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N:P:Si nutrient export ratios and ecological consequences in coastal seas evaluated by the ICEP approach

Josette Garnier,¹ Arthur Beusen,² Vincent Thieu,¹ Gilles Billen,¹ and Lex Bouwman^{2,3}

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[1] The Indicator for Coastal Eutrophication Potential (ICEP) for river nutrient export of nitrogen, phosphorus, and silica at the global scale was first calculated from available measurement data. Positive values of ICEP indicate an excess of nitrogen and phosphorus over silica and generally coincide with eutrophication. The sign of ICEP based on measured nutrient fluxes was in good agreement with the corresponding one calculated from the Global-NEWS models for more than 5000 watersheds in the world. Calculated ICEP for the year 2050 based on Global NEWS data for the four Millennium Ecosystem Assessment scenarios show increasing values particularly in developing countries. For further evaluation of the ICEP at the outlet of the rivers of the world based on measurements, there is a need for additional determination silica fluxes and concentrations, which are scarcely documented.

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1. Introduction

[2] In the past few decades, increased nutrient loading of coastal marine ecosystems has resulted in severe local, regional and global eutrophication problems [Billen and Garnier, 2007; Howarth, 2008; Glibert *et al.*, 2008]. The manifestation of these problems may be quite diverse, depending on local physiographical and hydrological conditions of the coastal marine system: (1) blooms of potentially toxic algae in the Seine Bight and the Baltic Sea [Cugier *et al.*, 2005; Vahtera *et al.*, 2007; Elmgren, 1989; Granéli, 2004]; (2) massive development of mucilaginous, unpalatable, algal species in the North Sea [Lancelot *et al.*, 1987, 2005, 2007], the Black Sea [Cociasu *et al.*, 1996], or the Adriatic Sea [Marchetti, 1991]; (3) increased deposition of organic material resulting in anoxic bottom waters as in the northern Adriatic and northern Gulf of Mexico [Justić, 1991; Justić *et al.*, 1995a], the open waters around Denmark and in Danish estuaries [Conley *et al.*, 2007], and in many places in the world [Diaz, 2001; Justić *et al.*, 1995b; Diaz and Rosenberg, 2008]. Profound changes in the structure of food webs are often observed as a result of eutrophication in coastal marine ecosystems, with changes in the structure of the benthic communities [Lim *et al.*, 2006] and a decline in

zooplankton affecting commercial fish production [Rousseau *et al.*, 2000].

[3] It is now recognized that these phenomena are not only caused by nutrient enrichment of the marine system *per se*, but rather by the changes in the nutrient stoichiometry. Coastal enrichment with nutrients, brought in proportion of the Redfield ratios [Redfield *et al.*, 1963] and characterizing the requirement of diatom growth, would only rarely result in eutrophication problems. New planktonic primary production is mostly ensured by diatoms, while nonsiliceous algae are restricted to regenerated production. On the contrary, when nitrogen (N) and phosphorus (P) are discharged in excess over silicon (Si) with respect to the requirements of diatoms, these will be limited, and nondiatoms, often undesirable algal species, will develop instead [Billen and Garnier, 1997, 2007; Conley *et al.*, 1993; Conley, 1999; Cugier *et al.*, 2005; Humborg *et al.*, 2000, 2008; Justić *et al.*, 1995a, 1995b; Officer and Ryther, 1980; Turner and Rabalais, 1994; Turner *et al.*, 1998].

[4] According to this view, an Indicator of Coastal Eutrophication Potential (ICEP) was proposed [Billen and Garnier, 2007]. ICEP is calculated on the basis of riverine N, P and Si deliveries, allowing to determine the possible problems resulting from a new production of nonsiliceous algae sustained by external inputs of N and/or P brought in excess over silica, *i.e.*, in limiting conditions for the diatom growth. Compared to N:P ratios which are often considered [e.g., Glibert *et al.*, 2008], the ICEP adds information about the role of Si in determining potential eutrophication impacts of changing element stoichiometry.

[5] The aim of this paper is to compare the ICEP values on the basis of the Global NEWS data [Seitzinger *et al.*, 2010]

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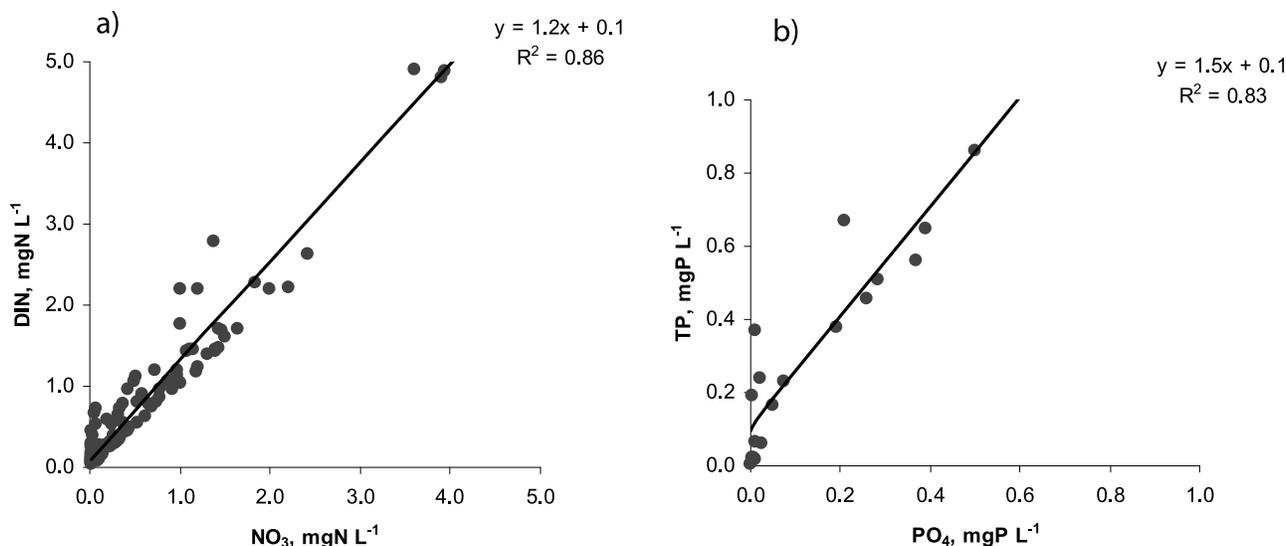


Figure 1. Relationship between (a) DIN and N-NO₃ and (b) TP and P-PO₄ for the data available in the GEMS-GLORI database.

and a data set of N, P, Si fluxes from the GEMS-GLORI database [Meybeck and Ragu, 1995, 1997] combined with other data from the literature. This data set is used to illustrate the ICEP values for the period from 1970–1990. Furthermore, the NEWS DSi model of global DSi export by world rivers [Beusen *et al.*, 2009], together with the Global-NEWS models of P and N [Seitzinger *et al.*, 2010], allows computing the ICEP for most of the drainage basins of the planet. Finally, we used the Global NEWS implementation of the Millennium Ecosystem Assessment scenarios [Alcamo *et al.*, 2006] to assess potential changes in eutrophication in coastal marine ecosystems based on ICEP for the period 2000 to 2050.

