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26 December 2024

UNIVERSITÀ DEGLI STUDI DI TORINO

Sources, factors, mechanisms and possible solutions to pollutants in marine ecosystems

by

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Abstract

Challenges. Algal toxins or red-tide toxins produced during algal blooms are naturally-derived toxic emerging contaminants (ECs) that may kill organisms, including humans, through contaminated fish or seafood. Other ECs produced either naturally or anthropogenically ultimately flow into marine waters. Pharmaceuticals are also an important pollution source, mostly due to overproduction and incorrect disposal. Ship breaking and recycle industries (SBRIs) can also cause release of various pollutants and substantially deteriorate habitats and marine biodiversity. Overfishing is significantly increasing due to the global food crisis caused by an increasing world population. It has severe impact on declining stocks, on various marine species and their breeding habitats. Organic matter (OM) pollution and global warming (GW) are key factors that exacerbate these challenges (*e.g.* algal blooms), to which acidification in marine waters should be added as well. Sources, factors and mechanisms of these challenges to marine ecosystems are discussed, including their eventual impact on all forms of life including humans.

Possible Solutions. Algal blooms and their toxins could possibly be controlled by reducing the OM pollution and GW, which would limit regeneration of various photoinduced and microbial products (CO_2, dissolved inorganic carbon, H_2O_2, NO_3^-, NH_4^+, etc). That could in turn reduce photosynthesis and, consequently, make algal blooms less likely. Release of ECs and other pollutants should be controlled in the respective sectors using proper techniques. Overfishing can be limited following ecosystem-based management strategies, restricting fishing (*i*) in essential habitats during the breeding period of marine species, usually for few days; and (*ii*) in some specific marine locations that have high biodiversity and target and non-target ecosystem components. Possible remedial measures are assessed, but their effective coming into force strongly depends on the public awareness of existing problems and on the priority level that such problems will reach in policy making.

Key words: Emerging contaminants; Pharmaceuticals; Algal blooms; Ocean acidification; Ship breaking and recycling industries; Overfishing.

Introduction

Marine ecosystems are adversely affected because of increasing demand from human activities and the effect of global warming (GW), thereby facing a number of challenges (Mostofa et al., 2012). The key problems in marine ecosystems can be summarized as follows:

- Emerging contaminants (ECs) are discharged into water environments, including seawater, because of human activities (Mottaleb et al., 2005, 2009; Richardson, 2007; Ramirez et al., 2009; Teijon et al., 2010; De Laender et al., 2011; Richardson and Ternes, 2011; Vidal-Dorsch et al., 2012; Mostofa et al., 2013a);
- Production of algal toxins or red-tide toxins during algal blooms is increasing due to the effects of organic matter (OM) pollution and GW (Landsberg, 2002; Moore et al., 2008; Prince et al., 2008; Castle and Rodgers Jr., 2009; Southard et al., 2010; Yates and Rogers, 2011; Mostofa et al., 2013b, 2013d);
- Marine surface waters are undergoing acidification (Brooks et al., 2007; Hansen et al., 2007; Doney et al., 2009; Yamamoto-Kawai et al., 2009; Byrne et al., 2010; Hofmann et al., 2010; Beaufort et al., 2011; Cai et al., 2011; Xiao et al., 2011), which is known to cause changes in marine chemistry and production of algal toxins (Gao et al. 2012a, and references therein)
- Ship breaking and recycle industries (SBRIs) along with oil exploration and transportation can have catastrophic effects on biodiversity due to OM, metals and other pollutants (Hossain and Islam, 2006; Reddy et al., 2007; Gbadebo et al., 2009; Demaria, 2010; Sarraf et al., 2010; Zou and Wei, 2010; Abdullah et al., 2012; Neşer et al., 2013a, 2013b; Pasha et al., 2012);
- Overfishing depletes ecosystems and has led to a global decline in fish catches (Jackson et al., 2001; Myers and Worm, 2003; Block et al., 2005; Fromentin and Powers, 2005; Dulvy et al., 2006; Rooker et al., 2008; Srinivasan et al., 2010).

Such problems are increasingly threatening the world's marine resources, and they are directly or indirectly linked with the world's growing population. It has been shown that > 40% of the world's oceans is highly affected by human activities (Halpern et al., 2008). Coastal areas are understandably suffering from the biggest impact, and human activities have depleted > 90% of formerly important species, destroyed > 65% of seagrass and wetland habitats, degraded water quality, and accelerated species invasions in diverse and productive estuaries and coastal seas (Lotze et al., 2006). A recent review showed that 63 % of assessed stocks are in need of rebuilding (Worm et al., 2009). At the same time, GW and related phenomena can accelerate the occurrence of algal blooms and acidification processes in marine ecosystems (Hare et al., 2007; Edmunds 2007; Cooper et al., 2008; Albright 2011; Anlauf et al., 2011; Mostofa et al., 2013b,

2013d). ECs are usually bioaccumulated into fish or other aquatic organisms and seafood, from which they can be transferred to humans and other organisms (Mottaleb et al., 2005, 2009; Ramirez et al., 2009; Richardson and Ternes 2011; van de Merwe et al., 2011).

Yet, marine ecosystems are also a key vital resource for fish and seafood, meeting the demand for fish proteins at relatively low prices (Meryl, 1996; FAO, 2008). More than half of the total animal proteins consumed in several small island states, as well as in Bangladesh, Cambodia, Equatorial Guinea, French Guiana, Gambia, Ghana, Indonesia and Sierra Leone comes from fish (FAO, 2008). Insufficient attention has been paid so far to the critical impacts of sequential declining in marine ecological communities (*e.g.* from ECs emissions, algal blooms, and overfishing), particularly by developing countries. Considering the importance of a sustainable use of marine resources and biodiversity, world communities should pay much more attention to the solution of current problems created by human activities on marine ecosystems.

This paper will provide an overview of important problems such as ECs, harmful algal blooms, acidification, ship breaking and recycle industries (SBRIs), and overfishing. The sources, factors, mechanisms and remedial measures of such challenges are discussed. As far as pharmaceuticals are concerned, the Chinese case of "100 tablets in a bottle" will be discussed as a major cause of ECs release into the environment, and as a suggestion for strategies aimed at the reduction of pharmaceutical pollution in other countries.

Emerging contaminants (ECs)

Emerging contaminants (ECs) are typically defined as a diverse group of both organic and inorganic compounds, which occur in very small amount (usually at concentration levels of nanograms to micrograms per liter), are persistent, have potential health effects on organisms including humans, fish and wildlife, and may have other adverse ecological effects (Mostofa et al., 2013a). ECs include: pharmaceuticals; personal care products (PCPs); endocrine-disrupting compounds (EDCs); steroids and hormones; drinking water disinfection byproducts (DBPs); perfluorinated compounds (PFCs); brominated flame retardants including polybrominated diphenyl ethers; sunscreens/UV filters; surfactants; fragrances; antiseptics; pesticides and herbicides; organotins; plasticizers; heavy metals including As, Sb, Pb, and Hg; algal toxins or red-tide toxins (Mottaleb et al., 2005, 2009; Richardson and Ternes, 2005; Richardson, 2007; Ramirez et al., 2009; Teijon et al., 2010 ; De Laender et al., 2011; Richardson and Ternes, 2011; Mostofa et al., 2012, 2013a; Vidal-Dorsch et al., 2012).

Most ECs in the aquatic environment originate from three major sources (Hirsch et al., 1999; Fent et al., 2006; Richardson and Ternes, 2011; Mostofa et al., 2013a): (*i*) anthropic emissions including atmospheric deposition, effluents of municipal, industrial and agricultural activities, aquaculture, livestock, and compounds excreted from the human body (*e.g.*

pharmaceuticals and their metabolites); (*ii*) natural production, including most notably algal (or phytoplankton) blooms in surface water; and (*iii*) photochemical and/or microbial origin, following alteration of primarily emitted organic substances by photoinduced and/or microbial processes during transport from rivers to lakes, oceans or other water sources (secondary pollution).

Moreover, point sources of pharmaceuticals and other drugs are (Jones et al., 2001; Fent et al., 2006; Richardson, 2007; Corcoran et al., 2011; Richardson and Ternes, 2011; Mostofa et al., 2013a, 2012):

- Discharge of expired and unused pharmaceuticals or drugs from household. The Chinese case of '100 tablets in a bottle' will be discussed later as a showcase example;
- Disposal of unused pharmaceuticals from hospitals;
- Wastewater and solid wastes discharged from pharmaceutical industries;
- Hormones and antibiotics used in aquaculture and livestock;
- Compounds excreted from the human body in the form of non-metabolized parent molecules or as metabolites, after drug ingestion and subsequent excretion. Note that in some cases there is an excretion of 50-80% of the parent compound (Hirsch et al., 1999).

All the above issues are strongly affected and exacerbated by the increase in world's population.

Adverse effects of pharmaceuticals to fish and aquatic life are typically detected in aquatic ecosystems (Grondel et al., 1985; Wishkovsky et al., 1987; Vos et al., 2000; Arukwe, 2001; Jobling et al., 2002, 2006; Cleuvers, 2003, 2004; Thorpe et al., 2003; Brooks et al., 2003, 2005; Schwaiger et al., 2004; Triebskorn et al., 2004; Hoeger et al., 2005; Flippin et al., 2007; Oaks et al., 2004; Caminada et al., 2006; Cunningham et al., 2006; Fent et al., 2006; Filby et al., 2007; Flippin et al., 2007; Johnston et al., 2007; Kim et al., 2007; Owen et al., 2007; Runnalls et al., 2007; Shved et al., 2008; Christen et al., 2010; Corcoran et al., 2010; Nassef et al., 2010; Santos et al., 2010; Cuthbert et al., 2011). Adverse effects include for instance the production of reactive oxygen species in fish (Gonzalez et al., 1998; Laville et al., 2004; Fent et al., 2006; Mostofa et al., 2013a, 2013b). Pharmaceuticals are detected in water at concentrations in the range of ng L^{-1} to µg L^{-1}, and they are also found in fish or other organisms (Jones et al., 2001; Laville et al., 2004; Fent et al., 2006; Ramirez et al., 2009; Corcoran et al., 2010; Santos et al., 2010; Mostofa et al., 2013a). The highest pharmaceutical concentrations (µg L^{-1} to mg L^{-1}) are found in river waters, in the effluents of hospitals/clinics and in those near pharmaceutical industries, and at the outlet of sewerage treatment plants (Qiting and Xiheng, 1988; Holm et al., 1995; Jones et al., 2001; Fent et al., 2006; Santos et al., 2010). Note that concentration levels at which toxic effects of pharmaceuticals on aquatic organisms have been observed are generally between ng L^{-1} and mg L^{-1} (Belfroid and Leonards, 1996; Schulte-Oehlmann et al., 2004; Crane et al., 2006; Corcoran et al., 2010; Santos et al., 2010).

