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Key Points:

- We show how climatic features affect the export of N and P by rivers throughout a broad range of conditions, from wet temperate to arid
- In arid environments water management plays a key role increasing inland retention, particularly for N
- The association between climate and water management affects asymmetrically the fluxes of N and P and the N:P ratios in water and soils

Supporting Information:

Supporting Information may be found in the online version of this article.

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The Mediterranean Region as a Paradigm of the Global Decoupling of N and P Between Soils and Freshwaters

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Abstract The global socio-economic and agricultural expansion is accompanied by large inputs of nitrogen (N) and phosphorus (P) on land and by a serious alteration of the water cycle and water quality. The Mediterranean basin represents a paradigmatic region to study the entangled nutrient and water challenges because the region, where many of the world's climatic and socio-economic gradients are present, is predicted to suffer severe water stress in the coming decades yet at the same time agricultural intensification and population are increasing in many rim countries. We here describe the biogeochemical budgets of N and P in 549 river basins for the 2000–2009 period, analyzing how the climatic gradient and water management practices affect the fluxes of N and P and their stoichiometric ratios. Average land inputs are 3,600 kg N km^{-2} yr^{-1} and 470 kg P km^{-2} yr^{-1}, with a significant variation between basins (>100 times) closely related to the stage of agricultural intensification. Moreover, the combination of aridity and water regulation can strongly alter the final balances, not only by changing the export of nutrients by rivers (riverine export is ca. 10% for N and 8% for P in arid basins), but also decoupling the N:P ratios between terrestrial and freshwater compartments.

1. Introduction

Socio-economic development and agricultural expansion are accompanied not only by larger inputs of N and P but also by a serious alteration of the water cycle (Grill et al., 2019). This coupling between water and nutrient resources underscores the need to address food security and water security as interrelated challenges. Agriculture accounts for 69% of annual water withdrawals worldwide (FAO Aquastat). Some of the countries currently making major efforts to expand irrigation systems are in arid and semiarid regions of the world, which entails intensive groundwater abstraction and the construction of dams and reservoirs to secure the access to water resources. Reservoir storage is also the classic solution applied to contend with water scarcity in areas experiencing urban and industrial expansion (Gleick, 2003). How human water management may affect the stocks and fluxes of N and P is a question that has received little attention so far. Studies that deal with water and nutrients at once are usually performed at the small-scale, analyzing specific agricultural patches (e.g., Bartoli et al., 2012; Törnqvist et al., 2015), and focus on (1) increasing crop yields without compromising water resources; or (2) agricultural practices as a means of reducing nutrient loads and shortcutting water eutrophication. Furthermore, the vast majority of these studies concern wet temperate systems, which are those for which the most data exist, but very few provide data specific to arid and semiarid conditions. Arid and semi-arid regions constitute a significant fraction of the Earth's terrestrial ecosystems, and are expected to increase in upcoming years (IPCC 2014), so filling this data gap is of concern in light of future climate scenarios.

The Mediterranean basin offers a unique test-bed to assess the coupling of N, P with water management for several reasons. First, because it comprises a gradient of climatological regimes (Figure 1) and different

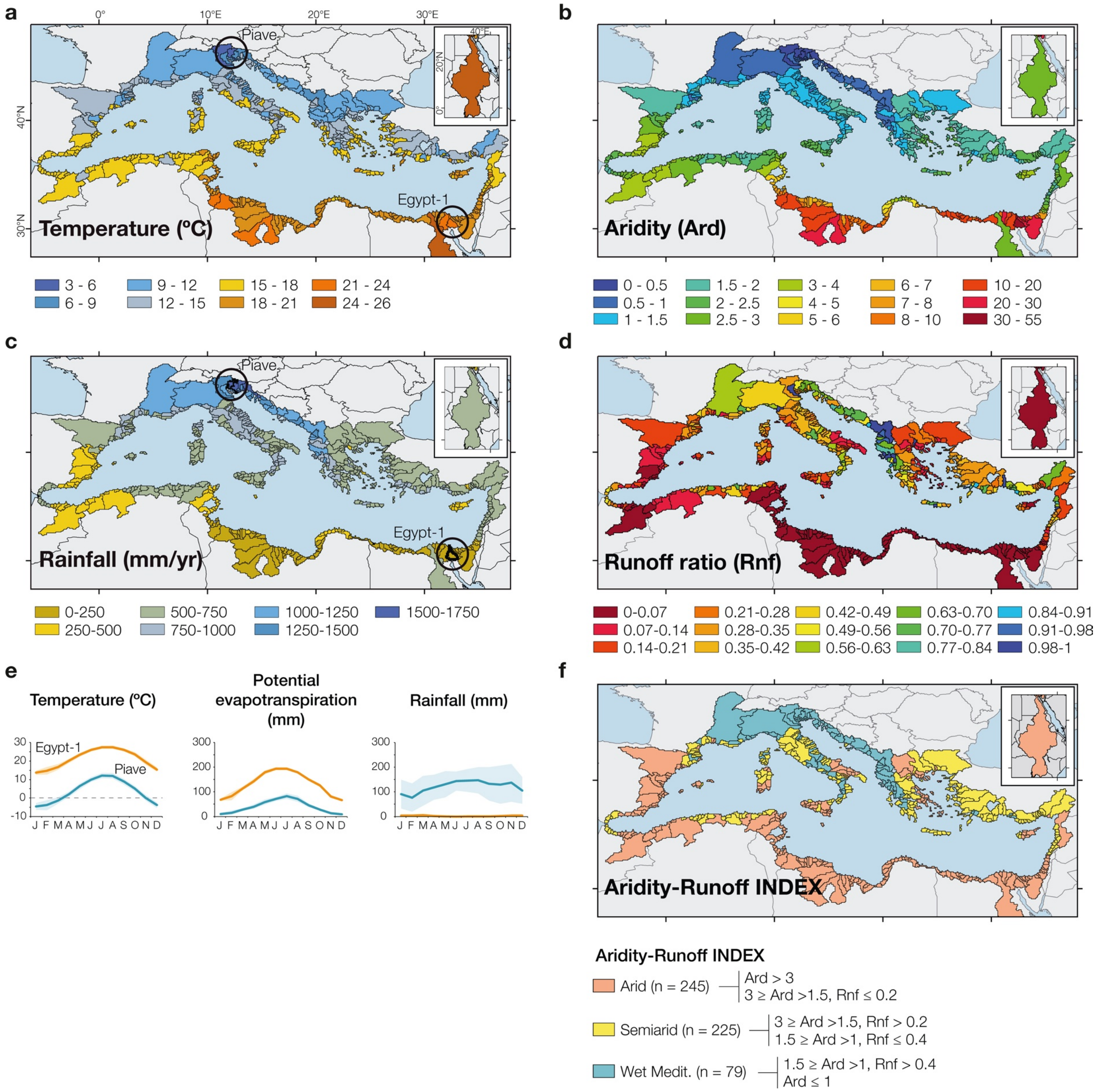

Figure 1. North-South climatic gradient. (a and c) Maps showing the area covered by the 549 Mediterranean river basins used in the study, and their corresponding temperature and rainfall values (annual averages for the 1960–2015 period). (e) The small plots show the temperature, potential evapotranspiration and rainfall during an average year in the coldest (Piave, blue line) and warmest (Egypt-1, orange line) basins (highlighted with circles), and therefore illustrate the maximum climatic range in the region. (b,d, and f) aridity (the ratio of potential evapotranspiration to rainfall), runoff ratio (the ratio of river flow to rainfall) and the combined aridity-runoff index for all Mediterranean basins. The thresholds used to categorize the rivers into arid, semiarid or wet Mediterranean are shown below. Further details on the calculations are described in the Methods section.

stages of industrial and agricultural development, lifestyles and diets, which allows analyzing the interplay between water and nutrients for a wide range of conditions. Second, Mediterranean countries are predicted to suffer severe water stress in the coming decades as a consequence of global warming (Cramer et al., 2018; Mekonnen & Hoekstra, 2016), yet at the same time population is rapidly increasing throughout the region. The growing water demand poses a threat to water resources and can severely alter nutrient loads in soils and freshwaters, and the transfers to the sea. Third, projected physical and socio-economic changes in the north and the south offer great opportunities to compare management options jointly addressing water and nutrient sustainability.