2. Observations of River Export of Nitrogen, Phosphorus, and Silica

[6] The documented DSi data were taken from the GEMS-GLORI database and literature, together with dissolved inorganic nitrogen (DIN) and total phosphorus (TP). DIN is calculated as the sum of dissolved nitrate and ammonium (DIN). When nitrate only is available, the relationship found between DIN and nitrate was used to calculate DIN (Figure 1a). Further, as the few total nitrogen (TN) data available (18) did not show any difference and trend compared with DIN (not shown), DIN has been taken as a good estimate of TN for calculating the ICEP, as we did in the initial paper by Billen and Garnier [2007]. Indeed, both organic and inorganic N forms are biologically available to marine plankton [Seitzinger and Sanders, 1999; Wiegner and Seitzinger, 2001]. Some studies showed that DON might represent over 20% of TN, but this occurs at low TN only [De Wit *et al.*, 2005; Ludwig *et al.*, 2010; P. Johnes, personal communication, 2009], so that DIN largely dominates in most N contaminated waters.

[7] Dissolved inorganic phosphorus (PO₄) is the P form for which data are mostly available. A relationship has been established between total phosphorus (TP) and PO₄ (Figure 1b), which was used when TP data were missing. We consider here that total phosphorus may be rapidly available as reported in several studies showing that phosphorus adsorbed onto particles is exchangeable [Conley *et al.*, 1995; Némery *et al.*, 2005; Némery and Garnier, 2007].

[8] According to Billen and Garnier [2007], we consider dissolved silica (DSi) fluxes only for the calculation of the ICEP; we ignore particulate amorphous silica (ASi) as only few data exist in the rivers of the world. Yet, ASi may be important in the total bioavailable silica delivery [Conley, 2002; Humborg *et al.*, 2000, 2002, 2006; Garnier *et al.*, 2002a; Sferratore *et al.*, 2006] with a share in total Si river global loads generally of the order of 10%–20% [Conley, 2002; Laruelle *et al.*, 2009]. Hence, we have to accept such an underestimation of total silica loads, and perhaps a slight overestimation of ICEP, if not compensated by a possible underestimation of the nitrogen load based on DIN only (see above).

[9] Our data set contains 189 observed data with both DIN and DSi, and 132 with TP and DSi. The GEMS-GLORI database includes data from rivers “still in a pristine state” and other “impacted by human activities” and a variety of rivers “affected at various stages of impacts, such as nutrient pollution, damming, salt pollution, land management, etc.” [Meybeck and Ragu, 1995, pp. 105–245]. The additional literature data include rivers from Asia [Zhang, 1996; Liu *et al.*, 2005; Le *et al.*, 2005], South America [Edmond *et al.*, 1981], Mediterranean rivers [El Boukhary, 2005; Gago *et al.*, 2005], temperate Europe [Marchetti *et al.*, 1989; Wafar *et al.*, 1989; Jaworski *et al.*, 1992; Billen and Garnier, 1999; Garnier *et al.*, 2002b; El Boukhary, 2005; Humborg *et al.*, 2006; Thieu *et al.*, 2009], North America [Turner and Rabalais, 1994; Justić *et al.*, 1995a,

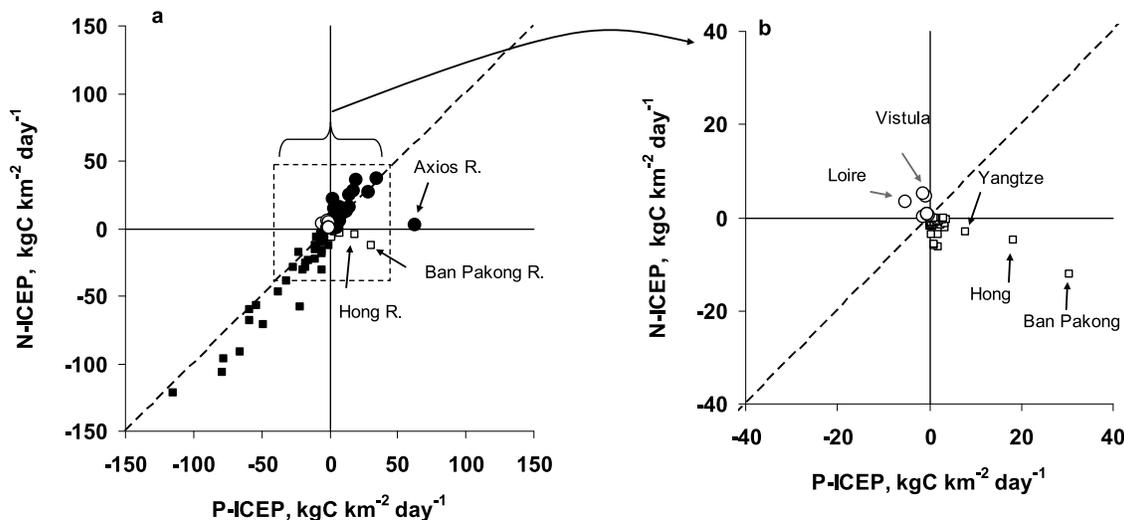


Figure 2. Relationship between the N-ICEP and P-ICEP (a) for all values calculated with DSi, TN (see section 2), and TP fluxes (section 2). Data from the GEMS- GLORI database [Meybeck and Ragu, 1995] and literature (references in the text). Black circles and squares for N-ICEP and P-ICEP values, both positive or negative, respectively. Open circles and squares when N-ICEP and P-ICEP values are mismatching (b) focus on the 42 mismatching ICEP data ranging from 0 to 30 kg C km⁻² d⁻¹.

1995b] and the Arctic [Telang *et al.*, 1991; Humborg *et al.*, 2004]. All these rivers are in different stages of human interference both in terms of hydrology and nutrient loading.

