



Nitrogen inputs by irrigation is a missing link in the agricultural nitrogen cycle and related policies in Europe



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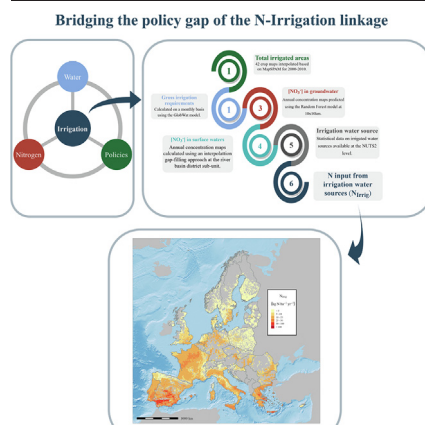
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HIGHLIGHTS

- Irrigation is an overlooked N input at the farm and policy levels.
- We quantify the N input from irrigation water (N_{Irrig}) in Europe for 2000–2010.
- Highly irrigated areas from nitrate contaminated groundwater are the main hotspots.
- N_{Irrig} is comparable to N from synthetic fertilisers in some regions.
- Not including N_{Irrig} overestimates N use efficiency and underestimates N pollution.

GRAPHICAL ABSTRACT



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ABSTRACT

Irrigation, one of the 28 agri-environmental indicators defined in the European Common Agricultural Policy, is often neglected in agricultural nitrogen (N) budgets, while it can be a considerable source of N in irrigated agriculture. The annual N input from irrigation water sources (N_{Irrig}) to cropping systems was quantified for Europe for 2000–2010 at a resolution of 10×10 km, accounting for crop-specific gross irrigation requirements (GIR) and surface- and groundwater nitrate concentration. GIR were computed for 20 crops, while spatially explicit nitrate concentration in groundwater was derived using a random forest model. We show that although GIR were relatively stable (46–60 km³ yr⁻¹), the N_{Irrig} in Europe increased over the 10-year period (184 to 259 Gg N yr⁻¹), approximately 68 % of which occurred in the Mediterranean region. The main hotspots appeared in areas with both high irrigation requirements and high groundwater nitrate concentration, reaching up to averaged values of 150 kg N ha⁻¹ yr⁻¹. These were mainly located

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in Mediterranean Europe (Greece, Portugal and Spain) and to a lesser extent in Northern Europe (The Netherlands, Sweden and Germany). By not including N_{irrig}, environmental and agricultural policies are underestimating the real extent of N pollution hotspots in European irrigated systems.

1. Introduction

Irrigated agriculture is responsible for 70 % of the global withdrawals from surface and groundwater sources (Siebert et al., 2010), playing a key role in agricultural productivity (Wang et al., 2021). The role of irrigation in agriculture largely depends on climate and the availability of water resources (Uniyal et al., 2019), which in turn have shaped local agricultural systems over centuries (Vila-Traver et al., 2021). There are marked climatic differences between Northern- and Southern Europe. During the summer in Southern Europe, especially in areas with a Mediterranean climate, irrigation is often necessary to achieve the break-even yield for some crops, particularly during the summer. In the rest of Europe, agricultural systems are mostly rainfed and only occasionally require supplementary irrigation (Salmon et al., 2015; Wriedt et al., 2009). For instance, about 80 % of the total irrigation demand in Europe occurs in Mediterranean Europe (30,000 km³ yr⁻¹) (Hoogeveen et al., 2015). It is expected that due to climate change, irrigation demand will increase not only in the current Mediterranean climate areas (Cramer et al., 2018; MedECC, 2020) but also in Northern Europe, with some Temperate areas potentially evolving to a Mediterranean climate (Klausmeyer and Shaw, 2009).

Agricultural water and nitrogen (N) cycles are tightly linked in irrigated cropping systems (Cameira et al., 2019). Greater water availability increases the response of crops to N fertilisation (Gabriel et al., 2016) but may lead to higher nitrate leaching (NO₃⁻) (Lassaletta et al., 2021; Quemada et al., 2013) and N₂O emissions (Cayuela et al., 2017). Agricultural NO₃⁻ leaching and surface runoff may contaminate ground- and surface waterbodies (Beusen et al., 2016) and cause subsequent N losses as N₂O and NO_x (Yao et al., 2019), which may, together with reactive N losses from other sources, cause severe public health and environmental problems (Erismann et al., 2013; de Vries, 2021). Aquifers and surface waterbodies are the two main water sources for irrigation, providing a mechanism for nutrient transport and recirculation in irrigated crops that is often overlooked. Given the considerable volume of irrigation water applied to crops, the NO₃⁻ content in irrigation water could represent a substantial N input to agricultural soils, with the potential to reduce the N demand for synthetic fertilisers.

Irrigation is here considered as a “missing link” in the agricultural N balances, since it is usually not accounted for in agricultural N balances in N use efficiency (NUE) studies, neither at global (Zhang et al., 2021a, 2021b; Bouwman et al., 2017) nor at the European scale (de Vries et al., 2021). Cameira et al. (2019) quantified the N input from irrigation water sources (N_{irrig}) in the Tagus Nitrate Vulnerable Zone (Portugal), showing an average N input of 20 kg N ha⁻¹ yr⁻¹ with variation up to almost 80 kg N ha⁻¹ yr⁻¹, accounting for 30–35 % of the total agricultural N inputs. In the dataset developed by Quemada et al. (2020) from farm surveys, N_{irrig} reached up to 159 kg N ha⁻¹ yr⁻¹, though the irrigation volume, nitrate concentration in the irrigation source nor the impact on the NUE was quantified. Cameira et al. (2014) reported similar values (up to 144 kg N ha⁻¹ yr⁻¹) in a field experiment in a peri-urban agricultural area in Lisbon. Zheng et al. (2015) estimated that N_{irrig} contributed 106–136 Gg N yr⁻¹ (about 3 % of N inputs) in the Haihe basin, one of the biggest grain producing areas in China. For mainland China, N_{irrig} estimates are about 700–1100 Gg N yr⁻¹, which represents between 3 and 5 % of N fertilisers (He et al., 2018; Wang et al., 2014).

Since N inputs by irrigation are not generally considered, the linkage between irrigation and nutrient dynamics of agricultural systems is barely addressed by agri-environmental policies. National and supra-national agricultural policies have focused mainly on improving water management through a more efficient use of irrigation in the context of water scarcity

(Kummu et al., 2016). Conversely, nutrient management approaches focus on a more efficient use of agricultural N inputs (e.g., synthetic fertilisers, livestock manure) in view of global food security (Domènech, 2015; Houlton et al., 2019; Kadiresan and Khanal, 2018) while excluding N inputs from irrigation water. In Europe, the Water Framework Directive and Nitrate Directive (e.g., EC-Nitrate D, 1991; EC-WFD–Water Framework Directive, 2000) give emphasis on water quality and N pollution without focusing specifically on the role of irrigation. In addition, the 28 agri-environmental indicators (AEI) developed within the scope of the Europe's Common Agricultural Policy (European Commission (EC), 2006) have separated the AEIs related to water (AEI 7 Irrigation, AEI 20 Water abstraction) and nutrient efficiency (e.g., AEI 15 Gross nitrogen balance).

A quantitative analysis of the impact of the N input from irrigation water sources over European cropping systems and its impact on the N cycle and agri-environmental indicators is thus lacking. We aim to contribute to closing this gap by estimating the gross irrigation requirements (GIR), the NO₃⁻ concentration in ground- and surface waterbodies and the N input from irrigation water sources for Europe in the period 2000–2010. These results are used to analyse the impact of including irrigation in agri-environmental indicators at different spatial scales.

2. Methods

A stepwise methodology was developed to derive N inputs from irrigation water sources, further denoted as N_{irrig}, from a multiplication of interpolated crop irrigated areas, the calculated GIR (related to crop irrigation demand and irrigation efficiency), and the predicted NO₃⁻ concentration in surface- and groundwater sources (Fig. 1), using input data as given in Table 1 and further discussed below. All calculations, including statistical analysis, were implemented and analysed using R statistical software (R Core Team, 2022).

2.1. Assessment of irrigated areas

Irrigated areas for different crops were derived for 2000, 2005 and 2010 from the MapSPAM dataset (<https://www.mapspam.info>). However, this dataset excludes irrigated pastures (both temporary and/or permanent), olives and vineyards, which are important components of irrigated agricultural systems in Mediterranean Europe. Despite its general global scope, MapSPAM 2010 attained a good agreement with Zajac et al. (2022) which disaggregated regional statistics on crop-specific irrigated areas from the EU Farm Structure Survey.

The crop maps were selected for Europe (including the EU-27 plus United Kingdom, Switzerland and Norway) and reprojected to ETRS89/LAEA Europe. Since the number of crop classes from MapSPAM 2000 differed from the 2005/2010 versions (20 and 42, respectively), a standardization procedure was applied using a linear extrapolation of the missing crop classes to 2000 (e.g., tropical and temperate fruits). This procedure implied extrapolating crops that were unavailable in 2000 based on data from 2005 and 2010. After standardization, the irrigated crop areas were linearly interpolated to the remaining years of the period 2000–2010 based on the years 2000, 2005 and 2010. Only crops with a total area exceeding 100 ha in Europe were selected.

2.2. Estimation of irrigation water requirements

Irrigation requirements were estimated with the model GlobWat, which is used by the Food and Agriculture Organization (FAO) to assess agricultural water use globally (Hoogeveen et al., 2015). The model takes a two-

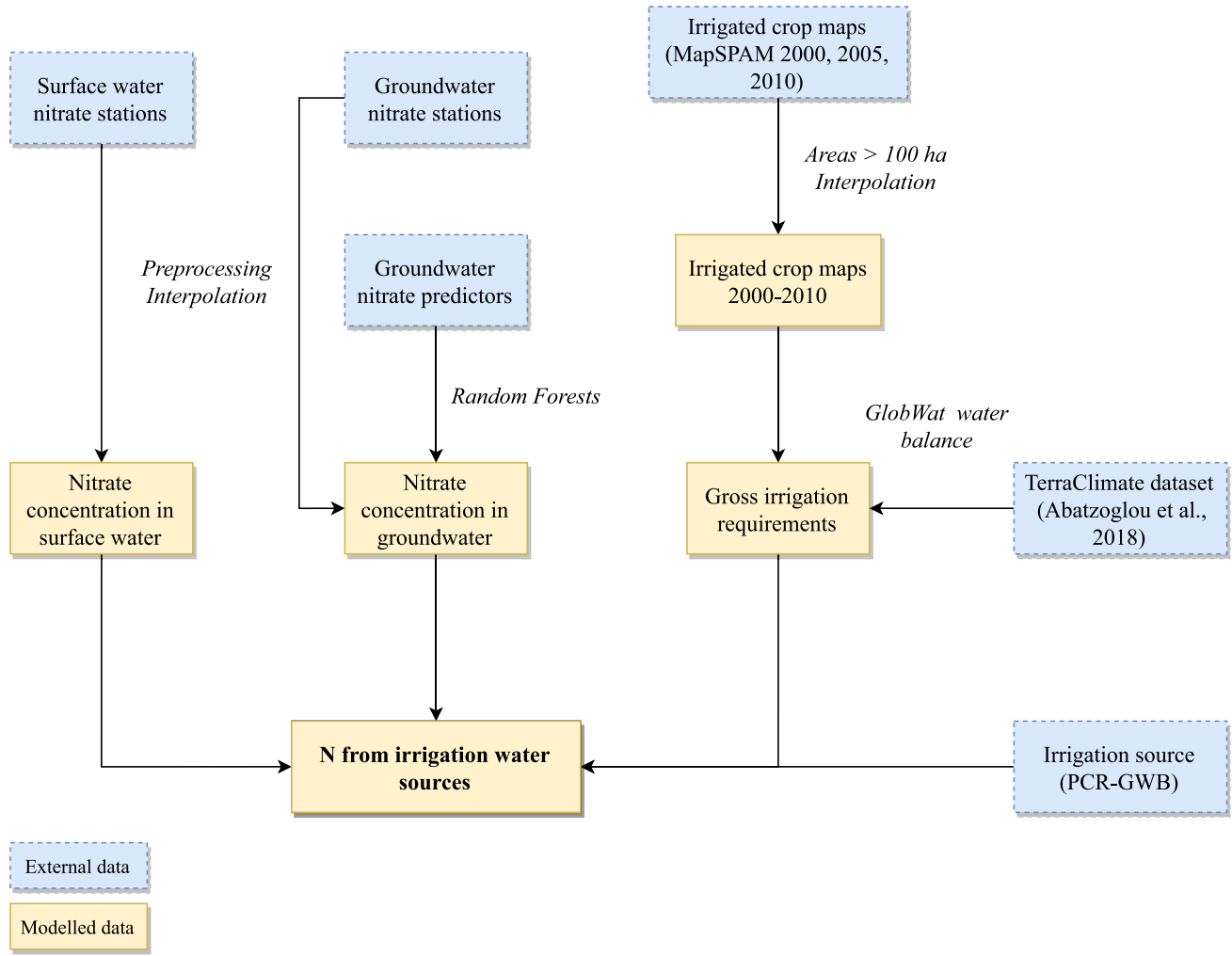


Fig. 1. Conceptual diagram of the methodology employed to compute N input from irrigation water sources. PCR-GLOBWB refers to the model developed by Sutanudjaja et al. (2018). GlobWat refers to the water balance model here employed (see below).

stepped approach to calculate a monthly soil water balance. Firstly, it computes a “vertical” water balance, used to estimate net irrigation water requirements (NIR), followed by a “horizontal” water balance that determines discharges from river basins. The first step was here implemented in R version 4.0.2 (R Core Team, 2022) to derive crop irrigation requirements for Europe during the period 2000–2010 at a spatial resolution of 3 × 4 km. An overview of the model is given below. The monthly soil water balance is given as:

$$P = E_{rain} + R_0 + GR + \Delta S, \quad (1)$$

where P is the precipitation (mm month⁻¹), E_{rain} is the rainfall-dependent evapotranspiration (mm month⁻¹), R₀ the (sub-)surface runoff (mm month⁻¹), GR is the groundwater recharge flux (mm month⁻¹), ΔS the change in soil moisture storage (mm). Precipitation and reference evapotranspiration (ET₀; mm month⁻¹) were collected from TerraClimate (Abatzoglou et al., 2018) (Table 1). All subsequent calculations in this section were proceeded on a monthly basis until stated otherwise.

