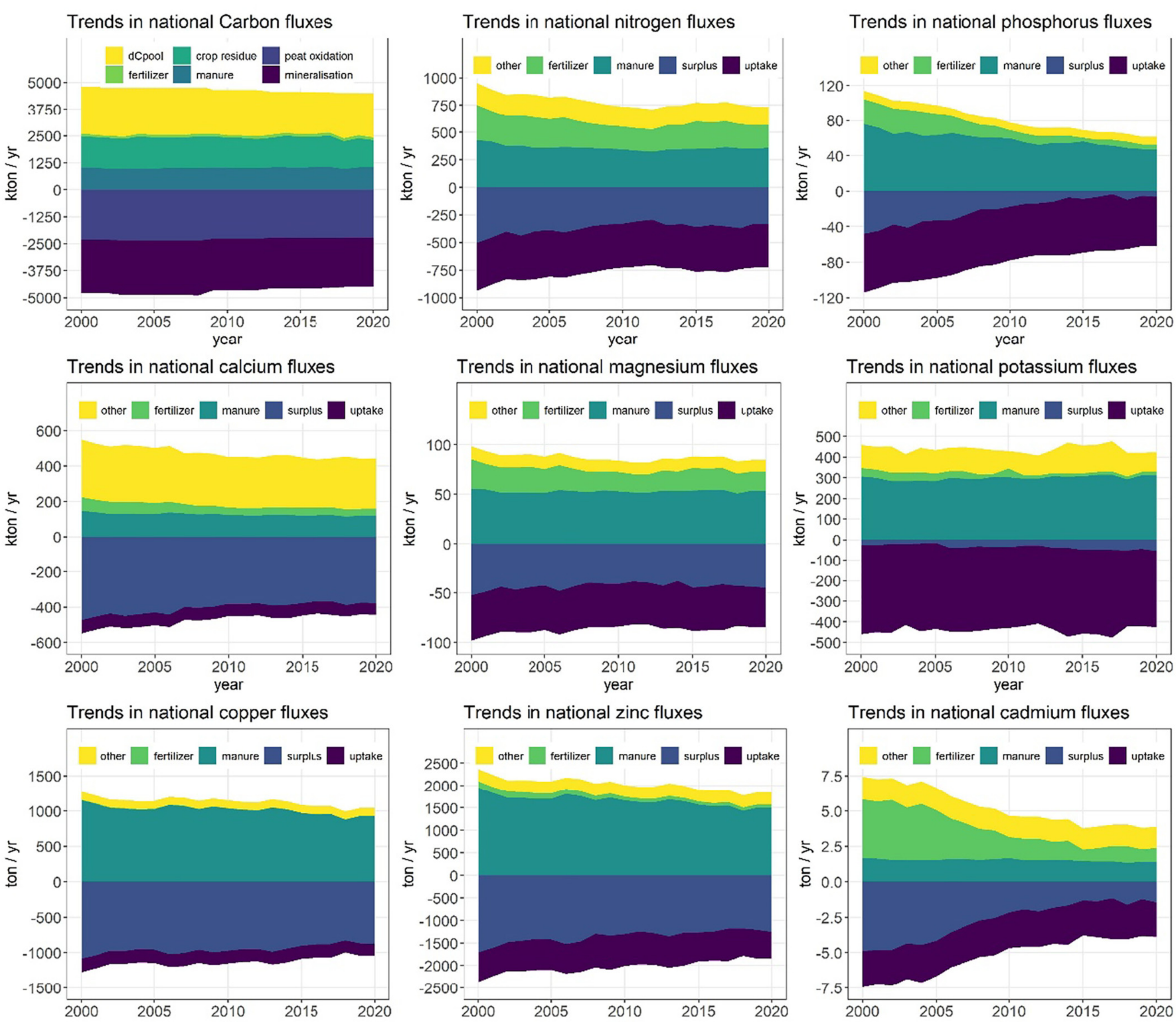

Fig. 2. National trends in inputs, uptake and surpluses of carbon (C), nitrogen (N), phosphorus (P), calcium (Ca), magnesium (Mg), potassium (K), copper (Cu), zinc (Zn) and cadmium (Cd) for 2000–2019 in the Netherlands. Units are in kton for C, N, P, Ca, Mg and K and in ton for Cu, Zn and Cd.
For carbon, the surplus equals the inputs by crop residues and manure minus the decomposition. For the nutrients (N, P, Ca, Mg, K) and metals (Cu, Zn and Cd) the surplus equals the inputs by fertilizer, manure, other organic products and deposition minus net crop removal). For N it also includes biological fixation.

constant in the years thereafter. The surplus of N and P declined over the whole period (33 % for N and 86 % for P) due to slightly increased crop yields and declining application standards. The base cation input varied over the years but there was a slightly declining trend for Ca (−19 %) and Mg (−13 %) over the last 20 years. Similarly, the surplus of cations declined with 20 % for Ca and 15 % for Mg and increased by >100 % for K. National trends in inputs in copper and zinc by fertilizer and manure also showed a gradual decline from 2000 to 2020 varying from 19 to 25 %, especially by Zn in fertilizer from 2005 onwards. For cadmium, there was an ongoing decline in input over the period 2000–2020 near 60 %, especially due to lower Cd concentrations in P fertilizers. As with the major nutrients, the metal uptake stayed constant or slightly increased, implying a clear decline of 20 to 25 % in Cu and Zn surpluses and a very significant (70 %) decline in Cd surplus (Fig. 3).

4.1.2. Trends in element emissions, accumulation and losses

Trends in element fluxes affecting air quality (atmospheric emissions of NH_3, NO_x and greenhouse gases, i.e. CO_2, N_2O and CH_4), soil quality (accumulation or release of C, P and Cd) and water quality (leaching and runoff of N, P and metals), for the Netherlands over the years 2000–2020 are shown in Fig. 3.

Compared to 2000, national atmospheric NH_3-N emissions have declined by 30 % to about 85 kton NH_3-N per year in 2020. Again, the largest reduction is realised in the period up to 2010 where strict legislation forced renewal of housing and storage systems as well as ammonia reducing application technologies. The NH3-N emissions didn't change much after 2010 and hence, the national emission in both 2010 as well as 2015 was >100 % higher than the proposed emission goals for the year 2030 of 50 kton NH_3 yr^{-1} (Fig. 3A). The emissions of N_2O and NO_x from agriculture declined with 29 % whereas the emission of CH_4 gradually increased with 4 % up to 10 kton CO_2-eq yr^{-1}. The overall decline in N_2O in the period 2000–2020, mainly being a decline up to ca 2010 and then a fluctuating constant value, compensated the slight increase in CH_4, leading to a net decrease of 13 % in GHG equivalents of CO_2. The decline in N_2O and NO_x corresponded to more sustainable housing and storage facilities as well more strict application technologies in the field. The increase in CH_4 is associated with the livestock density across the Netherlands; total animal numbers didn't change so much over the last 18 years. The CO_2 emission

