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Direct Discharges of Domestic Wastewater are a Major Source of Phosphorus and Nitrogen to the Mediterranean Sea

Helen R. Powley,[*,†] Hans H. Dürr,[†] Ana T. Lima,[†,‡] Michael D. Krom,[†,§,∥] and Philippe Van Cappellen[†]

[†]Ecohydrology Research Group, Water Institute and Department of Earth and Environmental Sciences, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada

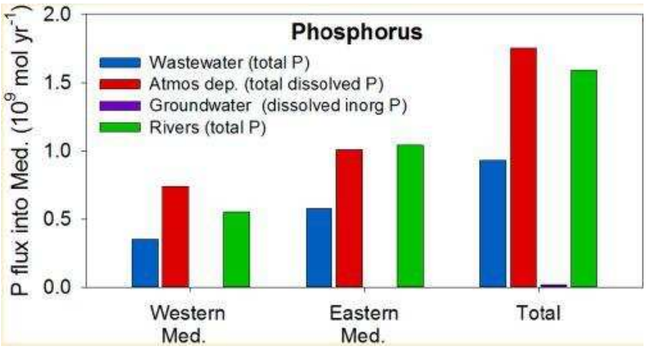
[‡]Department of Environmental Engineering, Universidade Federal do Espírito Santo, Av. Fernando Ferrari 514, CEP 29075-910 – Vitória, Espírito Santo Brazil

[§]School of Earth and Environment, University of Leeds, Leeds LS2 9JT, United Kingdom

[∥]Department of Marine Biology, Haifa University, Mt Carmel, Haifa, Israel

Supporting Information

ABSTRACT: Direct discharges of treated and untreated wastewater are important sources of nutrients to coastal marine ecosystems and contribute to their eutrophication. Here, we estimate the spatially distributed annual inputs of phosphorus (P) and nitrogen (N) associated with direct domestic wastewater discharges from coastal cities to the Mediterranean Sea (MS). According to our best estimates, in 2003 these inputs amounted to 0.9 x 10^9 mol P yr^{-1} and 15 x 10^9 mol N yr^{-1}, that is, values on the same order of magnitude as riverine inputs of P and N to the MS. By 2050, in the absence of any mitigation, population growth plus higher per capita protein intake and increased connectivity to the sewer system are projected to increase P inputs to the MS via direct wastewater discharges by 254, 163, and 32% for South, East, and North Mediterranean countries, respectively. Complete conversion to tertiary wastewater treatment would reduce the 2050 inputs to below their 2003 levels, but at an estimated additional cost of over €2 billion yr^{-1}. Management of coastal eutrophication may be best achieved by targeting tertiary treatment upgrades to the most affected near-shore areas, while simultaneously implementing legislation limiting P in detergents and increasing wastewater reuse across the entire basin.1.



INTRODUCTION

Wastewater discharges can be an important source of nutrients, contaminants and pathogens and, hence, may have a range of, often undesirable, impacts on the receiving water bodies, including harmful algal blooms and hypoxia.[1−4] Both treated and untreated wastewater from coastal cities are discharged directly into the Mediterranean Sea (MS), either at the surface or via submarine pipes.[5] Hereafter, these inputs are referred to as direct wastewater discharges. Phosphorus (P) and nitrogen (N) inputs associated with direct wastewater discharges have been invoked as possible drivers of eutrophication and hypoxia in coastal areas of the MS, for example the Nile delta region.[6] However, direct wastewater inputs have so far been neglected in existing P and N budgets of the MS.[7−12]

The MS is a semi-enclosed basin, with a coastline of more than 45 000 km (Table 1). The coastal urban population of the MS is rapidly growing, with a projected increase of over 30% from 2000 to 2025.[13] In addition to the permanent population of about 143 million, 176 million tourists visited the Mediterranean coast in 2000; this number is expected to almost double to 312 million per year by 2025.[13] Rising domestic wastewater loads accompanying population growth can drive increases in P and N delivery to the MS. For instance, according to Ludwig et al.,[14] by 2050 total riverine P discharge to the MS could be 18−42% greater than in year 2000. Changes in P and N emissions, however, are expected to vary significantly because of the large economic and demographic differences among the countries surrounding the MS. In particular, population and water stress are increasing faster in the southern Mediterranean countries, where there are generally less resources available to install treatment infrastructure to help mitigate the effects of the rising production of wastewater. Extensive wastewater reuse is currently limited to a few countries, including Cyprus, Israel and Tunisia, although the importance of “gray” water for use in agriculture is likely to increase in Mediterranean countries in the decades to come.[15,16]

able 1. Mediterranean Regions'