The value of E_{rain}, defined as the maximum evapotranspiration for a given land-use without water shortage, was calculated as:

$$E_{rain} = \begin{cases} k_{c,LU} \cdot ET_0, & S_{max} \geq S_{t-1} \geq S_{eav} \\ k_{c,LU} \cdot ET_0 \cdot S_{t-1} / S_{eav}, & S_{t-1} < S_{eav} \end{cases} \quad (2)$$

where k_{c,LU} is the land use coefficient (Allen et al., 1998; Hoogeveen et al., 2015) (dimensionless), ET₀ is the monthly potential evapotranspiration

(mm month⁻¹), S_{max} the maximum soil water storage (mm), S_{eav} the readily available soil water (mm) and S_{t-1} the soil water storage at the end of the previous month (mm month⁻¹). S_{max}, S_{eav} (half of S_{max}) and k_{c,LU} were collected from Hoogeveen et al. (2015). Annual land use maps were derived from European Space Agency Climate Change Initiative and resampled to a resolution of 10 × 10 km.

Soil moisture was initialized using S_{eav} following Hoogeveen et al. (2015) and updated monthly according to Eq. (1). If the soil moisture at timestep t was higher than S_{max}, soil moisture was set to S_{max} and (sub-)surface runoff was calculated as the difference of the soil moisture and S_{max}. If the soil was at field capacity or below, no (sub-)surface runoff was assumed to occur. Groundwater recharge (GR; mm month⁻¹) was calculated as a function of the maximum recharge flux and the fraction of soil water storage available to percolate. If there was insufficient water in the soil, GR was set to 0.

Monthly net crop irrigation requirements were calculated for all crops as the difference between the evapotranspiration of a crop under irrigated (ET_c; mm month⁻¹) and rainfed (E_{rain}; mm month⁻¹) conditions. For the ith crop, ET_c was calculated as:

$$ET_{c,i} = k_{c,crop i} \cdot ET_0, \quad (3)$$

where k_{c,crop i} are crop coefficients for four growing periods (i = 1 to 4): initial, development, mid and late stages (Allen et al., 1998). The initial and late phase stages were defined as the first and last month of a crop calendar, respectively. The development stage was the second month of the

Table 1
Overview of the key input data used in the calculations.

Input data	Spatial resolution	Source
Irrigated areas for 20 (year 2000) and 42 crop classes (years 2005 and 2010) (ha yr ⁻¹)	10 × 10 km	MapSPAM (You et al., 2009; Yu et al., 2020)
Water balance		
Precipitation (mm month ⁻¹)		
Reference evapotranspiration (mm month ⁻¹)		TerraClimate (Abatzoglou et al., 2018)
Maximum and initial soil moisture (mm mm ⁻¹)	3 × 4 km	
Maximum groundwater recharge (mm month ⁻¹)		Hoogeveen et al. (2015)
Land use (K _{c,LU}) factors ^a (-)		
Irrigation requirements		
Crop coefficients (k _{c, crop}) (-)		Allen et al. (1998), Hoogeveen et al. (2015)
Crop calendars (-)	Country	Portmann et al. (2008)
Fraction of irrigation systems (%)	10 × 10 km	Derived based on data from AQUASTAT (Food and Agriculture Organization (FAO), 2012)
Spatial prediction of nitrate in surface- and groundwater		
Data from surface- and groundwater NO ₃ ⁻ monitoring stations (mg NO ₃ ⁻ L ⁻¹)	Point	EEA Waterbase
Nitrogen input from irrigation water sources		
Fraction of irrigation water source (ground- and surface water; %)	10 × 10 km	PCR-GLOBWB (Sutanudjaja et al., 2018)

^a The land use factor introduces the effect of land use and vegetation type on the rainfed evaporation (Hoogeveen et al., 2015).

crop calendar, while the remaining months were assumed to be the mid-season stage. Monthly irrigation crop calendars were developed for each country based on Portmann et al. (2008), which were used to allocate k_{c,i} according to crop growing cycles. Thus, net irrigation requirements were computed for the *i*th crop (NIR_{*i*}; mm month⁻¹) as:

$$NIR_i = ET_{c,i} - E_{rain} \quad (4)$$

Following Hoogeveen et al. (2015), an additional water layer of 20 cm was added to the net irrigation requirements of rice paddies. The total (annual) irrigation requirements (TNIR; mm yr⁻¹) were then calculated for the *t*th month and *i*th crop:

$$TNIR = \sum_{t=1}^{12} \sum_{i=1}^I NIR_{i,t} \quad (5)$$

where *i* equals the number of crops (20).

Finally, the gross irrigation requirements (GIR; mm yr⁻¹) were calculated as:

$$GIR = TNIR / EF_{country} \quad (6)$$

where EF_{country} is the overall irrigation efficiency of a country depending on the irrigation systems applied. The efficiencies of the three main irrigation systems available in AQUASTAT (Food and Agriculture Organization (FAO), 2012) were included: gravity, sprinkler and localized (drip). Data regarding the acreage of these systems were collated from AQUASTAT (Food and Agriculture Organization (FAO), 2012) for the different countries and used to derive their respective fraction in total national irrigated areas (f_{irrig}; in %). The field irrigation efficiency of the irrigation systems (EF_{irrig}) was set at 60 % (gravity), 75 % (sprinkler) and 90 % (localized) according to Brouwer et al. (1989). The overall efficiency of each country was calculated using a weighted approach based on the three irrigation systems:

$$EF_{country} = \sum_{s=1}^3 EF_{irrig,i} \cdot f_{irrig,i} \quad (7)$$

2.3. Spatial prediction of N in irrigation water sources

Monitoring station data regarding mean NO₃⁻ concentrations in surface- and groundwater were collated from the European Environmental Agency Waterbase (<https://www.eea.europa.eu/data-and-maps/data/waterbase-water-quality-icm-2>) dataset for the period 2000–2010. The total number of stations with available data on NO₃⁻ concentration for the whole period was 60,377 for groundwater (mg NO₃⁻ L⁻¹; Fig. S1) and 137,715 for surface water (mg N-NO₃⁻ L⁻¹ Fig. S2). The approaches used to calculate spatially explicit NO₃⁻ concentration in surface and groundwater are described below.

2.3.1. Nitrate in the groundwater

A random forest model was developed to predict annual NO₃⁻ concentration in groundwater at a 10 × 10 km resolution in Europe. This algorithm was used in recent studies (Canion et al., 2019; Knoll et al., 2019; Ouedraogo et al., 2019; Rahmati et al., 2019; Spijker et al., 2021) outperforming other approaches such as multiple linear regressions, boosted regression trees and classification and regression trees. The random forest hyperparameters were fine-tuned using a grid search. The number of decision trees, the number of selected independent predictors from an out-of-bag sample, node size and two different sampling methods were the hyperparameters chosen (Probst et al., 2019). The out-of-bag Mean Square Error was used to evaluate the accuracy of the model and the best values for the hyperparameters. A detailed description of RF can be found in Breiman (2001) and Liaw and Wiener (2002). The dataset was partitioned into a training subset (80 %) and a testing subset (20 %).

Aquifer porosity, water table depth and unsaturated soil properties (e.g., fraction of sand), land use and the N surplus (difference of the N inputs and N yield, being a proxy for all N losses) are the most important variables when predicting groundwater NO₃⁻ concentrations (Knoll et al., 2019; Ouedraogo et al., 2019; Wick et al., 2012). Those data were collated from regional/global datasets and resampled to a spatial resolution of 10 × 10 km (SM1) and organized into three main categories: groundwater properties (e.g., porosity; Gleeson et al., 2011), crop management (e.g., soil cover; Panagos et al., 2015); crop fertiliser N (deposition, manure and synthetic fertilisers) (Mueller et al., 2012) and site properties (e.g., slope, soil pH; Panagos et al., 2012).

2.3.2. Nitrate in the surface water

Unlike groundwater, a simple interpolation gap-filling approach was used to derive NO₃⁻ concentrations in surface waters. The spatial unit to estimate surface water NO₃⁻ concentrations was the river basin district sub-unit. Other N compounds are in general much less important in stream waters (Garnier et al., 2010) so they have been neglected in this study. Annual NO₃⁻ concentrations were estimated for each river basin district sub-unit by averaging all the monitoring stations within their boundaries. Data gaps were filled according to the data availability for a given river basin district: if data was available for at least 5 years, the remaining years were estimated using a linear regression, otherwise annual country averages were used to populate the missing data.

2.4. Spatially explicit nitrogen input from irrigation water

For each grid cell and crop, N_{irrig} (kg N yr⁻¹) was calculated as the sum of the input from surface- and groundwater as:

$$N_{irrig} = \sum_{c=1}^{c=20} \sum_{s=1}^2 f_s \cdot GIR_c \cdot TIA_c \cdot NI_s \quad (8)$$

where *c* is the *c*th crop, *s* is the *s*th source (surface- and groundwater), *f_s* concerns the fraction of the irrigation requirements for the *s*th source, *GIR_c* are the gross crop irrigation requirements (m³ ha⁻¹ yr⁻¹), *TIA_c* are the total crop irrigated areas (ha) and *NI_s* is the N concentration in *s*th source (kg N-NO₃⁻ m⁻³). The *s*th source refers to two different water sources: surface- and groundwater. The fraction of irrigation from surface water

was derived from PCR-GWB (Sutanudjaja et al., 2018), while the complementary fraction was allocated to groundwater.

2.5. Impact of the nitrogen input from irrigation in nitrogen management and agri-environmental indicators

To assess the potential impact of crop-specific N_{Irrig} in balanced fertilisation schemes in irrigated systems, we computed the crop N offtake ($\text{kg N ha}^{-1} \text{ yr}^{-1}$) for six crops: rice, maize, wheat, oil crops, potatoes and vegetables:

$$N_{offake,crop} = \text{Yield } N_{content,crop} \quad (9)$$

where Yield is derived for the years 2005 and 2010 from the MapSPAM dataset for the respective irrigated crops ($\text{t ha}^{-1} \text{ yr}^{-1}$) and $N_{content}$ refers to crop-specific N contents (kg N t^{-1}). We recognise the regional variation of N contents in Europe, which is a key uncertainty in N budgets (Eurostat, 2013). The following N contents were used: 20 kg N t^{-1} (wheat), 22 kg N t^{-1} (rice), 16.7 kg N t^{-1} (maize), 30 kg N t^{-1} (oil crops), 3.5 kg N t^{-1} (potatoes) and 2.24 kg N t^{-1} (vegetables) (Leip et al., 2011). We computed two metrics to compare crop-specific N_{Irrig} with crop N offtake: (i) the ratio of N_{Irrig} and N_{offake} and (ii) a proxy of the N demand following the N application from irrigation, which was calculated as the difference of $N_{offtake}$ and N_{Irrig} .

To estimate the possible impact of (total) N_{Irrig} (kg N yr^{-1}) with the N input from synthetic fertilisers (N_{fert} ; kg N yr^{-1}), often the main source of N to crops, we calculated the fraction of N_{Irrig} relative to the N_{fert} at the NUTS3 (Nomenclature of territorial units for statistics) level from the INTEGRATOR (de Vries et al., 2021) model for the year 2010:

$$f_{N_{Irrig}} = N_{Irrig} / N_{fert} \quad (10)$$

We likewise computed the difference in terms of nitrogen use efficiency (NUE; %) using data from the INTEGRATOR, also for the year 2010 (Serra et al., 2023):

$$NUE_{dif} = NUE_{standard} - NUE_{Irrigation} = \frac{N_{Uptake}}{N_{man} + N_{fert} + N_{BNF} + N_{Deposition}} - \frac{N_{Uptake}}{N_{Irrig} + N_{man} + N_{fert} + N_{BNF} + N_{Deposition}} \quad (11)$$

where N_{man} is the N in manure, N_{BNF} the N from biological fixation and $N_{Deposition}$ the N from atmospheric deposition. All terms are given in kg N yr^{-1} . We calculated NUE_{dif} and $f_{N_{Irrig}}$ using the baseline and the lower/upper range of N_{Irrig} from the uncertainty analysis. Similarly, we applied the same equation (Eq. (11)) but for the national data for the period 2000–2010 using data from Eurostat.