An estimate of the fluxes of N and P in the Mediterranean and Black Sea region was presented by Ludwig et al. (2010), although the differences between this previous work and the current study are remarkable. While Ludwig et al. (2010) focused on the temporal evolution of riverine nutrient fluxes, we concentrate on the spatial patterns. We present a much finer spatial resolution of the terrestrial data (from 0.5° × 0.5° to 5′ × 5′), which includes the small streams from mountainous coastal areas that had hitherto been disregarded, and we have expanded the riverine data set. We include data from more than 100 rivers, including some from the south, so that the geographical coverage is broad and representative of the entire basin. Our main interest is not only to quantify the total export of nutrients from rivers to the Mediterranean Sea in a recent period, but especially to analyze land-based sources, how are those related with the N and P input-output balances, and how they change geographically as a function of climate, physical and socio-economic factors, and this is new with respect to previous approaches.

Finally, together with resource sustainability, the decoupling of the N and P cycles is of paramount importance. We use the concept of nutrient decoupling to refer to the increasing asymmetries in the flows and stocks of N and P in the different terrestrial compartments. This was shown at the global scale by Peñuelas et al. (2012, 2013), who attributed the growing imbalance between N and P to human activities, primarily to the mismatch between the use of N and P fertilizers. P is less mobile than N so its dispersal across surrounding natural ecosystems is more difficult (Bouwman et al., 2017; Peñuelas et al, 2012, 2013). On the other hand, P fluxes to rivers from erosion are a key and poorly quantified input to freshwaters (Mekonnen & Hoekstra, 2018). Advance in the knowledge of the links between different hydrological conditions and N, P fluxes is essential. Nutritional imbalances are already changing the structure and function of ecosystems, and can reduce their capacity to store CO_2, thus contributing to intensify global warming (Peñuelas et al., 2013, 2017; Sardans et al., 2012). The combined study of the water, N and P cycles in river basins in the Mediterranean area, where some of the world's climatic and socio-economic gradients are present, is a unique opportunity to advance in understanding the causes of this nutrient decoupling.

We here describe the biogeochemical budgets of N and P in 549 river basins for the 2000–2009 period. Our aims are threefold: (1) providing a high-resolution spatial characterization of the N and P fluxes in the entire Mediterranean area; (2) examining how the different climatic conditions and water management practices affect the fraction of N and P retained on land; and, importantly, (3) analyzing how these differences in retention and export of nutrients alter the N:P stoichiometric ratios of terrestrial and freshwater compartments, ultimately affecting the receiving coastal waters.

2. Materials and Methods

2.1. Study Scale and Working Units: Mediterranean River Basins

We used river basins as our main territorial units and studied all the basins draining to the Mediterranean Sea. River basins integrate natural, agricultural, and urban landscapes as well as water resources, and hence represent valuable units for comprehensive impact assessment studies. Moreover, as argued by Garnier et al. (2015), they are advantageous units from a management perspective, because (1) for historical reasons, river basins tend to correspond to the influence area of one or more large cities acting as hubs of regional economic activities; and (2) in many cases, river basins already count on administrative structures devoted to water resource management. This is the case in European Union countries (River Basin Management Plans are the instruments for implementing water policy as established in the EU Water Framework Directive, 2000/60/EC), but it is also the case in many North African countries (e.g., Morocco, Algeria or Tunisia), where water resources are customarily managed by river basin authorities.

We include a total of 549 river basins, covering an area of about 4.73 million km^2 (Figure 1). River basins were delineated at 5 arc-min resolution and correspond to those recently described by Sadaoui, Ludwig, Bourrin, and Romero (2018b), who provide thorough details on the spatial delimitation of the basins as well as the river routing procedure. In brief, the approach relies on digital elevation models and flow algorithms that link each grid cell to the lowest neighbor grid cell to produce drainage basin contours and hydrological networks. Special care was taken to ensure that river routing was correct and that all grid points in a given drainage basin ended up in one single grid cell—which represents the river mouth—, so a thorough verification involving hundreds of manual corrections was performed on the flow direction grids. The final spatial resolution is 5 × 5 arc-min, which is optimal for capturing the small coastal rivers that are so characteristic of the Mediterranean region and so often neglected in large-scale studies; excluding the Nile, about 40% of the total area in the Mediterranean basin is covered by coastal rivers of less than 5,000 km^2 (Sadaoui, Ludwig, Bourrin, & Romero, 2018b).

2.2. North-South Climatic Gradient

The climate of the studied region moves from a wet Mediterranean climate in the north (in transition to temperate climate) to an arid climate in the south, in all cases characterized by dry and hot summers, mild winter temperatures and recurrent drought periods (Lionello et al., 2006). Beyond these general patterns river basins show a wide range of temperature and rainfall conditions. Part of this variability is due to the high mountain ridges surrounding the Mediterranean Sea, which cause much sharper climatic features than those expected for geographically close areas.

Climatic data—monthly averages for precipitation, temperature and potential evapotranspiration, from 1960 to 2015—were derived from the Climatic Research Unit data sets (CRU TS v4.00, http://www.cru.uea.ac.uk/). CRU data sets are validated with real observations from an extensive network of weather stations. Detailed maps with the station coverage are shown in Harris et al. (2020).

There are remarkable differences between the north and the south. Temperature increases and precipitation decreases following a northwest-to-southeast direction, with temperate-like river basins being located close to the Alps and along the eastern coast of the Adriatic Sea, and the most extreme dry and hot conditions being found in Libya and Egypt. The Piave River, in Italy, is the coldest and wettest basin in the area. The Piave shows average temperatures of −5°C to 12°C and rainfall values of 77–147 mm per month, evenly distributed throughout the year; in contrast, the temperature range in the hottest basin (a small Egyptian river in the vicinity of the Nile Delta) goes from 11°C to 30°C—nearly 3 times the range of the Piave—, and precipitation barely reaches 0.1–5 mm per month, with very scarce or no rainfall at all during the summer months.

We used the precipitation, temperature and potential evapotranspiration data sets to derive two additional hydro-climatic variables: the aridity (Ard) and the runoff ratio (Rnf). We define Ard as the ratio of potential evapotranspiration to rainfall, and Rnf as the ratio of river flow to rainfall (all in mm/yr). Ard averaged 3.0 for all Mediterranean basins (median Ard = 1.9), and the mean Rnf value was 0.28 (median Rnf = 0.23), meaning that in most basins potential evapotranspiration is much larger than total rainfall and river exports represent less than 30% of the precipitation in the catchment. Additionally, to split the rivers into consistent hydro-climatic groups we established a joint aridity-runoff Index based on the combination of the Ard and Rnf values (Figure 1). We defined three categories: arid, semiarid, and wet Mediterranean. The runoff ratio implicitly includes the use of water for human activities, and therefore those rivers in which water management is very intensive can appear as arid even though their climatic conditions may not be so extreme (e.g., the Ebro River).

2.3. Terrestrial Inputs

We describe the biogeochemical budgets of N and P in the 549 basins, calculating how much N and P enters the territory and how much is exported by the rivers to the sea. Retention is defined as the difference between land inputs and river outputs (Lassaletta et al., 2012). This encompasses a fraction that may be stored, or partially or fully removed by subsequent processes within the soil and freshwater compartments, but it gives a good idea of the share of N and P inputs that is accumulated and processed on terrestrial systems versus that exported to marine systems.