North Mediterranean countries	NMCs	2003 EU countries (France, Greece, Italy and Spain) plus Albania, Croatia, Cyprus, Malta, Montenegro, Slovenia	34.2'	37.2	0.48 (0.15−1.07)	79	7.5 (4.2−14.9)	80
East Mediterranean countries	EMCs	Turkey, Syria, Lebanon, Israel, Gaza	5.8'	19.3	0.22 (0.13−0.32)	16	3.7 (2.2−4.8)	22
South Mediterranean countries	SMCs	Egypt, Libya, Tunisia, Algeria, Morocco	5.7'	19.5	0.23 (0.15−0.35)	36	3.9 (2.8−5.2)	38
Total			45.8	76.0	0.93 (0.44−1.74)	53	15.0 (9.2−24.8)	55

[a]Numerical values correspond to year 2003 (baseline values): coastal urban populations, model-derived direct wastewater discharges of TP and TN (values in brackets are minimum and maximum estimates), percentages of TP and TN discharges associated with treated wastewater. 'Plan-Bleu[13]

The climate in much of the Mediterranean basin is arid to semi-arid, leading to relatively low river flows. With climate change, river flows and runoff are projected to decrease even further in the future.[14,17−21] Thus, compared to riverine inputs and the previously overlooked source of submarine groundwater discharge,[22] direct discharges of treated and untreated wastewater could be a significant pathway delivering P and N to the MS, and become increasingly so in the future. Based on answers provided by local authorities to a United Nations survey questionnaire, the aggregated direct discharges of domestic plus industrial wastewater from Mediterranean coastal cities of over 100 000 inhabitants delivered 2.4 × 10^9 mol P yr^{-1} and 18.5 × 10^9 mol N yr^{-1} to the MS around the turn of the century.[23] For P, the estimated loading from direct wastewater discharges is on the same order of magnitude as the P input delivered by rivers.[24]

Other than the data collected by UNEP/WHO,[23] estimates of P and N inputs associated with wastewater effluents are only available for selected regions and local areas of the MS (e.g., refs 25−28 and SI Table S4). Furthermore, the variable reliability of data sources, and the lack of consistent estimation methods, complicates the assessment of uncertainties associated with reported wastewater inputs of P and N to the MS. Here, we use a systematic approach to quantify the spatially distributed fluxes of P and N associated with direct discharges of domestic wastewater to the MS. The method not only yields internally consistent estimates, but also enables projections of how the direct domestic wastewater P and N discharges may respond to scenarios of changing anthropogenic pressures or improved wastewater management practices.

2. MATERIALS AND METHODS

2.1. Modeling Approach. For any given coastal city, fluxes of total P (TP) and total N (TN) associated with direct discharges to the MS of both treated and untreated domestic wastewater effluents were estimated according to the flowchart in Figure 1, using the empirical formula proposed by Kristensen et al.:[29]

$$D_{P,N} = P,N_{capita} \times pop \times f_c \times (1 - f_R) \quad (1)$$

where $D_{P,N}$ is expressed in units of mol yr^{-1}, P,N_{capita} is the annual P or N domestic wastewater load per inhabitant (mol $capita^{-1}$ yr^{-1}), pop is the population of the city, f_c is the fraction of the city's population connected to the sewer system, and f_R is the fraction of P or N removed from the wastewater stream in the city's wastewater treatment plants (WWTPs), which is dependent on the type of treatment−primary, secondary or tertiary.

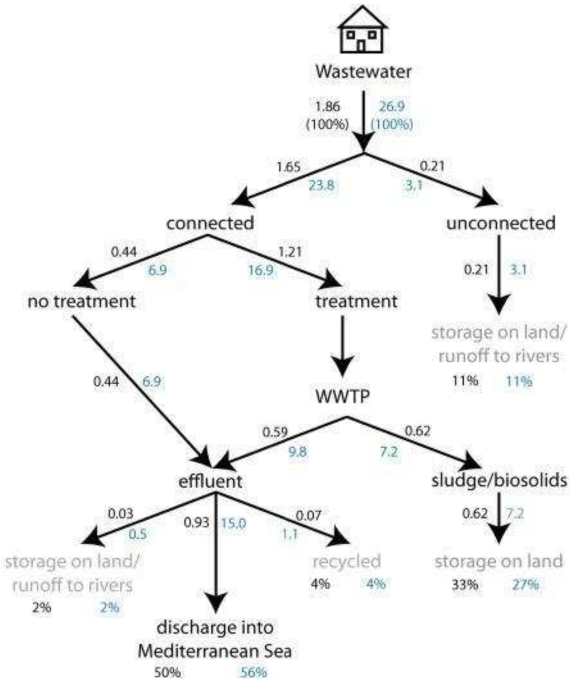


Figure 1. Flowchart for the calculation of direct domestic wastewater discharges of total P (TP) and total N (TN) into the Mediterranean Sea. The numerical values correspond to year 2003 (baseline values) fluxes of P (black) and N (blue) in units of 10^9 mol yr^{-1}, unless given as percentages of the corresponding total raw wastewater inputs.