2.6. Uncertainty, sensitivity and statistical analyses

The uncertainty assessment was carried out by implementing a Monte Carlo simulation analysis using a Latin Hypercube Sampling (e.g., Cameira et al., 2019) for the terms present in Eq. (8), performing 1000 simulations for each grid cell and year. We assumed a uniform distribution for the gross crop irrigation requirements (GIR) and the fraction of irrigation from groundwater (f_s) and a normal distribution for the nitrate concentration in groundwater (truncated) and in surface water, using CV values as given in Table S3, based on Cameira et al. (2019) and Zhang et al. (2021a, 2021b). We also performed a heuristic sensitivity analysis by recalculating N_{Irrig} after modifying different variables from Eq. (8) by $\pm 25\%$. The variables selected were the NO_3^- concentration in both surface- and groundwater irrigation (NI_s in Eq. (8)), and the fraction of irrigation from groundwater (f_s in Eq. (7)). All variables were simulated separately and in parallel. The reader is referred to the supplementary material for more information.

Spearman's rank correlation coefficient (ρ) was used as a nonparametric measure of rank correlation to identify monotonic

functions, i.e., statistically significant trends in the period 2000–2010. Significant differences were reported where $p < 0.05$.

3. Results and discussion

3.1. Spatial variability in irrigation requirements in Europe

GIR in Europe remained relatively stable between 2000 and 2010 ($46\text{--}60 \text{ km}^3 \text{ yr}^{-1}$) with a ρ of approximately 0 (Fig. 2a; annual maps are shown in Fig. S3). This occurred despite a statistically significant declining trend ($\rho: -0.96; p < 0.05$) found for the total irrigated areas (TIA) in Europe, decreasing from 11.5 to 8.7 million ha between 2000 and 2010. There was a considerable temporal variation in (total) GIR in Europe, varying in total from 46 to $60 \text{ km}^3 \text{ yr}^{-1}$ of water (Fig. 2a). By accounting for annual TIA, these values correspond to $400\text{--}600 \text{ mm yr}^{-1}$ (Fig. 2b). In percentage, the highest increases in GIR over time were attained in Spain ($38.2 \pm 2.3\%$ of total GIR), Greece ($16.5 \pm 2.1\%$) and Italy ($14.2 \pm 0.9\%$) in Mediterranean Europe, and in Romania ($7.1 \pm 3.8\%$) and Bulgaria ($1.7 \pm 0.9\%$) in the case of the rest of Europe. The spatial distribution of irrigation requirements also highlights the contrast between Mediterranean and other EU countries (Fig. 2a, b).

On average, GIR values were $374 \pm 240 \text{ mm yr}^{-1}$, but with marked annual variation as shown by dispersion of the values around the annual means and median (Fig. 2c). The highest annual average GIR values occurred in 2003, 2005 and 2009, i.e., 437 ± 243 , 434 ± 288 and $430 \pm 275 \text{ mm yr}^{-1}$, respectively, with annual variations of ± 32 , 13 and 16% around the mean (Fig. 2b, c). The two peaks observed in 2003 and 2005 correspond to heatwaves in Europe. The first peak is coincidental with the 2003 European heatwave (Ciais et al., 2005), which particularly affected Northern European countries, as shown by the increase in irrigation withdrawal of "Others" (e.g., Germany, the Netherlands; Fig. 2b). In 2005, the Iberian Peninsula also experienced a drought (García-Herrera et al., 2007; Spinoni et al., 2019), which led to a marked increase in irrigation withdrawals in Southern European countries (Fig. 2a). The increase observed in 2009 is explained by larger irrigation requirements in Spain and France (3.5 and $3 \text{ km}^3 \text{ yr}^{-1}$, respectively) despite the e.g., decrease of $1.2 \text{ km}^3 \text{ yr}^{-1}$ in Greece.

About $61.2 \pm 2.6\%$ ($\sim 33 \text{ km}^3 \text{ yr}^{-1}$) of the gross irrigation requirements occurred in the countries with a high share of crops under a Mediterranean climate. Indeed, irrigation requirements were highest ($>1000 \text{ mm yr}^{-1}$) in Southern Portugal, Southeast Greece and Greek islands, and Cyprus (Fig. 3), followed by the remaining regions of Portugal and Greece, Southern France and Italy, as well as most of Central/Southern Spain. In Eastern Europe, some regions of Bulgaria and Romania adjacent to the Black Sea also attained irrigation requirements between 500 and 750 mm yr^{-1} (Fig. 3). The peri-urban agricultural areas in London ($50\text{--}53^\circ\text{N}$) and in Southern Sweden, Finland and Latvia ($56\text{--}58^\circ\text{N}$) were the regions with highest irrigation requirements in Northern Europe (Fig. 3), particularly from irrigated cereals (wheat and the "other cereals" category), vegetables and potatoes (data not shown). The comparison of crop-specific and total GIR with local studies and global models is provided in the supplementary material.

3.2. Spatial variability of nitrate concentration in ground- and surface water

The average NO_3^- concentration in groundwater for the period 2000–2010 was $16.4 \pm 12.0 \text{ mg NO}_3^- \text{ L}^{-1}$, with annual fluctuations ranging from $14.2 \pm 10.1 \text{ mg NO}_3^- \text{ L}^{-1}$ in 2002 to $17.4 \pm 12.3 \text{ mg NO}_3^- \text{ L}^{-1}$ in 2001 (see SM for the validation of the random forest model; Fig. S6–8). For approximately $49,300 \text{ km}^2$ (ca. 1% of the study area), the averaged concentrations were above $50 \text{ mg NO}_3^- \text{ L}^{-1}$, which is the threshold used to identify Nitrate Vulnerable Zones by the EU Nitrates Directive. These areas were mainly located in Spain, with smaller hotspots scattered in The Netherlands, Western Germany, Czech Republic, Austria and Hungary (Fig. 4). Additional hotspots were found in Southern Portugal and Eastern Greece for other years (Fig. S9 shows the annual maps).

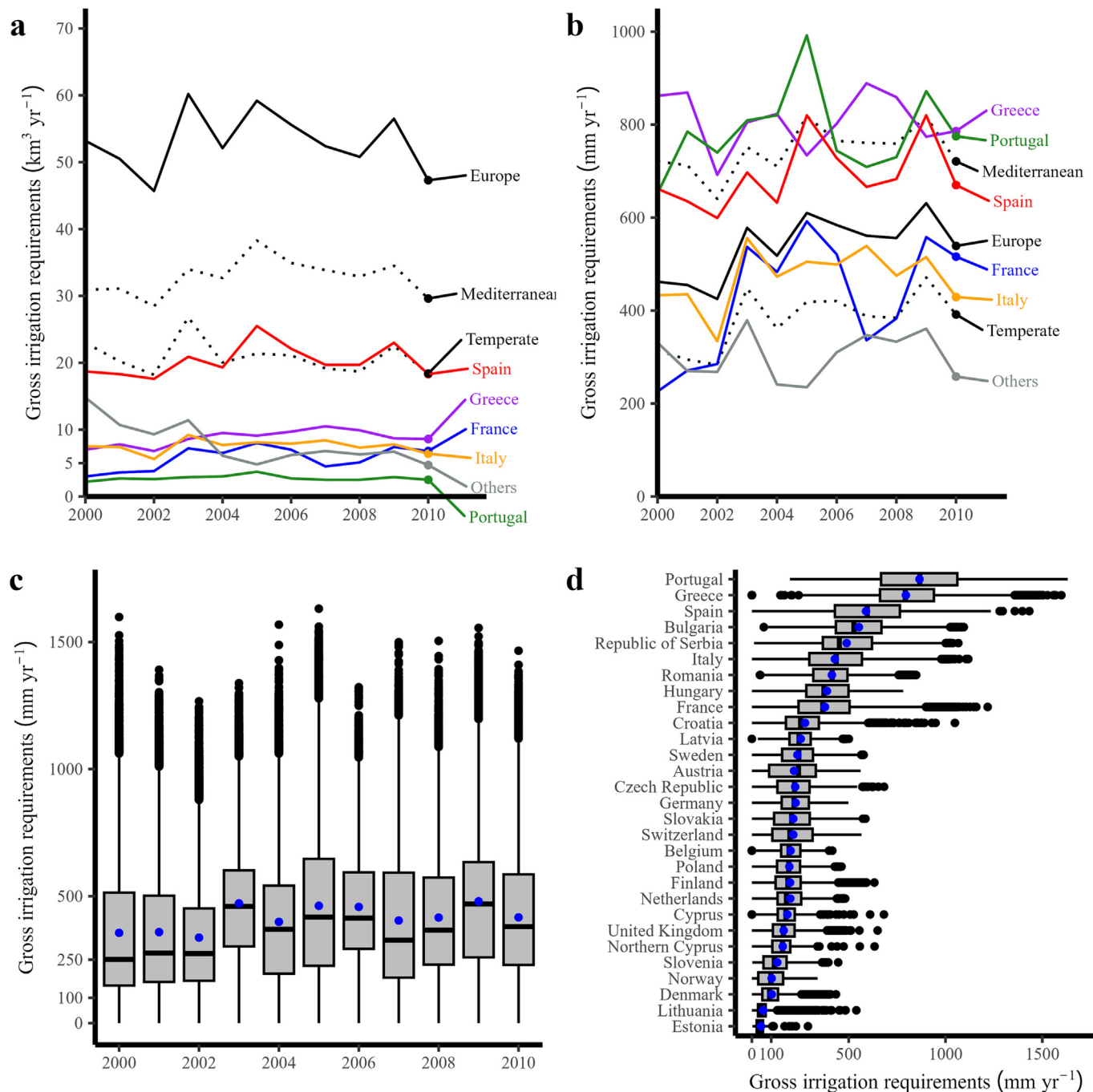


Fig. 2. a) Annual sum of gross irrigation requirements in Europe, relevant countries and in the temperate and Mediterranean climates, both a) in $\text{km}^3 \text{yr}^{-1}$ and b) in mm yr^{-1} ; c) temporal variation of total annual irrigation requirements at the grid level in Europe and d) in different countries for 2000–2010 (both in mm yr^{-1}). Differences in the unit displayed are useful to highlight the differences in the total amount of resources needed from water sources ($\text{km}^3 \text{yr}^{-1}$) versus the intensity in irrigated cropland (mm yr^{-1} , which is often the metric used to recommend irrigation rates at the farm level). A visual display of selected countries in Europe concerning annual and monthly variation in precipitation and reference evapotranspiration is available in the supplementary materials. The blue points represent the annual average.

Countries located in the North-western European plains, such as Belgium ($29 \pm 13 \text{ mg NO}_3^- \text{ L}^{-1}$), the Netherlands ($25 \pm 22 \text{ mg NO}_3^- \text{ L}^{-1}$) and Denmark ($23 \pm 13 \text{ mg NO}_3^- \text{ L}^{-1}$), attained among the highest average NO_3^- groundwater (Fig. S10), while Mediterranean (Spain, France, Italy, Cyprus) averaged $>20 \text{ mg NO}_3^- \text{ L}^{-1}$. Statistically significant increasing trends ($p < 0.05$, $\rho > 0.25$) were found over the European continent, particularly throughout Spain, the European plains and Central Europe (e.g., Hungary). Conversely, statistically significant declining trends ($p < 0.05$, $\rho < -0.25$) were mostly found in the mountainous regions of

Mediterranean Europe (e.g., Pyrenees, Alps), Poland, England and the Netherlands.

In general, the spatial patterns in the predicted NO_3^- concentration agree with the findings from Nistor (2019), insofar as the regions with the highest groundwater vulnerability (e.g., Northern European Plains, Southern Spain) also attained the highest NO_3^- concentration. In addition, the spatial distribution of NO_3^- concentrations are comparable to the predictions from the Netherlands (Spijker et al., 2021) and Germany (Knoll et al., 2019).