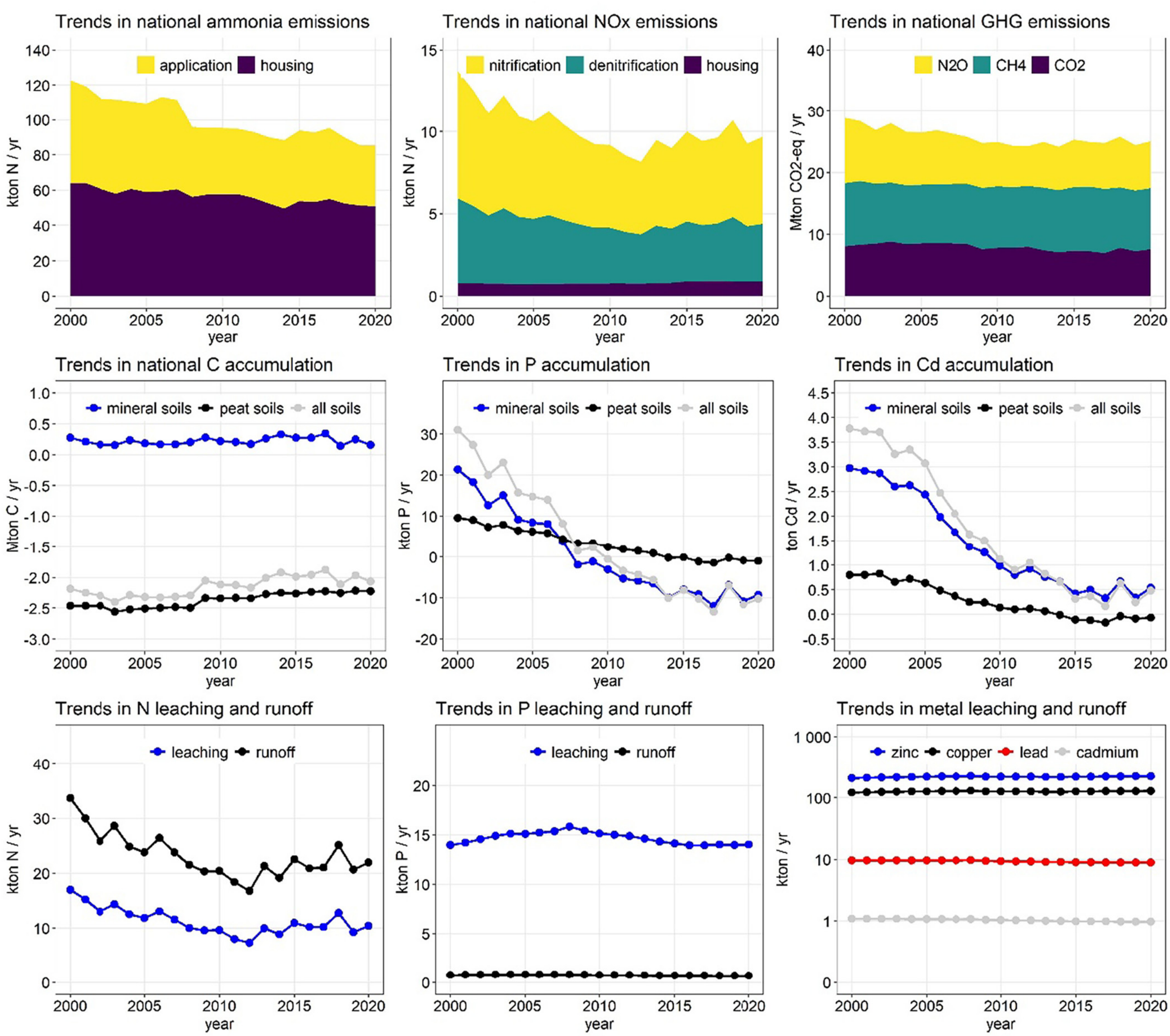

Fig. 3. National trends in element fluxes affecting air quality (top row) by emissions of NH_3, NO_x and greenhouse gases, soil quality (middle row) by accumulation or release of carbon, phosphorus and cadmium, and water quality (bottom row) for leaching and runoff of nitrogen, phosphate and copper and zinc for 2000–2020 in the Netherlands.

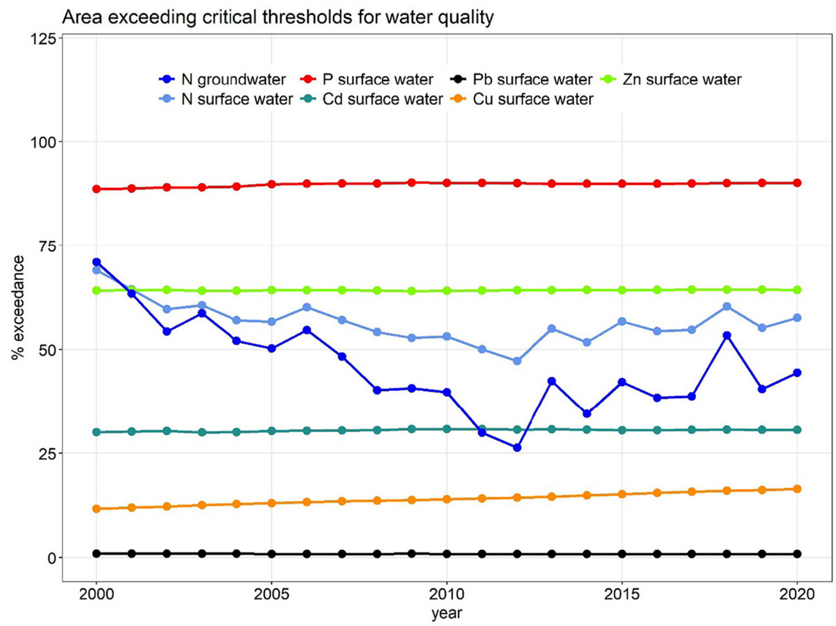

Fig. 4. National trends in areas (%) exceeding critical limits for concentrations of N, P, Cd, Pb, Cu and Zn in surface waters between 2000 and 2019. The used critical limits are (see introduction with references);N: 2.4 mg l^{-1}, P: 0.22 mg l^{-1}, Cd: 0.2 μg. l^{-1}, Pb: 7.2 μg·l^{-1}, Cu: 50 μg·l^{-1} and Zn: 15.6 μg·l^{-1}.

is mainly due to the oxidation of carbon in drained peat soils, partly counteracted by some accumulation in mineral soils, which stayed quit constant over the period 2000–2020.

Losses to the environment gradually declined for P runoff to surface waters (a decline of 10 % between 2000 and 2020) in particular in the period after introduction of the new manure application rules in 2006. Over the whole period 2000–2020 nitrogen losses to groundwater and surface water strongly reduced with almost 40 % down to 10 and 22 kton per year respectively. This decline particularly occurred up to the year 2012 after which it gradually increased. Agronomic measures implemented in the last 18 years have been more effective for nitrogen than for phosphorus. Net P surpluses have been declined down to zero (-6.5 kton P yr^{-1}) given the balanced fertilization strategies. Due to a decline in net metal surpluses, the levels in soil accumulation declined, especially for Cd, but this hardly affected the Cd leaching and, the leaching of Cu and Zn even slightly increased, due to the still ongoing soil accumulation.