To account for the terrestrial inputs we applied the soil budget (SB) approach (Bouwman et al., 2009; de Vries et al., 2011), a method that has been successfully used at both small-scale and large-scale studies (Bouwman et al., 2013, 2017). Briefly, the SB computes all the N and P inputs entering agricultural and natural soils, including synthetic fertilizers, manure application, biological fixation, and total atmospheric N deposition (the latter two only for N). The difference between these inputs and the outputs via harvested crops, hay and grass cutting, and grazing, corresponds to the soil nutrient surplus, which represents a potential source of diffuse pollution to the water and the atmosphere. Soil surpluses are added to the flux of N and P in sewage waters (point sources) to obtain the total amount of nutrients entering the catchments (Equations 1–4).

$$\begin{aligned}\text{N Soil surplus} &= \text{Synthetic Fertilizers} + \text{Manure} + \text{Biological N Fixation}\\ &\quad + \text{Atmospheric N Deposition} - \text{Crop harvest}\end{aligned} \tag{1}$$

$$\text{N Land Inputs} = \text{N Soil surplus} + \text{Sewage} \tag{2}$$

$$\text{P Soil surplus} = \text{Synthetic Fertilizers} + \text{Manure} - \text{Crop harvest} \tag{3}$$

$$\text{P Land Inputs} = \text{P Soil surplus} + \text{Sewage} \tag{4}$$

Data sources for each term of the budget are described below. To obtain spatialized fluxes at the desired 5 arc-min resolution SB calculations were performed via the IMAGE-Global Nutrient Model (Beusen et al, 2015, 2016; Bouwman et al., 2017), an integrated assessment model that combines nutrient data with population features and land use cover layers to provide spatially explicit results. Details on the model assumptions, the processing of data and the spatial distribution of the budget terms are provided in Stehfest et al. (2014). We focused on the 2000–2009 period. Results are always expressed per square kilometre to allow for comparisons between river basins regardless of their size.

A previous version of the model was used by Ludwig et al. (2010), but as mentioned in the Introduction, there are remarkable differences between this previous work and the current study. First, we have greatly improved the spatial resolution of the terrestrial data and the river network, from $0.5° \times 0.5°$ to a much finer $5' \times 5'$. This means that we were able to correctly represent many of the small coastal streams typical of the Mediterranean basin. Second, we have expanded the riverine data set, so instead of the 37 rivers that were previously used by Ludwig et al. (2010) to derive past and future trends, we include here actual data from more than 100 rivers to have a wide and representative geographical coverage. Specifically, we compiled discharge data for 130 rivers—of which 88 correspond to arid and semiarid basins—, and nutrient concentration measurements for 114 rivers (total nitrogen) and 107 rivers (total phosphorus).

2.3.1. Diffuse Sources (Soil Surplus)

Data on crop production and N, P fertilizer use by crop were retrieved from the Food and Agriculture Organization of the United Nations (FAOSTAT database collections). Animal manure inputs were calculated using FAOSTAT livestock production statistics and constant N, P excretion rates per head and animal type (Bouwman et al., 2009; Van der Hoek, 1998). Fertilizers and manure were distributed spatially over arable land and grassland as described in Bouwman et al. (2009). The model assumes that, in industrialized countries, 50% of the available manure is applied to arable land and the rest to grassland, while in developing countries, 95% of the available manure is applied to cropland and 5% to grassland. For EU countries the model sets maximum application rates of 17,000–25,000 kg N km^{-2} yr^{-1} based on existing regulations.

Biological N fixation (BNF) by leguminous crops (pulses and soybeans) was determined from crop production data and N content from the FAOSTAT database. In addition to leguminous crops, IMAGE-GNM considers an annual rate of BNF of 500 kg N km^{-2} for non-leguminous crops and grass, and 2,500 kg N km^{-2} for wetland rice. N fixation rates in natural ecosystems were based on the low estimates for areal coverage by legumes (Cleveland et al., 1999), as described by Bouwman et al. (2013).

Atmospheric N deposition rates (including dry and wet deposition of NH_x and NO_y) were obtained from an ensemble of atmospheric chemistry-transport models (Dentener et al., 2006). We do not consider atmospheric P deposition.

Nutrient uptake in harvested products was based on N, P content for each crop and crop production figures from FAOSTAT; data were then aggregated to broad crop categories (wetland rice, leguminous crops, upland crops and energy crops). IMAGE-GNM also accounts for uptake in grasslands and fodder crops; concerning the latter, N withdrawal through grazing and harvest is assumed to amount to 60% of all N inputs (manure, fertilizer, deposition, N fixation), and P withdrawal through grazing and harvest is calculated as a proportion of 87.5% of fertilizer and manure P input.

2.3.2. Point Sources (Sewage)

To calculate N and P in urban wastewater we used the model described in Morée et al. (2013). It is essentially an updated version of the one presented by Van Drecht et al. (2009) that uses FAO data on dietary protein consumption to estimate human excretion of N and P, population statistics at the national level, urban and rural population data, estimates on the fraction of population connected to sewage systems, and the percentage of N and P removal in wastewater treatment facilities (WHO/UNICEF, 2000). P-based detergents were assumed to be used exclusively in households connected to the sewerage network, and estimates were based on the calculations by Van Drecht et al. (2009).

2.4. River Outputs to the Coastal Area

2.4.1. River Flow

River discharge was calculated using a multiple regression model that considers hydro-climatic and environmental parameters (namely, the basin slope, the ratio between runoff and precipitation, and the Fournier index, an index that quantifies seasonality of precipitation during the year). Climatic data for the 1960–2015 period were extracted from the gridded data sets provided by the Climatic Research Unit (CRU TS v4.00, 0.5° resolution, http://www.cru.uea.ac.uk/). We applied linear interpolation techniques between the centers of the 0.5° grid cells to transform the data into 5′ grid point resolution layers. Further details on the method and the precise equations can be found in Sadaoui, Ludwig, Bourrin, and Romero (2018b). The model was tested and validated using data on river flow collected from the existing literature. Overall, long-term average flows for 130 rivers were assembled. These rivers were representative of the different Mediterranean regions and covered over 85% of the total freshwater discharge to the Mediterranean Sea. In particular, 88 basins corresponded to arid and semiarid conditions, and 39 of them were located in the eastern and southern Mediterranean. Most of the missing data are from areas of North Africa (Libya) where runoff is assumed to be almost zero.

The model proved to work well at both the basin scale and the individual grid cell scale. The coefficient of determination (R^2) between the observed and the modeled water discharge values is about 0.9 ($R^2 = 0.93$ at the basin scale, and $R^2 = 0.89$ at the grid cell scale). In addition, our model accurately captures the flow of small coastal streams: 85 out of the 130 river basins used for validation corresponded to coastal basins $<5{,}000$ km^2 (Sadaoui, Ludwig, Bourrin, & Romero, 2018b), contrary to what happens with most of the global river discharge models when applied to the Mediterranean region.

2.4.2. Freshwater Quality

Nutrient concentrations in rivers (nitrate, ammonium, phosphate, total nitrogen and total phosphorus) were gathered from the literature, official reports, and from different water authorities. The corpus of the data set corresponds to the compilation described in the PERSEUS reports (http://www.perseus-net.eu; Ludwig and Montreuil, D4.3), with additional updates. On the whole, we gathered information on water quality for over 100 Mediterranean rivers (114 for N, and 107 for P); approximately 60% of the measurements corresponded to arid and semiarid basins (71 for N; 64 for P), and 20% were obtained from basins in the East and South Mediterranean (21 for N; 14 for P), an area barely represented in the literature. Please notice that we used only actual nutrient measurements to calculate the riverine fluxes, so unlike the terrestrial inputs that are available for the 549 basins, the river outputs were available for a maximum of 114 and 107 basins (for N and P, respectively).

To have the best possible estimate of river export we systematically selected those monitoring stations that were closest to the river mouth and where the time series was at least a few years long. In most cases the data consisted of monthly measurements of inorganic nutrients. To be consistent with the terrestrial fluxes we used data from the 2000–2009 period. When this was not possible, the complete time series (from the 1970s to 2013) was used to calculate an average concentration. We removed a few values (7 in the case of P, 13 in the case of N) because the observed concentrations were disproportionately high and the resulting riverine fluxes were as high as, or higher than, the total terrestrial inputs.

We used the concentrations of total nitrogen (TN) and total phosphorus (TP) to calculate the nutrient export fluxes. In 41 rivers, however, only nitrate measurements were available. When that happened, we used the correlation between nitrate and TN for all Mediterranean rivers during the 2000–2009 period to estimate the TN value. We found that $TN = 1.28 \times N - NO_3 + 0.73$ ($N = 64$, $R^2 = 0.47$), which is very similar (slightly higher) to the formula obtained by Garnier et al. (2010) for the world's rivers. This is consistent with Romero et al. (2013), who found that the equation by Garnier et al. (2010) underestimated TN in Mediterranean rivers (concentrations of TN obtained with the equation accounted for approximately 75%–85% of measured concentrations). Nutrients exported by rivers were calculated as the product of average nutrient concentrations of TP and TN, and the average annual river flow.