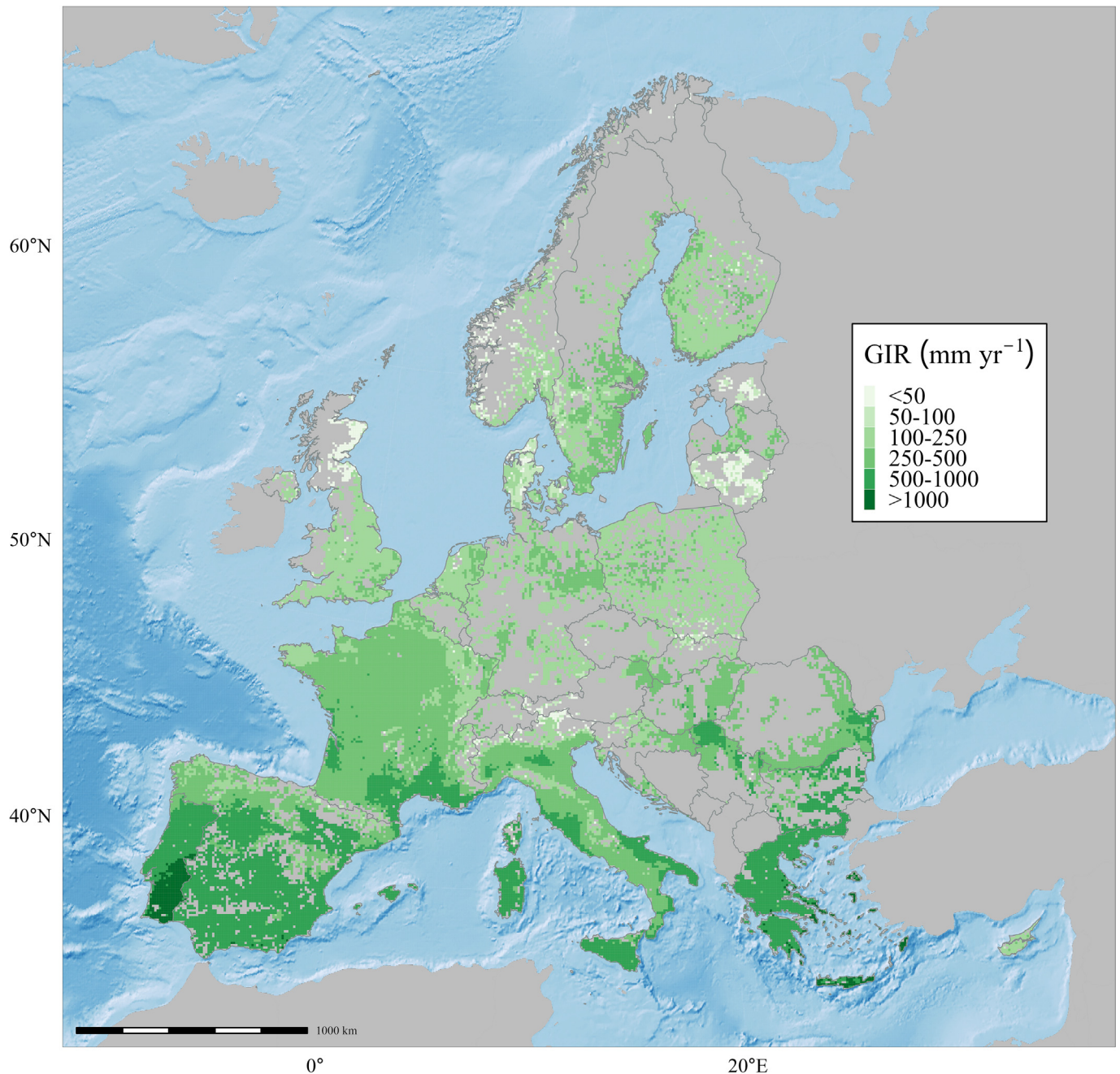


Fig. 3. Spatial distribution of the average annual gross irrigation requirements (GIR) for the period 2000–2010 (left).

In surface waters, the average annual NO_3^- concentration was $1.9 \pm 1.4 \text{ mg NO}_3^- \text{ L}^{-1}$. The concentrations were typically higher in Northern Europe (Fig. 4; Fig. S11 shows the annual maps), particularly in the United Kingdom (rivers Trent and Cam), France (Loire and Seine), Germany (Rhine) and Sweden (Lagan). In Southern Europe, the highest average concentration occurred in the Pineios River in Thessaly, Greece (ca. 4 mg N L^{-1}). Portugal, Greece, Southern France, Austria, and some regions in Germany/Italy attained increasing trends ($\rho > 0.5$; $p < 0.05$; Fig. 4).

3.3. Spatial and temporal variability in the nitrogen input from irrigation water in Europe

The N input from irrigation water sources (N_{Irrig}) in Europe showed an increasing trend ($\rho: 0.50$; $p = 0.12$), increasing from 184 to 259 Gg N between 2000 and 2010 (Fig. 5a; Fig. S12 shows the annual maps). This amount of N, in part recovered from the fertiliser application, is small

when compared with the 10 TgN of synthetic fertilisers used in EU in 2010 (FAOSTAT, 2023). However, the regional and local effects in some irrigated areas can be high. We found a strong increasing trend for N_{Irrig} when accounting for total irrigated areas (TIA; Fig. 5b), increasing from 16 to 30 $\text{kg N ha}^{-1} \text{ yr}^{-1}$ for the same period ($\rho: 0.9$; $p \approx 0$). Our estimates showed a considerable inter-annual variation with reductions up to -30 Gg N yr^{-1} between 2000 and 2001 and increases up to 89 Gg N yr^{-1} between 2004 and 2003 (Fig. 5a). Despite covering only 13 % of the land area in Europe, N_{Irrig} in the Mediterranean climate averaged $68 \pm 5 \%$ of the N_{Irrig} in Europe.

The value of N_{Irrig} was influenced by high irrigation requirements from NO_3^- contaminated water sources - specifically, about $70 \pm 5 \%$ of the N_{Irrig} in Europe came from groundwater. Our data shows the highest values of N_{Irrig} was about $102 \pm 26 \text{ Gg N yr}^{-1}$ in Spain ($\rho: 0.73$; $p = 0.015$) or $42 \pm 6 \%$ of N_{Irrig} in Europe. Greece showed an increasing trend ($\rho: 0.72$; $p = 0.017$), increasing more than twofold between 2000 and 2010

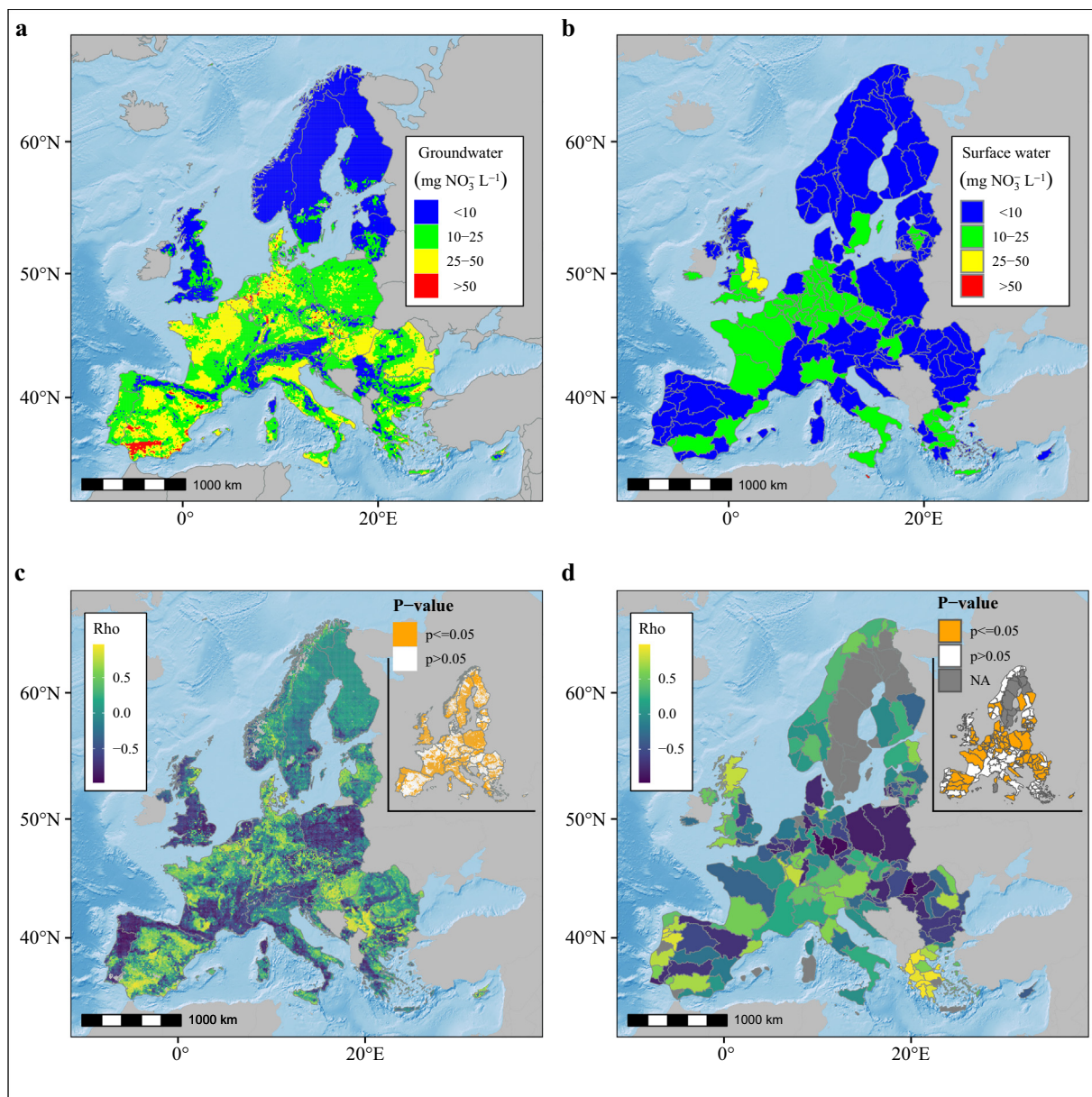


Fig. 4. Spatial distribution of the average NO_3^- concentration and rank correlation coefficient (ρ) and significance level (bottom panel) in groundwater (a, c) and surface waters (b, d) for the period 2000–2010. The inset maps in c) and d) refer to the p -value for each cell. ρ refers to a bivariate analysis of time and NO_3^- concentration for each cell.

(30 to 72 Gg N yr^{-1} , respectively). A contrasting situation was found for non-Mediterranean countries (“Others” in Fig. 5a), with N_{Irrig} decreasing from 44 Gg N yr^{-1} in 2000 (24 % of N_{Irrig} in Europe) to 21 Gg N yr^{-1} in 2010 (9 %). Our estimates for European countries are of the same magnitude as those from Hayashi et al. (2021) for Japan during 2000–2015 of 32–54 Gg N yr^{-1} . For China, national estimates from He et al. (2018) and Wang et al. (2014) are considerably higher around 1100–3730 Gg N yr^{-1} (ca. 13 kg N ha^{-1} yr^{-1} in irrigated cropland).

Grain maize (20 ± 4 %), oil crops (15 ± 2 %) and vegetables (12 ± 3 %) were the main contributors of N_{Irrig} during 2000–2010 (Fig. 5c) but with contrasting trends. Maize (ρ : 0.61; $p = 0.05$) and oil crops (ρ : 0.63; $p = 0.04$) showed strong increasing trends but the contribution of vegetables declined (ρ : -0.4 ; $p = 0.23$). The spatial distribution of crop-specific N_{Irrig} (Fig. S17–19) showed generally larger demand in Mediterranean Europe. We highlight Eastern Greece and Southern and Central Spain, particularly the Guadalquivir basin, where crop-specific N_{Irrig} was consistently larger than 75 kg N ha^{-1} yr^{-1} (e.g., vegetables, maize). For

instance, in some years N_{Irrig} applied to maize reached >200 kg N ha^{-1} yr^{-1} . In Northern Europe, the averaged contribution was comparatively lower (e.g., 25–50 kg N ha^{-1} yr^{-1} of maize in Denmark).

The spatiotemporal pattern of total N_{Irrig} showed marked variation in terms of the magnitude of the hotspots (Fig. 5d), which reached up to >500 kg N ha^{-1} yr^{-1} in 2007 (Eastern Greece). The main hotspots (Fig. 6a) were located at lower latitudes, particularly in Portugal (near the Tagus area and Alentejo), Spain (Guadalquivir basin in Andalucía and Castilla-La Mancha), Northeast Greece and its islands (Crete) and Cyprus. Minor and less common hotspots were also identified in the Northern Italy (Po watershed), Southern Netherlands, Northern Germany and Southern Sweden. These areas showed statistically significant increasing trends in N_{Irrig} , due to statistically significant increases in nitrate concentration in groundwater and/or GIR (Fig. S15). For instance, the large increase observed throughout Eastern Greece between 2005 and 2010 despite the relative stable GIR (Fig. 2a, b) was due to an increase in the NO_3^- concentration in groundwater.