4.1.3. Trends in areas exceeding critical limits for (air and) water quality

Trends in areas exceeding critical limits for NO_3 in groundwater, and of N, P and the metals Cd, Pb, Cu and Zn in surface waters are given in Fig. 4.

The area exceeding critical concentrations of P and metals stayed nearly constant over the period 2000–2020, being equal to ca 65 % for Zn, ca 30 % for Cd, ca 90 % for P, ca 12–15 % for Cu (a slight increase) and ca 1 % for Pb. The area exceeding critical N concentrations, however, declined in response to the decline in N surplus from ca 71 % in 2000 to ca 26 % in 2012 and increasing again to ca 45 % in 2020. The critical limits for total N and metals in surface waters are hardly exceeded on clay and peat soils whereas the exceedance is very large for N in the dry sandy soils, for P in the wet sandy soils and for Zn in the non-calcareous sandy soils.

4.2. National totals and spatial variation in inputs, uptake and losses of elements to air and water

4.2.1. National total nitrogen and phosphorus budgets

National total N and P budgets for the year 2015, including total N and P inputs by manure (excretion), fertilizer, deposition and fixation (in case of N), and total N and P outputs by removal by harvest (uptake), manure export and losses to air and water are given in Table 8.

Where P input is completely dominated by manure application, with an equal share of low inputs by fertilizer, compost/sludge and mineralization of peat soils, the N input is also strongly affected by addition of N fertilizer

Table 8
Total agricultural N and P budget for the Netherlands in 2015 (values in brackets are given in kg ha^{-1} yr^{-1}).

Input	Flux (kton yr^{-1})		Output	Flux (kton yr^{-1})	
	N	P		N	P
Animal manure	494 (281)	78.1 (44.4)	Uptake	406 (231)	60.5 (34.4)
- Cattle	329 (187)	47.4 (26.9)	- Grass	281 (298)	40.5 (43.0)
- Pigs	100 (57)	17.7 (10.1)	- Maize	35 (158)	5.4 (24.4)
- Poultry	65 (37)	13.0 (7.4)	- Arable	90 (151)	14.6 (24.6)
Fertilizer	252 (143)	4.4 (2.5)	N housing emission	64 (36)	
Deposition	37 (21)	0.9 (0.5)	Export	82 (46)	21.9 (12.5)
Fixation	15 (8)		Surplus	371 (211)	8.8 (5.0)
Compost and sludge	12 (7)	4.0 (2.3)	- NH_3 soil emission	40 (23)	
Mineralization	112 (64)	3.9 (2.2)	- NO_x soil emission	10 (6)	
			- N_2O soil emission	18 (10)	
			- N_2 soil emission	248 (141)	
			- Accumulation	22 (12)	-8.1 (-4.6)
			- Leaching/runoff	34 (19)	16.9 (9.6)
Total	922 (524)	91.2 (51.9)	Total	922 (524)	91.2 (51.9)

since the N/P ratio of manure is lower than of crops and because of higher N losses to air and water. Nitrogen emissions to air are dominated by N_2 emissions (being comparable to the N fertilizer input, both being near 250 kton yr^{-1}) followed by NH_3-N emissions, estimated at 104 kton N yr^{-1}, divided over emissions in the housing systems (near 60 %) and in the field (near 40 %). The high N2 emissions are due to the inclusion of denitrification in groundwater and ditches, also causing low runoff (23 kton N yr^{-1}) and leaching fluxes (11 kton N yr^{-1}). These fluxes are much higher before those processes occur, i.e. 71 kton N yr^{-1} for leaching below the rootzone (at 50 cm) and 35 kton N yr^{-1} for runoff.

The P balance is nearly closed (a P surplus of <10 % of the input), causing even mining of the soil since the P leaching and runoff is twice as high as the P surplus due to historic high soil P concentrations.

4.2.2. Spatial variation in uptake and losses of nitrogen, phosphorus and greenhouse gases

The geographic variation of N and P use efficiency, NUE and PUE, being the N or P crop removal (uptake) divided by total N or P input. Results show that the NUE varied from <50 % in the marine and river clays being used for arable cropping to above 70 % for the grassland systems on all soil types (Fig. 5A). Unlike NUE, the variation in PUE was much smaller and exceeds often the 100 % given the situation that crop uptake exceeded the P inputs (Fig. 5B). Lower P use efficiencies occurred in the arable regions on both sand, clay and loamy soils.

Fig. 5 also includes information on fluxes and concentration of N and P compounds affecting air and water quality, related to key directives, i.e. the NH_3 emissions (Birds and Habitats Directive: BHD), total GHG emissions (N_2O, CH_4 and CO_2; Paris Agreement; Dutch climate agreement), NO_3 leaching (Nitrates Directive, ND) and P runoff (Water Framework Directive, WFD) for the year 2015 (Fig. 5C–F).

Highest NH_3 emissions (above 80 kg N ha^{-1} yr^{-1}) occur in the Southern part and to a lesser extent in Central/Eastern part of the Netherlands (Fig. 5C), corresponding with the occurrence of intensive animal husbandry in these regions. Livestock farms located on peat soils in the Western and Northern regions are usually dairy farms and less intensive compared to the other regions (with less cows per hectare) resulting in lower NH_3 losses (40–80 kg N ha^{-1} yr^{-1}). Low emissions (below 40 kg N ha^{-1} yr^{-1}) occur typically in regions dominated by arable crops. Similar spatial patterns are also found in deposited NH_3 loadings. The total GHG emission, calculated as the total emission of nitrous oxide (N_2O), methane (CH_4), carbon dioxide (CO_2) expressed in CO_2 eq, is highest in the Western and Northern regions, in areas with agriculture (mostly grassland) on drained peat soils, and in Southern regions with high livestock density and related high CH_4 emissions (Fig. 5D; see also Fig. S3.1 for the geographic variation of the individual emissions of N_2O, CH_4 and CO_2 over agricultural soils). Most of the GHG emissions originate from pens fermentation and peat oxidation.

The NO_3-N leaching flux to groundwater (Fig. 5E) and the P runoff flux to surface water (Fig. 5F) show the classic distinction between the southern and eastern part of the Netherlands being vulnerable for nitrate leaching and the western and northern part being vulnerable for phosphorus losses to the surface water. The calculated NO_3 leaching fluxes are higher in the Central and Southern part of the country (Fig. 5E), not only due elevated manure doses but also due to more N inefficient arable crops like potatoes and vegetables (in contrast to grassland) and the occurrence of soils that are vulnerable for leaching, in particular the dry sandy soils. Sandy soils have relative low denitrification and high infiltration rates, implying that a relative large fraction of the N surplus is lost to ground - and surface water. Unlike NO_3, the runoff of P to surface water is high in the western and northern part of the country, dominated by moderately or poorly drained clay and peat soils, respectively, with a short delay time of P inflow and P runoff (Fig. 5F). Despite relative high P inputs and surpluses on well-drained sandy soils in the Eastern part of the country, runoff is relatively limited.