The adverse effects of pharmaceuticals on aquatic life can be summarized as follows:

- Compounds such as estrogens and diclofenac, ibuprofen, propranolol, sulphonamides, fibrates, beta blockers, antibiotics, carbamazepine, serotonin, synthetic steroids, and antineoplasics have an additive acute and chronic toxicity. Among the observed effects, there are: reduction in growth, sperm count, egg production, and reproduction; sexual disruption; inhibition of settlement of larvae; disruption in mitochondrial function, intestine, and immune systems; impaired spermatogenesis; disruption in energy metabolism; cytotoxicity in liver, kidney, and gills; oxidative stress in membrane cells; changes in appetite (Webb, 2001; Jobling et al., 2002, 2006; Cleuvers, 2003, 2004; Thorpe et al., 2003; Schwaiger et al., 2004; Triebskorn et al., 2004; Hoeger et al., 2005; Caminada et al., 2006; Crane et al., 2006; Fent et al., 2006; Filby et al., 2007; Flippin et al., 2007; Johnston et al., 2007; Kim et al., 2007; Owen et al., 2007; Runnalls et al., 2007; Shved et al., 2008; Christen et al., 2010; Corcoran et al., 2010; Nassef et al., 2010; Santos et al., 2010; Cuthbert et al., 2011). As far as ecosystems are concerned, effects may include a decline in biodiversity at different trophic levels such as bacteria, algae, zooplankton, fish, crustaceans, and invertebrates. The intersex condition in the most severely affected fish is associated with reduced fertility (Jobling et al., 2002).

- Endocrine-disrupting pharmaceuticals can adversely affect the reproductive organs and the thyroid system, with population-level consequences. The latter include impact on reproduction of fish and other organisms, increase of exotic species, habitat loss, and lethal diseases in aquatic organisms including fish, amphibians and reptiles (Vos et al., 2000; Arukwe, 2001; Sumpter, 2005; Orlando and Guillette, 2007; Ankley et al., 2009; Kloas et al., 2009; Santos et al., 2010).

- The antidepressants fluoxetine and sertraline, as well as their metabolites, have been detected in effluents of wastewater treatment plants, at concentration levels shown to cause abnormalities in development and endocrine function of Japanese medaka (*Oryzias latipes*) (Brooks et al., 2003, 2005).

- Antibiotics such as tetracycline at environmental concentrations and laboratory conditions (up to mg L^{-1}) may induce development of resistance in microbial assemblages, may have adverse effect on immune systems, and may inhibit growth and reproduction in fish, microorganisms, algae and aquatic plants (Grondel et al., 1985; Wishkovsky et al., 1987; Thomulka and McGee, 1993; Pro et al., 2003; Crane et al., 2006; Yamashita et al., 2006; Santos et al., 2010). Antibiotics may affect fish indirectly, by modulating microbial functions in aquatic ecosystems and by subsequently affecting processes such as denitrification, nitrogen fixation, and organic breakdown (Constanzo et al., 2004).

- Azole antifungal drugs can cause structural changes and functional impairment of cell membranes, ultimately inhibiting fungal growth, decreasing egg production and plasma vitellogenin concentration in fish, inhibiting ovarian growth, and causing reproductive effects in both male and female fish (Ankley et al., 2002, 2006; Panter et al., 2004; Villeneuve et al., 2007; Corcoran et al., 2010).

- Diclofenac residues are responsible for the decline of vulture populations, and for the inhibition of growth of marine phytoplankton species *Dunaliella tertiolecta* (Oaks et al., 2004; DeLorenzo and Fleming 2008; Nassef et al., 2010; Cuthbert et al., 2011).

- Considering different trophic levels, algae are usually more sensitive to specific pharmaceuticals than *Daphnia magna*, which is in turn more sensitive compared to most fish (Webb, 2001; Ferrari et al., 2004; Crane et al., 2006; Fent et al., 2006). Therefore, phytoplankton would be more affected by pharmaceuticals than zooplankton and other aquatic organisms (Ferrari et al., 2004: Fent et al., 2006).

- Benthic species are likely more exposed than pelagic species to pharmaceuticals bound to sediment (Corcoran et al., 2010).

Most effect concentrations of pharmaceuticals in fish have been determined under relatively short-term exposure (*e.g.*, days to weeks). Because fish may be chronically exposed to many pharmaceuticals over time (*e.g.*, for months or possibly years), sufficient concentrations could accumulate in their bodies to cause adverse effects (Corcoran et al., 2010).

Incorrect drug disposal is particularly important as a cause of pollution by pharmaceuticals (Daughton and Ternes, 1999; Hirsch et al., 1999; Jones et al., 2001; EMEA, 2006; Fent et al., 2006; Islam et al., 2010). It is in this context that a popular initiative by Chinese pharmaceutical manufacturers (the so-called '100 tablets in a bottle') comes into play as a case study of what should be avoided. The manufacture and commercialization of widely sold drugs in relatively big tablet stocks, with relatively low cost per single tablet, was initially welcome as a way to decrease expenditure for medicines. Following commercial success, at least 88 pharmaceuticals have been sold in China in the '100 tablets in a bottle' format, with a wide variety of active principles (see Table 1; Mostofa et al., 2012). The main environmental drawback of this initiative is that only a fraction of the tablets is actually used before the expiration period, while the remaining ones are often discharged into household wastes or (even worse) wastewater. In the cases of paracetamol (anti-inflammatory and antipyretic) and prednisone acetate (used for allergic or autoimmune inflammatory diseases), the structures of which are reported in Figure 1, it has been estimated that the ratio of consumed *vs*. disposed-of tablets would be around 10-20% *vs*. 80-90% (Mostofa et al., 2012). It should be highlighted that incorrect drug disposal is a worldwide problem (EMEA, 2006; Roig, 2008). In Europe, the disposal of waste pharmaceuticals is bound by strict control in the cases of manufacturers,

wholesalers, retailers and hospitals (EU 1994). However, the general public is under no obligation to do such action (Daughton and Ternes, 1999). Therefore, most people will either flush unused pharmaceuticals down the drain, or dispose of them in household wastes. The latter will ultimately enter waste landfill sites or, to a lesser degree, be incinerated (Jones et al., 2001). Similar situations are observed in Japan and North America, whereas specific legislative requirements are introduced to ensure that any pharmaceutical reaching the market is assessed for its likely environmental fate and biological effects (EMEA, 2006). There is more limited regulation concerning the environmental impact of pharmaceuticals, and of effluents released from pharmaceutical industries in China, India, Bangladesh, and other developing countries (EMEA, 2006; Islam et al., 2010).

The combination of over-production and excessive disposal of pharmaceuticals can cause environmental pollution *via* several pathways. First of all, unused pharmaceuticals can mix up with natural waters, either through leaching of household wastes by rainwater or upon direct input of household wastes into natural waters (Mostofa et al., 2012; Jones et al., 2001; Fent et al., 2006). Second, residues of pharmaceuticals are present in manufacturers' wastewaters resulting from production processes (Holm et al., 1995; Mostofa et al., 2012). The released compounds are transmitted to fish or other aquatic organisms and seafood, from which they can reach humans through food consumption (Mostofa et al., 2012, 2013a; Mottaleb et al., 2005, 2009; Richardson 2007; Ramirez et al., 2009). Furthermore, over-manufacturing of drugs (such as in the '100 tablets in a bottle' case) has additional production costs as well as environmental impact, due to raw materials, electricity, gases, organic solvents, which are all needed in the process. Finally, it may also result in increased expenditure for medicines by the people and contribute to increasing medical costs, because the purchase of excess drugs would compensate for their lower unit cost (Mostofa et al., 2012).

Fortunately, the Chinese government is now considering taking initiative to enforce a modification of production processes and reduce the bulk amount of tablets per envelope, in particular shifting to paper or plastic sheet that would make tablet management easier and reduce the wasted amount (Mostofa and Liu, 2012). This story tells us that governments should (and could) take measures to prevent and discourage manufacturing policies such as the '100 tablets in a bottle' one, or to modify them in case they have been started. Indeed, commercialization of drug tablets in formats that allow efficient use and reduce waste would provide a benefit both for the environment in terms of reduced pollution, and for the consumer in the form of lower expenditure in medicines, despite the higher cost per tablet that smaller formats would entail.