TN concentration at the outlets of Mediterranean rivers during the 2000–2009 period is ~3.6 mgN l^{-1} (median = 2.7, Q1–Q3 range = 1.5–4.3), while the concentration of TP is 0.25 mgP l^{-1} (median = 0.13, Q1–Q3 range = 0.06–0.28). The highest values correspond to rivers in Algeria, Turkey, Israel, and Egypt, where TN and TP concentrations can reach 3 to 4-fold the Mediterranean average. Nitrate represents about 62% of TN, while phosphate accounts for ca. 58% of TP. Although nitrate makes up the majority of TN, the fraction of ammonium is large (>50%) in river basins that drain extensive urban areas or in basins that host cities located very close to the coast.

The average concentrations here reported for the entire Mediterranean basin are partly biased by the European rivers, where monitoring is more frequent and therefore where most of the available data comes from (approx. 70% of the total measurements). Freshwater quality variables in many southern catchments are either not measured or completely inaccessible, and despite the existence of some quality measurements in stations at the outlet of reservoirs, these are located upstream in the catchment so that the values cannot be used to assess the export fluxes to coastal seas. On the basis of the available data, however, we believe that nutrient concentrations in southern Mediterranean catchments are rather high (of the order of the concentrations found in Turkish and Moroccan streams) since there are numerous spills of untreated waters coming from urban and rural areas. Ultimately, this means that the average nutrient export fluxes calculated below are likely to be at the lower bound of actual values.

2.5. Water Management Features

Calculations on water storage capacity were derived from the data presented in Sadaoui, Ludwig, Bourrin, and Romero (2018b). Essentially, information on dams and reservoirs in Mediterranean rivers come from the Global Reservoir and Dam (GRanD) database of Lehner et al. (2011). GRanD comprises 378 dams for the entire Mediterranean drainage basin, yet due to missing information and/or too small reservoir sizes, only 319 were retained. Specifically, we excluded the reservoirs with a storage capacity smaller than 1.6 E+06 m^3 because (1) their contribution to the total storage capacity in the basins was minor; and (2) several small reservoirs fell within one single 5-arc-min cell, in which case we only retained the largest reservoir in a given grid point cell. This means that the water storage capacity here considered is on the lower range, because the Mediterranean is characterized by a remarkable amount of weirs that divert water to small impoundments (Sadaoui, Ludwig, Bourrin, & Bissonnais, et al., 2018). In the Ebro, for instance, the present study considers 38 dams and an overall storage capacity of 7.22 E+09 m^3, yet in a previous work where accurate regional information could be retrieved (cf., Romero et al., 2016a) we calculated a total of 193 dams and weirs (5-fold the number of large infrastructures) and an accumulated storage capacity of 7.40 E+09 m^3 (i.e., only +2.6% higher than the capacity considered by Lehner et al., 2011). Given that the total water storage capacity is well-represented by the large reservoirs and information on small-size infrastructures is very difficult to obtain for most river basins, to be consistent across basins and allow for comparisons we have used the information provided in Lehner et al. (2011).

Water storage capacity was used to calculate the impounded runoff (IR) index, an indicator developed by Batalla et al. (2004) to assess the degree of hydrologic alteration caused by water management practices. The IR index is computed as the total capacity divided by the river's mean annual discharge. According to Batalla et al. (2004), unimpaired rivers with natural flow conditions have IR = 0 years; rivers with a few dams, where the regulatory capacity does not exceed the mean annual water yield (water stocked is effectively used on an annual cycle) present IR < 1 year; and rivers with IR > 1 year are exposed to strong flow regulation, with multi-year regulatory strategies to face recurring and persistent droughts. Although following the description by Batalla et al. (2004), the IR index is expressed as a dimensionless decimal fraction, the index

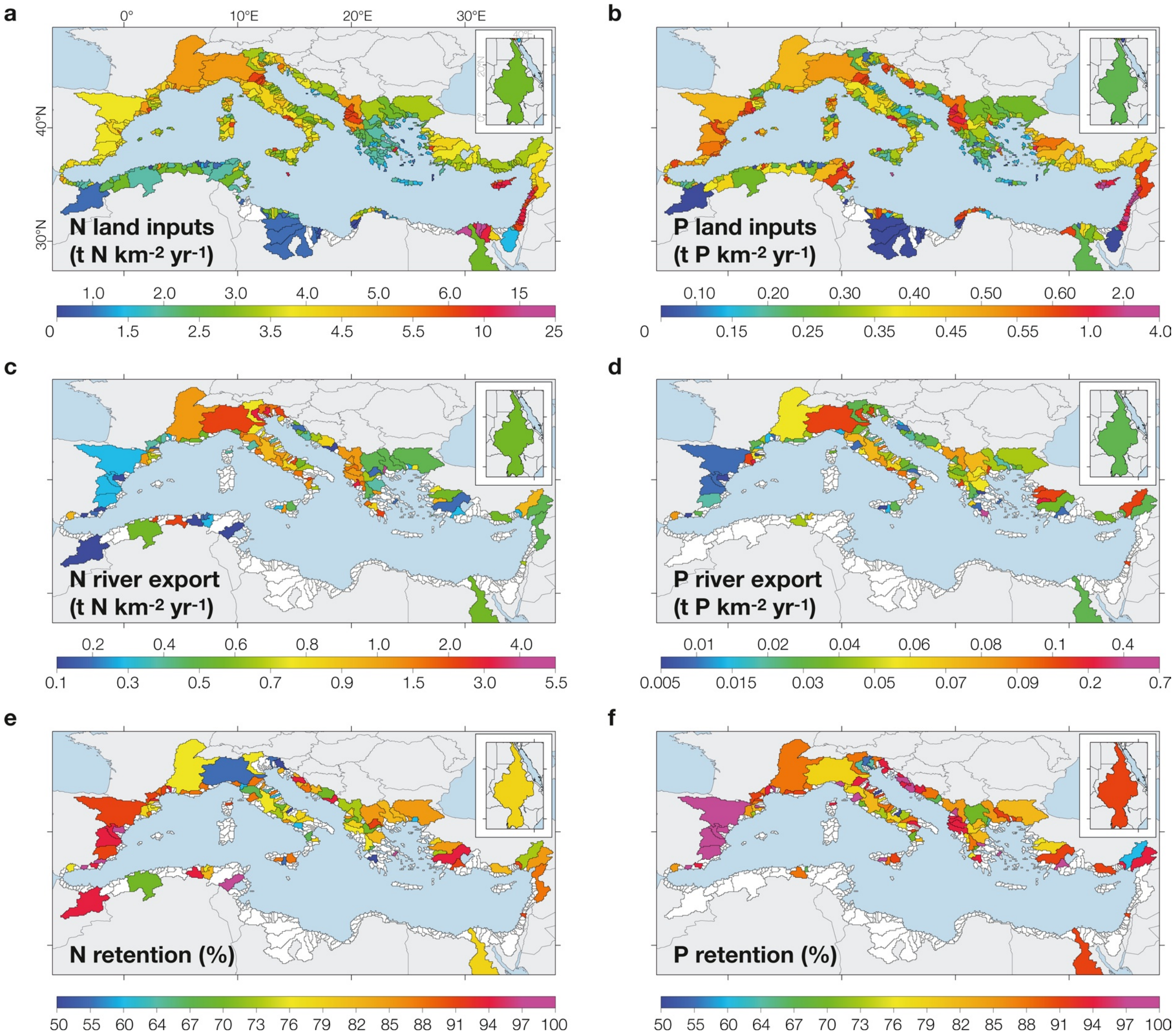

Figure 2. N and P budgets for the 2000–2009 period. (a and b) Nitrogen and phosphorus land inputs per basin (land inputs = fertilizers + manure + N_2-fixation + atmospheric deposition + sewage − crop uptake), (c and d) riverine export to coastal seas, and (e and f) the corresponding retention on land (land inputs − river export). Data are spatialized per square km so that basins can be directly compared.

is indeed a proxy of the holdup capacity of the basin, and it is calculated based on annual variables, meaning that years are the underlying units.