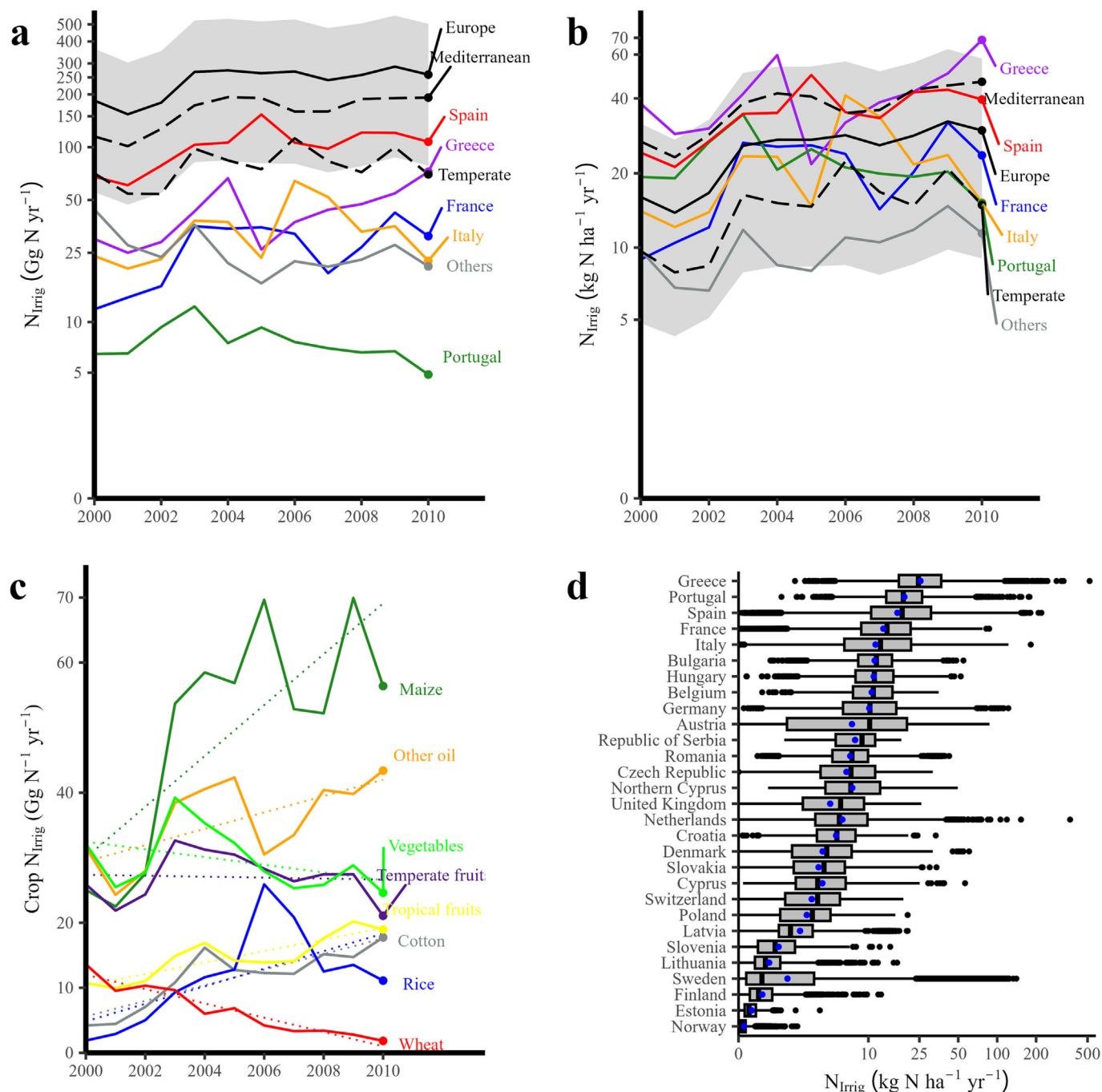


Fig. 5. a) Total N input from irrigation water sources (N_{Irrig}) per country and climatic region (Temperate, Mediterranean) and b) per total irrigated area using a logarithmic scale – the grey shade refers to the uncertainty range. c) Crop-specific N input from irrigation water sources in Europe for the period 2000–2010 and (d) variation within all countries in the study area. The black points in c) and d) represent the annual averages.

3.4. Uncertainties in the results

The mean uncertainty of N_{Irrig} was 25 % (Fig. 6b). The uncertainty ranged from 23 % to 518 % for the period 2000–2010 at the grid level, but the range between the 10th and 90th percentile was rather close (24–27 %) (Fig. S13). The uncertainty was generally lower (ca. 24 %) in the Mediterranean hotspots (e.g., Eastern Greece, Southern Spain and Italy). Our data from the uncertainty analysis shows that N_{Irrig} ranged between 47 and 560 Gg N yr⁻¹ or 4 and 62 kg N ha⁻¹ yr⁻¹ (min-max) during the period 2000–2010 in Europe (Fig. 5a). This wide variation of >500 Gg N yr⁻¹ (a factor of ~10) has wide implications for the extent to which N_{Irrig} can impact N management and policies (see

Section 3.5). The heuristic sensitivity analysis showed a range of ± 43 % relative to the baseline N_{Irrig} by varying ± 25 % the parameters simultaneously; varying the ground- and surface water concentrations yielded a variation of ± 15 and ± 7 %, while the variation was ± 10 % by changing the fraction of irrigation from groundwater (Fig. S14). GIR attained the largest influence (21 %).

3.5. Implications to nitrogen management practices and policies

The marked differences in the spatiotemporal variation of N_{Irrig} during the period 2000–2010 in Europe resulted in very different results for the different countries. Our data shows that N_{Irrig} had a minor impact at the

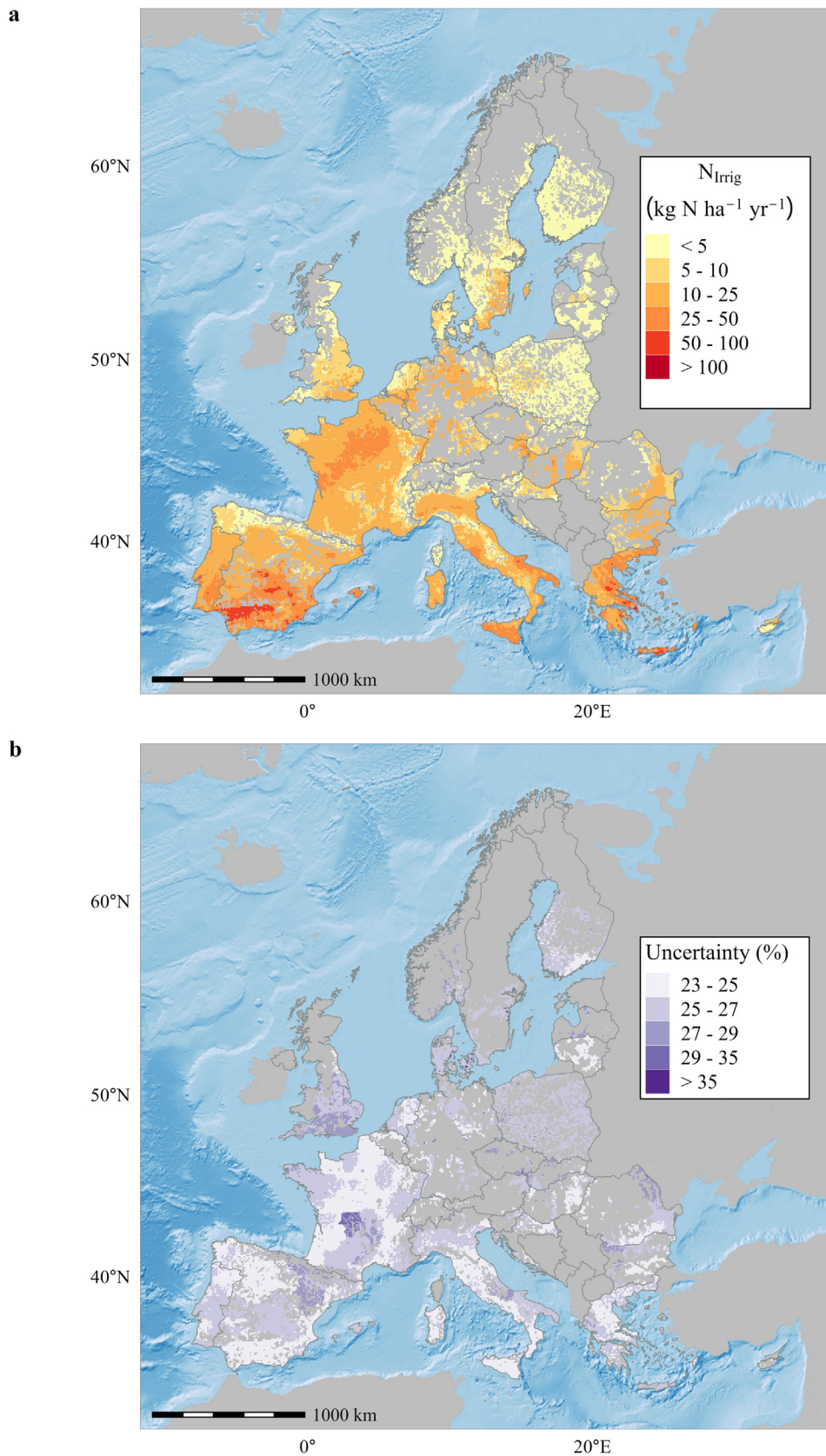


Fig. 6. Spatial distribution of the a) average annual N_{Irrig} and b) uncertainty for the period 2000–2010 in Europe.

European scale, while its contribution was much larger in some areas when studied at finer spatial scales, especially in Southern Europe. While N_{Irrig} had a major contribution in Mediterranean countries irrespective of scale,

the national comparisons masked local hotspots in Northern European countries. In the latter countries, our data suggests that given the magnitude of N_{Irrig} (Fig. 6a), this input may be substantial relative to other

agricultural N inputs in certain areas (e.g., Southern Sweden, some regions in The Netherlands and Northern Germany).

In general, we show that N_{Irrig} is a considerable N input in regions where irrigation requirements are high, mostly from the NO_3^- contaminated groundwater used for irrigation. This is mainly the case for Mediterranean countries where irrigation requirements are large (Fig. 3) and where the larger NO_3^- contaminated areas ($>50 \text{ mg L}^{-1}$) are located. This also occurs over North-western European countries, where NO_3^- contaminated groundwater (Fig. 4) is the main irrigation water source (60–100 %). In Central and parts of Eastern Europe (Romania, Bulgaria), although surface water is the main irrigation source, the high annual NO_3^- concentrations in groundwater can also lead to substantial N_{Irrig} in some regions (Fig. 6). Sweden is the exception as the large N_{Irrig} contribution is due to high NO_3^- concentration in surface waters although the temporal coverage of monitoring stations in this country was insufficient to reliably estimate the contribution. Our data also suggest that the statistically significant increasing trend found for N_{Irrig} is driven by the high NO_3^- groundwater concentration in specific European regions, despite the slight declining trend of irrigation requirements.

Our results show that (i) by not accounting for N_{Irrig} , farmers are not always optimizing balanced fertilisation schemes and (ii) agricultural N studies underestimate the magnitude and extent of agricultural N hotspots while overestimating their N use efficiency (NUE) in European irrigated systems. This also has implications for regional and EU N boundaries (de Vries et al., 2021; Schulte-Uebbing and de Vries, 2021). Indeed, the N_{Irrig} of all crops (e.g., grain maize) seems to surpass the N demand in the hotspots in Mediterranean Europe, i.e., in Southern/Central Spain, Portugal, Southern Italy and Eastern Greece (Figs. S20–S26). Intensively managed crops, such as grain maize, have been the cause of groundwater over-exploitation and contamination in these regions (e.g., Perego et al., 2012; Malik and Dechmi, 2019). In Northern Europe, the contribution of N_{Irrig} to fulfil N demand is generally lower (1–10 %) except in some regions where N_{Irrig} contributes to 75 % of N demand (e.g., maize in Central Denmark). The overall ratio for all crops declined from 50 to 24 % between 2005 and 2010, respectively, as crop N offtake were about 8 % higher in 2010 but N_{Irrig} was 2 % lower.

The implications of including N_{Irrig} on the ratio of N_{Irrig} relative to N from synthetic fertilisers (N_{Fert}) and on the NUE is shown in Fig. 7. N_{Irrig} showed a reduction in the NUE of European agricultural systems of $0.6 \pm 0.1 \%$ (range of 0.2–1.1 %) with a larger relative importance in countries where this input was substantial (Fig. 6). In NW European countries the impact of the upper range of N_{Irrig} reduced the NUE by $<0.2 \%$ with a lower importance than biological N fixation or N deposition (Fig. 7a). Conversely, the average reduction in the NUE was higher in Mediterranean countries, such as Greece (0.8–4.5 %), Spain (0.7–4 %) or Portugal (0.4–2.5 %). These values are comparable to Hayashi et al. (2021), which estimated that the NUE of paddy rice for Japan (2000–2015) without including irrigation was 44 %, almost 7 % higher than when accounting for irrigation.

The effect of N_{Irrig} was more pronounced at the NUTS3 level for 2010 (Fig. 7b) where in some regions the NUE declined $>30 \%$ in Southern Spain (Guadalquivir basin) and Eastern Greece (Thessaly). A similar spatial distribution was also found for the ratio of N_{Irrig} and N_{Fert} (Fig. 7c) where N_{Irrig} has more than doubled N_{Fert} in Eastern Greece. Even at the lower uncertainty range, N_{Irrig} reached up to 75 % of N_{Fert} and reduced the NUE by almost 25 % while the median of all estimate ranges is well below (1 % of N_{Fert} and a reduction of 1 % in the NUE). This suggests that in some regions, particularly in NW Europe, the implications of including N_{Irrig} in balanced fertilisation schemes or AELs are likely to be very low. In Mediterranean Europe, however, N_{Irrig} may range from being a considerable source of N to likely being a key input, more important than synthetic fertilisers.