3. ICEP: An Indicator for Coastal Eutrophication Potential of Rivers

[10] The indicator for coastal eutrophication potential (ICEP) represents the new production of nonsiliceous algal biomass potentially sustained in the receiving coastal water body by either nitrogen or phosphorus delivered in excess over silica [Billen and Garnier, 2007]. ICEP is based on the Redfield molar C:N:P:Si ratios 106:16:1:20 [Redfield *et al.*, 1963; Conley *et al.*, 1989]. For the purpose of river to river comparison, it is scaled to the river watershed area, thus expressed in kg C km⁻² d⁻¹. According to the nutrient considered, either N-ICEP or P-ICEP can be defined, following relationships (1) or (2), respectively:

$$N - ICEP = [NFlx / (14 \times 16) - SiFlx / (28 \times 20)] \times 106 \times 12 \quad (1)$$

$$P - ICEP = [PFlx / 31 - SiFlx / (28 \times 20)] \times 106 \times 12 \quad (2)$$

where PFlx, NFlx, and SiFlx are the mean specific fluxes of total nitrogen, total phosphorus, and dissolved silica, respectively, delivered at the outlet of the river basin, expressed in kg P km⁻² d⁻¹, kg N km⁻² d⁻¹, and kg Si km⁻² d⁻¹.

[11] A negative value of the ICEP indicates that silica is present in excess over the other nutrient and would thus characterize the absence of eutrophication problems. Positive values indicate an excess of either N or P over the requirements for diatom growth, thus a condition for potentially

harmful nonsiliceous algal development. The ICEP represents the potential impact of the riverine delivery to the coastal zone, and does not take into account the particular morphological, climatic and hydrological conditions that locally determine the response of the marine algae in the receiving coastal zone [cf. Rabouille *et al.*, 2008; Nixon, 2009].

[12] Depending on the N:P ratio of nutrient loading, either N or P is the potential limiting nutrient. The lowest value between N-ICEP and P-ICEP should thus theoretically be considered. However, owing to high biochemical adaptation capacity of phytoplankton to low phosphate availability (induction of algal phosphatase), P may be actively and rapidly remobilized in marine waters (bacterial and zooplankton mineralization), e.g., where the N:P ratios of either the nutrient stocks or fluxes are generally above the Redfield ratio of 16 [Currie and Kalff, 1984; Labry *et al.*, 2005]. We compared the data set of 132 couples of N-ICEP and P-ICEP values (Figure 2). For the rivers with negative N-ICEP values, 60% of the P-ICEP are negative too, whereas among the N-ICEP positive values, 83% of P-ICEP values are also positive. In total, for 68% of the rivers, the sign of the N-ICEP and P-ICEP values match. Within the 32% (42 cases) of the rivers for which the ICEPs differs by sign, the absolute values of both indicators are very close to zero; the mismatch appears to be important for 5 rivers only, the Loire (France) and the Vistula (Poland) in western Europe (P limited) and the Yangtze (China), the Hong (Vietnam) and the Ban Pakong (Thailand) in Asia (N limited). These observations mean that for most of the available cases, P and N are simultaneously in excess relative to Si. We will therefore concentrate our analysis on N-ICEP values, because 70% of the river examples in the data set are N-limited and also because we have a larger data set for N-ICEP (189) than for P-ICEP

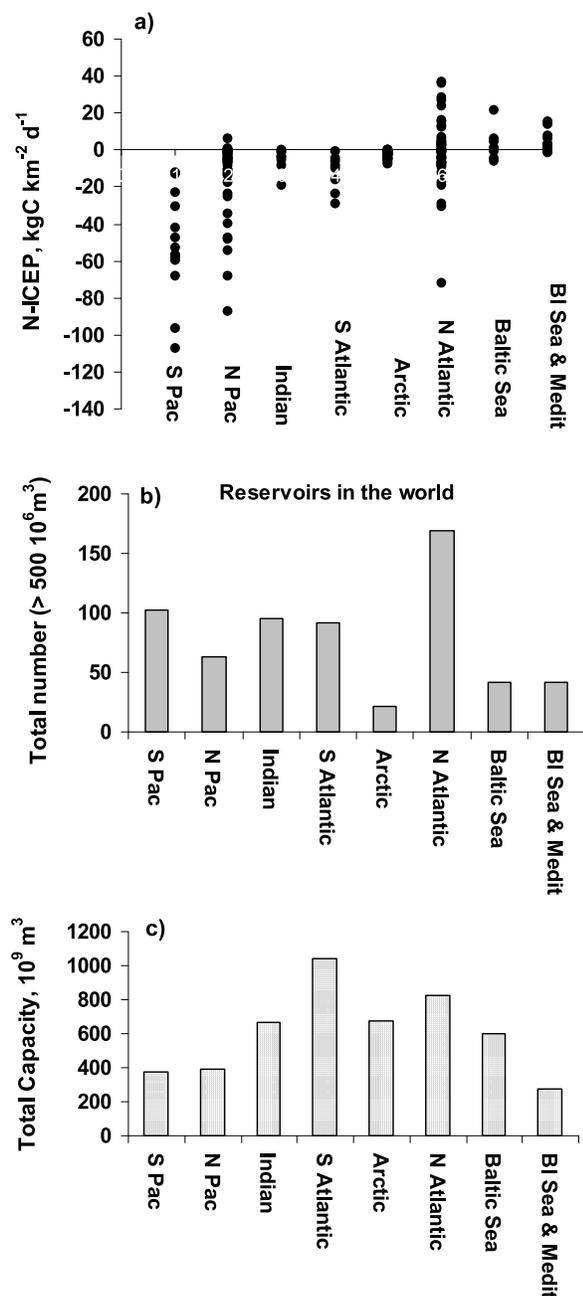


Figure 3. (a) N-ICEP values calculated on the basis of DIN and DSi fluxes for the documented rivers in the GEMS-GLORI database and literature, grouped by oceans/seas. (b) Corresponding total number of reservoirs in the concerned watersheds. (c) Corresponding capacity of the reservoirs.

(132). We recognize however, that both P- and N-ICEP should be considered to strengthen the ICEP approach, even though P may not systematically be limiting in marine waters where the N:P ratios are lower than 16 due to its rapid recycling.

[13] On the basis of the measured data set, we see positive N-ICEP values for river basins draining to the Mediterra-

nean Sea, Black Sea, Baltic Sea and the North Atlantic. N-ICEP values for rivers draining to the South Atlantic, South and North Pacific, the Arctic and the Indian Ocean are most often negative (Figure 3).