3. Results

3.1. Land Inputs in the Mediterranean Region

The North-South hydro-climatic gradient is paralleled by differences in the N and P terrestrial inputs. Total land inputs are the sum of diffuse and point sources minus the nutrient removal by crop harvest. Overall, total land inputs of N are remarkably higher in the northern and eastern basins than in the south, with ~3,500 kg N km^{-2} yr^{-1} in European countries and ~10,000 kg N km^{-2} yr^{-1} around Israel, Lebanon or Egypt (Figures 2, S1, and S2). The lowest inputs are consistently found in Libya and Algeria, where total N fluxes hardly exceed the 2,000 kg N km^{-2} yr^{-1} and can be as low as 150 kg N km^{-2} yr^{-1}. The geographical patterns are similar for P, although north-south differences are less marked and P inputs are roughly 10-fold lower than those of N.

Of the total inputs, agriculture, and in particular the use of synthetic fertilizers and manure, is the main terrestrial source of N and P in the entire Mediterranean basin. In the east, synthetic fertilizer inputs (hereafter, referred to as fertilizers) average 3,026 kg N km^{-2} yr^{-1} and 452 kg P km^{-2} yr^{-1} (ca. 65 kg N ha^{-1} and 15 kg P ha^{-1} of arable land), but the range is huge, from <100 kg N km^{-2} yr^{-1} and <50 kg P km^{-2} yr^{-1} in north Sinai basins to 32,000 kg N km^{-2} yr^{-1} and 2,350 kg P km^{-2} yr^{-1} (>300 kg N ha^{-1} and >25 kg P ha^{-1} cropland) in the Nile floodplain. In the northern rim countries –including Spain, France, Italy, and the Balkan Peninsula—fertilizer inputs are ca. 2,000 kg N km^{-2} yr^{-1} and 350 kg P km^{-2} yr^{-1} (67 kg N and 12 kg P ha^{-1} cropland). In the south (i.e., Morocco, Algeria, Tunisia, and Libya) fertilizer inputs average 678 and 224 kg km^{-2} yr^{-1} of N and P, respectively (~23 kg N and 7 kg P ha^{-1} of arable land), and maximum values are found in Tunisia (2,300 kg N km^{-2} yr^{-1} and 700 kg P km^{-2} yr^{-1}). The inputs of manure are on the same order than synthetic fertilizers (about 2,400 kg N km^{-2} yr^{-1} and 400 kg P km^{-2} yr^{-1} on average for the entire region), and the large-scale spatial patterns are also consistent with those of fertilizer inputs: values are maximum in the east, medium in the north, and much lower in the south (Figures S1 and S2).

Interestingly, the ratio between manure and fertilizers differs across regions: in the intensive agricultural regions of the eastern Mediterranean and around the Nile the use of fertilizers is 3-fold that of manure for both N and P, while in European countries the ratio of fertilizers to manure in 2005 was around 0.8 for both N and P, and this value is likely to be lower at present with the implementation of agricultural practices favoring manure recycling. In the south, where total inputs are low, the ratio is <1 for N and P in most basins.

There are also large north-south differences in the atmospheric deposition of N, which is much greater in the north of the Mediterranean than anywhere else, and in the biological N_2-fixation, which is high in European countries but reaches its maximum around the Nile delta (~1400 kg N km^{-2} yr^{-1}) due to a higher share of fodder and grain legumes in the crop mix. The crop mix and the type of agronomic practices also influence the crop uptake of N and P—a removal term in the nutrient budget—. Crop uptake averages 44% for N and 54% for P in the entire Mediterranean region, and is highest in Italy, France, Greece and around the Nile (Figures S1 and S2), where it can represent the equivalent of up to 84% (N) and 100% (P) of all diffuse nutrient inputs to the soil.

Point sources of N and P do not show clear regional patterns but rather track major Mediterranean cities. Sewage typically represents a small part of the total inputs per basin (the median value is 5% for N and 9% for P), but it can be important in small coastal basins close to large urban areas, such as some in Greece, Libya or Algeria, where sewage makes up >80% of the total N, P inputs in the catchment.

3.2. River Export and Inland Retention

Mediterranean basins export, on average, approx. 1030 kg N km^{-2} yr^{-1} and 63 kg P km^{-2} yr^{-1}, which correspond to about 25% and 12% of the N and P terrestrial inputs, respectively (Figure 2). These average export values are on the order of the export fractions reported for other areas of the world (e.g., Bennett et al., 2001; Billen et al., 2011; Hong et al., 2012; Howarth et al., 1996, 2012; Russell et al., 2008). Two other features stand out from the results: (1) the retention of P is consistently higher than that of N—as shown, for instance, in wetter basins of the Baltic (Hong et al., 2017 and references therein); and (2) the fraction of the total inputs exported by rivers is heterogeneous throughout the Mediterranean: high nutrient inputs around the Adriatic, for instance, are coincident with high riverine fluxes, but high N and P inputs in Spanish and Turkish basins concur with very low export values.

Differences in the percentage of inland retention of N roughly follow the climatic gradient. For the same amount of N inputs, export fluxes decrease (and retention increases) as we move to warmer and drier conditions, while these differences are not apparent in the case of P, for which river exports are systematically low regardless of the amount of land inputs and the hydro-climatic characteristics (Figure 3).

Flow regulation had a moderate effect on the retention of N—that is, highly regulated rivers had higher percentages of N retention—whereas no effect was observed for P. Interestingly, when splitting the basins into arid, semiarid, and wet Mediterranean it was clear that regulation practices were principally affecting retention in arid basins, while their role was minor in the other conditions (Figures 3c and 3d). The results of principal component analysis (PCA) confirmed a major role of climate and water management practices for N retention, and to a lower extent, for P retention (temperature, aridity, the IR index and the percentage of irrigated croplands have all positive loadings on component 1, opposite to the runoff ratio), and pointed

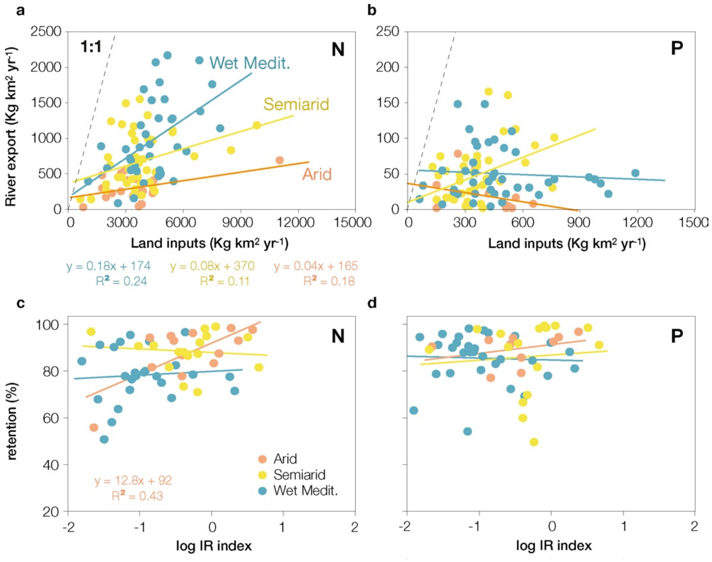

Figure 3. Retention in relation to the hydro-climatic characteristics of the basins. (a and b) River exports of N and P versus the land inputs of both elements. (c and d) The percentage of N and P retention in the river basins plotted against the impounded runoff (IR) index. The IR index is a measure of the total capacity of the reservoirs with regard to the average annual discharge, and therefore represents a proxy of the degree of flow regulation in the basin. The equations of significant regressions are included below. River basins are colored according to their hydro-climatic features (see text for details) and the gray dotted line corresponds to the line 1:1.

at agricultural issues as a secondary determining factor (component 2 is driven by the percentage of agricultural land within the basin, and fertilizers) (Figure 4).

3.3. N to P Mass Ratios in the Different Compartments

On average for the entire Mediterranean basin, the N:P mass ratio of the land inputs is 11. The range of variation is narrow (Q1–Q3 = 7–11), although large N:P values (>50) appear in some southern basins (Figure 5). The N:P mass ratios of the total inputs are spatially fully consistent with the N:P of the soil surplus (average N:P = 15), again pointing at the prevailing role of agricultural practices in the overall N and P budgets. The N:P of fertilizer and manure inputs is around 5, and the N:P ratio in harvested crops (crop uptake, Figure 5) is around 9. There are some peak values (N:P~90) in harvests in Libya, likely related to lower P fertilization rates and the abundant presence of nitrogen-fixation crops; the lowest N:P ratios in harvested crops (N:P <5) are found in Western European basins, Israel, Lebanon, and Egypt, all of them heavily fertilized areas.