4.2.3. Spatial variation in element fluxes affecting soil quality and in water quality

Soil quality: The spatial variation of carbon (C) and cadmium (Cd) accumulation in agricultural soils affecting soil quality for the year 2015 is shown in Fig. S3.2. The spatial variation in carbon accumulation (Fig. S3.2A) map mirrors the spatial variation in CO_2 emission (Fig. S3.1C) with high losses (negative accumulation over 5 ton C ha^{-1} yr^{-1}) occurring in regions with drained peatlands in the western and northern part of the country, and limited carbon sequestration (mostly below 5 ton C ha^{-1} yr^{-1}) in large part of the southern, eastern and Northern part of the country. Carbon accumulation particularly occurs in arable systems dominated by crops with either high carbon inputs via roots and residues or with high manure and compost inputs. This increase is in line with measured SOC trends in these regions and is related to the significant C inputs by manure, apart from crop residues. The spatial variation in Cd accumulation is mainly determined by soil type, with accumulation occurring in clay and peat soils due to low leaching rates because of high adsorption on clay and organic matter. Net losses occur in in regions with sandy soils and specifically in the southern part with high Cd levels due to historic contamination.

4.2.4. Water quality

The geographic variation of calculated NO_3 concentrations in leachate to groundwater (Fig. S3.3A) and the P concentration in runoff to surface water (Fig. S3.3B) mimics the spatial patterns of the NO_3-N leaching flux to groundwater (Fig. 5E) and the P runoff flux to surface water (Fig. 5F). As with calculated N leaching, NO_3 concentrations in leaching water are higher in the Central and Southern part of the country (Fig. S3.3A), with nitrate concentrations often above a critical concentration of 50 mg NO_3 l^{-1}. Similarly, as with P runoff fluxes the P concentration in runoff of to surface water is high in the western part of the country, dominated by moderately or poorly drained clay and peat soils, respectively (Fig. S3.3B) with P concentrations above a critical concentration of 0.15 mg P l^{-1}.

The calculated geographic variation of calculated Cu and Zn concentrations in leaching water from agricultural soils in the Netherlands for the year 2015 (Fig. S3.4) also reflects the impacts of soil type with relative low concentrations in the western part, dominated by clay and peat soils and higher concentrations in the eastern part, dominated by sandy soils. In those regions critical limits for surface water of 50 $\mu g \cdot l^{-1}$ for Cu and 15.6 $\mu g \cdot l^{-1}$ for Zn, respectively, were often exceeded.

4.3. Impacts of measures and livestock reduction on element accumulation and losses

The emissions of N compounds and greenhouse gases to air and the leaching and runoff of N and P from agricultural soils in the year 2015 and in response to five scenarios are given in Figs. 6 and 7, respectively. Five different scenarios are evaluated. The first two scenarios include the combined application of all sets of measures being implemented and fully effective on all farms (S1F) or being implemented/effective for 50 % only (S1H). The other three scenarios include a 50 % livestock reduction on all farms without additional measures (S2), with a combination of measures being implemented/effective for 50 % and livestock 25 % (S2H) or 100 % (S3F). Details on the results for each separate set of measures, as defined in Table 7, are given in S4. Below, results are evaluated in view of the environmental targets that need to be reached in 2050 (Table 1).

4.3.1. Nitrogen losses and greenhouse gas emissions to air

Compared to the year 2015 (while not taking excess manure application into account), the combined full implementation of all measures (S1F) reduced annual total ammonia emissions from 94 to 44 kton N yr^{-1}, a reduction of 53 %. The impacts were much lower for NO_x where the emissions declined by 15 % from 10 to 8.5 kton N yr^{-1}. When the measures are effectively implemented by 50 % only (S1H) then NH_3 can be reduced by 31 % and NO_x with 13 %. Reducing animal numbers by 50 % (S2) reduces the ammonia emission with 34 kton N yr^{-1} leading to a 36 % reduction, being less than the desired reduction of 50 % since manure export form the Netherlands is also reduced. The reduction in NO_x emission is only 14 %, since N fertilizer use, being a large source of NO_x is hardly affected by the measures applied. Combining S1H and S1F with 25 % and 50 %