These findings should, in principle, be underestimated since the N flows from the INTEGRATOR model refer to total utilised agricultural area (sum of permanent crops, grasslands and arable crops) while N_{Irrig} was quantified only for irrigated cropland (with some permanent crops). The fertilisation potential of N_{Irrig} and its impact on the NUE is likely much higher (Fig. 7)

if we were to assess only irrigated areas. This is mainly the case for regions with small patches of irrigated crops, while in regions where irrigated systems are predominant our values should be relatively representative. It is thus important for models to disaggregate rainfed and irrigated crops to assess nutrient flows in agriculture. The extent to which N_{Irrig} can satisfy crop N demand and affects AELs depends not only on the uncertainty of our estimates, but in other factors analysed below.

The quantification of N_{Irrig} should, in principle, differ according to different spatial scales. This is due to the implications of the interactions between hydrogeologic and biochemical processes that may result in N recycling through irrigation water sources (e.g., from fertilisers) depending on the reference spatial unit. At the regional level, the recycled fraction of N_{Irrig} depends on whether the water is retained for less or equal than one year in either source (the reference time for most agri-environmental indicators). In surface water, this fraction depends on the NO_3^- losses through runoff from upstream farms due to the fast NO_3^- dynamics (i.e., low residence time). In lakes/dams, the residence time is likely much higher (Heidbüchel et al., 2012) and thus also the recycling fraction. The water residence time in groundwater is more heterogeneous, depending on aquifer thickness, water table depth and recharge (Wang et al., 2016). Irrigation from aquifers with a residence time less than one year is considered a form of NO_3^- reuse in irrigation. Conversely, aquifers with a residence time of more than one year represent a form of soil legacy effect, comparable to phosphorus (e.g., Pavinato et al., 2020). Ascott et al. (2017) showed the potential NO_3^- storage in the vadose zone, which eventually reaches the aquifer and is available to be transported through irrigation.

It would be appropriate if irrigation related policies in Europe set different regional targets for the different agro-climatic zones according to the prevalence of irrigation practices and NO_3^- contamination in water sources. Including the fertiliser value of N_{Irrig} would enable farmers to somewhat reduce their expenditure on synthetic fertiliser N and reduce the environmental impact of irrigated crop production (Fig. 7). In Europe, accounting for N_{Irrig} in balanced fertilisation plans can be mandatory according to Action Programmes defined for Nitrate Vulnerable Zones according to the Nitrates Directive (Cameira et al., 2019) but this should be expanded to avoid over-fertilisation in other regions where N_{Irrig} is substantial. Outside the Nitrate Vulnerable Zones, it is possible that farmers are unaware of irrigation as a source of nutrients. Even if they do, they may not have been given the technical knowledge to calculate its input and balance synthetic fertilisers accordingly. Thus, to achieve optimal fertilisation, unintentional N inputs through irrigation should universally be estimated and included in the N crop budgets (Villalobos et al., 2020).

Climate change will play a major role in N_{Irrig} dynamics (Cayuela et al., 2017). Firstly, climate change will affect precipitation and temperature in Europe, and the frequency and magnitude of extreme events (Valentini et al., 2014). The impact will vary regionally and seasonally as, for instance, precipitation is projected to increase in Northern Europe and decline in Southern Europe. While an overall increase in irrigation demand is projected for the future (Wada et al., 2013), it is not yet known the extent to which that changes in irrigation systems and technologies as well as the expansion of irrigated agriculture (AEI 9 Land use change) can optimize irrigation demand and crop production. In addition, the future magnitude of N_{Irrig} will be influenced by the effectiveness of nutrient (and water) policies in reducing nutrient loads (depending on residence times) and improving water quality (AEI 27.1 Nitrate pollution).

3.6. Research limitations

There are several uncertainties and gaps associated with the estimates here detailed, all data related (Eq. (7)). Spatiotemporal data on irrigated crop areas are unavailable outside of the three years from MapSPAM (2000, 2005 and 2010). Recently, new datasets have been created to increase the accuracy and reliability of crop irrigated maps in the European Union (Zajac et al., 2022), to expand its temporal coverage to the year 2015 (Grogan et al., 2022) or its temporal development during 2001–2015 (Nagaraj et al., 2021). Those data were insufficient to extent

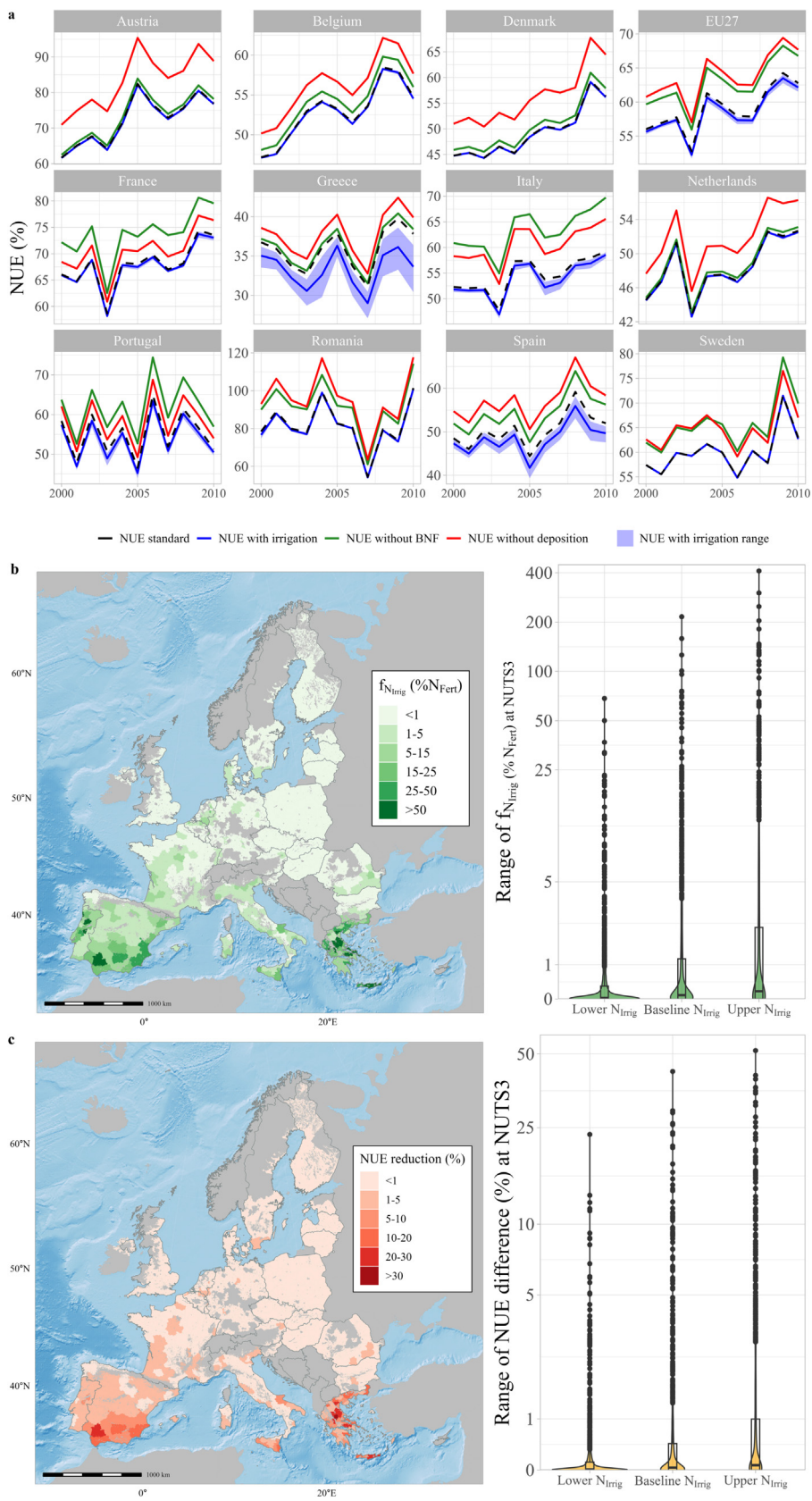


Fig. 7. a) Impact of including the range of N_{Irrig} in the nitrogen use efficiency (NUE), also including N deposition and N fixation for comparison. b) Spatial variation of the fraction of the baseline N_{Irrig} relative to N_{Fert} at the NUTS3 level (left) and respective variation for the range of N_{Irrig} (right). c) Spatial variation of the NUE reduction (difference of NUE without and with irrigation) at the NUTS3 level (left) and respective variation for the range of N_{Irrig} (right). The lower and upper range of N_{Irrig} were calculated from the uncertainty analysis.

our modelling period beyond 2010, due to a reduced number of crops (26; Grogan et al., 2022) or the inherent data structure, e.g., in Nagaraj et al. (2021). Puy et al. (2022) already highlighted how mismatches in the different irrigated crop datasets greatly influence GIR, as well as other key uncertainties in large-scale models.

To estimate GIR, we tried to balance the large-scale implementation of this model and the degree of spatiotemporal detail. We chose a simple water balance model that required small input data relative to other process-based models (GlobWat; Hoogeveen et al., 2015). Although the model was able to simulate comparable GIR to other datasets, this decision is not without caveats. The irrigation allocation from GlobWat is unrestrained, representing ideal conditions in terms of water availability. Farmers may decide to use less irrigation and thus our model can overestimate GIR in these situations. An alternative to offset this limitation would be to use a more detailed model such as AquaCrop, which has already been used at a global scale (Mialyk et al., 2022). This would enable to calibrate the model against statistical crop yields and adjust irrigation accordingly.

The lack of high spatial and temporal resolution data on irrigation statistics affected our approach to derive GIR (Eq. (7)), particularly in terms of irrigation efficiencies, a known challenge in large-scale irrigation models (Puy et al., 2022). Specifically, (i) we were unable to disaggregate the fraction of irrigation systems per crop; (ii) we also assumed the fraction for each irrigation system (f_{irrig}) at the national rather than at the grid level and (iii) assumed the same irrigation efficiencies for the period 2000–2010. In truth, there are regional variations in terms of the overall efficiency depending on cropping systems and predominant irrigation systems as well as temporal variations as it is likely that the efficiencies have improved over time due to better irrigation technologies.

Lastly, the spatial and temporal distribution of the nitrate monitoring stations, both in ground- and surface waters, played a large role in the spatially explicit nitrate concentrations. We used a random forests model to estimate mean NO_3^- concentrations in groundwater at 10x10km resolution and a simple interpolation gap-filling approach for the mean NO_3^- concentrations in surface waters at a river basin district subunit. For groundwater, the spatial distribution of the stations influences the predictive power of the random forests model in regions with different conditions (e.g., management, climate). For instance, in the 2000–2002 period the coverage of the Mediterranean climate was reduced (Fig. S1), which may have influenced the overall magnitude of the predictions for this area. More importantly, the spatial resolution employed (10x10km and watershed level) may be insufficient to accurately reflect the heterogeneity at the local level, particularly in surface waters. Irrigation water from surface waters should only be used near surface waterbodies (e.g., dams, lakes, rivers) and not spatially uniform across a watershed. This is also true for groundwater though it is more readily available.

4. Conclusions

This study presents the first spatiotemporal assessment of the N input, in the form of NO_3^- , from irrigation water sources in Europe. The N input from irrigation water sources was larger in areas with high irrigation requirements and where the irrigation water is abstracted from NO_3^- contaminated groundwater. While there are marked differences between Mediterranean and Northern/Temperate Europe in terms of irrigation requirements, the differences are less clear concerning groundwater NO_3^- concentration. Hotspots with high groundwater NO_3^- concentration were the main driver for the increase in the total N input from irrigation water sources.

The assessment of the N input from irrigation water sources at the national scale highlighted the differences between Temperate and Mediterranean countries, with much higher values in the latter. At a finer spatial scale, the impacts of this N input were magnified, allowing the identification of regions where it can substantially impact agricultural N flows and agri-environmental indicators. Although these regions were mainly located in the Mediterranean region, our data also showed that this input also impacted irrigated systems in North-western Europe. Neglecting N from irrigation water sources in agricultural studies results in an over-estimation of the

nitrogen use efficiency derived from crop N budgets. Conversely, including the N input in irrigation water as a fertiliser resource would permit a better optimization of N fertilisation.

CRedit authorship contribution statement

João Serra: Conceptualization, Data curation, Methodology, Software, Visualization, Writing – original draft. **Cláudia Marques-dos-Santos:** Writing – review & editing, Supervision. **Joana Marinheiro:** Writing – review & editing. **Eduardo Aguilera:** Writing – review & editing. **Luis Lassaletta:** Writing – review & editing. **Alberto Sanz-Cobeña:** Writing – review & editing. **Josette Garnier:** Writing – review & editing. **Gilles Billen:** Writing – review & editing. **Wim de Vries:** Writing – review & editing. **Tommy Dalgaard:** Writing – review & editing, Supervision. **Nicholas Hutchings:** Writing – review & editing, Supervision. **Maria do Rosário Cameira:** Formal analysis, Methodology, Writing – review & editing, Supervision.

Data availability

Nitrogen in irrigation water sources is a missing link in the agricultural nitrogen cycle and related policies in Europe - Supplementary data (Original data) (Figshare)

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

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.164249>.