In rivers, N:P mass ratios average 31 (median = 20; Q1–Q3 = 11–31). The lowest N:P values are found in rivers around the Aegean Sea, and many of them correspond to basins that drain extensive urban areas and receive a disproportionate amount of poorly treated sewage at their lowermost stretches (wastewaters have N:P ratios of 4–7 and represent ca. 80% of the total inputs in those basins).

On average, the N:P of the retained fraction is ca. 10% lower than the N:P of the land inputs (N:P ~ 10) (Figure 5e). Interestingly, unlike the PCA results for N and P, PCA analysis for the N:P ratio in the retained fraction show that it is not directly related to the climatic conditions but it is instead mainly determined by the inputs of fertilizers, sewage and manure (the latter, related to low N:P ratios), and by the presence of large reservoirs in the basins.

4. Discussion

The Mediterranean basin is home to an exceptional combination of densely populated areas, with some of the world's oldest historic human settlements, and extraordinarily diverse ecosystems. This close intimacy

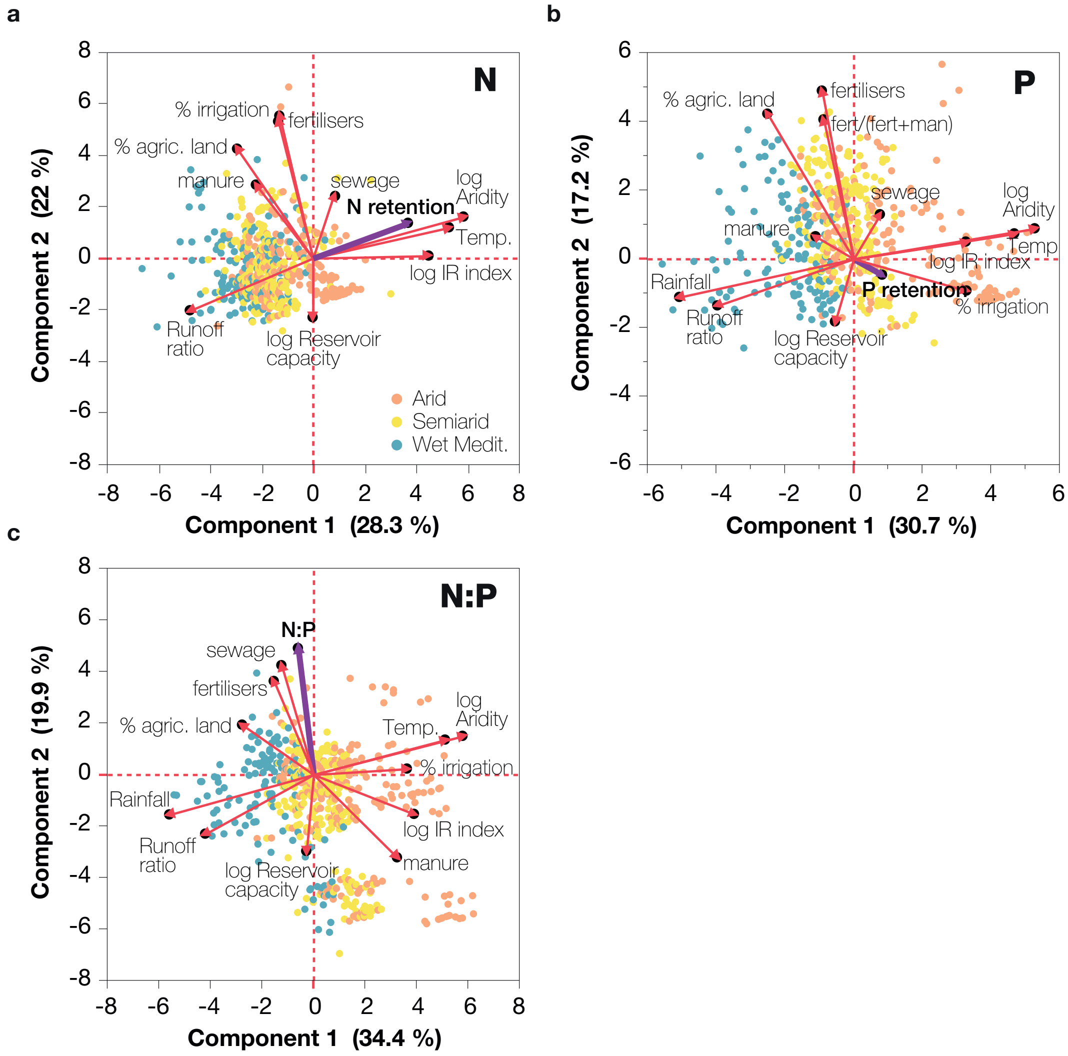

Figure 4. Main factors related to N and P retention. Principal Component Analysis for (a) N data, (b) P data, and (c) the two N, P data sets, and the N:P ratio of the inland retained nutrients. Climatic factors (temperature, rainfall, runoff ratio, aridity), water management factors (reservoir capacity, IR index, % irrigation), and terrestrial factors (synthetic fertilizers, manure, % of agricultural land, sewage) were considered in the analyses. River basins are colored according to their hydro-climatic features.

between humans and nature has led to many modifications in natural systems that are of great interest for environmental studies. Moreover, climate change in the Mediterranean Basin exceed global trends for most variables: basin-wide, annual mean temperatures are now 1.4°C above late-nineteenth-century levels (Cramer et al., 2018), and future warming is expected to exceed global rates by 25%, notably with summer warming at a pace 40% larger than the global mean (Lionello & Scarascia, 2018).

In the southern part of the Mediterranean basin, current water withdrawals can reach up to >75% of the total renewable water resources (WWAP, 2016), and projected warming together with urban development and agricultural intensification will likely require larger amounts of water and higher nutrient inputs.

4.1. Agriculture Intensification as a Driver of N, P Geographical Differences

Overall, we calculate that the N, P land inputs to the entire Mediterranean basin are 13.9 Tg N/yr and 1.4 Tg P/yr, and circa 2.6 Tg N/yr and 0.12 Tg P/yr are exported to the Mediterranean Sea by rivers. We consider only those rivers for which we have quality data (that cover 84% of the Mediterranean basin and account for