[14] The N, P, Si fluxes analyzed here for the period after the 1960s are necessarily influenced by dams constructed in a large number of watersheds; silica fluxes are probably affected by retention in reservoirs, resulting from DSi transformation into amorphous silica after diatom uptake under N and P enrichment conditions and subsequent sedimentation. As reservoirs trap sediment, they also retain silica and phosphorus. However, sedimented phosphorus and silica have a different behavior, sedimented silica being remobilized by a slow dissolution (from 2 to 0.2 10⁻² % h⁻¹ in freshwaters [Garnier *et al.*, 2002a]), whereas adsorbed phosphorus can be instantaneously desorbed [cf. Némery *et al.*, 2005; Némery and Garnier, 2007]. The number of reservoirs (>500 10⁶ m³) in the Global NEWS data [Vörösmarty *et al.*, 1997] may therefore be an indicator of nutrient retention (Figure 3b). The total number of reservoirs is the highest for rivers discharging into the North Atlantic Ocean where the highest number of positive N-ICEP values is found (Figures 3a and 3b). In contrast, total reservoir capacity is not related to N-ICEP (Figure 3c). This is possibly related to differences in the location of reservoirs. An even small reservoir close to the mouth of a river may retain more sediment and nutrients than one located upstream and affecting only a small portion of the discharge.

[15] The N-ICEP values from the whole data set were plotted as a function of population density and the percentage of agricultural land (including cropland and grassland) of the corresponding watershed provided by the Global-NEWS database (Figure 4). The data suggest an increase of N-ICEP in function of population density, thus a higher risk of eutrophication in the North Atlantic coast, the Baltic Sea, the Mediterranean and the Black Sea coastal zones (Figure 4a). The data also indicates a shift from negative to positive N-ICEP values in river basins with >30% agricultural land, with generally lower N-ICEP values in coastal marine ecosystems in the Southern Hemisphere (Figure 4b).

4. Assessing the ICEP at the Global Scale

4.1. Current 2000 Situation and Hindcast (1970)

[16] We calculated the ICEP indicator for over 5000 rivers systems in the world on the basis of the Global-NEWS model results for the years 1970 and 2000, more or less corresponding to the time period of our measurement data set. An overview of the Global-NEWS models and data is provided by Seitzinger *et al.* [2010]. The Global-NEWS system includes river-basin-scale models for predicting export of TN and TP, as well as their various forms. These models have been first described in various papers [Beusen *et al.*, 2005; Dumont *et al.*, 2005; Harrison *et al.*, 2005a, 2005b], and an update is presented by Mayorga *et al.* [2010]. The DSi model was recently developed by Beusen *et al.* [2009]. All NEWS models were calibrated with the GEMS-GLORI database. Here we used the total TN, TP and DSi fluxes results from the Global-NEWS models for

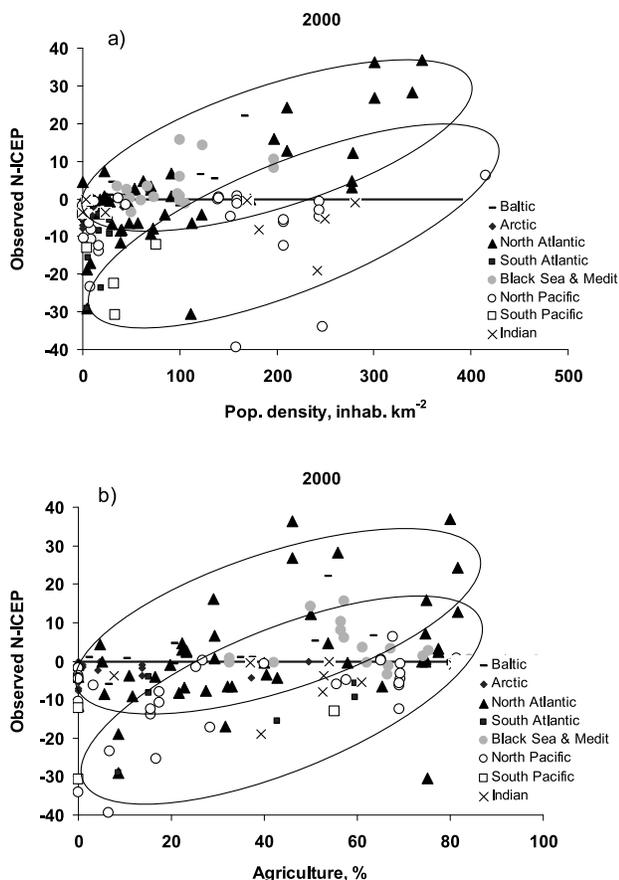


Figure 4. Relationships between the N-ICEP values identified by the adjacent oceans/seas and (a) the population density (Pop. Density) and (b) the percentage of agricultural area in the corresponding watershed.

all individual watersheds of the world [Fekete *et al.*, 2002] and calculated their N-ICEP. The NEWS-DSi model [Beusen *et al.*, 2009] represents natural conditions for the predam situation; DSi retention was calculated with the sediment trapping efficiency for all global rivers [Fekete *et al.*, 2002, 2010] proposed by Vörösmarty *et al.* [2003]. This is based on the assumption that sedimentation rates of suspended solids and diatom frustules are similar.

[17] The calculated N-ICEP values from Global NEWS data vs. the ones obtained from the observations are significantly correlated ($R^2 = 0.4$, $n = 189$), with a lower slope (0.34) compared to the 1:1 line (Figure 5). However, the global-scale model matches the observations in terms of sign in about 70% of the cases (i.e., calculated and observed N-ICEP being both negative or both positive) (Figure 5). Most mismatching data correspond to a potential of eutrophication due to silica limitation (positive calculated N-ICEP) when measured silica would not be limited (negative observed N-ICEP). The calculated ICEP can be a warning indicator in these cases. The calculated N-ICEP for 1970 and 2000 shows the changes during the last 30 years (Figure 6). In the Northern Hemisphere, e.g., the North Atlantic and North Sea, the Baltic Sea and the Black Sea in Central Europe, the

Hudson in Canada and the Gulf of Mexico in the United States, have high N-ICEP values. Although the Southern Hemisphere generally appears to be less sensitive to eutrophication, some hot spots can be identified for South Pacific, South Atlantic and Indian Oceans (Figure 6). The comparison shows that eutrophication risk has decreased in Europe, and eastern Canada, and increased in some other places in the period 1970–2000 (e.g., Japanese and Chinese seas, Indian and South Africa coasts). This increase of N-ICEP values is related to rapidly increasing agricultural production and fast urbanization and development of sewerage systems without sufficient treatment of sewage water. In contrast, in industrialized countries in Europe and the United States, N-ICEP values are constant or show a decrease as a result of major efforts to remove N and P in wastewater treatment, and increases in the efficiency of agricultural N and P use [Bouwman *et al.*, 2009].