Per capita domestic P and N inputs to WWTPs were obtained following the approach of Morée et al.[30] The total dietary P or N consumption rates were calculated as

$$P,N_{diet} = (\text{protein supplied} - \text{retail losses}) \times P,N_{protein} \times (1 - f_H) \quad (2)$$

where protein supplied is the yearly per capita protein supply quantity for a given country as compiled by Faostat,[31] retail losses are the average regional P or N losses by retail businesses and households,[30] $_{\mathrm{P,Nprotein}}$ represents the average P or N content of dietary protein,[30] and f_H represents the fraction of the dietary P or N consumed that does not end up in the wastewater stream.[30] For $_{\mathrm{Ncapita}}$, we assumed that $N_{\mathrm{capita}} = N_{\mathrm{diet}}$ for $_{\mathrm{Pcapita}}$, we also accounted for P inputs resulting from the use of laundry (P_L) and dishwasher detergents (P_D):[2]

$$P_{\mathrm{capita}} = P_{\mathrm{diet}} + P_D + P_L \quad (3)$$

Table 2. Scenarios Used in the Year 2050 Projection[a]

scenario	acronym	description
A	NMIT	no mitigation: projected change in direct TP inputs associated with domestic wastewater discharge due to (a) population growth, (b) per capita dietary P of 115 g capita^{-1} day^{-1}, plus (c) connecting at least 75% of the population to the sewage network in all coastal cities
B	REC50	at least 50% of treated wastewater is recycled in all countries bordering the Mediterranean Sea and, hence not discharged to the MS
C	ETER	all cities in eutrophic areas identified in Figure 3 have minimum of tertiary treatment
D	LEG	EU legislation to reduce P in laundry (92% reduction) and dishwasher detergent (82% reduction) is applied to all countries[38]
E	ETER+LEG	scenarios C and D combined
F	MSEC	all discharged wastewater has a minimum of secondary treatment; all current and projected WWTPs are assumed to be operational.
G	MSEC+LEG	scenarios D and F combined
H	MSEC+REC50	scenarios B and F combined
I	MTER	all wastewater discharged to the MS has a minimum of tertiary treatment; all current and projected WWTPs are assumed to be operational

[a]In scenarios B-I all conditions are identical to those in scenario A, except for the imposed mitigation strategy.

The values of N_{capita}, P_{capita}, P_D, and P_L for each of the countries around the MS are compiled in SI Table S7.

Data on population, sewerage, WWTPs and wastewater recycling of Mediterranean coastal cities were gathered from two surveys, one for cities of more than 10 000 inhabitants, the other for cities with 2000−10 000 inhabitants,[5,32] supplemented with data for Gaza[33] and Lake Manzella (or Manzala) along the coast of Egypt.[34,35] For the survey of coastal cities with more than 10 000 inhabitants,[5] city-specific parameter values were used in eq 1. Because of data limitations, for coastal cities of 2000−10 000 inhabitants[32] average parameter values for the corresponding countries were entered in eq 1 (SI Table S12).

The fraction of domestic wastewater collected by the sewage system, f_c, is divided into the fraction of wastewater that is treated (f_t) and the fraction that is untreated (f_u), or

$$f_c = f_t + f_u \quad (4)$$

Together with the fraction of the population that is not connected to the sewage network (f_n), we then have

$$f_t + f_u + f_n = 1 \quad (5)$$

Equation 1 assumes that the fraction f_n of the coastal urban population does not contribute to direct wastewater discharges entering the MS (see also Van Drecht et al.[2]). Some of the P and N associated with unconnected wastewater, however, may ultimately reach the MS through other delivery pathways, such as riverine discharge and submarine groundwater outflow.

Details on how the values on the right-hand side of eq 1 were assigned are given in the Supporting Information. Values representative for the early years of the 21st century were used to calculate baseline P and N input fluxes associated with direct wastewater outflow into the MS for the nominal year 2003. Note that, when information on treatment for a given WWTP could not be obtained, we assumed secondary treatment, as it is the most common treatment type in the countries around the MS. If a WWTP was under construction or out of order in 2003, we assumed that no wastewater treatment occurred. In addition, if f_c for a given city was unavailable, the average f_c of the host country for the year closest to 2003 was imposed (SI Table S8). Wastewater treatment costs were estimated taking into account the capital, operational and maintenance costs (COM_i in € per m^3) for each treatment type i (primary, secondary or tertiary; SI Table S14). The total wastewater treatment costs for a given coastal city were then computed as

$$\mathrm{cost} = \sum (V_i \times \mathrm{COM}_i) \quad (6)$$

where V_i (m^3 yr^{-1}) is the annual volume of wastewater undergoing treatment type i.