A major issue concerning the disposed-of drugs is that the active principles and/or metabolites are not efficiently removed by traditional wastewater treatment plants (Castiglioni et al., 2006). Research activity is currently under way to try to upgrade existing technologies, so

that removal efficiencies are improved (Ternes, 2004). In the meanwhile, the reduction of wastes is the more reasonable approach. This is all the more true, considering that even the most traditional technologies for wastewater treatment are very far from having a worldwide distribution.
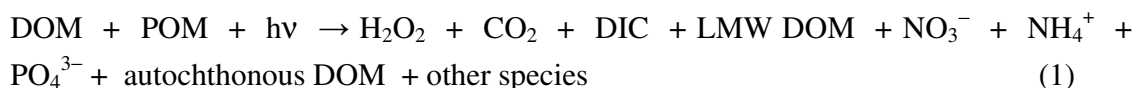
Algal toxins or red tide toxins

Algal toxins or red tide toxins are naturally-derived and toxic ECs produced during harmful algal blooms in surface waters (Tester et al., 1991; Falconer, 1993; Landsberg, 2002; Imai et al., 2006; Imai and Kimura, 2008; Moore et al., 2008; Prince et al., 2008; Castle and Rodgers Jr. 2009; Southard et al., 2010; Yates and Rogers 2011). The occurrence, abundance and geographical distribution of toxin-producing algae or cyanobacterial blooms have substantially increased during the last few decades, because of increased anthropogenic input of organic matter pollution and nutrients as well as global warming (Van Dolah, 2000; Phlips et al., 2004; Yan and Zhou, 2004; Glibert et al., 2005; Luckas et al., 2005; McCarthy et al., 2007; Moore et al., 2008; Mostofa et al., 2013b, 2013d). Algal toxins or red tide toxins produced during algal blooms in surface waters are responsible for physiological, ecological and environmental adverse effects (Hayman et al., 1992; Falconer, 1993; Bricelj and Lonsdale, 1997; Pilotto et al., 1999; Glibert et al., 2001; Fleming et al., 2005; Imai et al., 2006; Álvarez-Salgado et al., 2007; Backer et al., 2005, 2008; Erdner et al., 2008; Imai and Kimura, 2008; Moore et al., 2008; Prince et al., 2008; Sekiguchi and Aksornkoae, 2008; Castle and Rodgers Jr., 2009; Yates and Rogers 2011; Mostofa et al., 2013b, 2013d):

- Deterioration of water quality with high eutrophication.
- Depletion of dissolved oxygen below the pycnocline.
- Loss of seagrasses and benthos.
- Loss of phytoplankton competitor motility.
- Inhibition of enzymes and photosynthesis.
- Cell and membrane damage.
- Mortality of fish, coral reefs, livestock and wildlife.
- Shellfish or finfish poisoning caused by neurotoxic compounds (brevetoxins), produced by blooms of red-tide dinoflagellates such as *Karenia brevis* or other algae.
- Illness or even death of higher organisms or humans, associated with consumption of contaminated fish, seafood and water, inhalation of contaminated aerosol, and contact with contaminated water during outdoor recreational or occupational activities.
- Adverse health effects (*e.g.* eczema or acute respiratory illness) from direct contact with, ingestion, or inhalation of cyanobacteria or various toxins, during recreational or occupational activities (*e.g.* water skiing, water craft riding, swimming, fishing). Such
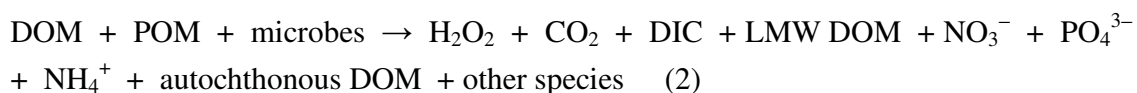
effects can be observed when algal scum appears on the water surface, in coastal sea beaches or in freshwater ecosystems.

The mechanism behind the increasing occurrence of harmful algal blooms is apparently an effect of global warming on waters with high content of DOM and POM through high photosynthesis (Mostofa et al., 2013d). Organic matter (OM) including DOM and POM (*e.g.* phytoplankton or algae) is one of the key factors that can fuel production of additional DOM (autochthonous), nutrients and various photochemical and microbial products (Bushaw et al., 1996; Granéli et al., 1998; Bertilsson and Tranvik, 2000; Moran et al., 2000; Kopáćek et al., 2003; Ma and Green, 2004; Smith and Benner 2005; Stedmon et al., 2007a, 2007b; Zhang et al., 2008; Mostofa and Sakugawa, 2009; Zhang et al., 2009; Cai et al., 2011; Zepp et al., 2011; Mostofa et al., 2011, 2013b, 2013d; Letscher et al., 2013). The complex photoinduced processes can be summarized as follows (Mostofa et al., 2013b, and references therein):

$$DOM + POM + h\nu \rightarrow H_2O_2 + CO_2 + DIC + LMW\ DOM + NO_3^- + NH_4^+ + PO_4^{3-} + autochthonous\ DOM + other\ species \qquad (1)$$

where DIC is usually defined as the sum of an equilibrium mixture of dissolved CO_2, H_2CO_3, HCO_3^-, and CO_3^{2-}, while LMW DOM means low molecular weight DOM formed upon photoinduced fragmentation of larger organic compounds (Bushaw et al., 1996; Granéli et al., 1998; Bertilsson and Tranvik, 2000; Moran et al., 2000; Kopáćek et al., 2003; Smith and Benner 2005; Stedmon et al., 2007a, 2007b; Mostofa et al., 2011; Ma and Green 2004; Mostofa and Sakugawa 2009; Remington et al., 2011; Zepp et al., 2011). Increased stability of the water column as a consequence of warming may enhance the photoinduced degradation of DOM and POM, by combination of high temperature and longer exposure of the water surface layer to sunlight (Huisman et al., 2006; Mostofa et al., 2013d).

Despite very different mechanisms involved, microbial processes have several analogies with photochemical ones as far as the final products are concerned (Köhler et al., 2002; Ma and Green, 2004; Millero, 2007; Moran et al., 2000; Mostofa et al., 2007; Mostofa and Sakugawa, 2009; Zhang et al., 2009; Letscher et al., 2013; Mostofa et al., 2011, 2013b, 2013d and references therein):

$$DOM + POM + microbes \rightarrow H_2O_2 + CO_2 + DIC + LMW\ DOM + NO_3^- + PO_4^{3-} + NH_4^+ + autochthonous\ DOM + other\ species \qquad (2)$$

The compounds formed from DOM and POM because of photochemical and microbial processes would be substantially increased due to increased temperature following global warming. These compounds act as nutrients, enhancing photosynthesis and, as a consequence, primary production as summarized in earlier reports (Fig. 2; Mostofa et al., 2013b, 2013d). This phenomenon has the consequence of increasing the worldwide incidence of harmful algal blooms, in waters with high contents of DOM and POM (*e.g.* algae or phytoplankton) in the presence of sufficient light or high water temperature. This can lead to further eutrophication of DOM-rich waters. Indeed, more extensive eutrophication and hypoxia have been observed in river-dominated ocean margins because of climate and land use changes (Bianchi et al., 2009; Greene et al., 2009; Howarth et al., 2011).

Regeneration of autochthonous DOM and nutrients from POM in DOM-rich waters (Goldman et al., 1972; Carrillo et al., 2002;Vähätalo et al., 2003; Kopáček et al., 2003, 2004; Vähätalo and Zepp, 2005; Bronk et al., 2007; Stedmon et al., 2007a, 2007b; Li et al., 2008; Zhang et al., 2008; Stedmon et al., 2007a, 2007b; Li et al., 2008; Zhang et al., 2009; Fu et al., 2010; Mostofa et al., 2011, 2013a; Letscher et al., 2013) is a key factor for the enhancement of photosynthesis or primary production and for the subsequent harmful algal blooms (Goldman et al., 1972; Vähätalo et al., 2003; Bronk et al., 2007; Mostofa et al., 2013b, 2013d). The process is favored in surface waters by the increase of water temperature and of the vertical stratification period, and by the extension of the euphotic zone that is expected to take place because of global warming (Huisman et al., 2006; Mostofa et al., 2013d). The self-powering potential of this process is of particular concern, and could possibly have a role in the increasing occurrence of harmful algal blooms or toxic phytoplankton populations (Richardson and Jorgensen, 1996; Harvell et al., 1999; Davis et al., 2009; Mostofa et al., 2013b, 2013d). In contrast, global warming can affect waters with low contents of DOM in the opposite direction, by inhibiting the production and regeneration of various compounds. This would ultimately limit photosynthesis and primary production and, as a consequence, reduce algal blooms (Mostofa et al., 2013b, 2013d). The process can proceed either by gradually decreasing the total contents of DOM and nutrients, or by reducing the nutrients at equal DOM.

Introduction of allochthonous nutrients can have negligible effects in waters with high contents of DOM and POM, which often regenerate nutrients on their own (Vähätalo et al., 2003; Kopáček et al., 2003, 2004; Vähätalo and Zepp, 2005; Li et al., 2008; Fu et al., 2010; Jiang et al., 2011; Mostofa et al., 2011, 2013b; Letscher et al., 2013). For example, riverine delivery of both inorganic and organic nitrogen has only a minor (<15%) impact on Arctic shelf export production (Letscher et al., 2013). In addition, regeneration of DOM and nutrients could severely worsen the quality of DOM- and POM-rich waters, particularly in lakes, estuaries, coastal waters and in the Arctic and Antarctic regions (Vähätalo et al., 2003; Vähätalo and Zepp, 2005; Larsen

et al., 2011; Hessen et al., 1990; Imai et al., 2006; Zhang et al., 2008; Ask et al., 2009; Karlsson et al., 2009; Elofsson, 2010; Letscher et al., 2013; Mostofa et al., 2011, 2013d and references therein). A conceptual model for the generation and occurrence of harmful algal blooms by global warming, linked to photochemical and microbial processing of DOM and POM is presented in Fig. 2 (Mostofa et al., 2013d).

Remedial measures are needed for controlling algal blooms, particularly in lakes and coastal seawaters (McCarthy et al., 2007; Prince et al., 2008; Zhang et al., 2008; Castle and Rodgers Jr., 2009; Southard et al., 2010; Jiang et al., 2011; Yates and Rogers, 2011). Prevention measures are basically centered on avoiding eutrophication (Ollikainen and Honkatukia, 2001; Imai et al., 2006; Gren, 2008; Sekiguchi and Aksornkoae, 2008; Elofsson, 2010 and references therein). In fact, control of organic matter inputs including both DOM and POM can reduce the regeneration of photoproducts, microbial products and nutrients (NH_4^+, NO_3^- and PO_4^{3-}). Such measures would reduce photosynthesis and, as a consequence, primary production in natural waters, also limiting the positive-feedback processes described above. Unfortunately, such measures could work less well in already eutrophic environments, due to nutrient regeneration phenomena. In such cases, removal of algae or phytoplankton during algal blooms using fine, small-mesh nets and removal of sediments (when feasible) could reduce the further photoinduced and microbial release of DOM and nutrients from primary production.