4.2. Millenium Ecosystems Assessment Scenarios

[18] Apart from the current and hindcast analysis of the ICEP values, the Global-NEWS implementation of the Millenium Ecosystem Assessment (MEA) scenarios has been used to calculate the possible future nutrient discharge of N, P and Si by world rivers to coastal marine ecosystems. Although the MEA scenario results are reported for 2030 and 2050, we limit here our analysis to the data for 2050. The four MEA scenarios differ in terms of management of the environment and the degree and scale of connectedness among and within institutions across country borders [Alcamo *et al.*, 2006]. Technogarden (TG) and Adapting Mosaic (AM) were developed assuming proactive approach to environmental management, while Order from Strength (OS) and Global Orchestration (GO) assume reactive environmental management. GO and TG reflect trends toward

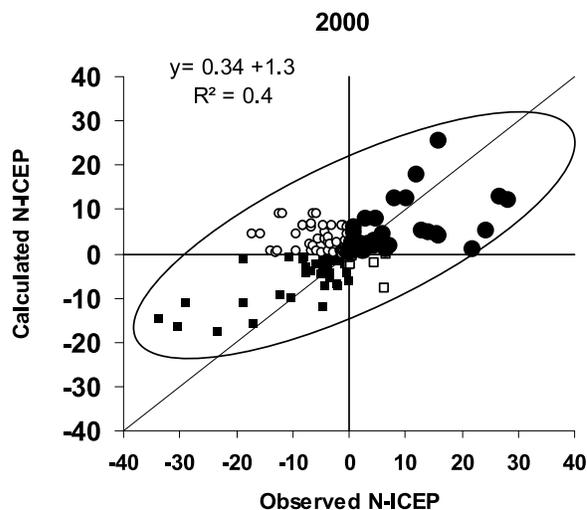


Figure 5. Relationship between observed and calculated N-ICEP for the year 2000. Black circles and squares for calculated and observed N-ICEP values both positive or negative, respectively. Open circles and squares when calculated and observed N-ICEP values are mismatching.

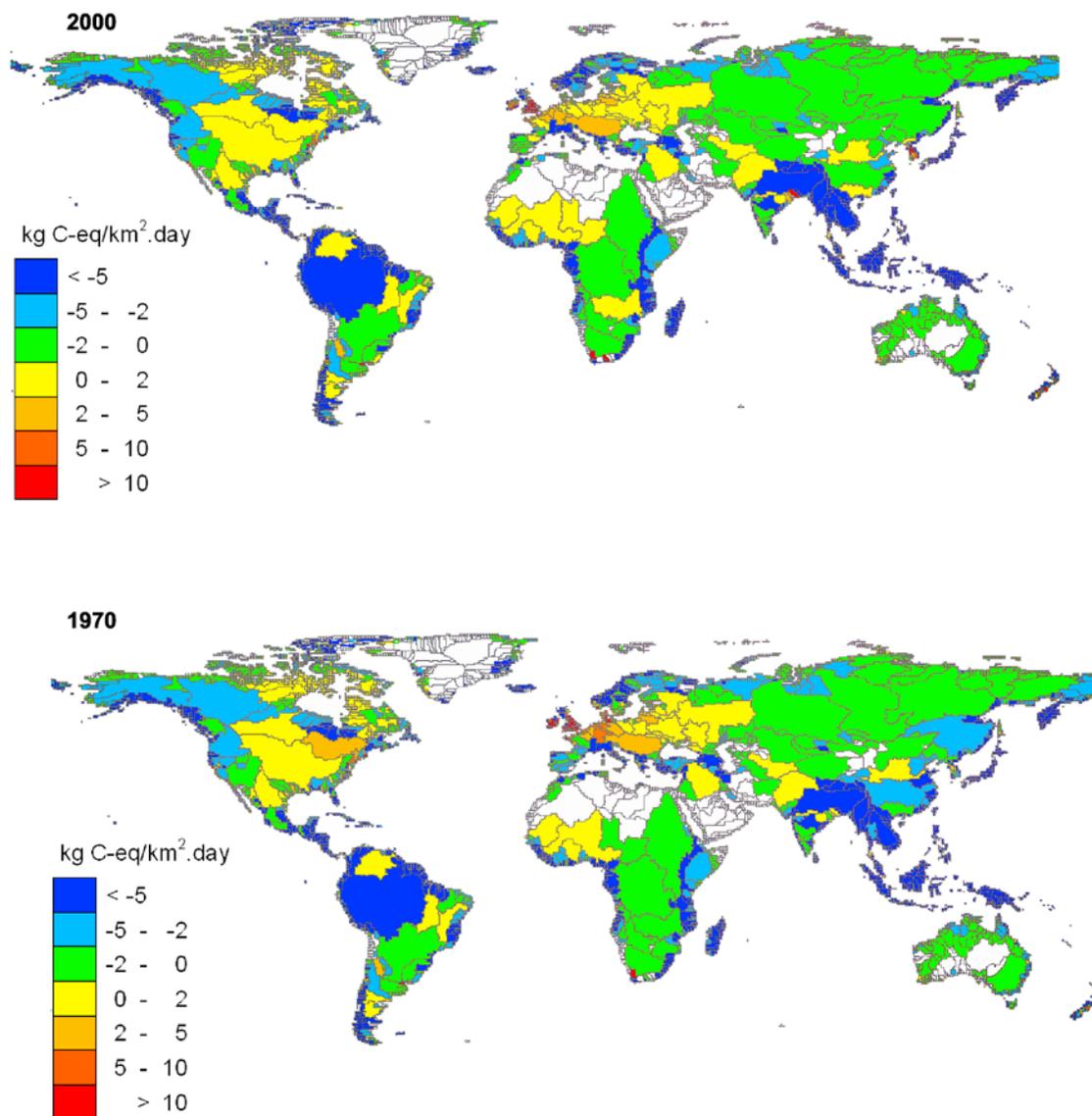


Figure 6. Global N-ICEP values calculated from (top) Global NEWS data 2000 and (bottom) 1970.

globalization, while regionalization is assumed in OS and AM. Further details on the assumptions made with respect to nutrient management are given by *Seitzinger et al.* [2010], *Bouwman et al.* [2009] and *Van Drecht et al.* [2009].