The UNEP-MAP surveys provide the most comprehensive data set on the distribution of WWTPs along the Mediterranean coastline in the early 21st Century.[5,31] Nonetheless, there are significant gaps in the data set, for example, lack of information on the connectivity to the sewage system or wastewater treatment type. In addition, the data quality varies from country to country. For instance, in the UNEP-MAP survey of cities with more than 10 000 inhabitants,[5] data for Spain do not include information on the level of connectivity of the population to the sewage network (f_c), while for Italy no treatment type is reported for about 50% of WWTPs (SI Table S1a). In order to account for the uncertainties associated with the data, as well as with the assumptions made in our estimates, we calculated high and low values for the TP and TN discharges from individual WWTPs (see SI Tables S10 and S11 for the imposed uncertainty ranges in the calculations). The uncertainties on the per capita P and N inputs to the sewerage system were assessed by assigning an uncertainty to each parameter in eqs 2 and 3 (SI Table S11) and applying the average uncertainties on $_{\mathrm{P,Ncapita}}$ to eq 1 across all Mediterranean countries.

A full validation of the empirical method used to estimate the direct domestic wastewater TP and TN discharges into the MS is currently impractical, primarily because of the lack of open reporting of direct monitoring data for surface and submarine outfalls. Nonetheless, measured outflows of P and N from a number of WWTPs in Spain and Italy[36] are in general agreement with values predicted based on eq 1 (Nash Sutcliffe efficiencies, E, of 0.328 and 0.862 for P and N, respectively, SI Figure S1). Differences in observed versus modeled discharge values mainly reflect uncertainties in treatment type and retention efficiencies. (For instance, for

50% of the WWTPs of Italian coastal cities with a population greater than 10 000 no treatment type is reported.)

2.2. Projections (Year 2050). Projections of direct domestic wastewater P discharges to the MS in year 2050 were carried out taking into account the potential effects of population growth, changes in dietary habits, regulatory measures and upgrades in sewerage and water treatment infrastructure. The projections focus on P, because primary

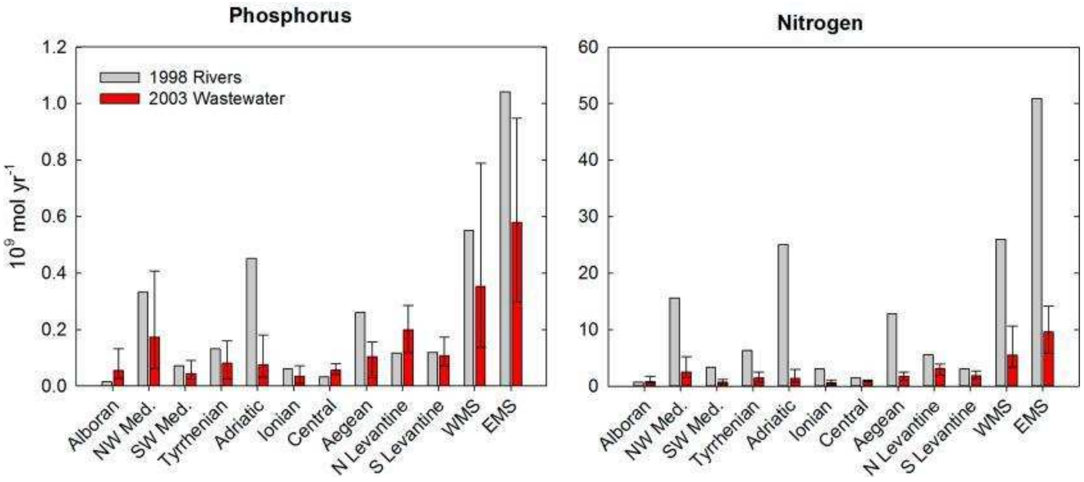


Figure 2. Inputs of TP and TN to the Mediterranean Sea: direct domestic wastewater versus riverine discharges. Wastewater inputs are those calculated in this study for year 2003 (baseline), riverine inputs are those reported in Ludwig et al.[24] for year 1998. Error bars represent estimated flux ranges (see SI Tables S10 and S11 for details). WMS = Western Mediterranean Sea, EMS = Eastern Mediterranean Sea. For the definition of the sub-basins: see Figure 3.