References

- Abatzoglou, J.T., Dobrowski, S.Z., Parks, S.A., Hegewisch, K.C., 2018. TerraClimate, a high-resolution global dataset of monthly climate and climatic water balance from 1958–2015. *Sci. Data* 5, 1–12. <https://doi.org/10.1038/sdata.2017.191>.
- Allen, R.G., Pereira, L.S., Raes, D., Smith, M., 1998. *Crop evapotranspiration – guidelines for computing crop water requirements*. FAO Irrigation and Drainage Paper, No. 56. FAO, Rome.
- Ascott, M.J., Gooddy, D.C., Wang, L., Stuart, M.E., Lewis, M.A., Ward, R.S., Binley, A.M., 2017. Global patterns of nitrate storage in the vadose zone. *Nat. Commun.* 8, 1–6. <https://doi.org/10.1038/s41467-017-01321-w>.
- Beusen, A.H.W., Bouwman, A.F., Van Beek, L.P.H., Mogollón, J.M., Middelburg, J.J., 2016. Global riverine N and P transport to ocean increased during the 20th century despite increased retention along the aquatic continuum. *Biogeosciences* 13, 2441–2451. <https://doi.org/10.5194/bg-13-2441-2016>.

- Bouwman, A.F., Beusen, A.H.W., Lassaletta, L., van Apeldoorn, D.F., van Grinsven, H.J.M., Zhang, J., Ittersum Van, M., K., 2017. Lessons from temporal and spatial patterns in global use of N and P fertilizer on cropland. *Sci. Rep.* 7–11. <https://doi.org/10.1038/srep40366>.
- Breiman, L., 2001. ST4_Method_Random_Forest. *Mach. Learn.* 45, 5–32. <https://doi.org/10.1017/CBO9781107415324.004>.
- Brouwer, C., Prins, K., Heibloem, M., 1989. Irrigation water management: irrigation scheduling. Annex I: Irrigation Efficiencies. FAO Retrieved from <http://www.fao.org/3/t7202e/t7202e08.htm>.
- Cameira, M.R., Tedesco, S., Leitão, T.E., 2014. Water and nitrogen budgets under different production systems in Lisbon urban farming. *Biosyst. Eng.* 125, 65–79. <https://doi.org/10.1016/j.biosystemseng.2014.06.020>.
- Cameira, M.R., Rolim, J., Valente, F., Faro, A., Dragosits, U., Cordovil, C.M.D.S., 2019. Spatial distribution and uncertainties of nitrogen budgets for agriculture in the Tagus river basin in Portugal – implications for effectiveness of mitigation measures. *Land Use Policy* 84, 278–293. <https://doi.org/10.1016/j.landusepol.2019.02.028>.
- Canion, A., McCloud, L., Dobberfuhl, D., 2019. Predictive modeling of elevated groundwater nitrate in a karstic spring-contributing area using random forests and regression-kriging. *Environ. Earth Sci.* 78, 1–11. <https://doi.org/10.1007/s12665-019-8277-1>.
- Cayuela, María L., Aguilera, E., Sanz-Cobena, A., Adams, D.C., Abalos, D., Barton, L., Ryals, R., Silver, W.L., Alfaro, M.A., Pappa, V.A., Smith, P., Garnier, J., Billen, G., Bouwman, L., Bondeau, A., Lassaletta, L., 2017. Direct nitrous oxide emissions in Mediterranean climate cropping systems: emission factors based on a meta-analysis of available measurement data. *Agric. Ecosyst. Environ.* 238, 25–35. <https://doi.org/10.1016/j.agee.2016.10.006>.
- Ciais, P., Reichstein, M., Viovy, N., Granier, A., Ogée, J., Allard, V., Aubinet, M., Buchmann, N., Bernhofer, C., Carrara, A., Chevallier, F., De Noblet, N., Friend, A.D., Friedlingstein, P., Grünwald, T., Heinesch, B., Keronen, P., Knohl, A., Krinner, G., Loustau, D., Manca, G., Matteucci, G., Miglietta, F., Ourcival, J.M., Papale, D., Pilegaard, K., Rambal, S., Seufert, G., Soussana, J.F., Sanz, M.J., Schulze, E.D., Vesala, T., Valentini, R., 2005. Europe-wide reduction in primary productivity caused by the heat and drought in 2003. *Nature* 437, 529–533. <https://doi.org/10.1038/nature03972>.
- Cramer, W., Guiot, J., Fader, M., Garrabou, J., Gattuso, J.P., Iglesias, A., Lange, M.A., Lionello, P., Llasat, M.C., Paz, S., Peñuelas, J., Snoussi, J., Toret, A., Tsimplis, M.N., Xoplaki, E., 2018. Climate change and interconnected risks to sustainable development in the Mediterranean. *Nat. Clim. Chang.* <https://doi.org/10.1038/s41558-018-0299-2>.
- de Vries, W., 2021. Impacts of nitrogen emissions on ecosystems and human health: a mini review. *Curr. Opin. Environ. Sci. Health*, 100249. <https://doi.org/10.1016/j.coesh.2021.100249>.
- de Vries, W., Schulte-Uebbing, L., Kros, H., Voogd, J.C., Louwagie, G., 2021. Spatially explicit boundaries for agricultural nitrogen inputs in the European Union to meet air and water quality targets. *Sci. Total Environ.* 786, 147283. <https://doi.org/10.1016/j.scitotenv.2021.147283>.
- Domènech, L., 2015. Improving irrigation access to combat food insecurity and undernutrition: a review. *Glob. Food Secur.* 6, 24–33. <https://doi.org/10.1016/j.gfs.2015.09.001>.
- EC-Nitrate D, 1991. In: 8 pp. Last access 10-08-2022 (Ed.), Official Journal of the European Communities (31-12-91). No L 375 / 1. EU Nitrate Directive 91/676/CEE. <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:31991L0676&from=EN>.
- EC-WFD –Water Framework Directive, 2000. Directive 2000/60/EC of the European Parliament and of the council, of 23 October 2000, establishing a framework for community action in the field of water policy. Official Journal of the European Commission Last access 10-08-2022 <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32000L0060&from=EN>.
- Erismann, J.W., Galloway, J., Seitzinger, S., Bleeker, A., Dise, N.B., Petrescu, R., Leach, A.M., de Vries, W., 2013. Consequences of human modification of the global nitrogen cycle. *Philos. Trans. R. Soc. B* 368, 20130116. <https://doi.org/10.1098/rstb.2013.0116>.
- European Commission (EC), 2006. Communication From the Commission to the Council and the European Parliament – Development of Agri-environmental Indicators for Monitoring the Integration of Environmental Concerns Into the Common Agricultural Policy. Accessed from <https://eur-lex.europa.eu/legal-content/en/ALL/?uri=CELEX%3A52006DC0508> 14 June 2021.
- Eurostat, 2013. Methodology and Handbook Eurostat/OECD Nutrient Budgets. version 1.02 pp. 1–112.
- FAOSTAT, 2023. Fertilizers by Nutrient. FAOSTAT database Retrieved from <https://www.fao.org/faostat/#data/RFN>.
- Food and Agriculture Organization (FAO), 2012. AQUAMAPS – global spatial database on water and agriculture. <http://www.fao.org/nr/water/aquamaps/index.html>. (Accessed 20 March 2020).
- Gabriel, J.L., Alonso-Ayuso, M., García-González, I., Hontoria, C., Quemada, M., 2016. Nitrogen use efficiency and fertilizer fate in a long-term experiment with winter cover crops. *Eur. J. Agron.* 79, 14–22. <https://doi.org/10.1016/j.eja.2016.04.015>.
- García-Herrera, R., Paredes, D., Trigo, R.M., Trigo, I.F., Hernández, E., Barriopedro, D., Mendes, M.A., 2007. The outstanding 2004/05 drought in the Iberian Peninsula: associated atmospheric circulation. *J. Hydrometeorol.* 8, 483–498. <https://doi.org/10.1175/JHM578.1>.
- Garnier, J., Billen, G., Némery, J., Sebilo, M., 2010. Transformations of nutrients (N, P, Si) in the turbidity maximum zone of the seine estuary and export to the sea. *Estuar. Coast. Shelf Sci.* 90, 129–141. <https://doi.org/10.1016/j.ecss.2010.07.012>.
- Gleeson, T., Smith, L., Moosdorf, N., Hartmann, J., Dürr, H.H., Manning, A.H., Van Beek, L.P.H., Jellinek, A.M., 2011. Mapping permeability over the surface of the earth. *Geophys. Res. Lett.* 38, 1–6. <https://doi.org/10.1029/2010GL045565>.
- Grogan, D., Frolking, S., Wisser, D., Prusevich, A., Glidden, S., 2022. Global gridded crop harvested area, production, yield, and monthly physical area data circa 2015. *Sci. Data* 9. <https://doi.org/10.1038/s41597-021-01115-2>.
- Hayashi, K., Shibata, H., Oita, A., Nishina, K., Ito, A., Katagiri, K., Shindo, J., Winiwarter, W., 2021. Nitrogen budgets in Japan from 2000 to 2015: decreasing trend of nitrogen loss to the environment and the challenge to further reduce nitrogen waste. *Environ. Pollut.* 286. <https://doi.org/10.1016/j.envpol.2021.117559>.
- He, W., Jiang, R., He, P., Yang, J., Zhou, W., Ma, J., Liu, Y., 2018. Estimating soil nitrogen balance at regional scale in China's croplands from 1984 to 2014. *Agric. Syst.* 167, 125–135. <https://doi.org/10.1016/j.agsy.2018.09.002>.
- Heidbüchel, L., Troch, P.A., Lyon, S.W., Weiler, M., 2012. The master transit time distribution of variable flow systems. *Water Resour. Res.* 48. <https://doi.org/10.1029/2011WR011293>.
- Hoogetveen, J., Faurès, J.M., Peiser, L., Burke, J., van de Giesen, N., 2015. GlobWat - a global water balance model to assess water use in irrigated agriculture. *Hydrol. Earth Syst. Sci.* 19, 3829–3844. <https://doi.org/10.5194/hess-19-3829-2015>.
- Houlton, B.Z., Almaraz, M., Aneja, V., Austin, A.T., Bai, E., Cassman, K.G., Compton, J.E., Davidson, E.A., Erismann, J.W., Galloway, J.N., Gu, B., Yao, G., Martinelli, L.A., Scow, K., Schlesinger, W.H., Tomich, T.P., Wang, C., Zhang, X., 2019. A world of Cobenefits: solving the global nitrogen challenge. *Earth's Future* 7, 865–872. <https://doi.org/10.1029/2019EF001222>.
- Kadiresan, K., Khanal, P.R., 2018. Rethinking irrigation for global food security. *Irrig. Drain.* 67, 8–11. <https://doi.org/10.1002/ird.2219>.
- Klausmeyer, K.R., Shaw, M.R., 2009. Climate change, habitat loss, protected areas and the climate adaptation potential of species in mediterranean ecosystems worldwide. *PLoS One* 4. <https://doi.org/10.1371/journal.pone.0006392>.
- Knoll, L., Breuer, L., Bach, M., 2019. Large scale prediction of groundwater nitrate concentrations from spatial data using machine learning. *Sci. Total Environ.* 668, 1317–1327. <https://doi.org/10.1016/j.scitotenv.2019.03.045>.
- Kummu, M., Guillaume, J.H.A., de Moel, H., Eisner, S., Flörke, M., Porkka, M., Siebert, S., Veldkamp, T.I.E., Ward, P.J., 2016. The world's road to water scarcity: shortage and stress in the 20th century and pathways towards sustainability. *Sci. Rep.* 6, 1–16. <https://doi.org/10.1038/srep38495>.
- Lassaletta, L., Sanz Cobena, A., Aguilera, E., Quemada, M., Billen, G.F., Bondeau, A., Cayuela, M., Cramer, W., Eekhout, J.P., Grizzetti, B., Intrigliolo, D., Romero, E., Ruiz-Ramos, M., Vallejo, A., Gimeno, B.S., 2021. Nitrogen dynamics in cropping systems under Mediterranean climate: a systemic analysis. *Environ. Res. Lett.* 18, 119–123. <https://doi.org/10.1088/1748-9326/ac002c>.
- Leip, A., Britz, W., Weiss, F., de Vries, W., 2011. Farm, land, and soil nitrogen budgets for agriculture in Europe calculated with CAPRI. *Environ. Pollut.* 159 (11), 3243–3253. <https://doi.org/10.1016/j.envpol.2011.01.040>.
- Liaw, A., Wiener, M., 2002. Classification and regression by randomForest. *R News* 2, 18–22.
- Malik, W., Dechmi, F., 2019. DSSAT modelling for best irrigation management practices assessment under Mediterranean conditions. *Agric. Water Manag.* 216, 27–43. <https://doi.org/10.1016/j.agwat.2019.01.017>.
- MedECC, 2020. Climate and environmental change in the Mediterranean basin – current situation and risks for the future. In: Cramer, W., Guiot, J., Marini, K. (Eds.), First Mediterranean Assessment Report. Union for the Mediterranean, Plan Bleu, UNEP/MAP, Marseille, France <https://doi.org/10.5281/zenodo.4768833> 632pp.ISBN 978-2-9577416-0-1.
- Mialyk, O., Schyns, J.F., Booi, M.J., Hogeboom, R.J., 2022. Historical simulation of maize water footprints with a new global gridded crop model ACEA. *Hydrol. Earth Syst. Sci.* 26, 923–940. <https://doi.org/10.5194/hess-26-923-2022>.
- Mueller, N.D., Gerber, J.S., Johnston, M., Ray, D.K., Ramankutty, N., Foley, J.A., 2012. Closing yield gaps through nutrient and water management. *Nature* 490, 254–257. <https://doi.org/10.1038/nature11420>.
- Nagaraj, D., Proust, E., Todeschini, A., Rulli, M.C., D'Odorico, P., 2021. A new dataset of global irrigation areas from 2001 to 2015. *Adv. Water Resour.* 152. <https://doi.org/10.1016/j.advwatres.2021.103910>.
- Nistor, M.M., 2019. Groundwater vulnerability in Europe under climate change. *Quat. Int.* <https://doi.org/10.1016/j.quaint.2019.04.012>.
- Ouedraogo, I., Defourny, P., Vanclouster, M., 2019. Application of random forest regression and comparison of its performance to multiple linear regression in modeling groundwater nitrate concentration at the African continent scale. *Hydrogeol. J.* 27, 1081–1098. <https://doi.org/10.1007/s10040-018-1900-5>.
- Panagos, P., Van Liedekerke, M., Jones, A., Montanarella, L., 2012. European Soil Data Centre: response to European policy support and public data requirements. *Land Use Policy* 29, 329–338. <https://doi.org/10.1016/j.landusepol.2011.07.003>.
- Panagos, P., Borrelli, P., Meusburger, K., Alewell, C., Lugato, E., Montanarella, L., 2015. Estimating the soil erosion cover-management factor at the European scale. *Land Use Policy* 48, 38–50. <https://doi.org/10.1016/j.landusepol.2015.05.021>.
- Pavinato, P.S., Cherubin, M.R., Soltangheisi, A., Rocha, G.C., Chadwick, D.R., Jones, D.L., 2020. Revealing soil legacy phosphorus to promote sustainable agriculture in Brazil. *Sci. Rep.* 10. <https://doi.org/10.1038/s41598-020-72302-1>.
- Perego, A., Basile, A., Bonfante, A., de Mascellis, R., Terribile, F., Brenna, S., Acutis, M., 2012. Nitrate leaching under maize cropping systems in Po Valley (Italy). *Agric. Ecosyst. Environ.* 147, 57–65. <https://doi.org/10.1016/j.agee.2011.06.014>.
- Portmann, F., Siebert, S., Bauer, C., Döll, P., 2008. Global dataset of monthly growing areas of 26 irrigated crops. *Frankfurt Hydrology Paper*. 06, p. 400.
- Probst, P., Wright, M.N., Boulesteix, A.L., 2019. Hyperparameters and tuning strategies for random forest. *Wiley Interdiscip. Rev.: Data Min. Knowl. Discov.* 9, 1–15. <https://doi.org/10.1002/widm.1301>.
- Puy, A., Sheikholeslami, R., Gupta, H.V., Hall, J.W., Lankford, B., Lo Piano, S., Meier, J., Pappenberger, F., Porporato, A., Vico, G., Saltelli, A., 2022. The delusive accuracy of global irrigation water withdrawal estimates. *Nat. Commun.* 13, 3183. <https://doi.org/10.1038/s41467-022-30731-8>.
- Quemada, M., Baranski, M., Nobel-de Lange, M.N.J., Vallejo, A., Cooper, J.M., 2013. Meta-analysis of strategies to control nitrate leaching in irrigated agricultural systems and their effects on crop yield. *Agric. Ecosyst. Environ.* 174, 1–10. <https://doi.org/10.1016/j.agee.2013.04.018>.
- Quemada, M., Lassaletta, L., Jensen, L.S., Godinot, O., Brentrup, F., Buckley, C., Foray, S., Hvid, S.K., Oenema, J., Richards, K.G., Oenema, O., 2020. Exploring nitrogen indicators