Changes of pH and acidification in marine surface waters

The pH value of the surface water layer is substantially increased during the summer stratification period or during algal blooms, particularly if water is rich in DOM and POM (Fig. 3; Mostofa et al., unpublished data; Paerl and Ustach, 1982; Hinga, 1992; Brezonik et al., 1993; Gennings et al., 2001; Köhler et al., 2002; Engelhaupt et al., 2003; Kopáćek et al., 2003; Lundholm et al., 2004; Ishida et al., 2006; Blackford and Gilbert, 2007; Brooks et al., 2007; Hansen et al., 2007; Minella et al., 2011; Xiao et al., 2011; Minella et al., in press). In the case of Lake Biwa (Japan) a pH increase of ~ 1.6 units has been observed in the surface layer (0-10 m), and a decrease of 0.3-0.6 units in deeper layers (20-80 m) during the summer stratification period. During the vertical mixing period, pH = 7.6-7.7 was uniform throughout the water column (Fig. 3; Mostofa et al., unpublished data; Mostofa et al., 2005). Similar results have been observed across the southern North Sea, where annual pH ranges varied from <0.2 in areas of low biological activity to >1.0 in areas influenced by riverine inputs (Blackford and Gilbert, 2007).

The pH increase in surface stratified waters during summer is mostly related to products of photoinduced degradation or respiration of DOM and POM and to photosynthesis, which consumes CO_2. Moreover, microbial degradation products could account for the pH decrease in deeper water layers (Mostofa et al., 2005; Mostofa et al., 2013a, 2013e; Feely et al., 2008, 2010;

Byrne et al., 2010; Cai et al., 2011). DOM photodegradation experiments have shown a pH increase after a certain irradiation time (Gennings et al., 2001; Köhler et al., 2002; Brinkmann et al., 2003; Kopáćek et al., 2003; Mostofa and Sakugawa, unpublished data). In contrast, microbial degradation of DOM and POM (dark incubation experiments or field observations in subsurface layers) is commonly found to decrease pH (Mostofa et al., unpublished data; Feely et al., 2008, 2010; Byrne et al., 2010; Cai et al., 2011).

A complex chain of interrelationships would be operational between water pH and several photoinduced processes involving DOM and POM, including the production of nutrients (NH_4^+, PO_4^{3-}, NO_3^-, NO_2^-), DIC, or other substances arising from DOM and/or POM photobleaching (Bushaw et al., 1996; Bertilsson and Tranvik, 2000; Moran et al., 2000; Osburn et al., 2001; Zeebe and Wolf-Gladrow, 2001; Köhler et al., 2002; Kopáćek et al., 2003; Smith and Benner, 2005; Li et al., 2008; Sulzberger and Durisch-Kaiser, 2009; Remington et al., 2011; Mostofa et al., 2011, 2013b, 2013d). On the one side, DOM and POM photoprocessing could affect water pH. On the other side, pH can alter nutrient speciation including for instance the proportion of NH_3 to NH_4^+ and of PO_4^{3-} to HPO_4^{2-}, which are very sensitive to small pH variations around 8 (Zeebe and Wolf-Gladrow, 2001). Variation of pH would also modify the photochemical production of low molecular weight acids, DIC and nutrients from DOM and POM, acidified waters usually being more photoreactive (Tranvik et al., 1999; Bertilsson and Tranvik 2000; Vione et al., 2009; Remington et al., 2011; Mostofa et al., unpublished data). Moreover, nitrification rates can decrease to zero at pH ~ 6.0–6.5, as the NH_3 substrate disappears from the system (Huesemann et al., 2002).

Seawater acidification that would be linked to buildup of atmospheric CO_2 is a key challenge and could have significant consequences for marine ecosystems (D'Hondt et al., 1994; Evans et al., 2001; Skjelkvåle et al., 2001; Doney, 2005; Orr et al., 2005; Blackford and Gilbert 2007; Doney et al., 2007; Andersson et al., 2008; Fabry et al., 2008; Feely et al., 2008, 2010; Guinotte and Fabry, 2008; Doney et al., 2009; Hofmann and Schellnhuber, 2009; Yamamoto-Kawai et al., 2009; Byrne et al., 2010; Hofmann et al., 2010; Beaufort et al., 2011; Cai et al., 2011;Gao et al. 2012a). Note that the average pH in surface ocean has dropped by approximately 0.1 units globally, which is about a 30% increase in $[H^+]$ (Orr et al., 2005; Fabry et al., 2008). Under the IPCC emission scenario (A1F1) (Houghton et al., 2001), the average surface-ocean pH could decrease by 0.3–0.4 units from pre-industrial values, by the end of this century (Caldeira and Wickett, 2005).

Global warming is operational at the same time and can induce modifications in the euphotic zone as well as lengthen the summer stratification period as discussed earlier. Such changes can impact the vertical O_2 profiles, particularly in deep marine ecosystems (Brewer and Peltzer 2009; Byrne et al., 2010; Mostofa et al., 2013b, 2013d), where lower O_2 availability can

have important effects on marine organisms. Moreover, high [CO_2] is found to enhance the release of dissolved organic carbon from phytoplankton cells (Riebesell, 2004). Released DOM could undergo degradation with formation of various products and nutrients, which might favor algal blooms according to the conceptual model shown in Fig. 2 (Mostofa et al., 2013b). Acidification is thus expected to act in a complex system (Beaufort et al., 2011), where it might be difficult to completely distinguish which effects can be purely attributed to pH decrease, and which ones to other consequences of GW and other ocean changes that are simultaneously active. The scenario could be made even more complex by interactions between several factors (Gao et al. 2012a).

Ocean acidification would decrease the saturation states of carbonate minerals and subsequently change the calcification rates of some marine organisms, thereby affecting aquatic food chains (Barker and Elderfield, 2002; Fabry et al., 2008; Kurihara, 2008; Cohen et al., 2009; Cohen and Holcomb, 2009; Moy et al., 2009; Buck and Folger, 2010; Morita et al., 2010; Cooley et al., 2010; Hofmann et al., 2010; Kroeker et al., 2010; Beaufort et al., 2011; Albright, 2011). The most likely effects of ocean acidification are predicted and identified on coral reefs, shellfish and other aquatic organisms, and can be summarized as follows:

- Shellfish or marine calcifyers are particularly sensitive to increase in acidity/ decrease in pH. This phenomenon can cause dissolution of magnesium calcite, which is an important component of these organisms (*e.g.*, echinoderms and some coralline algae). Impact on the calcification rates of marine calcifying organisms can affect early developmental stages, which include fertilization, sexual reproduction, cleavage, larval settlement, survival and growth, finally causing a substantial population decline (Barker and Elderfield, 2002; Caldeira and Wickett, 2003; Gazeau et al., 2007; Andreas et al., 2008; Kurihara, 2008; Ries et al., 2008; Arnold et al., 2009; de Moel et al., 2009; Moy et al., 2009; Hofmann et al., 2010; Kroeker et al., 2010; Albright, 2011). Shells made of high magnesium or amorphous calcium calcite would be more impacted, because they tend to be dissolved at lower concentrations of carbonic acid compared to shells made of less soluble forms such as calcite and aragonite (Brečević and Nielsen, 1989; Politi et al., 2004; Buck and Folger, 2010; Kroeker et al., 2010). For instance, amorphous calcium carbonate is 30 times more soluble than calcite (Brečević and Nielsen, 1989; Politi et al., 2004). A significant reduction of shell mass and thickness has been observed for several Southern Ocean marine algae and animals, the most likely reason being the recent decrease in seawater pH (Mapstone, 2008).
- Coral reefs are extremely sensitive to acidification, which can: dissolve reef carbonate; reduce the development of coral larvae into juvenile colonies; decrease growth rates of juvenile scleractinian corals; increase sperm mortality; cause a decline in the early

developmental stages (fertilization, sexual reproduction, metabolism, cleavage, larval settlement and reproductive stages); reduce algal symbiosis and post-settlement growth; delay the onset of calcification and alter crystal morphology and composition; increase juvenile mortality because of slower post-settlement growth; reduce effective population size and fecundity, and disrupt the generation of sturdy skeletons and the resilience of reef-building corals (Done, 1999; Langdon and Atkinson, 2005; Edmunds, 2007; Albright et al., 2008; Kuffner et al., 2008; Kurihara, 2008; Cohen et al., 2009; de Moel et al., 2009; Cohen and Holcomb, 2009; Buck and Folger, 2010; Morita et al., 2010; Albright, 2011; Albright and Langdon, 2011; Nakamura et al., 2011). Such effects have an impact on the overall growth and reproduction, and on populations of corals as a whole. The synergistic effects of elevated seawater temperature and of CO_2-driven ocean acidification are responsible for coral bleaching, and for the decline in growth and calcification rates (Hare et al., 2007; Edmunds, 2007; Cooper et al., 2008; Albright, 2011; Anlauf et al., 2011). Differently from corals, it has been shown that calcareous algae that also contribute to build the reef frame can recruit, grow, and calcify under lower pH conditions (Kuffner et al., 2008).

- In some marine invertebrates, calcification of larval and juvenile or smaller individuals is often more sensitive to acidification compared to adults or larger individuals (Kurihara, 2008; Maier et al., 2009; Waldbusser et al., 2010).

- Any decline in shellfish and coral reefs, which constitute the foundation of marine ecosystems, would substantially affect food webs and marine population dynamics, including fish and other organisms (Doherty and Fowler, 1994; GCRMN, 2002; Riegl et al., 2009; Cooley et al., 2010; Albright, 2011). In fact, coral reefs are generally used as habitats by many marine organisms and are a center for biodiversity, where nearly one-third of all fish species live (NMFS, 2004). Changes in food chains could significantly alter global marine harvests, which in 2006 provided 110 million metric tons of food for humans and were valued at US$160 billion (Cooley et al., 2010).

- Photosynthesis and nitrogen fixation of some coccolithophores, prokaryotes and cyanobacteria are either unmodified or increased or decreased in high-CO_2 water (Doney et al., 2009; Gao et al. 2012b; Mostofa et al., 2013b, 2013d). Toxins produced by harmful algae might increase due to ocean acidification (Tatters et al. 2012). Interaction of ocean acidification and solar UV-B radiation decrease the growth and photochemical yield of the red tide alga of Phaeocystis globosa (Chen and Gao 2011).