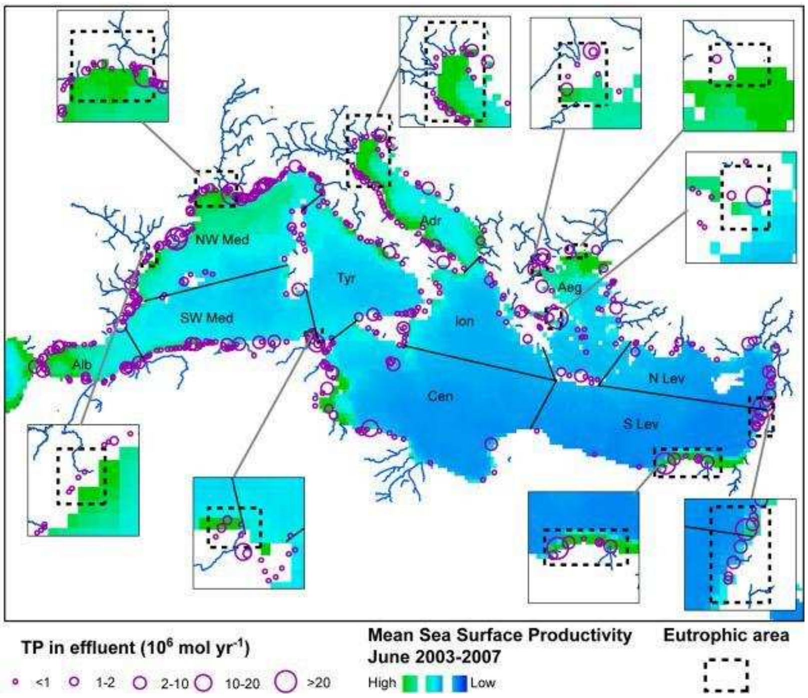


Figure 3. Direct domestic wastewater discharges of TP into the Mediterranean Sea from cities with more than 10 000 inhabitants (purple circles) in year 2003 (baseline). Also shown are the spatial distribution of the mean primary productivity across Mediterranean surface waters,[56] and coastal areas with 2 or more classified eutrophic sites between 1960 and 2010[6] (dashed boxes). Alb = Alboran Sea; NW Med = North-West Mediterranean; SW Med = South West Mediterranean; Tyr = Tyrrhenian Sea; Cen = Central Mediterranean; Ion = Ionian Sea; Adr = Adriatic Sea; Aeg = Aegean Sea; N Lev = North Levantine; S Lev = South Levantine.

production in the MS tends to be P rather than N limited.[9] The 2050 populations of coastal cities were estimated by extrapolating for each country the reported coastal urban population growth rate between 2000 and 2025[13] to 2050. In addition, we assigned a constant value of 115 g capita^{-1} day^{-1} to the 2050 protein intake in all the Mediterranean countries, that is, the combined average value of France, Greece, Italy, and Spain in 2003.[30] Thus, protein intake decreased slightly in France, Greece, and Israel in 2050 relative to 2003. We further imposed a minimum of 75% connectivity to all the coastal cities, which is the current EU average. Note that in all 2050 projections we assumed that cities maintain enough wastewater treatment capacity to accommodate the growth in wastewater inflow. Projections for the 2050 direct P and N discharge fluxes carry the same uncertainties as those in 2003.

The scenarios considered for the 2050 projections are summarized in Table 2 and their implementation described in detail in the Supporting Information. Scenarios B to I were designed to assess the potential effectiveness of various mitigation strategies in reducing the increases in direct domestic wastewater TP discharges relative to the no-mitigation scenario A (Figure 4). Mitigation measure includedupgrading WWTPs using the average P retentions in SI Table S9, extending EU legislation limiting the use of P in detergents to all Mediterranean countries,[37] which would reduce P laundry detergent inputs by 92% and dishwasher detergents by 85%,[38] and imposing a minimum of 50% reuse of treated wastewater— a reasonable future average recycling rate for the Mediterranean basin.[39] We assumed that reused wastewater does not contribute to the direct wastewater discharges of P and N to the MS.

3. RESULTS

3.1. Direct Domestic Wastewater P and N Inputs: Baseline (Year 2003). For 2003, our best estimates of the aggregated inputs of domestic TP and TN discharged directly into the entire MS, by 534 cities with population ≥10 000 plus 950 cities with 2000−10 000 inhabitants, are 0.93 × 10^9 mol P yr^{-1} and 15 × 10^9 mol N yr^{-1} (lower and higher bounds are 0.44−1.74 × 10^9 mol P yr^{-1} and 9.2−24.8 × 10^9 mol N yr^{-1}, see SI Tables S1, S10, and S11 for details). For P, our best direct wastewater input estimate is comparable to the riverine input to the MS, while for N it is distinctly lower (Figure 2). According to Ludwig et al.,[24] in 1998 riverine input fluxes to the entire MS were 1.6 × 10^9 mol P yr^{-1} and 77 × 10^9 mol N yr^{-1}. A more detailed comparison shows that Ludwig et al.'s 1998 riverine TP input is 1.6 times greater than our best estimate of direct domestic wastewater TP input to the Western Mediterranean (WMS), and 1.8 times greater for the Eastern Mediterranean Sea (EMS). Direct domestic wastewater TP inputs, however, exceed riverine inputs in the Alboran, Central, and North Levantine basins. For TN, riverine inputs are systematically higher than our best estimates of direct domestic wastewater inputs in every sub-basin of the MS, except in the Alboran Sea. (note: see Figure 3 for the definitions of the Mediterranean sub-basins.)