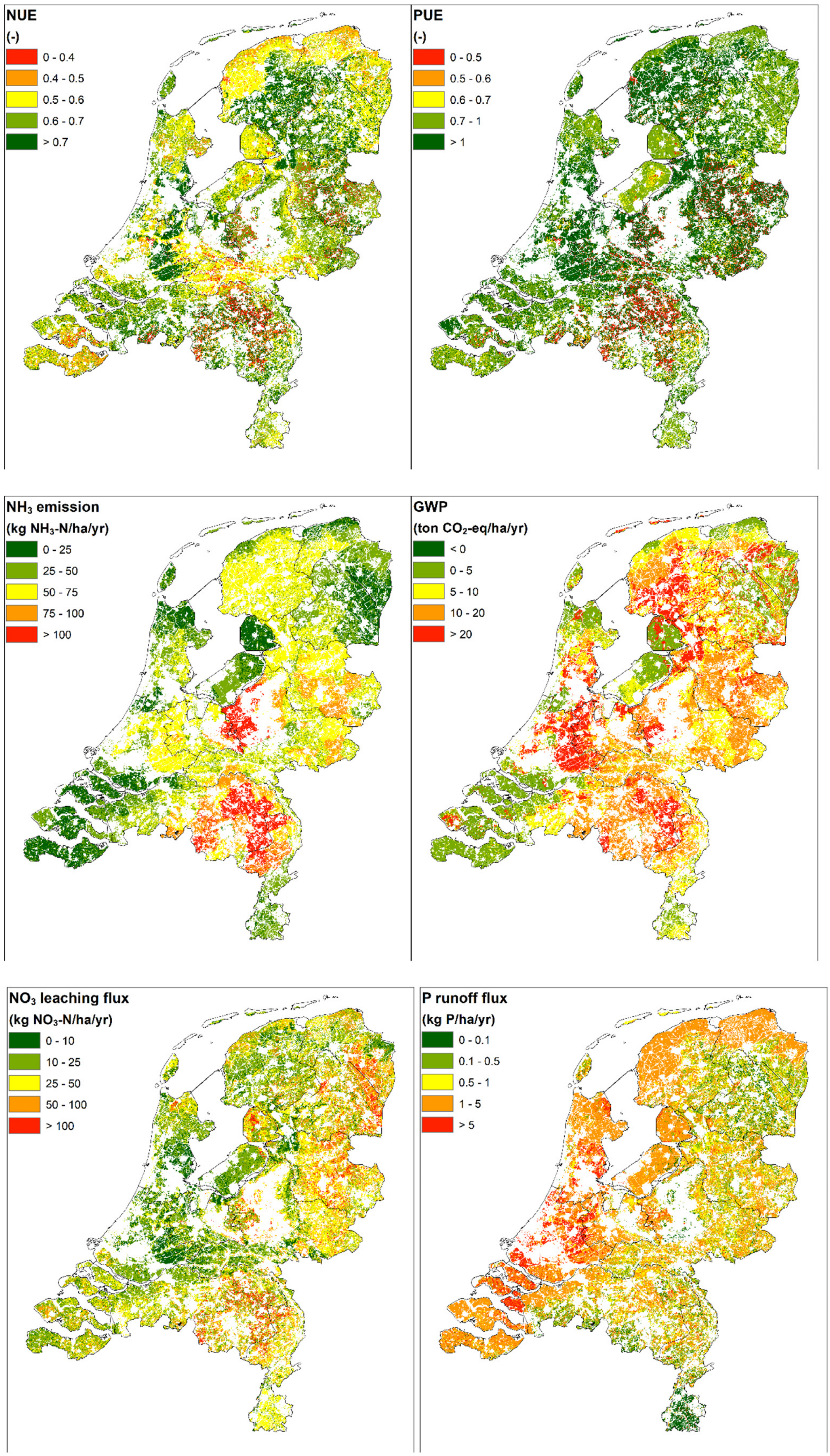

Fig. 5. Spatial variation in simulated N use efficiency, NUE (A), P use efficiency, PUE (B), NH_3 emissions (C), total GHG emissions (expressed in CO_2 eq) (D), NO_3 leaching to groundwater (E) and $H_2PO_4^-$ (expressed as P) runoff to surface water (F) from Dutch agricultural soils, based on the application of INITIATOR for the year 2015.

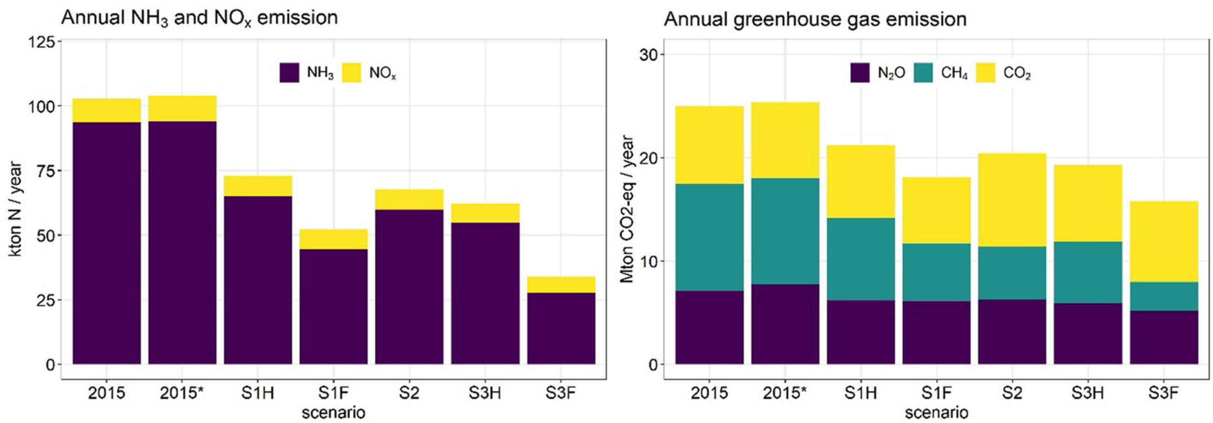

Fig. 6. Calculated annual total NH_3-N and NO_x-N emissions (left) and annual total N_2O, CH_4 and CO_2 emissions (the sum being the total GHG emissions in CO_2 eq) (right) for the year 2015 and for five scenarios, i.e. after the implementation of all sets of measures, both assuming 50 % implementation (S1H) and full implementation (S1F), 50 % livestock reduction (S2) and the combination of all measures and 50 % implementation (S3H) and full implementation (S3F) of measures and livestock reduction.

livestock reduction (S3H and S3F) leads to a reduction in ammonia emissions of 42 % and 71 %, respectively (Fig. 6).

Compared to the year 2015 (while not taking excess manure application into account), the combined application of relevant measures (S1H and S1F) reduced annual total greenhouse gas emissions by 17 % and 29 %, while 50 % livestock reduction (S2) caused a reduction of 21 %, mainly due to reduced CH_4 emissions. The combination of both full implementation of all measures and 50 % livestock reduction (S3F) resulted in GHG (CH_4 + N_2O) reduction of 40 %, but this was only 25 % when measures are effective for 50 % and livestock reduction is 25 % (S3H) (Fig. 6). A more likely 50 % implementation/effectiveness of all measures (S1H) reaches the GHG emission reduction target for 2030 of 17 %. Full implementation of measures with 50 % livestock reduction, does not lead to the desired decline in GHG emissions by 2050 (40 % vs 50 %).The strongest decline in GHG emissions was observed for optimized ration and housing systems, directly affecting the main sources of CH_4. Improving nutrient, soil and crop management had only a limited impact (<5 %) on the total GHG emission (Fig. S4.1).

4.3.2. Nitrogen and phosphorus leaching and runoff

Compared to the year 2015, the combined full implementation of all measures (S1F) reduced total N emission via runoff and leaching by 40 %, while 50 % livestock reduction (S2R) only caused a reduction near 15 % for both fluxes (Fig. 7), thereby contributing to the desired 20 % reduction in N emissions to surface water and groundwater (Table 1). The combination of full implementation of measures and 50 % livestock reduction (S3F) caused a reduction near 40 %, but the 50 % variant (S3H) caused a reduction near 29 % of both fluxes, with a slightly higher impact on nitrate leaching (Fig. 7). The P runoff in S3F declined from 2.1 to 1.95 kg P ha^{-1} yr^{-1}, corresponding to a decline from 3.7 to 3.4 kton P $year^{-1}$.

Measures resulting in a decline of N surpluses were mainly related to improved fertilization techniques. The use of buffer strips had a comparable impact on the N runoff as all measures improving crop production, but most progress originated from optimized fertilization techniques. This might be related that the effectiveness of buffer strips is highly dependent on soil, geohydrology, drainage and land use. Reducing animal numbers leads on average to 35 % reduction in ammonia and 13 % reduction in N losses to the aquatic environment.

The combination of all measures lead to a significant reduction in the area exceeding critical NO_3 concentrations (50 mg NO_3 l^{-1}) in leachate to groundwater (from 6.8 % in 2015 to 1.3 % for all agricultural arable land) but much less in the area exceeding critical N concentrations (2.4 mg N l^{-1}) in runoff to surface water (from 57 down to 44 %). Even when all measures are combined with a significant livestock reduction, the concentrations are such that a reduction in these areas is 'only' near 36 %. Even though the area exceeding critical NO_3 concentrations is very low, there are still fields exceeding the critical limit for nitrate in groundwater, specifically below well-drained sandy soil, where concentrations are generally highest (e.g. Fraters et al., 2001).