[19] All four scenarios show an improvement of the ICEP indicator compared to the year 2000 in most areas where the situation was already improving in 2000 with respect to 1970 (Figure 7). Results for 2050 for the OS, TG and AM scenarios indicate a decrease of the ICEP for the rivers draining into the North Atlantic (European Union and United States) and the Baltic Sea, zones that are presently the most eutrophicated, whereas for the Mediterranean and Black Seas, the Arctic and South Atlantic, ICEP remains stable around zero; for the Pacific and Indian coastal zones it is kept largely negative (Figure 7). In the GO scenario the number of river basins with ICEP > 0 will increase in the Arctic, Mediterranean and Black Seas, Indian Ocean, Gulf

of Mexico and Caribbean Sea, South and North Pacific. Globally, 41% of the land area had a positive N-ICEP in 1970, increasing to 45% in 2000 up to 49%–52% in 2050 (depending on the scenario). GO leads to contrasting results in terms of ICEP values, and whereas the coastal zones of the northern countries (except the Arctic) show a decrease of the ICEP values, i.e., a decreasing risk of eutrophication, those of the south reveal an increase (Figure 7).

5. Discussion

5.1. ICEP Approach

[20] Although nutrient deliveries from river basins are ecologically vital to the survival and diversity of these ecosystems, increasing amount of unbalanced anthropogenic riverine inputs have greatly disturbed the coastal zones that are complex and fragile ecosystems. Many coastal zones are

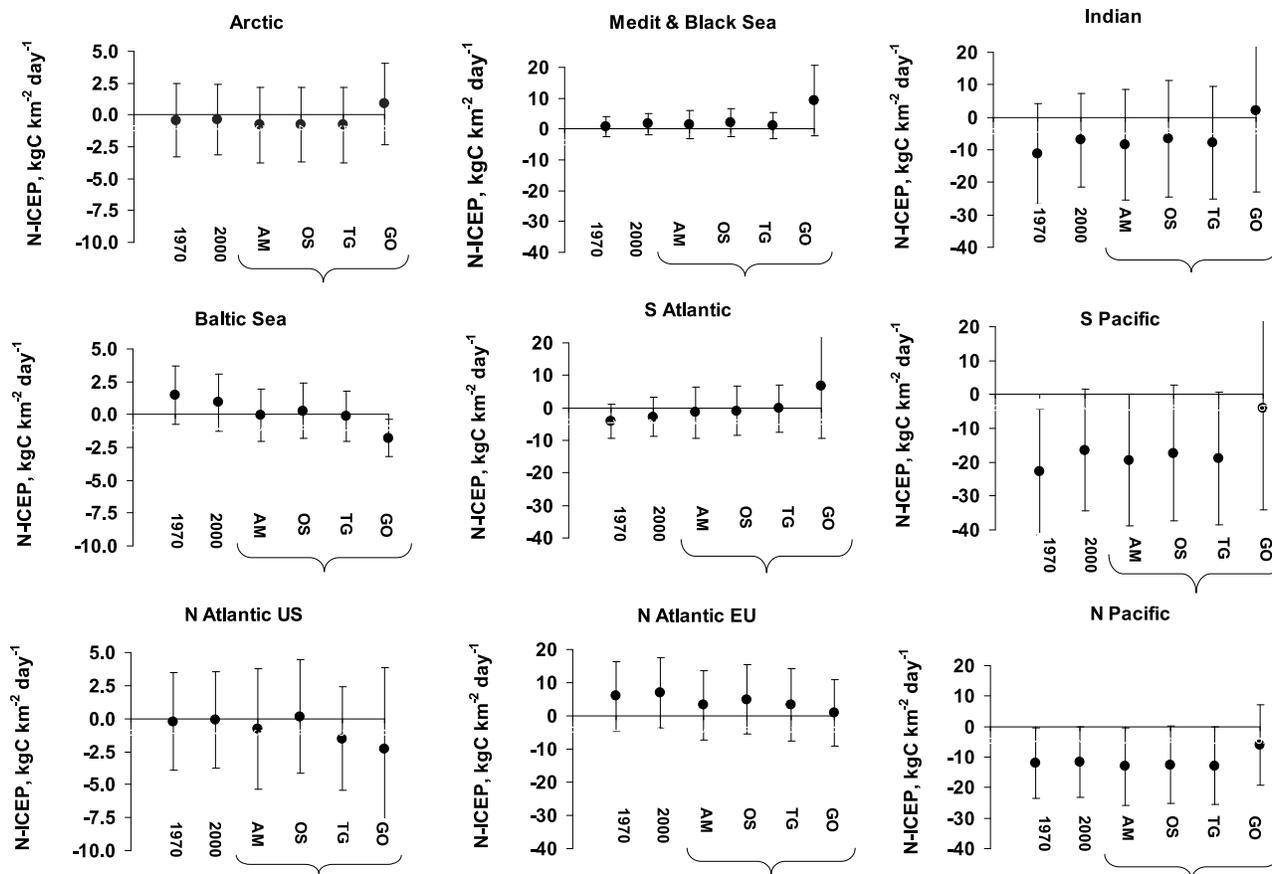


Figure 7. Aggregated N-ICEP calculated from Global NEWS data for different oceans and seas for the years 1970 and 2000 and for 2050 according to the four MEA scenarios (AM, Adapting mosaic; OS, Order from Strength; TG, Techno Garden; GO, Global Orchestration). Black circles are mean values of the ICEP; error bars indicate standard deviations.

affected by an increase in riverine nutrient loading [Howarth *et al.*, 1996; Boyer *et al.*, 2002] caused by human activities including agricultural runoff, domestic and industrial sewage and atmospheric deposition, as well as groundwater seepage [Slomp and Van Cappellen, 2004]. Silica from rock weathering is not affected by human activity (or only indirectly by climate change and dam construction). Si may therefore become limiting if anthropogenic N and P loading increases, which may lead to harmful algal blooms, hypoxia and negative impacts on fisheries as reported by many studies [Lancelot *et al.*, 2002; Cugier *et al.*, 2005; Diaz and Rosenberg, 2008; Rabouille *et al.*, 2008; Rabalais *et al.*, 2009]. The nonsiliceous algae that replace the diatoms as soon as N and P are in excess over Si may indeed form mucilaginous blooms, such as *Phaeocystis* in the North Sea [Lancelot, 1995; Lancelot *et al.*, 1987], toxin-producing *Cyanobacteria* in the Baltic Sea [Granéli and Granéli, 2008] and *Dinophysis* in the Seine Bight [Cugier *et al.*, 2005]. On the other hand, there is evidence that toxin production by both nonsiliceous and siliceous algae is enhanced at high N levels and high N:P ratio [Moestrup *et al.*, 2004; Leong *et al.*, 2004; Murata *et al.*, 2006; Zingone *et al.*,

2006], making it necessary to examine both N and P deliveries together with Si.