Among the three regions of the MS defined in Table 1, North Mediterranean Countries (NMCs) contribute the highest direct domestic TP and TN wastewater discharges: 0.48 × 10^9 mol P yr^{-1} and 7.5 × 10^9 mol N yr^{-1}. Mainly, this reflects the higher coastal population of NMCs (37.2 million) compared to East Mediterranean Countries (EMCs, 19.3 million) and South Mediterranean Countries (SMCs, 19.5 million). Per person, NMCs discharge more P than EMCs and SMCs: 12.9, 11.5, and 11.8 mol P yr^{-1}, respectively, while the N input per capita in NMCs is similar to SMCs and higher than EMCs (SI Table S2). Incomplete data for Spain and Italy, however, represent a major source of uncertainty on the direct domestic wastewater inputs of NMCs (SI Table S1a). As a consequence, the lower bound estimates for NMCs (0.15 × 10^9 mol P yr^{-1} and 4.2 × 10^9 mol N yr^{-1}) fall within the ranges calculated for EMCs and SMCs.

Treated wastewater contributes most to the direct domestic wastewater P and N inputs from NMCs (79% and 80% of the TP and TN total inputs, respectively); for EMCs and SMCs untreated wastewater is the main source, with only 16 and 36% contributions from treated wastewater, respectively (Table 1). Lebanon, Libya, and Syria, in particular, lack adequate wastewater treatment facilities: 95−100% of all P and N in effluent outfalls into the MS comes from untreated wastewater. Of all the sub-basins of the EMS, the North Levantine basin yields the highest per capita TP and TN inputs from direct domestic wastewater discharges (SI Table S2).

3.2. Direct Domestic Wastewater P Inputs: Projections (Year 2050). Population growth is generally the main driver of the projected increases in TP inputs. The total population of Mediterranean coastal cities is predicted to increase from 76 million in 2003 to 130 million in 2050 (SI Table S5). For the entire MS basin, population growth alone would result in a 72% higher direct domestic wastewater TP input in 2050, relative to 2003. For EMCs and SMCs, the corresponding increases are 103% and 159%, respectively, but only 15% for NMCs (see scenario A in Figure 4, and SI Table S5).

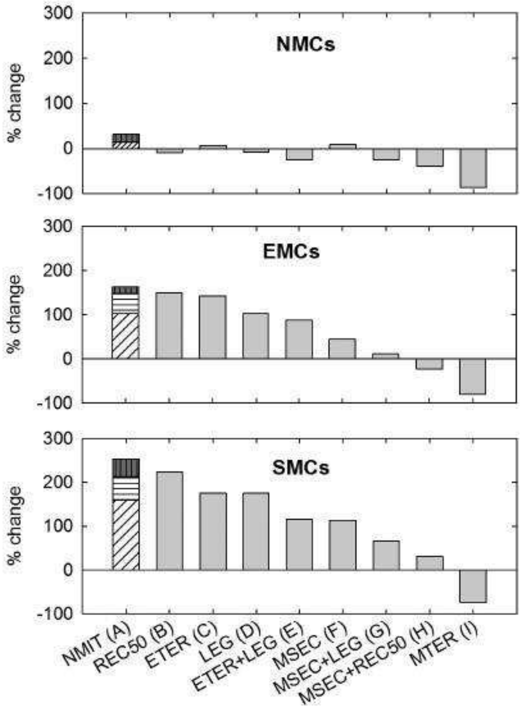


Figure 4. Changes in direct domestic wastewater TP inputs to the Mediterranean Sea in 2050, expressed as percentages relative to the corresponding 2003 inputs, for the scenarios defined in Table 2. For the non-mitigation scenario A (NMIT), the diagonal, horizontal and vertical lines identify the relative contributions of population growth, changes in diet, and connecting at least 75% of all inhabitants of coastal cities to the sewage network. See text for further details.

The projected changes in direct domestic wastewater P inputs to the MS are highly sensitive to the assumed per capita wastewater loads. If, besides the projected population growth, protein intake in all Mediterranean countries would reach 115 g capita^{-1} day^{-1} by 2050, direct domestic TP discharges, would increase by an additional 46% for EMCs, 53% for SMCs and 0.4% for NMCs. Connecting more people to the sewer system has the greatest relative impact in SMCs, where direct TP discharges would further increase by 41%, compared to 17 and 14% in NMCs and EMCs, respectively. Overall, if no mitigation measures are implemented (scenario A, Figure 4, SI Table S6), population growth, together with higher per capita protein intake and increased sewerage, would result in direct domestic TP wastewater inputs in 2050 that are 254, 163, and 32% higher for SMCs, EMCs and NMCs, respectively, relative to the corresponding 2003 values.