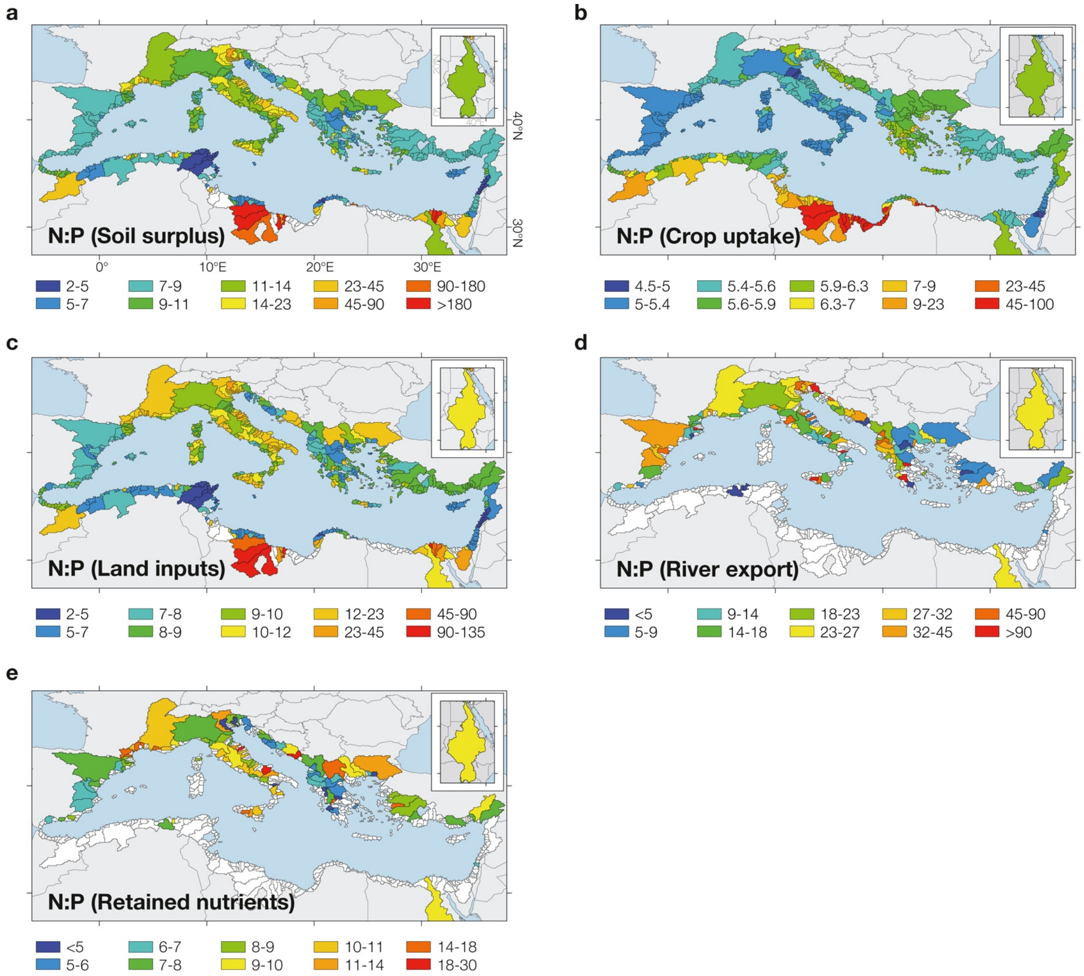

Figure 5. N:P (mass) ratios of several budget components. The N:P of the soil surplus refers to the N:P of the diffuse sources. The N:P of the retained nutrients refers to the total amount of nutrients that remains and/or is processed on land, that is, the land inputs (diffuse + point sources) minus the river export to coastal seas.

~85% of the total water discharge), so the actual export figures could be somewhat higher. Our calculated fluxes are in fair agreement with the recent estimations by Malagò et al. (2019), who applied the GREEN model and calculated 11.4 Tg N/yr and 1.1 Tg P/yr as total inputs, 1.9 Tg N/yr and 0.11 Tg P/yr as river exports, which makes us feel confident on the large-scale balance calculations and the related ratios here discussed.

Fertilizers and manure are the largest sources of nutrients on land, as it has been shown for many other world regions (Bouwman et al., 2009; Potter et al., 2010 and references therein), but our spatial results show great differences in the N, P inputs between the basins of the north and the south Mediterranean, and especially those of the east, and we deem they are tightly linked to agricultural practices and their evolution over time. During the early stages of agricultural intensification there is an increase in machinery, a peak in the application of chemical fertilizers—which are used more often than manure and are often overly applied without taking into account the plant and soil requirements—, and excessive tillage (Lassaletta et al., 2014; Zhang et al., 2015). This is what happens in many eastern river basins and around the Nile Delta, where enormous fertilizer and manure inputs (on average, fertilizer inputs are two to three-fold higher than manure) match only moderate values of crop withdrawal, resulting in very high agricultural surpluses—and also suggesting large nutrient losses. In Europe, the high nutrient status of soils as a result of many years of over-fertilization and the implementation of successive common agricultural policy (CAP) reforms has encouraged the adoption of good practices and has allowed a progressive reduction in the use of chemical fertilizers (Good & Beatty, 2011). In fact, current values of fertilizer application are likely to be somewhat lower than the ones here reported for the 2000–2009 period. In the southern Mediterranean basins, our data indicate that fertilization rates are very low and that both manure application and the use of N fixation crops play an important role in providing sufficient nutrients for plant growth. Accordingly, soil surpluses of both nutrients are low, and even negative in the case of P, which would explain the low crop yields often reported in these areas (Mueller et al., 2012).

4.2. Water Management Enhances Nutrient Retention in Arid Basins

While the north-south gradient of N, P land inputs can be largely attributed to the development stage of intensive agricultural systems, a number of factors are involved in the different retention and export values, and they do not equally apply to N and P. Climate, and specifically the combination of rainfall and surface runoff, plays a part: the lowest retention percentages occur in wet regions, where regular precipitation contributes to the continuous leaching of nutrients from the soil, mainly of N, while drier watersheds present longer residence times and favor nutrient retention (Howarth et al., 2006; Sobota et al., 2009). The strong particle affinity of phosphates favors P rapid adsorption onto soil particles and binding to organic compounds (Holtan et al., 1988; Mogollón et al., 2018; Sharpley & Syers, 1979), where it may remain during several tillage cycles, while N is more easily drained by runoff due to the higher solubility of N compounds. Runoff and precipitation patterns had been previously identified as important factors determining N and P retention (e.g., Goyette et al., 2019; Green & Finlay, 2010; Howarth et al., 2012) but these studies were focused on temperate watersheds and none of them included a broad climate gradient like the one presented here. The wide range of climatic conditions in the Mediterranean—including very arid basins where precipitation is <50 mm/yr and streams run dry during part of the year—, along with the heterogeneity in terms of freshwater quality data—with different techniques and nutrient measurement protocols—may explain the lower relationships between land inputs and river outputs found in the Mediterranean with regard to other regional studies (e.g., Howarth et al., 2012; Swaney et al., 2012). Despite this variability, our results fit nicely with previous findings concerning the higher retention of nutrients (especially, N) under dry conditions. More data from water quality surveillance programs—notably from southern Mediterranean basins—would be required to validate some of the assumptions (e.g., the relationship between nitrate and TN), but most of all, further studies should consider intra-annual variability to explore to what extent the seasonal patterns in precipitation and river flow explain most of the variability in river export within arid and semiarid basins.

Beyond the direct effect of rainfall and runoff, we are particularly concerned about the effects of water management, because rising temperatures are expected to increase flow regulation and water control practices throughout the Mediterranean. The intensive use of water resources had been pointed out to be a crucial factor underpinning the high N retention values under warm, semiarid conditions (Lassaletta et al., 2012; Romero et al., 2016a). In arid and semiarid zones high water demand drives intensive water management practices that (1) divert large quantities of water away from the main river course, making the amount finally exported to the coast much smaller; and (2) favor nutrient removal and retention processes such as biological consumption in reservoirs, leaching to groundwater and denitrification in the extensive networks of irrigation channels (Castaldelli et al., 2015; Hitomi et al., 2006; Powers et al., 2015; Törnqvist et al., 2015). Our data confirm the degree of impoundment has a moderate effect on the retention, and separating into climatic groups it is clear that the effect of reservoirs is particularly important in arid environments. River damming and reservoirs are known to act as efficient traps of nutrients (Van Cappellen & Maavara, 2016), and their effect may be disproportionately large in arid basins because hydraulic residence times are long and reservoirs dampen the high export episodes associated to flashy rainfall that typically occur in arid regions. Interestingly, in our data reservoirs and flow regulation have a stronger effect on N than on P retention. This is possibly because P retention is already very high and shows less variability, but also due to the fact that (1) in arid regions the presence of reservoirs is closely related to that of irrigation infrastructures, which largely favor N removal processes; and (2) anoxia is common in bottom waters of deep reservoirs, which also contributes to increase denitrification.

Goyette et al. (2019) found that the number of dams in temperate basins was positively related to the export of N and attributed this relationship to legacy effects and saturation of the retention capacity of the reservoirs. Although this may seem contradictory to our results, it is possibly an interesting difference also linked to the association of water regulation with aridity. Small reservoirs from temperate watersheds may switch from nutrient sinks to nutrient sources as nutrients accumulate over time, while this tipping point is harder to reach in arid basins because they have large reservoirs that enable multi-year water storage strategies and potentially allow for higher, longer-term nutrient retention.