- of farm performance among farm types across several European case studies. *Agric. Syst.* 177, 102689. <https://doi.org/10.1016/j.agry.2019.102689>.
- R Core Team, 2022. R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. <https://www.R-project.org/>.
- Rahmati, O., Choubin, B., Fathabadi, A., Coulon, F., Soltani, E., Shahabi, H., Mollaefer, E., Tiefenbacher, J., Cipullo, S., bin Ahmad, B., Tien Bui, D., 2019. Predicting uncertainty of machine learning models for modelling nitrate pollution of groundwater using quantile regression and UNEEC methods. *Sci. Total Environ.* 688, 855–866. <https://doi.org/10.1016/j.scitotenv.2019.06.320>.
- Salmon, J.M., Friedl, M.A., Frohling, S., Wisser, D., Douglas, E.M., 2015. Global rain-fed, irrigated, and paddy croplands: a new high resolution map derived from remote sensing, crop inventories and climate data. *Int. J. Appl. Earth Obs. Geoinf.* 38, 321–334. <https://doi.org/10.1016/j.jag.2015.01.014>.
- Schulte-Uebbing, L., de Vries, W., 2021. Reconciling food production and environmental boundaries for nitrogen in the European Union. *Sci. Total Environ.* 786, 147427. <https://doi.org/10.1016/j.scitotenv.2021.147427>.
- Serra, J., Paredes, P., Cordovil, C.M.D.S., Cruz, S., Hutchings, N.J., Cameira, M.R., 2023. Is irrigation water an overlooked source of nitrogen in agriculture? *Agric. Water Manag.* 278, 108147. <https://doi.org/10.1016/j.agwat.2023.108147>.
- Siebert, S., Burke, J., Faures, J.M., Frenken, K., Hoogeveen, J., Döll, P., Portmann, F.T., 2010. Groundwater use for irrigation - a global inventory. *Hydrol. Earth Syst. Sci.* 14, 1863–1880. <https://doi.org/10.5194/hess-14-1863-2010>.
- Spijker, J., Fraters, L., Vrijhoef, A., 2021. A machine learning based modelling framework to predict nitrate leaching from agricultural soils across the Netherlands. *Environ. Res. Commun.* 3, 045002. <https://doi.org/10.1088/2515-7620/abf15f>.
- Spinoni, J., Barbosa, P., De Jager, A., McCormick, N., Naumann, G., Vogt, J.V., Magni, D., Masante, D., Mazzechi, M., 2019. A new global database of meteorological drought events from 1951 to 2016. *J. Hydrol.: Reg. Stud.* 22, 100593. <https://doi.org/10.1016/j.jehrs.2019.100593>.
- Sutanudjaja, E.H., van Beek, R., Wanders, N., Wada, Y., Bosmans, J.H.C., Drost, N., van der Ent, R.J., de Graaf, I.E.M., Hoch, J.M., de Jong, K., Karssenberg, D., López López, P., Peßenteiner, S., Schmitz, O., Straatsma, M.W., Vannamete, E., Wisser, D., Bierkens, M.F.P., 2018. PCR-GLOBWB 2: a 5 arcmin global hydrological and water resources model. *Geosci. Model Dev.* 11 (6), 2429–2453. <https://doi.org/10.5194/gmd-11-2429-2018>.
- Uniyal, B., Dietrich, J., Vu, N.Q., Jha, M.K., Arumí, J.L., 2019. Simulation of regional irrigation requirement with SWAT in different agro-climatic zones driven by observed climate and two reanalysis datasets. *Sci. Total Environ.* 649, 846–865. <https://doi.org/10.1016/j.scitotenv.2018.08.248>.
- Valentini, R.S., Bouwer, L.M., Georgopoulou, E., Jacob, D., Martin, E., Rounsevell, M., Soussana, J.F., 2014. Europe. In: Barros, V.R., Field, C.B., Dokken, D.J., Mastrandrea, M.D., Mach, K.J., Bilir, T.E., Chatterjee, M., Ebi, K.L., Estrada, Y.O., Genova, R.C., Girma, B., Kissel, E.S., Levy, A.N., MacCracken, S., Mastrandrea, P.R., White, L.L. (Eds.), *Climate Change 2014: Impacts, Adaptation, and Vulnerability. Part B: Regional Aspects. Contribution of Working Group II to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 1267–1326.
- Vila-Traver, J., Aguilera, E., Infante-Amate, J., González de Molina, M., 2021. Climate change and industrialization as the main drivers of Spanish agriculture waters. *Sci. Total Environ.* 760. <https://doi.org/10.1016/j.scitotenv.2020.143399>.
- Villalobos, F.J., Delgado, A., López-Bernal, Á., Quemada, M., 2020. FertilizerCalc: a decision support system for fertilizer management. *Int. J. Plant Prod.* 14, 299–308. <https://doi.org/10.1007/s42106-019-00085-1>.
- Wada, Y., van Beek, L.P.H., Wanders, N., Bierkens, M.F.P., 2013. Human water consumption intensifies hydrological drought worldwide. *Environ. Res. Lett.* 8. <https://doi.org/10.1088/1748-9326/8/3/034036>.
- Wang, L., Stuart, M.E., Lewis, M.A., Ward, R.S., Skirvin, D., Naden, P.S., Collins, A.L., Ascott, M.J., 2016. The changing trend in nitrate concentrations in major aquifers due to historical nitrate loading from agricultural land across England and Wales from 1925 to 2150. *Sci. Total Environ.* 542, 694–705. <https://doi.org/10.1016/j.scitotenv.2015.10.127>.
- Wang, X., Feng, A., Wang, Q., Wu, C., Liu, Z., Ma, Z., Wei, X., 2014. Spatial variability of the nutrient balance and related NPSP risk analysis for agro-ecosystems in China in 2010. *Agric. Ecosyst. Environ.* 193, 42–52. <https://doi.org/10.1016/j.agee.2014.04.027>.
- Wang, X., Müller, C., Elliot, J., Mueller, N.D., Ciais, P., Jägermeyr, J., Gerber, J., Dumas, P., Wang, C., Yang, H., Li, L., Deryng, D., Folberth, C., Liu, W., Makowski, D., Olin, S., Pugh, T.A.M., Reddy, A., Schmid, E., Jeong, S., Zhou, F., Piao, S., 2021. Global irrigation contribution to wheat and maize yield. *Nat. Commun.* 12, 1–8. <https://doi.org/10.1038/s41467-021-21498-5>.
- Wick, K., Heumesser, C., Schmid, E., 2012. Groundwater nitrate contamination: factors and indicators. *J. Environ. Manag.* 111, 178–186. <https://doi.org/10.1016/j.jenvman.2012.06.030>.
- Wriedt, G., van der Velde, M., Aloe, A., Bouraoui, F., 2009. A European irrigation map for spatially distributed agricultural modelling. *Agric. Water Manag.* 96, 771–789. <https://doi.org/10.1016/j.agwat.2008.10.012>.
- Yao, Z., Yan, G., Wang, R., Zheng, X., Liu, C., Butterbach-Bahl, K., 2019. Drip irrigation or reduced N-fertilizer rate can mitigate the high annual N₂O + NO fluxes from Chinese intensive greenhouse vegetable systems. *Atmos. Environ.* 212, 183–193. <https://doi.org/10.1016/j.atmosenv.2019.05.056>.
- You, L., Wood, S., Wood-Sichra, U., 2009. Generating plausible crop distribution maps for sub-Saharan Africa using a spatially disaggregated data fusion and optimization approach. *Agric. Syst.* 99, 126–140. <https://doi.org/10.1016/j.agry.2008.11.003>.
- Yu, Q., You, L., Wood-sichra, U., Ru, Y., Joglekar, A.K.B., Fritz, S., Xiong, W., Lu, M., Wu, W., Yang, P., 2020. *Production Maps* 1–40.
- Zajac, Z., Gomez, O., Gelati, E., van der Velde, M., Bassu, S., Ceglaz, A., Chukaliev, O., Panarello, L., Koeble, R., van den Berg, M., Niemeier, S., Fumagalli, D., 2022. Estimation of spatial distribution of irrigated crop areas in Europe for large-scale modelling applications. *Agric. Water Manag.* 266. <https://doi.org/10.1016/j.agwat.2022.107527>.
- Zhang, X., Ren, C., Gu, B., Chen, D., 2021a. Uncertainty of nitrogen budget in China. *Environ. Pollut.* 281, 117216. <https://doi.org/10.1016/j.envpol.2021.117216>.
- Zhang, X., Zou, T., Lassaletta, L., Mueller, N.D., Tubiello, F.N., Lisk, M.D., Lu, C., Conant, R.T., Dorich, C.D., Gerber, J., Tian, H., Bruulsema, T., Maaz, T.M.C., Nishina, K., Bodirsky, B.L., Popp, A., Bouwman, L., Beusen, A., Chang, J., Havlík, P., Leclère, D., Canadell, J.G., Jackson, R.B., Heffer, P., Wanner, N., Zhang, W., Davidson, E.A., 2021b. Quantification of global and national nitrogen budgets for crop production. *Nat. Food* 2, 529–540. <https://doi.org/10.1038/s43016-021-00318-5>.
- Zheng, M., Zheng, H., Wu, Y., Xiao, Y., Du, Y., Xu, W., Lu, F., Wang, X., Ouyang, Z., 2015. Changes in nitrogen budget and potential risk to the environment over 20 years (1990–2010) in the agroecosystems of the Haihe Basin, China. *J. Environ. Sci. (China)* 28, 195–202. <https://doi.org/10.1016/j.jes.2014.05.053>.