Almost all measures improving the efficiency of nitrogen had limited impact on the total P input to soils. The P input declined with 3 % at maximum. When all measures are applied, the P surplus declined from −0.4 to −7.8 kg P ha^{-1}, suggesting that it might take >30 years to substantially reduce the high P level in soil. High P saturation for in particular the sandy soils and high risk for surface runoff at clay and peat soil caused that none of the measures had a substantial impact on P runoff, declining

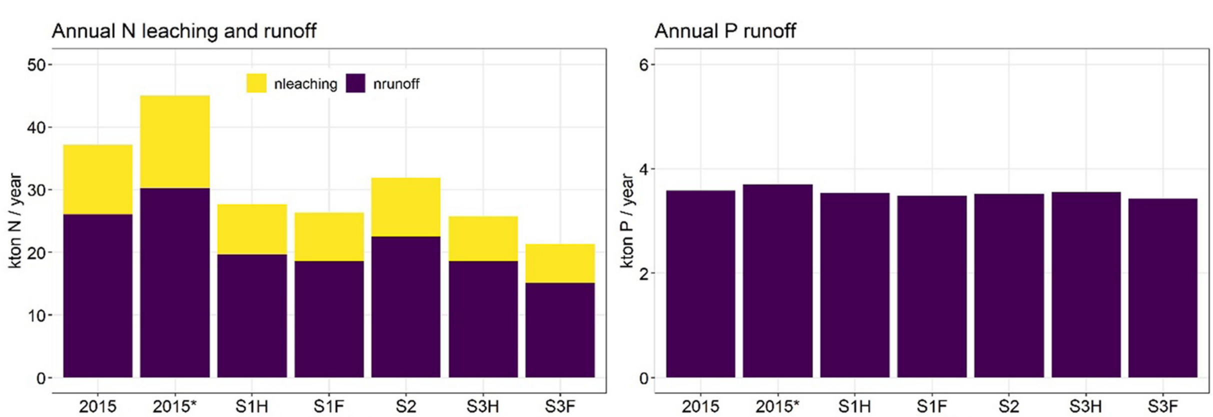

Fig. 7. Calculated annual total NO_3-N leaching and N runoff (left) and P runoff (right) for the year 2015 and for five scenarios, as described below Fig. 6.

from 0.6 to 0.5 kton year^{-1}. The runoff was reduced by at maximum 15 %. Lowering the number of animals in combination with all measures had only an impact of 4 % at maximum. The low reductions of P in runoff to surface water are in line with other studies (e.g. Schoumans and Groenendijk (2000)) since the fate of P is mainly governed by the P soil pool, and even after a 30-year period, this pool is not very strongly depleted in the top 50 cm. Apparently, it takes a very long period to mine the soil with P. In addition, the actual P concentrations in the surface water are also affected by seepage and runoff of particulate P as well as erosion.

5. Discussion and conclusions

5.1. Plausibility of model results

A detailed overview of the plausibility of model results, in view of results from other models and observations is given in S5 and summarized below.

Comparability of national scale trends in nitrogen and phosphorus flows with literature.

A comparison of national scale trends in inputs, uptake and surpluses over the period 2000–2020 calculated with INITIATOR with estimates derived from national statistics (CBS results) for N (Fig. S5.1) and P (Fig. S5.2) shows generally comparable results. Overall, N inputs by fertilizer are comparable (Fig. S5.1A), while P inputs are generally higher in INITIATOR (Fig. S5.2A). This difference is caused because CBS accounts for the consumption rates (bought fertilizer) whereas INITIATOR calculates the applied fertilizer, thus implying differences in farm storage. The results of INITIATOR and CBS for the national scale trends in N and P inputs by manure (Fig. S5.1B and S5.2B) over the period 2000–2020 are, however, strikingly similar. Overall, the calculated N and P uptake by INITIATOR is significantly (10–15 %) higher (Fig. S5.1C and S5.2C), causing a lower N ?thyc=5?> surplus, (Fig. S5.1D) whereas the difference in P surplus is limited (Fig. S5.2D) due to the calculated higher P fertilizer input.

The calculated national trends in NH_3 emissions due to housing and grazing and due to fertilizer and manure application (Fig. S5.3) and in NO_x emissions (Fig. S5.4B) for the period 2000–2020 are also comparable to national CBS but those for total N_2O emissions, due to housing, grazing, fertilizer and manure application, are systematically ca. 15–30 % higher than the national CBS data (Fig. S5.4A). This is likely due to the fact that INITIATOR includes (indirect) N_2O emissions due to denitrification between the rootzone and upper groundwater and in ditches, not included in emission estimates by CBS.

5.1.1. Comparability of model predictions with observations

To model results were also compared with (i) large scale observations of ammonia concentrations in air, (ii) observed nitrous oxide emissions at field scale fi and (ii) NO_3 and $H_2PO_4^-$ concentrations in upper groundwater and ditch water at national scale. Average calculated NH_3 depositions by INITIATOR-OPS with depositions derived from passive sampler concentration measurements at 60 locations corresponded very well but the correlation between the calculated and observed individual NH_3 depositions was weak (Fig. S5.5). However, more recent predicted NH_3 concentrations with the INITIATOR-OPS model with >300 observations at national scale in the year 2018, shows overall a good correlation (R^2 = 0.93–0.96) despite high local under- and overestimates (Fig. S5.6). The comparisons indicate that the national scale uncertainty for NH_3 emissions is near 20 %.

A comparison of modelled and measured N_2O emissions from experiments in four Dutch grassland sites (perennial ryegrass) with different soil types (sand, clay and two peat soils) with three different treatments (mown and unfertilized, mown and N fertilized and grazed and N fertilized) showed a good overall correlation. The variation in treatment was also well covered, but the uncertainty at local scale was high, showing the intrinsic uncertainty of N_2O emissions, since the same result was found when applying the more detailed model DNDC (Fig. S5.7).

Median NO_3 concentrations - below the root zone compared well with observations in upper groundwater and ditch water below sandy soils and clay soils, while they were overestimated in peat soils in a national monitoring network (LMM) for the period 2000–2014 (Table S5.1, Fig. S5.6). The overall uncertainty in predicted NO_3 concentrations was near 30 %.

A comparison of model results and observations of sorbed and water extractable P in long term experimental field studies of grassland on sand, clay and peat including situations with P mining, equilibrium P fertilization and P surpluses showed good agreement (Van der Salm et al., 2016). The same holds for a comparison of the predicted adsorbed and dissolved metal (Cd, Pb, Cu and Zn) concentrations, with and observed reactive and dissolved metal concentrations in three independent soil datasets (Groenenberg et al., 2012). However, P concentrations were generally overestimated when compared to large scale observations in ditch water. Arguments for causes in the differences, including limitations of the measurements for comparison with the predictions, are given in the supplementary material (S5). Despite, the limitations of the measurements, the results indicate that overall the predicted concentrations are plausible and large scale spatial concentration patterns over the Netherlands seem reliable but at local scale, the calculated concentrations can be very uncertain.