An important issue that should be taken into account, as far as acidification is concerned, is that dissolution of atmospheric CO_2 into ocean water is not the only possible cause of pH modification. Therefore, it might not be easy to experimentally determine the exact contribution

of CO_2 buildup to the pH decrease. For instance, significant variations of the partial pressure of CO_2 in seawater (pCO_2) have been observed along the P16 N transect at 152 °W in the North Pacific (Fabry et al., 2008). Moreover, it has been shown that acidification of seawater can also be caused by eutrophication, or by several factors including photosynthesis, respiration, temperature, light, and nutrients (Paasche, 2001; Bollmann et al., 2002; Colmenero-Hidalgo et al., 2002; Blackford and Gilbert 2007; Bollmann and Herrle 2007; Zondervan 2007; Feng et al., 2008; Beaufort et al., 2011; Cai et al., 2011). All these factors could significantly modify water alkalinity and, therefore, the variation of pH upon CO_2 dissolution. Another example, although referred to inland freshwaters, is the recovery of acidification in waters of European countries (Curtis et al., 2005; Battarbee et al., 2012; Murphy et al., 2012), which suggests that several processes with the potential to modify water pH can be operational at the same time.

Here an account is given of some additional processes that could potentially modify the pH of oceanic waters. Of course, it should be considered that CO_2 dissolution is operational on a global scale, while other processes have a more local impact. Anyway, in limited locations the pH changes due to local processes can be significant compared to those caused by CO_2 dissolution, which should be taken into account in the interpretation of pH data.

In addition to enhanced dissolution of atmospheric CO_2, pH at the seawater surface can be modified by the following processes:

(i) Photoinduced and microbial processes in low-DOM waters can produce relatively low amounts of products such as CO_2, DIC, H_2O_2, low molecular weight (LMW) substances, other acid-containing organic photoproducts, and autochthonous DOM (Mostofa et al., 2013b, 2013d). In the absence of allochthonous nutrients, the low amount of such compounds would give a limited support to primary productivity and cause limited CO_2 consumption, which could keep pH values lower compared to more productive sites. Such a behavior works in the opposite direction than the mechanism causing algal blooms in water with high contents of DOM and POM (Mostofa et al., 2013b, 2013d). Interestingly most freshwater cyanobacteria, and in particular the species associated with harmful algal blooms are poor competitors with other phytoplankton at low pH (Shapiro, 1973; Paerl and Ustach, 1982).

(ii) Atmospheric acid deposition and acid rain (involving most notably HNO_3 and H_2SO_4) can have an impact on the pH values and the geochemistry of surface waters (Beamish, 1976; Worrall and Burt, 2007). Model results suggest that acid rain could also affect the pH of seawater (Doney et al., 2007). Agricultural activities, through the oxidation of nitrogen fertilizers to nitrate, can further contribute to the decrease of seawater alkalinity (Mackenzie, 1995; Doney et al., 2007) which, as a consequence, can decrease water pH. Seawater acidification due to acid rains or agriculture would be very limited on a global scale, but it could be quite important in coastal

areas where the impact of human activities is considerably higher than in the average ocean (Doney et al., 2007).

(*iii*) Global warming can substantially increase the surface-water temperature, which can enhance the rate of photoinduced and microbial degradation of DOM and POM. It may modify seasonal patterns in chlorophyll or primary production, contents of nutrients, carbon cycling, pH values, microbial food web stimulation, and the depth of the mixing layer (Mostofa et al., 2013b; 2013d). More stable stratification during the summer period would favor photochemical processes in the euphotic zone, thereby leading to more extensive photoprocessing of DOM. In some cases these processes have been shown to increase the pH of the surface water layer, while the opposite effect (pH decrease) has been observed in sub-surface water (Mostofa et al., 2005; Byrne et al., 2010; Cai et al., 2011; Mostofa et al., 2013a, 2013e).

(*iv*) Worldwide increase in harmful algal blooms may be connected with regeneration of CO_2, DIC and nutrients from such algae or phytoplankton, thereby enhancing autochthonous DOM (Fig. 2; Zhang et al., 2008; Zhang et al., 2009; Ballare et al., 2011; Zepp et al., 2011; Mostofa et al., 2011, 2013d and references therein). Such effects might induce high production of CO_2 in surface waters, with a potential role in acidification.

The possible remedial measures for acidification are not easy to be implemented and, in some cases, they could have uncertain effects. On a global scale, limitation of the ocean acidification is clearly a part of the important task of fighting against global warming. Therefore, it implies the difficult goal of limiting CO_2 emissions which, as far as acidification is concerned, would be more important compared to other greenhouse gases. Other remedial actions against acidification could work on a local scale, provided that the effects they are intended to control are important in decreasing the pH and alkalinity of water. In some coastal waters, a beneficial action could be achieved by controlling the anthropogenic emissions of SO_2 and NO_x in the atmosphere and the discharge of N-containing fertilizers from agricultural activities. In this way one could reduce acidic depositions and acid rain, as well as the input of compounds that take part to transformation reactions that lower the pH of water.

Waters with low primary production are more exposed to acidification processes. From this point of view, a way to decrease the susceptibility of oligotrophic water to acidification could be to favor the primary production, *e.g.* by enhancing the release of terrestrial DOM in natural waters through increased runoff (Evans et al., 2001). However, partial eutrophication of very oligotrophic waters could be a risky procedure if not properly controlled. As shown before, elevated primary productivity could start a self-augmenting process that could lead to increased probability of harmful algal blooms (Bianchi et al., 2009; Greene et al., 2009; Howarth et al., 2011; Mostofa et al., 2013b, 2013d).

Ship breaking and recycling industries (SBRIs)

Ship breaking is the process of cutting and breaking apart old ships to recycle scrap metals, along with simultaneous scrapping or disposal of expired or unused ships (Demaria, 2010; Abdullah et al., 2012). Ship breaking is currently carried out mostly by developing countries, and the level of activity in terms of light displacement ton (LDT) by country from 1994 to 2009 is: India (42%), Bangladesh (23%), China (15%), Pakistan (8%), Vietnam (1%), Turkey (1%), and others (10%) (NCSG 2011; Abdullah et al., 2012). SBRIs are important sources of hazardous contaminants along the coastal seashore, most notably in the case of old oil tankers, bulk carriers, general cargo, container ships and passenger ships. On the other hand, SBRIs are also key sources of cheap iron and steel, for construction and other development purposes in the respective countries. SBRIs during ship breaking or demolition can produce three kinds of pollutants (Islam and Hossain, 1986; Zhijie, 1988; Bhatt, 2004; Hossain and Islam, 2004; UNESCO, 2004; Hossain and Islam, 2006; Reddy et al., 2007; Gbadebo et al., 2009; Demaria, 2010; Sarraf et al., 2010; Zou and Wei, 2010; Abdullah et al., 2012; Neşer et al., 2013a, 2013b; Pasha et al., 2012):

(*i*) liquid wastes, which include for instance
- oils and oil products (engine oil, bilge oil, hydraulic and lubricant oils and grease);
- persistent organic pollutants including polychlorinated biphenyls (PCBs, used *e.g.* in transformers);
- polycyclic aromatic hydrocarbons (PAHs);
- ozone depleting substances (ODSs) (*e.g.* CFCs and Halons);
- preservative coatings;
- organotins including monobutyltin (MBT), dibutyltin (DBT) and tributyltin (TBT);
- waste inorganic liquids (*e.g.* sulfuric acid);
- waste organic liquids; reusable organic liquids;
- miscellaneous (mainly sewage);

(*ii*) solid wastes, which include for instance
- various types of asbestos;
- paint chips;
- heavy metals such as mercury (Hg), cadmium (Cd), lead (Pb), arsenic (As), chromium (Cr), copper (Cu), manganese (Mn), iron (Fe), zinc (Zn), nickel (Ni) and aluminum (Al);
- polyvinyl chloride (PVC);
- solid ODSs (*e.g.* polyurethane);
- solid PCB-contaminated wastes (*e.g.* capacitors and ballasts);
- plastic;

- sludge;
- glass;
- cuttings;
- ceramics

(*iii*) gaseous wastes, which include for instance
- sulfur fumes;
- dioxins produced during burning of chlorine-containing products such as PCBs and PVC;
- ODSs, when they are released into the atmosphere;
- volatile (at high temperature) toxic components of marine paints and anti-fouling paints (such as lead, arsenic and pesticides), during the furnace of cutting ships in re-rolling mills;
- miscellaneous gases during ship demolition.

These toxic and hazardous materials from yards or waste dumping sites of SBRIs are often released into the surrounding environment, thereby polluting water and adversely affecting living organisms as well as humans. Impacts on both marine areas and nearby land environments can be summarized as follows (Desai and Vyas, 1997; Majumdar, 1997; Mehta, 1997; Soni, 1997; Trivedi, 1997; Bhatt, 2004; Mandal, 2004; UNESCO, 2004; Hossain and Islam, 2006; Reddy et al., 2007; Demaria, 2010; Subba Rao, 2011; Abdullah et al., 2012; Mostofa et al., 2013b):
- Decline in fish communities, size and standing stock, along with fish contamination in nearby marine waters (up to 100 km distance, or more);
- Decline in fish habitats, eggs and larvae;
- Decline in primary production (*e.g.* algae or phytoplankton), zooplankton communities and their standing stock, which affects all the aquatic food web;
- Decline in benthic invertebrates and their standing stock;
- Decline in terrestrial vegetation, and of agricultural productivity in soils near SBRIs;
- Contamination of agricultural crops and vegetables due to use of polluted water;
- Contamination of groundwater aquifers;
- Adverse health problems for workers and nearby village people, including diseases (*e.g.* throat burning, kidney diseases, respiratory disorders, endocrine disruption, reproductive abnormalities, neurological problems, asthma, angiosarcoma, cancer, diarrhea), upon exposure to contaminated environments including water, soil, air, seafood, flora and fauna;
- Noise pollution to people living very close to SBRI yards;
- Unsuitable and harmful coastal sea waters for recreational purposes;

- Toxic effects and population decline for marine birds, mammals, crustaceans, turtles and reptiles, through uptake of contaminated fish, polluted waters and other seafood;
- Decline in flora, fauna and other aquatic plants or mangroves;
- Deaths of cattle upon feeding on contaminated food;
- Contribution to acid rains from atmospheric emissions during the furnace of cutting ships in re-rolling mills;
- Overall loss of biodiversity (species diversity, genetic diversity and ecosystem diversity) of nearby marine and terrestrial ecosystems.