[21] However, simultaneous measurements of Si, N and P concentrations or fluxes are rather scarce in the literature. On the one hand, dissolved Si is rarely measured systematically together with N and P in the routine surveys of most water agencies and in eutrophication studies [Smith *et al.*, 1999]. On the other hand, dissolved Si is generally measured with N (DIN) only in the hydrogeochemical research, when the aim is to study the chemistry of major elements or rock weathering [Roy *et al.*, 1999; Dupré *et al.*, 2003; Zakharova *et al.*, 2005]. The southern North Sea [Cugier *et al.*, 2005; Lancelot *et al.*, 1987; Lancelot, 1995; Lancelot *et al.*, 2005], the Baltic Sea [Humborg *et al.*, 2006, 2008], the Gulf of Mexico [Turner *et al.*, 1998; Turner and Rabalais, 2003; Turner *et al.*, 2006] and some Asian rivers [Zhang *et al.*, 2004; Li *et al.*, 2007; Rabouille *et al.*, 2008] are among the best documented examples for simultaneous Si, N, P delivery measurements.

[22] In our analysis, we ignored river export of amorphous silica (ASi), that might be bioavailable for the diatoms and therefore should be taken into account in the ICEP calculation. However, data on ASi river export are scarce [Conley,

2002; Humborg *et al.*, 2006; Garnier *et al.*, 2002a; Sferratore *et al.*, 2006]. This may cause an underestimation of the total Si flux and thus an overestimation of ICEP. In particular, because ASi dissolution has been shown to increase when it passes the salinity gradient of the estuarine zones, ASi can be of major importance in coastal marine ecosystems [Ragueneau *et al.*, 2002; Roubeix and Lancelot, 2008]. This indicates the need for data on ASi fluxes for a better understanding and forecasting of eutrophication. Another drawback of the ICEP approach is that it is based on the Redfield ratio concept to assess N or P limitation. This may be a problem where, for example, phosphorus may be low, but made rapidly available for the algae by microbiological processes [Currie and Kalff, 1984; Labry *et al.*, 2005].

[23] As a whole, a strong advantage of the ICEP approach is to provide a functional and warning indicator, as opposed to other indices of ecological status proposed based on the works conducted in the scope of the European Water Framework Directive [European Commission, 2000]. In addition, the ICEP can be calculated either from observations, at the scale of the year or the season, or from modeling. Furthermore, ICEP values calculated with a validated model allow for reconstructing the ICEP trajectories for the past [Billen *et al.*, 2007] or in projected futures based on scenarios, like those of the MEA discussed in this paper. The ICEP concept should help the researchers and the managers, not to only consider P and N, with a propensity to reduce the P:N ratio in order to obviate eutrophication (see the debate initiated by Conley *et al.* [2009]), but rather to consider the Si:P and Si:N ratios and imagine scenarios that tend to converge toward the physiological requirement of algae in terms of N:P:Si.

5.2. Features of ICEP for Documented Rivers

[24] The number of rivers where the ICEP could be calculated was limited, because in many cases N, P and Si are not measured simultaneously. In the data set compiled here, only 189 can be used in this N-ICEP framework (132 for P-ICEP). It is interesting to mention that the deviations between the two values of the ICEP are larger around the ICEP thresholds value of zero, e.g., when silica is close to a limitation with regard to N and P.

[25] The N-ICEP grouping by sea basin confirms some well-known coastal zone behavior with respect to eutrophication, such as the North Atlantic Ocean, both on European and American sides. Whereas the North Sea is well known for its *Phaeocystis* developments [Lancelot *et al.*, 1987; Lancelot, 1995] and its *Dinophysis* blooms in the Seine Bight [Cugier *et al.*, 2005], eutrophication of the Baltic has been often discussed for its *Cyanobacteria* blooms [Elmgren, 1989; Vahtera *et al.*, 2007; Granéli and Granéli, 2008]. Most rivers connected to these coastal zones have positive ICEP values and thus eutrophication risk (Seine, Scheldt, Rhine for the North Sea; Oder, Elbe, Vistula, Daugava, Neva for the Baltic Sea). Eutrophication manifestations have also been reported in the western Black Sea [Lancelot *et al.*, 2002], and in some part of the Mediterranean, as the Adriatic [McQuatters-Gollop *et al.*, 2009] where the largely positive ICEP suggests a high eutrophication risk (Figure 5). Within the data set of North Atlantic on the U.S. side, the

positive ICEP of the Mississippi is consistent with the severe eutrophication state, leading to trophic chain disturbance as a result of excess in N and P over silica fluxes delivered by the Mississippi [Turner *et al.*, 1998; Turner and Rabalais, 2003; Turner *et al.*, 2006].

[26] Most rivers draining to the North Pacific have a negative ICEP, such as the Huanghe (Yellow River), while a number of smaller rivers such the Lunanhe and the Fuchun Jian rivers show positive ICEP values. Recently, however, the Huanghe, discharging into the Bohai Sea, has experienced a dramatic reduction of freshwater and sediment discharge and export of N, P and Si since the 1980s [Fan and Huang, 2008]. According to these authors, changes in river water quality impacted negatively primary productivity so that the number of species, the values of density and biomass of fish community dropped dramatically.

[27] Hot spots of eutrophication may occur in areas with high population density and large coverage of agricultural area. The tropics and subtropics seem to have a lower risk to eutrophication based on their negative ICEP values. The distinction between the tropics and subtropics and the northern oceans can be explained from the lithology, soils, hydrology and temperature, all factors enhancing rock weathering and leading to elevated silica concentrations and fluxes [Conley, 2002; Garnier *et al.*, 2002a; Humborg *et al.*, 2004, 2006; Garnier *et al.*, 2006; Sferratore *et al.*, 2006; Beusen *et al.*, 2009].

5.3. Role of Reservoirs in Silica Trapping

[28] During the 20th century, according to the International Rivers Network an estimated \$2 trillion was spent for dams' construction [International Rivers Network, 2003]. Analysis of the Global-NEWS database of the large reservoirs (>500 10⁶ m³) [Vörösmarty *et al.*, 1997] shows that their construction increased significantly in the 1950s (a total capacity of 900 10⁹ m³ impounded between 1940 and 1960), accelerated between 1960 and 1980 with an additional installed capacity of 2500 10⁹ m³, then leveled off during the 1990s (700 10⁹ m³ new capacity impounded).