5.1.2. Uncertainty in model predictions

Apart from comparing model predictions with observations, insight in the model uncertainty can underpin the robustness, accuracy and reliability of the model output. Such insight can be derived by an uncertainty quantification of the values of state variables at the start of the simulation and model parameters including the: (i) uncertainty in terms of coefficient of variation or standard deviation, distribution type (normal or lognormal), minimum and maximum at the plot level and (ii) spatial correlation coefficients and cross correlation coefficients for certain pairs of model inputs (see e.g. Kros et al., 2012). Such an analysis has been carried out for all included N processes and N losses (De Vries et al., 2003b) The 90 % confidence interval for the fluxes of N compounds to air, groundwater and surface water thus derived was 31 % for ammonia emission, 48 % for N_2O emissions, and 54 % for N inflow to groundwater and surface water. It should be noted, however, that spatial correlation coefficients were not included in the study by De Vries et al. (2003b), implying that the uncertainty is overestimated since uncertainties at local scale average out at larger scale (Kros et al., 2012). Unfortunately, spatial correlation coefficients are hardly known and the best insight in the accuracy of model predictions remains a comparison with large scale data sets, such as those on NH_3 concentrations in air and NO_3 concentrations in water (see above). These comparisons indicate that the national scale uncertainty is near 20 % for NH_3 emissions and near 30 % for NO_3 concentrations, being indeed slightly lower than results derived by the above mentioned uncertainty analysis. An in-depth analysis for all element fluxes in INITIATOR requires a separate study and is foreseen in the future.

5.2. Impacts of measures and livestock reduction

5.2.1. Animal feeding and low emission housing and application

Ammonia emissions from livestock present a major challenge for the Dutch agriculture, causing even a nitrogen crisis in view of the policy ambition to reduce emissions by 50 %. Animal feeding measures, such as optimizing the ration of dairy cows, and their housing systems, with innovative technical solutions as well as optimized injection techniques, thus mainly focus on the needed reduction in the emission of ammonia. On national level the ammonia losses can decline with >40 % based on our first estimates of the effectiveness of the technical solutions considered. There is even potential for further reduction since deep injection of dairy slurry might reduce the ammonia emissions down to 2 % (Bussink and Bruins, 1992), controlled-release fertilizers might reduce volatilization by 20 to 40 % (Tian et al., 2021), feeding strategies reducing urine-N production and related ammonia by 7 to 21 % (Bussink et al., 2017; Groenestein et al., 2017; Anonymous, 2019) and slurry acidification and aeration in pig and poultry housing systems can reduce the ammonia up to 80 to 95 % (Bussink and van Rotterdam-Los, 2011; Groenestein et al., 2011; Bussink et al., 2014), but the impact in dairy stables is usually below

40 % (Groenestein et al., 2017). In all these cases, the actual implementation on farm level determines the effectiveness for diminishing NH_3 emissions. We did not include application of urease inhibitors in housing systems in view of potential impacts on animal health. However, by including the impact of low-emission stables, we already accounted for a high reduction in ammonia emissions. Overall, t is unlikely that technical innovations only can lead to the required emission reduction of 50 %, considering since full application of measures with the assumed attainable reduction percentage is highly unlikely, considering the costs of low emission stables and the fact that emission reductions of such stables were generally lower in practice in the past.

Regarding greenhouse gas emissions, results showed that full application of all relevant measures reduced GHG emissions by 29 %, which is more than enough for the target of 2030 but by far not for the target of 50 % in 2050. A possible measure that has not been implemented is manure fermentation, reducing CH_4 emissions from manure storage by 95–99 %, considering CH_4 emission losses of 1–5 % from manure fermentation installations (Groenestein et al., 2020). However, considering that we included already the effect of closed manure storages with thermal oxidation assuming a related CH_4 emission of 63 % for manure from dairy cows and 87 % from pigs, the additional impact of manure fermentation is limited, also because CH_4 emissions from manure storage are near 31 % of the total emissions only. Inclusion included the overall reduction by 2–3 % only.

5.2.2. Improved nutrient, soil and crop management

Measures aiming to improve nutrient management, such as site-specific precision farming, soil management, such as reduced soil tillage, and crop management, such as increased use of cover crops, play a key role in protecting and enhancing water quality, but the impact on NH_3 emissions was small (see Fig. S4.1 and S4.2A). Improved nutrient, soil and crop management had a consistent impact on the fate of nitrogen: improving nutrient uptake, reducing the surplus and subsequent losses to water, Improved nutrient management had a stronger impact on N leaching and runoff, than soil management and crop management.

This is in line with several long-term trials showed that cover cropping, restoring soil health and the four principles of sustainable fertilization management (selecting the right fertilizer for the right dose and applied at the right location and right time) boost crop production, nutrient efficiency and reduce nutrient losses to air and water (Johnston and Bruulsema, 2014; Pan et al., 2016; Young et al., 2021). In fact, reduction in nitrate loss to groundwater is generally due to plant uptake by cover crops and improved crop nitrogen uptake, and reduced risk of leaching by lower and split N inputs (Carstensen et al., 2020; Nicholson et al., 2020; Velthof et al., 2020). In addition fertilizer recommendations can be improved by accounting for nitrogen that will become available from soil N mineralization, often originating from organic manure and residues from earlier years (Van Es et al., 2020). However, current N fertilizer regulations in the Netherland are already tight, minimizing the N dose based on the agronomic N requirement and ensuring that N losses to groundwater do not cause an exceedance of the limit, further lowering will inevitably lead to a decline in crop production unless the efficiency of the application is increased.

Our study showed that measures improving nutrient, soil and crop management resulted in a decline of the NO_3 losses to groundwater and surface water by ca. 30 %, but the ammonia emission declined with <15 %, and the P surplus declined with 2 kton P (Fig. S4.1 and S4.2). The impact is specifically clear from the reduction in N losses to water, varying from 10 to 28 % (Fig. S4.2A), while P losses to water are hardly affected (Fig. S4.2B) since the P input to surface water is largely originating from water originating from the subsoil, which is hardly affected by the mining of P in agricultural fields with certain measures. Furthermore, the impact of improved nutrient management, soil management and crop management on GHG emissions (Fig. S4.1B) was also very small, with the total GHG emission declining with almost 1 Mton CO_2-eq per year. Due to the assumption that the total nutrient input remained equal, the modelled environmental impact might underestimate the potential of agronomic (soil and fertilizer related) measures and are partly biased to direct animal induced emissions.