Fisheries of Bombay Ducks (*Harpodon neherius*), Hilsa fish, prawns and other species have declined by approximately 50-100% at three places that are 50 km away from ship-breaking industries in India (Dholakia, 1997; Demaria, 2010). Heavy metals and other toxic contaminants have been detected in fish communities, various kinds of sea food, suspended particulate matter (SPM), marine sediments and soils, with values that can be several times higher than the maximum standard level (Mehta, 1997; Khan and Khan, 2003; Kulshrestha et al., 2004; Tripathi et al., 2004; Reddy et al., 2005; Hossain and Islam, 2006; Basha et al., 2007; Mitra et al., 2012; Neşer et al., 2013b). Therefore, all organisms including humans are susceptible to adverse health effects through uptake of contaminated sea food.

Because a substantial part of world's SBRI activity is carried out in developing countries, most of the pollutants released from SBRIs are usually discharged into the surrounding coastal marine ecosystems, often without any pretreatment. Main reasons for such an alarming level of pollution are: (*i*) lack of knowledge about environmental impacts of those pollutants; (*ii*) lack of technology to treat or recycle pollutants released from SBRIs; (*iii*) search for profit by SBRI owners, who are often unwilling to take remedial measures; (*iv*) lack of proper rules and regulations to control SBRIs in developing countries. The latter issue is closely linked to the fact that SBRIs are often closely related to other important economic activities such as construction, which slows down strict implementation and enforcement of regulations.

It is vital to make a safe and environmentally sound yard of all SBRIs, along with solving their pollution problems. SBRI evolution toward sustainable development can be provided through reasonable and enforceable legislative and judicial action, which takes a balanced approach but does not diminish the value of coastal conservation (Abdullah et al., 2012). Each country should take initiatives for sustainable development and follow certain obligations, which can be listed as follows (Mostofa et al., 2012; Pasha et al., 2012; Hassan, 2010; Hossain and Islam, 2006):
- Each SBRI yard should be conducted in an exclusive zone, from which pollutants could not be directly released into marine and terrestrial ecosystems;

- Advanced techniques from developed countries should be supplied to each SBRI, so that they can treat or recycle pollutants;
- Awareness should be raised among workers, SBRI owners, the general population, as well as people who are directly involved, on the release of various pollutants and their hazardous impact on surrounding environments, organisms and humans.

Overfishing

The decline in fish stocks, habitats and the biodiversity of marine waters can have several causes, but overfishing is certainly a major one. Unfortunately, the substantially increasing demand of fish proteins for an increasing world's populations is being met by a combination of industrial-scale commercial fishing, various netting techniques, as well as illegal and unregulated or unreported fishing (Table 2; Rotschild et al., 1994; Pauly and Christensen, 1995; Tegner and Dayton, 1999; Jackson et al., 2001; Burkhardt-Holm et al., 2002; Daskalov, 2002; Pauly et al., 2002; Myers and Worm, 2003; Platt et al., 2003; Bascompte et al., 2005; Block et al., 2005; Fromentin and Powers, 2005; Dulvy et al., 2006; Rooker et al., 2008; Srinivasan et al., 2010). The decline of fish communities also has other causes, including toxic algal blooms (Landsberg, 2002; Etheridge, 2010; Mostofa et al., 2013b, 2013c), emission of emerging contaminants by agriculture and industry (including SBRIs, see above), as well as other human activities (Richardson and Ternes, 2011; Richardson, 2007; Sarraf et al., 2010; Zou and Wei, 2010; Abdullah et al., 2012; Mostofa et al., 2013a; Pasha et al., 2012). Last but not least, there are the effects of global warming with associated water stratification, depletion of dissolved O_2 and acidification (Matear and Hirst, 2003; Ben Rais Lasram et al., 2010; Keeling et al. 2010; Mostofa et al., 2013b, 2013d). These problems add to overfishing by impacting water quality and/or enhancing the deterioration of food resources for fish, with consequences that span from disease and mortality of fish communities to severe reduction of fish breeding in marine ecosystems.

Industrialized fisheries can typically reduce community biomass by 80% within 15 years of exploitation (Myers and Worm, 2003). Overfishing has affected 36–53 % of fish stocks in more than half of the world's exclusive economic zones (EEZs), from 1950 to 2004 (Srinivasan et al., 2010). Overfishing is not exclusive of saltwater: fish catch, particularly of brown trout, has decreased by approximately 50% over a 15-year period in many Swiss rivers and streams (Burkhardt-Holm et al., 2002). The catch per unit effort (CPUE) of trawl shrimp has decreased by approximately ~52% in 2000-2001 (284.23 kg day^{-1}) compared to 1992-1993 (592.78 kg day^{-1}) in coastal waters of Bay of Bengal (Nurul Amin et al., 2006). However, during the same period the total fishing effort has increased by approximately 58%, from 7,065 fishing days in 1992-1993 to 11,160 fishing days in 2000-2001 (Table 2). More intensive exploitation is not

without effect: the maximum sustainable yield (MSY) of trawl shrimp has declined by approximately 54% in 2001 (3,441 tons) compared to 1989 (7000-8000 tons) (Nurul Amin et al., 2006).

A rare anadromous species in tropical water is the Hilsa fish (*Clupeidae Tenualosa ilisha*), the catches of which have increased by approximately 101% from 1983-1984 (144,438 tons) to 2007-2008 (290,000 tons) in Bangladesh, and by 567% from 1966-1975 (1457 tons) to 1995-2004 (9726 tons) at Hooghly – Matlah estuary in India (Table 2; BOBLME, 2010). Moreover, there has been a 6-13% increase from 2005-2006 (15,836 tons) to 2007-2008 (17,952 tons) and 2008-2009 (16,744 tons) at the Ayeyarwaddy and Yangon Division of the Irrawaddy Delta, southwest coast, Myanmar (BOBLME, 2010). Hilsa fish (locally known as Ilish) migrates for spawning from the Bay of Bengal into estuaries and into most of the upstream rivers up to 100 km in Bangladesh/India/Myanmar, during the monsoon (July –November) and the spring warming (February – May) (Pandit and Hora, 1951; Ghosh and Nangpal, 1970; FAO, 1971; UNDP, 1985; BOBLME, 2010). Landings of Hilsa fish have significantly declined, by up to 100% in different river mouths and several coastal locations. This is due to decrease in freshwater discharge from upstream rivers or international rivers, and to cross-dam construction either for electricity or for the Irrigation and Flood Control Project in both Bangladesh and India (Ganapati, 1973; Ghosh, 1976; Haldar et. al., 1992; Mahmood et. al., 1994; Haldar and Rahman, 1998; BOBLME, 2010). Such a decline might also be due to deterioration of water quality, because of environmentally driven changes (*e.g.* pollutants released from ship breaking and recycle industries, agricultural pesticides, sewage and other industries), loss of habitat, overfishing (Farakka Barrage on the Bhagirathi River), and global warming (Dholakia, 1997; Haldar et al., 2001; BOBLME, 2010; Demaria, 2010). The traditional habitat of the Hilsa fish is the Bengal delta in the Bay of Bengal, the world's largest flooded wetland that includes the combined basin of three main river systems: Bhagirathi, Padma (two tributaries of Ganges) and Meghna, a tributary of Brahmaputra, along with the river Hooghly of India and the Irrawaddy of Myanmar. Moreover, Hilsa is also found in Satil Arab, Tigris and Euphrates of Iran and Iraq, and in Indus of Pakistan. River water nurses millions of larvae, which become juvenile and adult Hilsa and then migrate towards the sea. Overfishing and human-driven environmental changes can significantly affect Hilsa populations (Dholakia, 1997; Haldar et al., 2001; BOBLME, 2010; Demaria, 2010).

Top pelagic predators such as bluefin tuna, *Thunnus thynnus*, are often found in the Mediterranean Sea, Black Sea, Atlantic Ocean, and Pacific Ocean, but they have undergone a substantial decline (Farley and Davis, 1998; Fromentin and Powers, 2005; Carlsson et al., 2007; Rooker et al., 2008; Yamada et al., 2009; Kimura et al., 2010; Kitagawa et al., 2010; Riccioni et al., 2010; Teo and Block, 2010; Muhling et al., 2011a; MacKenzie and Mariani, 2012). Atlantic

bluefin tuna (ABFT) is a highly migratory species that feeds in cold waters in North Atlantic and migrates to tropical seas to spawn (Muhling et al., 2011b). Mediterranean fisheries have been the main source of bluefin tuna since mid 1990s, but the total reported catch data are at the same time interesting and alarming. Yearly catches have declined by approximately 43% (20,000 tons) in the late 1980s from the 35,000 tons in the 1950s and 60s (ICCAT, 2009; Marion et al., 2010). However, there has been an increase as high as 150% (50,000 tons) in 1995, after which a ~30% decrease (35,000 tons) was observed in 2005. The most recent (2010) stock assessment showed a global decline of between 29% and 51% over the past 21–39 years, based on summed spawning stock biomass from both the Western and Eastern Mediterranean stocks (Collette et al., 2011). The bluefin has also declined globally, including the eastern and western Atlantic because of over-harvesting (ICCAT, 2003; Myers and Worm, 2003; ICCAT Scientific Commitee, 2010; MacKenzie and Mariani, 2012). The spawning stock biomass of the western Atlantic has collapsed by approximately 75-80%, which could even entail a danger of extinction (ICCAT, 2003; Block et al., 2005; MacKenzie et al., 2009). The reason for such a decline is for the most part commercial overfishing, because bluefin tuna is a highly prized fish and it is the favorite one for sushi and sashimi in Japan and, to some extent, also in other countries such as USA, EU and Russia (Bestor, 2000; Hutchings, 2000; Myers and Worm, 2003; Teo and Block, 2010). The record price set in 2011 was $396,000 for a single large specimen (Frayer, 2011). Moreover, as demand and fish prices rise, exports of fish products from developing nations will tend to rise, leaving fewer fish for local consumption and putting fish protein increasingly out of reach for low-income families (Meryl, 1996).