4.3. Securing Water Quantity May Compromise Water Quality

The impact of water regulation practices on the already high retention of nutrients in arid and semiarid areas is of concern in light of the swift increase of water demand predicted in many southern and eastern Mediterranean regions, where both population growth and agricultural intensification are already occurring at a fast pace (World Bank Group data sets). In the case of P, which is largely accumulated in croplands, higher retention percentages could mean a progressive build-up of the nutrient status in agricultural soils (Bennett et al., 2001; Bouwman et al., 2017; MacDonald et al., 2012). This would not be a problem in many of the southern basins considering that they currently show soil nutrient balance deficits, but the P retention capacity of soils is uncertain, and over a specific threshold (which can be as low as 0.03 t P km^{-2}) P inputs can cause a dramatic acceleration of P loss in runoff (Goyette et al., 2018). High P concentrations in reservoirs and ponds, where P-rich particles are massively retained, cause eutrophication and a loss of water quality. The case of N is even more complex, because its high solubility causes a large part of the inputs to be dispersed by water to several terrestrial compartments. Irrigation channels and impounded waters enhance removal processes, but they also increase contact surface and promote infiltration and percolation to groundwater, potentially exacerbating N pollution problems (Barakat et al., 2016; Hansen et al., 2018; Valé et al., 2007). The extent of the two processes is poorly constrained. Some studies suggest that denitrification could remove up to 50% of the total N retained in the basin (Bartoli et al., 2012), although this was questioned by Burgin and Hamilton (2007), who state that we may have overemphasized the role of denitrification in aquatic systems and that N processing by other pathways can result in transformation of the nitrate to something other than dinitrogen gas or nitrous oxide—including other bioavailable N forms that are less mobile in soils and sediments.

Even considering the most favorable removal estimates, an increase in the retention of nutrients calls to the importance of N and P legacy issues and to the fact that we may be postponing and extending pollution problems in time, as has occurred in many western European countries where present poor groundwater quality is linked to past agricultural practices (e.g., Romero et al., 2016b). In a recent study, Van Meter et al. (2018) reviewed all aspects related to N legacy and how it may hamper the achievement of good quality status for several decades, and similar conclusions were reached by Goyette et al. (2018) analyzing P legacies. The long-term degradation of water quality linked to high retention percentages can represent a very serious problem in many of the countries of the Mediterranean Basin, where water supply is already challenging. The system can hardly afford an increase in water stress and, least of all, compromise the quality of groundwater, a vital resource in arid and semiarid regions that may take decades to recover. It is estimated that people facing problems from excessive N and P loads will increase to one third of the global population by 2050 (Veolia & IFPRI, 2015), with some Mediterranean regions included among those with the highest risk exposure. Warmer temperatures, higher evapotranspiration rates and less precipitation would all contribute to decreasing river discharge and, unless nutrient inputs on land are simultaneously reduced, increase nutrient concentrations in surface streams and aquifers. The importance of considering water quality when addressing scarcity issues was highlighted by Van Vliet et al. (2017), who noted that future scenarios of water availability should take into account not only the direct decrease of river flow but also the lower dilution capacity and the decline in water quality—and the constraints on usability that this may have. Lower flows and higher concentrations of nutrients in rivers can ultimately affect the receiving marine waters, particularly in shallow coastal locations with a limited exchange with offshore waters. Marine eutrophication problems and hypoxia are not a widespread phenomenon in the Mediterranean and are currently restricted to a few specific sites (Macias et al., 2018), but very high nutrient loads concentrated in a few episodes—as it may occur in arid basins—can foster the occurrence of harmful algal blooms, particularly if stoichiometric ratios are unbalanced with regard to Redfield values (Glibert, 2017; Justić et al., 1995).

Investments in efficient irrigation systems such as drip irrigation and the use of closed conduits, for example, can increase crop yields while avoiding losses of water, N and P, and can be just as or less costly than the large reservoirs and long-distance transfers that are most often foreseen. Particularly suitable for arid and semiarid zones are those practices that improve water-use efficiency considering their interaction with nutrient management, such as the development of crops with greater drought tolerance, cover crops, rainwater harvesting, mulching and preserving soil organic matter (Aguilera et al., 2013; Quemada & Gabriel, 2016; Wallace, 2000), and their use in combination with N- and P-efficient crop genotypes (Vandamme et al., 2016). Structural transitions such as the return to the so-called Mediterranean diet, which is less resource demanding and is adapted to the local conditions, can play an important role to reduce environmental pressure (Sanz-Cobena et al., 2017).

4.4. Nutrient Decoupling Between Soils and Water

Human-induced changes of nutrient and water resources have also implications in terms of N:P ratios in the different terrestrial compartments. Changes in N:P ratios have been reported to produce a cascade of effects, from altered organisms' growth rates to shifts in community composition and function, changes in the rate of energy transfer through food webs, proliferation of harmful algal blooms (Glibert, 2017), and even reductions in the capacity of terrestrial systems to store CO_2 (Peñuelas et al., 2013, 2017; Sardans et al., 2012).

At the large scale, the growing mismatch between the use of N and P fertilizers is usually recalled as one of the main drivers of the globally increasing N:P ratios (Peñuelas et al., 2012, 2013). While this is true for global budgets, at the regional scale other patterns emerge. First, intensive agriculture and sustained high fertilization rates may not increase but rather decrease the N:P mass ratio of 14–16 typical of terrains with wild or less disturbed plants. The ratio between manure and synthetic fertilizers also affects the N:P ratio. Animal manure applied to crops has a higher relative P content than mineral fertilizers mainly due to gaseous N losses during manure management (Bouwman et al., 2017). In the Mediterranean, the average N:P of soil surpluses (i.e., all diffuse sources) is 15, but basins in the east and west where very intensive agricultural exploitations are located (e.g., Spain, Morocco, Israel) have N:P<10. The N:P ratio of croplands tends to further decrease as a consequence of the different N, P dispersal rates and the higher relative export of N by rivers, so the average N:P of inland retained nutrients is 9–10. Assuming that 30% of the retained N is denitrified (which is a moderate removal estimate, Bartoli et al., 2012), the N:P in crop soil compartments subject to intensive agricultural practices could be as low as 6.

Second, the opposite effect occurs in freshwaters: higher leaching of N compounds leads to much higher N:P ratios in rivers than in croplands. The average N:P of Mediterranean river exports is 31, twice as high the N:P of soil surpluses. This suggests that intensive agricultural practices are not only increasing the total amount of nutrients in terrestrial and freshwater systems but they are also creating a gap in the N:P ratios of the two compartments: croplands progressively decrease their N:P (Lun et al., 2018; Peñuelas et al., 2009), while rivers show increasing N:P trends (Ibáñez & Peñuelas, 2019). High N:P values were also described by Ludwig et al. (2010) and by Beusen et al. (2016), who found that N:P ratios have steadily increased throughout the 20th century in most world streams.

Third, the climatic gradient also plays a role in the divergence of N:P ratios between water and soils. Water regulation practices in arid regions promote N retention more than P retention, thereby increasing the N:P of the terrestrial fraction and decreasing the N:P of river exports, and contributing to narrowing the water-soil gap in N:P ratios. Increased erosion and flashy precipitation in arid basins also contribute to lower the N:P ratio of freshwaters, because storms disproportionately favor the transport of P sorbed on to particles (Goyette et al., 2019; Green & Finlay, 2010). The N:P of river exports in arid and semiarid regions can decrease further with a reduction of inhabitant specific water resources: the population has only limited surface water resources to dispose of waste, which causes enrichment of P from urban sewage. This is already happening in the basins surrounding the Aegean Sea, and the area has been identified as one of the future major contributors of P to the Mediterranean Sea (Ludwig et al., 2010). P is a key element for changes in marine productivity in the Mediterranean, and water stress in arid and semiarid regions is therefore one of the drivers to consider. The stoichiometry of river inputs is also an important factor to ponder when addressing the state of coastal waters because, as mentioned above, nutrient imbalances have been related to changes in the planktonic communities and the proliferation of harmful algal blooms (Glibert, 2017; Justić et al., 1995; Romero et al., 2013).

The results provide strong evidence of the human contribution to the decoupling between the N and P cycle and highlight the fact that global N:P increasing trends may affect asymmetrically the shifts in N:P ratios in different environmental compartments. As a consequence of human activities, we observe an increase of N:P ratios in the river exports to coastal waters, but a concomitant decrease of N:P in soils within the same basins, and the two can be modified by (globally increasing) water regulation practices.

Data Availability Statement

All data on nutrient inputs and outputs, climatic and hydrologic features for the 549 river basins are available online in the public repository GLONUTECO (http://glonuteco.creaf.cat/data/).

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