The results of scenarios B−I illustrate the large differences among the three Mediterranean regions (Figure 4). For NMCs, all mitigation scenarios bring the 2050 direct domestic TP inputs below that in 2003, except for scenarios C and F. The wastewater treatment upgrades imposed in scenarios C and Fare not highly effective for NMCs, because most direct urban wastewater discharge already undergoes secondary or tertiary treatment. This is not the case for EMCs and SMCs and, in these regions, scenario F in particular results in large relative decreases of the direct P discharges. Recycling of treated wastewater alone has a greater relative impact for SMCs than EMCs (scenario B), because much wastewater goes untreated in EMCs and over 50% of treated wastewater in most EMCs was already being recycled in 2003 (ref 39 and SI Table S15). However, when recycling of treated wastewater is combined with a minimum secondary treatment (scenario H), the predicted 2050 direct wastewater TP discharges decrease significantly relative to the no-mitigation scenario A. In EMCs, scenario H even yields 2050 TP inputs below the 2030 value. Introducing EU legislation limiting P in detergents to all Mediterranean countries strongly reduces direct domestic TP inputs in EMCs (scenarios D and G), because of the current high laundry and dishwasher detergent use in Israel (SI Table S7). For the three regions, major relative reductions in TP inputs, to at least 74% below 2003 values, would be achieved by switching to 100% tertiary wastewater treatment (scenario I).

4. DISCUSSION

The calculated TP and TN inputs show that direct discharges of treated and untreated domestic wastewater is an important, so far mostly ignored, pathway for transporting nutrients to the MS (Table 1, Figure 2). This is more pronounced for P than N. The estimated 2003 direct domestic wastewater TP input to the entire MS (0.93×10^9 mol P yr^{-1}) is comparable to atmospheric deposition of total dissolved P, $1.0-2.5 \times 10^9$ mol yr^{-1},[40] and an order of magnitude greater than estimates of the freshwater dissolved inorganic P (DIP) delivered by submarine groundwater discharge, 0.02×10^9 mol yr^{-1}.[22] The direct domestic wastewater input of TN to the MS (15×10^9 mol yr^{-1}) is relatively less important, but still significant compared to total dissolved N deposited from the atmosphere, $67-176 \times 10^9$ mol yr^{-1},[40] or dissolved inorganic N (DIN) delivered via fresh submarine groundwater discharge, 30×10^9 mol yr^{-1}.[22] Thus, direct wastewater discharges into the MS need to be accounted for in the N and P biogeochemical budgets of the MS.

The inputs summarized in Table 1 are low-end estimates, for the following reasons. (i) They are derived from the permanent coastal population sizes (when the data are available), rather than population equivalents, hence neglecting the increase in summer population due to tourism. (ii) They apply high-end estimates for P and N retention by WWTPs (SI Table S9). (iii) They assume that P and N associated with wastewater not collected by sewers remain on land.[2] (iv) They do not include the contributions of direct industrial wastewater discharges. Thus, the total inputs of P and N associated with direct disposal of wastewaters into the MS could be considerably larger than the estimates presented here.

According to Moree et al.,[30] industrial P and N discharges globally amount to about 15% of the corresponding domestic sources. For the Baltic Sea, industrial sources of P and N make up 14−16% of the total−municipal plus industrial−wastewater inputs,[41] while industrial inputs may represent up to 50% of the direct wastewater P releases to the Laurentian Great Lakes.[42] Data reported by UNEP/WHO[23] also imply that industrial effluents are a large source of nutrients in direct wastewater discharges to the MS. The available data, however, are

insufficient to produce spatially explicit estimations of industrial P and N inputs to the MS, similar to those of the domestic inputs in Figure 3. More complete estimations will require countries sharing the MS to more consistently and openly report discharges from WWTPs and industrial sources.

In contrast to most other large marine basins, primary production in the MS is P, not N, limited.[9,43,44] A unique characteristic of the water masses of the MS are the high inorganic molar N:P ratios, on the order of 20−28,[8,11,45,46] that systematically exceed the Redfield ratio of 16:1.[47] Averaged over the entire MS, the N:P ratio of domestic wastewater inputs is close to the Redfield value (16.1:1). Thus, overall, wastewater inputs would reduce the degree of P limitation of the MS, relative to N. However, the predicted N:P ratios of domestic wastewater discharges are highly variable from one country to another, ranging from 11:1 to 23:1 (SI Table S1b). This variability reflects differences in the P and N concentrations of the raw inputs to the sewage network and the degree of wastewater treatment. The efficient removal of P in tertiary treatment leads to molar N:P ratios exceeding 75 in the outflow, while untreated wastewater typically exhibits N:P values less than Redfieldian. Wastewater inputs may thus locally modify the N:P ratio of nearshore waters, potentially altering nutrient limitation patterns in coastal zones of the MS.[48]

In particular for coastal areas where riverine inputs are minimal, for example offshore Lebanon and the Athens metropolitan area, direct wastewater discharges are a probable driver of observed eutrophication (Figure 3). However, the ecological impacts of P and N delivery to aquatic environments not only depend on the total input fluxes, but also on the speciation of the nutrient elements.[12,49] Based on average effluent compositions for the three wastewater treatment types (SI Table S13), we estimate that domestic wastewater discharges supply TP to the MS in approximately the following proportions: 43% dissolved inorganic P, 22% particulate inorganic P, 25% particulate organic P, and 10% dissolved organic P (SI Table S3). A similar calculation yields 65% of discharged TN in the form of dissolved NH_4, which tends to be preferentially assimilated by phytoplankton compared to other N species.[50]