It is demonstrated that Atlantic bluefin tuna (ABFT) has two main stocks, with spawning grounds in the Gulf of Mexico and in the Mediterranean Sea. It has a high degree of spawning site fidelity, as found from field observations of electronically tagged specimens (Block et al., 2005; Carlsson et al., 2007; Rooker et al., 2008; Westneat, 2009; Froese and Pauly, 2010). Moreover, new studies using satellite tags show that some parts of ABFT (up to ~44%) can spawn in distant oceanic regions, other than the two main breeding grounds (Block et al., 2005; Galuardi et al., 2010; Muhling et al., 2011a). Mortality of bluefin tuna during spawning is quite elevated (Block et al., 2005; Teo and Block, 2010) and might be caused by spawning grounds and conditions, because of increased thermal and hypoxic stress induced by longevity in warm surface waters (Block et al., 2005). As suggested before, global warming can increase surface water temperature, lead to a longer summer stratification period and increase the occurrence of harmful algal blooms through high photosynthesis (Huisman et al., 2006; Mostofa et al., 2013d). All these issues can alter the food web in surface waters (Huisman et al., 2006; Mostofa et al., 2013d) and might be responsible for increased mortality of larvae, juveniles and adult bluefin

tuna in the spawning grounds (Kimura et al., 2010; Chapman et al., 2011; Muhling et al., 2011b; MacKenzie et al., 2012). These effects would add to overfishing in inducing population decline.

Total landings of cod in the ICES sub-divisions 22-24 in the western Baltic Sea have declined by approximately 63% in 2011 (16,332 tons) compared to 1970 (43,959 tons) (WGBFAS, 2012). The estimate is based on fish catching by several countries including Denmark, Finland, Germany, Estonia, Lithuania, Latvia, Poland, and Sweden. Moreover, the global diffusion of industrial-scale commercial fishing has caused a 90% decline of the oceans' populations of large predatory species, such as blue marlin and cod, in the past half century (Myers and Worm, 2003). Such global changes in large predatory fish may have severe consequences on the food web in marine ecosystems (Steele et al., 2000; Jackson et al., 2001; Worm et al., 2002). In fact, the stability of ecological communities significantly depends on the strength of interaction between predators and preys (Bascompte et al., 2005). The disruption of existing interactions on two consecutive levels of a trophic chain can potentially alter the structure and dynamics of the entire food web, through trophic cascades (Paine, 1980; Carpenter and Kitchell 1993; Pace et al., 1999; Pinnegar et al., 2000; Shurin et al., 2002). For instance, a food web model has shown that overfishing of sharks may have contributed to the depletion of herbivorous fish through trophic cascades, thus enhancing the degradation of Caribbean reefs (Bascompte et al., 2005). Strongly interacting tritrophic food chain (TFC) includes species at the base, such as parrotfishes (*Scaridae*) and other herbivores, which are important grazers of macroalgae (Randall, 1967). The removal of herbivores by fishing is partly responsible for the shift of Caribbean reefs from coral- to algae-dominated (Hughes, 1994). These interaction strength combinations can reduce the likelihood of trophic cascades after the overfishing of top predators (Bascompte et al., 2005). A TFC can be exemplified by the case in which a top predator P (*e.g*., the shark) eats a consumer C (*e.g.* parrotfish), which in turn eats a resource R (*e.g*., algae and corals) (Block et al., 2005). Therefore, any decline in the shark may substantially increase the parrotfish, thereby decreasing algae or corals in water.

The forbidding of fishing activities at specific times in specific areas, termed as 'time and area closures' might be a common management tool to protect the spawning fish or parent populations. In this way, one can protect or restore proper age and sex distribution, spawning stocks, and aid the most vulnerable fish populations to recover from overfishing (Beets and Friedlander, 1998; Sala et al., 2001; Heyman et al., 2005; Pelletier et al., 2008; Druon, 2010; Teo and Block, 2010). It is also vital to create marine reserves or protected areas in each country, within the territorial coastal marine waters, in which fishing is banned. In this way one can protect sea plants, animals and habitats, thereby preserving marine biodiversity. Some countries have already taken initiatives, and other countries should follow them. For example:

(1) Australia has created the world's largest Marine Reserve Network of reef and marine life, covering nearly 1.2 million square miles -a third of the nation's waters- around the country's borders (Edyvane, 1999; McGuirk, 2012). This is an example to be followed, if one wants to save biodiversity and avoid overfishing.

(2) With the goal of establishing an "ecologically coherent" network of marine protected areas within Northeast Atlantic waters, the Convention for the Protection of the Marine Environment of the North-East Atlantic (the 'OSPAR Convention') has been signed by 16 Parties including Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, UK, Northern Ireland and the European Union (Ardron, 2008).

(3) Since 1999, China has banned fishing in different areas of the Bohai Sea, Yellow Sea and East China Sea, beginning on June 1 for three to three-and-a-half months, as well as in northern parts of the South China Sea, including waters around Huangyan Island, for the next two and a half months (Cheng et al., 2006). The aim is to protect fishery resources and to preserve their sustainable growth and productivity.

(4) To increase the popular Hilsa fish in different parts of the Bay of Bengal and its coastal rivers, Bangladesh has banned fish catching during the peak breeding period from September 25 to October. Moreover, it has banned catch, transportation, marketing, selling and possessing of juvenile Hilsa (*jatka*, up to 23.0 cm size), between 1 November and 31 May every year (BOBLME, 2010).

In the case of the highly exploited Mediterranean Sea, the diffusion of aquaculture has played a significant role in compensating for declining catches and in providing an alternative economic activity to struggling fisheries (Grigorakis and Rigos, 2011). This is an example that could be followed in other parts of the world, but high attention should be paid at the environmental impact of aquaculture, including water pollution by pharmaceutical compounds such as antibiotics (Rico et al., 2012).

Effect of world's populations on marine problems

The first issue is how humans relate with problems in marine ecosystems. An increasing world population has an increasing demand of food, medicines, goods and habitats. All these issues are directly or indirectly associated with marine problems, such as overfishing, increasing emission of pharmaceuticals and other ECs, increasing activity of SBRIs, and increase in plastic wastes, oil exploration and transportation, and algal blooms (Fig. 4; Mostofa et al., 2012). In particular, algal blooms are closely connected to the increase in OM (DOM and POM) inputs and to the effect of global warming.

The second issue is the way problems in marine ecosystems affect humans and other

organisms. The fast depletion of fish stocks by overfishing and environmental deterioration could constitute an economic as well as an ecological problem, ruining fishing communities and seriously damaging the whole fishing-based supply chain. An example in this sense is constituted by recent difficulties of fisheries in the Mediterranean, which has undergone overfishing for decades (Grigorakis and Rigos, 2011). Release of pollutants to marine environments is a serious threat to human health, because food consumption including most notably seafood is a major route of transmission of ECs to both humans and other organisms (Fig. 5; Mostofa et al., 2012).

To have an idea of the pollution load, one can consider that world's population was 3 billion in 1960, 7 billion in 2012 and will be approximately 10.6 billion in 2050 (UNFPA, 2011). The present pollution of marine waters by human activities can be roughly assessed by considering that each person can pollute 20 L/day, which makes approximately 5.1×10^4 billion L/year worldwide (Mostofa et al., 2012). This volume might seem small when compared to the total volume of waters in oceans, $\sim 1.37 \times 10^{12}$ billion L (Garrison, 2007), but one should consider that a considerable fraction of the pollution is concentrated in coastal or estuarine zones that can be key breeding areas for some marine species. Considering the demands of the world's population (7 billion in 2012 + 10.5 billions in next 50 years), marine ecosystems could be polluted approximately three times more in the next 50 years compared to the last 50 years (Mostofa et al., 2012). At equal technology, there seems to be little doubt that some control of the world's population could be important to solve problems in marine ecosystems.

Awareness among citizens of all countries

Awareness is an important factor to make citizens understand problems such as the effect of pollution on water environments and the loss of marine biodiversity, and it should be raised in people engaged in all relevant sectors as well as in the general population. There are three series of arguments that could be used to raise awareness. The first is how marine pollution affects humans and other organisms through the food chain, which has been discussed previously. The second is how the loss of marine biodiversity affects the food chain. There is a close connection between marine biodiversity and the availability of food for fish and other organisms, which are further linked with humans. This can be expressed as follows:

Loss of marine biodiversity $\rightarrow$ Loss of food for fish and other organisms $\rightarrow$ Loss of food for humans and other organisms

The third issue is connected with the way problems in marine ecosystems can be mitigated or solved. The highly trans-boundary nature of marine environments requires that in each marine problematic sector, recommended solutions should be followed by many or all countries. One-

country initiatives may be largely insufficient (Mostofa and Liu, 2012), which accounts for the key role played by awareness. Moreover, there is wide space for action by aware citizens in everyday life to limit avoidable pollution in marine ecosystems, drug disposal being one such example. There should also be increased awareness that if immediate protection measures for marine ecosystems are not taken, there will be danger to future generations though contaminated fish or lack of fish and other seafood. The increasing demand of food by the increasing world's populations is in fact a two-sided issue. One the one side, it places a huge burden on marine resources. On the other side, any drastic changes in marine resources may severely impact availability of *e.g.* fish protein, which could further exacerbate the food demand problem. Therefore, among the many needs, a key one is that of raising awareness among citizens of all countries on 'save the marine resources and biodiversity from the devastating consequences of unavoidable changes, save the future generations'.

Conclusion and recommendations

(1) Pollution caused by ECs will require a huge technological effort to be decreased. However, at least in the case of pharmaceuticals, a huge benefit could derive from a relatively simple action. By modifying the commercialization of drugs in many countries, from '100 tablets in a bottle'-like solutions into paper or plastic sheet that can be bought one by one, one could drastically reduce the amount of disposed-of pharmaceuticals.