[29] Dams are known to be a major factor of silica retention through (1) sedimentation of suspended solids [Vörösmarty *et al.*, 1997, 2003; Chen *et al.*, 2001, 2008] including particulate silica, amorphous and lithogenic [Chen *et al.*, 2001; Li *et al.*, 2007]; (2) uptake of DSi by diatoms when residence time is compatible with their growth rates, and subsequent sedimentation of ASi incorporated in their frustules [Humborg *et al.*, 1997]; and (3) reduction of DSi fluxes when rivers are regulated by the effect of hydrological alterations affecting ground-surface water interactions [Humborg *et al.*, 2002, 2006, 2008]. Increased loading of N and P may also lead to increasing production, and thus sedimentation, of diatoms in reservoirs [Conley, 2002].

[30] A budget for small reservoirs with a residence time of 2–3 months revealed a DSi retention of 30% [Garnier *et al.*, 1999], whereas at the scale of the Baltic, the DSi fluxes would have decreased for 30%–40% [Humborg *et al.*, 2008], and up to 22% in the Yangtze River [Li *et al.*, 2007] since the 1960s as a result of increased damming.

[31] The direct influence of the reservoirs cannot be shown with the data presented here. However, there is evidence of a

decrease of Si following damming, and associated eutrophication problems [Cociasu *et al.*, 1996; Humborg *et al.*, 2008, and references therein]. Data on DSi concentration and discharge are scarce, both for water entering and leaving the reservoirs. Further research is needed for better evaluating the role of reservoirs in the global silica budget. Also, the impact on Si river export and eutrophication should be considered when planning construction of water diversion for agricultural, domestic and industrial use, as well as dam construction for hydroelectric power development.

5.4. Global Modeling of Past, Current, and Future ICEP

[32] The N-ICEP was selected for analyzing the model results on the 1970 and 2000 as well as the scenarios of the Millennium Ecosystem Assessment (MEA). The changes in ICEP values for the scenarios depend on many different changes in society and in the environment. For example, DSi export may increase as a result of increasing precipitation, and decrease as a result of dam construction. The net result may be no change. River export of N and P is influenced by the volume of agricultural production and the efficiency of nutrient use. River N and P export is also determined by the increase in sewage effluent as a result of increasing numbers of people with a sewerage connection and the degree of sewage water treatment. In general, in industrialized countries as well as in China and India, there is a rapid increase in nutrient use efficiency in agriculture; this increase is rapid in Technogarden and Global Orchestration, and less so in Adapting Mosaic and Order from Strength. However, in some countries (e.g., China and India) the increasing efficiency may not be sufficient to prevent an absolute increase of nutrient transport to rivers, because the agricultural production volume increases more rapidly. Adapting Mosaic is a special case. In this scenario there is recycling of human excreta and better integration of animal manure in agricultural systems, leading to reduced fertilizer use and high nutrient use efficiency. In most developing countries, there is a rapid increase in fertilizer use. This development is most pronounced in Technogarden and Global Orchestration. Globally, the treatment of sewage water is much more advanced in Technogarden and Global Orchestration than in Adapting Mosaic and Order from Strength. The resulting transport of nutrients through river systems to coastal seas may thus be a constant DSi and decreasing N and P in most industrialized countries. In developing countries the scenarios generally indicate a nearly constant DSi export, and increasing N and P from agriculture and sewerage systems. The magnitude of these changes depends on the scenario. More details on the developments in agriculture can be found in the work of Bouwman *et al.* [2009] (agriculture) and Van Drecht *et al.* [2009] (urban wastewater).

[33] The N-ICEP analyzed by oceans/sea shows a same trend for observed and modeled values during the period 1970–2000 (see Figures 3 and 7). The calculated ICEP from the Global-NEWS models indicates an improvement in the Baltic, and no change in the North Pacific. The ICEP markedly increased in northern Europe, the Mediterranean Sea and Black Sea. The 1970s is typically the critical period

for eutrophication, although particularly well documented for lakes (see the pioneer work by Vollenweider [1968]). Between 1970 and 2000, as shown by both observations and modeled scenarios, emissions from point sources markedly decreased in Europe as a result of advanced wastewater treatment and the banning of P-based detergents in laundry machines [Billen *et al.*, 1999; Garnier *et al.*, 2005; Van Drecht *et al.*, 2009]. However, the diffuse sources (especially nitrate, but also phosphorus) markedly increased due to intensification and modernization of the agriculture in the industrialized countries of North America and Europe, so that the excess of N (and P) over silica increased, especially when rivers are controlled by dams with high residence time.

[34] The demand for nutrients in agriculture will increase in all scenarios, associated to the future population growth [Bouwman *et al.*, 2005, 2009]. However, in the AM scenario, N and P use in agriculture is more efficient [Bouwman *et al.*, 2009] so that N leakage can be reduced. Turning to the wastewater flows, we see increasing population, urbanization, sewerage connection, per capita N and P emissions in all scenarios. At the same time, wastewater treatment is lagging, particularly in developing countries. Among the four MEA scenarios, the AM is one where societies develop a strong proactive approach to the management of ecosystems based on simple technologies: animal manure is better integrated in agricultural production systems, and human N and P is recycled to substitute N and P fertilizers [Bouwman *et al.*, 2009]. As a result, river N and P export appears to be slightly reduced by the year 2050 compared to the year 2000. In the GO scenario, TN export increases by 10% in the period 2000–2050, while river P export shows only a small change. Hence, river TN is 12% lower in AM than in GO, and TP export is very similar in GO and AM. The small difference for P is related to the wastewater P flows; wastewater treatment is less advanced in AM than in GO. The reduction of agricultural P flows is comparable to the increase in P flows from households.

6. Concluding Remarks

[35] Humans have been occupying and using the large drainage network and coasts for long and their impact has been severe in some areas of the world and will increase in the future in some other areas [Nixon, 1995]. Because of the negative consequences of this impact, many coastal ecosystems have lost their functionality and their capacity to deliver ecosystem services. The services offered by coastal ecosystems have been estimated to represent about 1/3 of the total economic value of all services offered by natural systems in the world [Costanza *et al.*, 1997; Martínez *et al.*, 2007].

[36] The MEA scenario analysis shows that it will be difficult to reduce the human impact, and consequently the river export of N and P at the coastal marine systems. In addition, increasing the number of reservoirs in the world's river systems will decrease Si river export globally as a result of retention and eutrophication. These simultaneous changes of N:P:Si will result in an increasing ICEP value, indicating an increasing risk that severe problems associated with eutrophication will occur in many places. For the future

decades, the proactive scenarios which involves more sustainable agricultural practices (e.g., AM) is the one which minimize globally the risks of eutrophication in the regions of the southern continents not yet facing eutrophication problems, whereas GO scenarios lead to better improve the situation in the industrialized countries of the north, probably because rehabilitation efforts have already been undertaken there, successfully.

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