The large economic contrasts between Mediterranean regions are reflected in the differences in domestic wastewater nutrient inputs discharged into the sea. The higher TP input per capita in NMCs, compared to EMCs and SMCs, largely reflects higher contributions from laundry and dishwashers detergents (SI Table S2a and S7). A surprisingly large number of European countries bordering the MS still consumed P-containing laundry detergents in 2003, with only Italy, Monaco and Slovenia reporting 100% P-free detergents (SI Table S7). Legislative measures, intended to be implemented by 2017, should reduce P inputs from laundry and dishwasher detergents in EU countries by 92% and 85%, respectively (refs 37 and 38 and EU regulation 259/2012). These regulatory changes alone would reduce the 2003 direct P input from NMCs by 33% (SI Figure S3) and, if implemented by 2050, would result in no additional P entering the MS from NMCs via direct wastewater discharges relative to 2003 (scenario D). Enforcement of the same regulations by all countries would achieve an overall lowering of wastewater discharges of P by 30% for the entire MS (SI Figure S3). Phosphorus-limiting legislation is thus an attractive strategy to help reduce direct wastewater P inputs (Figure 4, scenario D), also given that phosphate-free detergents do not increase manufacturing costs substantially,while a reduction in phosphorus loading to WWTPs lowers their operational costs.[38]

Reuse of treated wastewater can also effectively curb P inputs from direct wastewater discharges (scenarios B and H; Figure 4). In the early 2000s, significant reuse of treated wastewater was already occurring in many Mediterranean countries, including Syria (100% reuse), Libya (100%), Lebanon (50%), Cyprus (95%), Israel (88%), Egypt (28%), Tunisia (23%), Spain (13%) and France (11%) (SI Table S15; refs 5,15, and 39). With growing populations and increasingly arid conditions in the future, water reuse is bound to become more important in North Africa and the Middle East.[15,18,51] Additional incentives are (i) the cost, which is approximately half that of creating usable water by desalination plants,[51] and (ii) the recovery of P and N, under the form of biosolids, sludge and wastewater itself, which can be used as fertilizer for agricultural production. The latter also represents a mitigation measure to deal with the projected depletion of P mining reserves within the next 50−400 years.[52−54] Increasing the reuse of treated wastewater in SMCs and EMCs, however, will require further investment in WWTPs coupled to stringent regulations and monitoring.[55]

Of all mitigation measures considered, only the complete upgrade of all WWTPs to tertiary treatment results in 2050 P wastewater discharges from SMCs that are below the corresponding 2003 values (scenarios I, Figure 4). However, the added costs associated with the complete conversion to tertiary treatment may be prohibitive: relative to the 2050 no-mitigation scenario, the additional costs are estimated to be on the order of €930 million yr^{-1} for SMCs alone, and €2.2 billion yr^{-1} for the entire MS basin (Table 3). These estimations are

Table 3. Annual Costs of Wastewater Treatment (in 10^6 € yr^{-1}) for the 2003 Baseline and 2050 Projections'

	2003	2050: scenario A	2050: scenario C	2050: scenario F	2050: scenario I
NMC	682	861	984	1052	1452
EMC	96	244	339	715	960
SMC	110	362	658	920	1296

[a]Scenario A: no mitigation; scenario C: minimum tertiary treatment in eutrophic areas; scenario F: minimum secondary treatment of all wastewater; scenario I: minimum tertiary treatment of all wastewater. See Table 2 for complete definitions of scenarios.based on the differences

in average costs for primary, secondary, and tertiary treatment (SI Table S14). Therefore, a more achievable course of action to manage coastal eutrophication caused by direct wastewater discharges may be to limit upgrades to tertiary treatment to recognized eutrophic areas, while accelerating the transition to phosphate-free detergents, and promoting the reuse of treated wastewater throughout the Mediterranean region.

A unique aspect of the Mediterranean basin is that it includes countries of widely different levels of economic development. Hence, trends observed for the MS may provide lessons for other parts of the world. In coastal areas with rapidly growing populations a first public health response is typically to expand the sewerage system. If this is not matched by increased wastewater treatment or other mitigation measures, discharges of nutrients and other contaminants to the coastal zone may actually increase. Results for EMCs and SMCs indicate that for less developed countries curbing coastal wastewater nutrient inputs may represent a major financial challenge (Table 3). Direct discharges of wastewater are thus likely to continue to threaten many coastal areas around the world.

ASSOCIATED CONTENT

*S Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b01742.

Detailed methods, country-by-country input data, and calculated wastewater discharges of P and N at country and regional scales (PDF)

AUTHOR INFORMATION

Corresponding Author

*Phone: +1-519-888- 4567, ×37789; fax: +1 519-746-7484; email: hrpowley@uwaterloo.ca.

Notes

The authors declare no competing financial interest.

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