5.2.3. Livestock decline

Declining the animal numbers over the whole of the Netherlands had a direct impact on the fate of all nutrients and the emission of all nitrogen and greenhouse gases. It reduces NH_3 emissions and N leaching and runoff, but the impact is less than one might expect due to current manure export, avoiding N losses from field application, while this manure export will stop with livestock reduction. The CH_4 emissions, however, are strongly reduced as most CH_4 stems from enteric fermentation but net CO_2 emissions are predicted to increase. This is because in the scenario with livestock reduction, we assumed that the area of agricultural land stayed constant, implying a lower livestock density on the current land area. Due to this assumption, the carbon input by manure per hectare declined thus slightly reducing carbon sequestration. Given the high initial soil fertility and the substantial contribution of deep rooting crops (grassland, beets, cereals) and default management practices stimulating carbon inputs to the soils, there is, however, only a limited adverse impact on soil organic carbon content. In the livestock reduction scenarios, the reduced N input by animal manure was further compensated by N fertilizers to limit the risk for N deficiencies, and consequently, the N_2O emissions only reduced by approximately 20 %.

The reduction in livestock could also, at least partly, be accompanied by a reduction in agricultural lands. A logic option would then be to reduce the livestock on drained peatlands and stop lowering the groundwater level in these areas. This would lead to less CO_2 emissions from drained peatlands and more C sequestration per hectare on mineral since the same carbon input from manure is then applied on a reduced agricultural land area. Sufficient soil organic matter is of key importance for water and nutrient availability, trafficability, carbon sequestration, resilience against diseases and plagues and crop production, all leading to higher yields and fertilizer efficiency (Hijbeek et al., 2017; Oldfield et al., 2019). Maintaining and, where needed, increasing soil organic matter content serves to meet challenges that intensively used agricultural lands face, like dealing with extreme precipitation and drought, both occurring more frequently due to climate change.

5.2.4. Trade-offs and spatial variation in impacts

Note that the presented analysis summarizes the total effect of all measures combined, in which synergies and trade-offs can counterbalance each other. For example, an increase in grazing, as included in measure set 1, causes less emission of NH_3 and CH_4, considering the housing systems used in 2015, but it increases N leaching (see Fig. S4.1). Such trade-offs are not visible in the total set of measures but can be considered in guiding an optimal set of measures to be applied.

Though not visible in the total nutrient balances for the whole of the Netherlands, substantial differences in impacts occurred due to the impact of soil type, crop rotation plan and geohydrology. Given the landscape of the Netherlands there is a clear distinction between the clayey and peaty soils in the Western regions (struggling mainly with surface water quality and GHG emissions) and the Eastern and Southern regions (struggling mainly with groundwater quality, soil health, and ammonia emissions). The actual impact of the measures thus has to be evaluated in a spatially explicit regional approach, since reductions in national total losses are only an average indication of the impact.

5.3. Challenges and outlook

Currently, no spatial explicit models exist that assess the carbon and nutrient budgets across scales and farming systems in a holistic approach. Our model INITIATOR is among the first that bridges the gap from national and regional policies to farm and field management across the Netherlands while integrating all challenges in the targeted transition to a zero pollution agriculture. The multi-nutrient approach allows the holistic assessment of the flow of carbon and nutrients within the agricultural system, linking nutrient inputs to agronomic desired crop production and minimized environmental targets, also accounting for impacts on greenhouse gas emissions and metal pollution. The model thereby allows agricultural sectors and policy makers to evaluate the impact of management strategies and policies, linking action-focused research to scientific underpinned pathways to sustainability.

There is a strong need for moving farming systems to a more efficient and sustainable future, where carbon and nutrient inputs are optimized given critical thresholds for ammonia and GHG emissions, nitrate in groundwater and nitrogen and phosphorus in surface water. Despite societal benefits, agronomic measures optimizing nutrient budgets for environmental issues are hindered by a range of drawbacks. One drawback is the long timespan before effects become apparent, especially with respect to changes in soil carbon, phosphorus and metal pools, together with uncertainty as to whether the impacts do not cause trade-offs. In this context, it is key to have insights in existing opportunities of operational and strategic management in different landscapes, using robust large scale models that produce plausible results.

INITIATOR model addresses this temporal barrier by gathering and analysing long-term evidence to demonstrate effects at high resolution for the different farming systems across the Netherlands. In addition, perceived risks, user preferences and barriers of farmers can often hinder the uptake of optimized practices for agronomic sustainability as this is often perceived as not economically attractive given the perceived negative trade-offs on crop yield and long timespan before effects become apparent. Identifying the effectiveness of a series of measures will certainly help to adopt them to the location conditions on farm level while meeting the desired regional and national environmental goals.

5.4. Conclusions

The INITIATOR model appeared to be a tool that can well illustrate the impacts of improved feeding, housing, nutrient, soil and crop management on nutrient losses to air and water. Calculated national trends in nutrient losses over 2000–2020 compared well with independent estimates and showed a reduction in N and P input of 26 % and 33 %, respectively, whereas the surplus declined by 33 % for N and 86 % for P due to slightly increased crop yields at reduced inputs. This was accompanied by a reduction of 30–35 % in atmospheric emissions of ammonia and nitrous oxide as well a decline in N and P runoff of 35 and 10 %, respectively, whereas the emission of methane increased with 4 %.

Overall, model results compared well with (i) large scale observations of ammonia concentrations in air and nitrate concentrations in upper groundwater and ditch water, (ii) with nitrous oxide emissions and phosphorus adsorption in experiments at field scale and (iii) with metal adsorption in large scale soil datasets. The large scale comparisons indicate that the national scale uncertainty (95 % confidence interval) is near 20 % for NH_3 emissions and near 30 % for NO_3 concentrations in water.

Mitigation measures related to improved feeding, housing, nutrient, soil and crop management can reduce losses of NH_3 and GHG to air and of N and P to water but reaching the policy ambitions is hard, especially for GHG emissions and P losses to water. Our integrative analysis showed that combined full application of relevant measures focused on emission reduction (source), increased nutrient efficiency (path) and mitigation (route) pathways can reduce NH_3 emission, N leaching and N runoff by 53 %, 47 % and 39 %, respectively, but there is less potential to reduce GHG emissions and specifically P runoff, being 29 % and 6 %, respectively. The combination of a more likely 50 % implementation of measures with 25 % livestock reduction leads to a comparable reduction, i.e. NH_3 emission by 42 %, N leaching by 51 %, N runoff by 49 %, GHG emissions by 25 % and P runoff by 4 %. The potential to reduce P runoff remains small, being near 7 %, since it takes a very long period to mine the soil with P and due to a large background P flux from groundwater.

This study illustrates that reduction targets for the year 2030 for N losses to water can be reached with severe innovation implementations, while it is not possible to reach both NH_3 and GHG emission targets by innovation. The emission reduction ambition of 50 % by 2050 for greenhouse gases is currently impossible, even with improved management and livestock reduction by 50 %. In this context, it seems logic that GHG reductions should come specifically from society and less form agriculture, also considering the dominant share of energy production to GHG emissions.

CRediT authorship contribution statement

Wim de Vries: Conceptualization, Methodology, Writing - Original Draft, - Review & Editing, Hans Kros: Conceptualization, Methodology, Review & Editing, Jan Cees Voogd: Software. Formal analysis, Gerard Ros: Conceptualization, Writing - Review & Editing.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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