(2) An attempt to limit the consequences of overfishing can be made by following ecosystem-based management strategies, restricting fishing (*i*) in essential habitats during the breeding period of marine species; and (*ii*) in some specific marine locations that have high biodiversity.

(3) Algal blooms and their produced toxins could be influenced differentially by different ocean change factors (GW, OA, increased UV exposures), and the effects of ocean changes on harmful algae would depend on latitudes, which is almost unknown. SBRIs should be conducted in an exclusive zone, and developed countries should supply state-of-the-art techniques to control and/or recycle pollutants produced from such industries.

(4) Despite a substantial improvement in environmental technologies, unless a real technological revolution occurs, control of world's population appears as a key issue in limiting the problems of marine ecosystems.

(5) The success of many remedial actions is critically dependent on the awareness of citizens of all countries, who should understand that saving marine resources and biodiversity from the consequences of unavoidable changes is vital for future generations.

(6) When considering all previous issues, it is clear that international organizations could potentially play an essential role in raising awareness and coordinating policies aimed at

the protection of marine ecosystems. Comprehensive research is also vital, to achieve sustainable management and share the developed techniques among all countries.

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Table 1. List of pharmaceuticals or medicines sold as '100 tablets in a bottle' in China.

No	Name of the pharmaceuticals (in Chinese)	Name of the pharmaceuticals (in English)	Specifications
1	制霉菌素片	Nystatin Tablets	50万u×100
2	左旋咪唑片	Levomisole Tablets	25 mg×100
3	去痛片	Somedon Tablets	0.5 g×100
4	罗痛定片	Rotundine Tablets	30 mg×100
5	吲哚美辛片	Indometacin Tablets	25 mg×100
6	地西泮片	Diazepam Tablets	2.5 mg×100
7	谷维素	Oryzanol Tablets	10 mg×100
8	阿托品片	Atropine Tablets	0.3 mg×100
9	山莨菪碱片	Anisodamine Tablets	5 mg×100
10	尼群地平片	Nitrendipine Tablets	10 mg×100
11	地高辛片	Digoxin Tablets	0.25 mg×100
12	硝酸异山梨酯片	Isosorbide Dinitrate Tablets	5 mg×100
13	速效救心丸	Available Save Heart Tablets	40 mg×100
14	卡托普利片	Captopril Tablets	25 mg×100
15	螺类酯	Snails Ester Tablets	20 mg×100
16	溴乙新片	Bromhexine Tablets	8 mg×100
17	复方甘草片	Compound Liquorice Tablets	100
18	硫糖铝片	Sucralfate Tablets	0.25 g×100
19	碳酸氢钠片	Sodium Bicarbonate Tablets	0.3 g×100
20	甲氧氯普胺片	Metoclopramide Tablets	5 mg×100
21	马来酸多潘立酮	Domperidone Maleate Tablets	30 mg×100
22	酚酞片	Phenolphthalein Tablets	100 mg×100
23	复方地芬诺酯片	Compound Diphenoxylate Tablets	2.5 mg×100
24	护肝片	Liver-Protecting Tablets	0.35 g×100
25	金胆片	Jindan Tablet Tablets	0.32 g×100
26	消炎利胆片	Nflammation-Resolving Gall-Bladder-Excreting Tablets	0.24 g×100
27	呋塞米片	Furosemide Tablets	20 mg×100
28	氢氯噻嗪片	Hydrochlorothiazide Tablets	25 mg×100
29	阿司匹林肠溶片	Aspirin Tablets	25 mg×100
30	氯苯那敏片	Chlorphenamine Tablets	4 mg×100
31	地塞米松片	Dexamethasone Tablets	0.75 mg×100
32	强的松片	Prednisone Tablets	5 mg×100
33	安宫黄体酮	Medroxyprogesterone Acetate Tablets	2 mg×100
34	乙烯雌酚片	Diethylstilbestrol Tablets	0.5 g×100
35	炔诺酮片	Norethindrone Tablets	0.625 mg×100
36	苯乙双胍片	Phenformin Tablets	25 mg×100
37	呋喃硫胺	Thiamine Tetrahydrofuryl Disulfide Tablets	25 mg×100
38	维生素B2片	Vitamin B2 Tablets	5 mg×100
39	维生素C片	Vitamin C Tablets	0.1 g×100
40	千柏鼻炎片	Qingrejiedu Oral Tablets	100
41	乳癖消	Breast Mass Resolving Tablets	0.32 g×100
42	刺五加片	Acanthopanax Root Tablets	100
43	茶苯海明片	Dimenhydrinate Tablets	25 mg×20
44	磷酸川芎嗪片	Ligustmzine Phosphate Tablets	50 mg×100

(Table 1 continued)

Table 1 (continued)			
45	盐酸普罗帕酮片	Propafenone Hydrochloride Tablets	50 mg × 100
46	呋塞米片	Furosemide Tablets	20 mg × 100
47	复方芦丁片	Compound Rutin Tablets	20 mg × 100
48	青霉胺片	Penicillamine Tablets	0.125 g × 100
49	呋喃妥因肠溶片	Nitrofurantoin Enteric-Coated Tablets	50 mg × 100
50	四环素片	Tetracycline Tablets	25 mg × 100
51	土霉素片	Terramycin Tablets	25 mg × 101
52	复方磺胺甲恶唑片	Compound Sulfamethoxazole Tablets	100
53	氯霉素片	Chloramphenicol Tablets	100
54	制霉素片	Nystatin Tablets	100
55	灰黄霉素片	Griseofulvin Tablets	10 mg × 100
56	盐酸吗啉胍片	Moroxydine Hydrochloride Tablets	10 mg × 100
57	醋酸地塞米松	Dexamethasone Acetate Tablets	
58	氢氯噻嗪片	Hydrochlorothiazide Tablet	25 mg × 100
59	叶酸片	FolicAcid Tablets	5 mg × 100
60	葡醛内酯片	Glucurolactone Tablets	50 mg × 100
61	安乃近片	Metamizole Sodium Tablets	0.5 g × 100
62	吡拉西坦片	Piracetam Tablets	0.4 g × 100
63	盐酸苯海索片	Benzhexol Hydrochloride Tablets	2 mg × 100
64	吡罗昔康片	Piroxicam Tablets	10 mg × 100
65	盐酸美西律片	Mexiletine Hydrochloride Tablets	50 mg × 100
66	桂利嗪片	CinnarizineTablets	25 mg × 100
67	布洛芬片	Ibuprofen Tablets	0.1 g × 100
68	氨茶碱片	Aminophylline Tablets	0.1 g × 100
69	磷酸川芎嗪片	Ligustmzine Phosphate Tablets	50 mg × 100
70	硝酸甘油片	Nitroglycerin Tablets	0.5 mg × 100
71	盐酸赛庚啶片	Cyproheptadine Hydrochloride Tablets	2 mg × 100
72	富马酸酮替芬片	Ketotifen Fumarate Tablets	1 mg × 60
73	盐酸金刚烷胺片	Amantadine Hydrochloride Tablets	0.1 g × 100
74	戊四硝酯片	Pentaerithrityl Tetranitrate Tablets	10 mg × 100
75	呋喃唑酮片	Furazolidone Tablets	0.1 g × 100
76	马来酸氯苯那敏片	Chlorphenamine Maleate Tablets	4 mg × 100
77	呋喃妥因肠溶片	Nitrofurantoin Enteric-coated Tablets	50 mg × 100
78	咳必清	Pentoxyverine Citrate Tablets	25 mg × 100
79	醋酸泼尼松片	Prednisone Acetate Tablets	5 mg × 100
80	鱼腥草素钠片	Sodium Houttyfonate Tablets	30 mg × 100
81	白葡萄球菌片	Staphylococcus Albus Tablets	40 mg × 100
82	双氯芬酸钠肠溶片	Diclofenac Sodium Enteric-coated Tablets	25 mg × 100
83	二羟丙茶碱片	Diprophylline Tablets	0.2 g × 50
84	呋塞米片	Furosemide Tablets	20 mg × 100
85	复方罗布麻片I	Compound Kendir Lenves Tablets	100
86	复方妥英麻黄茶碱片	Compound Phenytoin Sodium，Ephedrin Hydrochloride and Theophylline Tablets	100
87	胱氨酸片	Cystine Tablets	50 mg × 100
88	甲氧氯普胺片	Metoclopramide Tablets	5 mg × 100

Table 2. Changes in the catch per unit effort (CPUE), fishing effort and catches of various fishes in marine ecosystems.

| Fishes	Year		% Changes	References
	from	to		
CPUE for trawl shrimp (kg day^{-1})	1992-1993 (592.8)	2000-2001 (284.2)	(—) 52	Nurul Amin et al. 2006
Fishing effort for trawl shrimp (days)	1992-1993 (7065)	2000-2001 (11160)	(+) 58	Nurul Amin et al. 2006
Hilsa in total catch at the Bay of Bengal, Bangladesh (t)	1983-1984 (144,438)	2007-2008 (290,000)	(+) 101	BOBLME 2010
Hilsa in total catch at around the Hooghly estuary (t)	1966-1975 (1,457)	1995-2004 (9,726)	(+) 567	BOBLME 2010
Hilsa in total catch at the Irrawaddy Delta in Myanmar (t)	2005-2006 (15,836)	2007-2008 (17,952)	(+) 13	BOBLME 2010
Hilsa in total catch at the Irrawaddy Delta in Myanmar (t)	2005-2006 (15,836)	2008-2009 (16,744)	(+) 6	BOBLME 2010
Catches western Baltic cod in subdivision 22-24 (t)	1970 (43,959)	2011 (16,332)	(—) 63	WGBFAS Report 2012
Large predatory fishes (blue marlin, cod)	1950	2000	(—) 90	Myers and Worm 2003
Europe/Asia/N. America/S. America/Africa	1950	2000	(—) 7.0-50	Srinivasan UT et al. 2010
The numbers in parentheses are the amounts for different units.				
t in parentheses indicates the 'tons'				

(a)

(b)
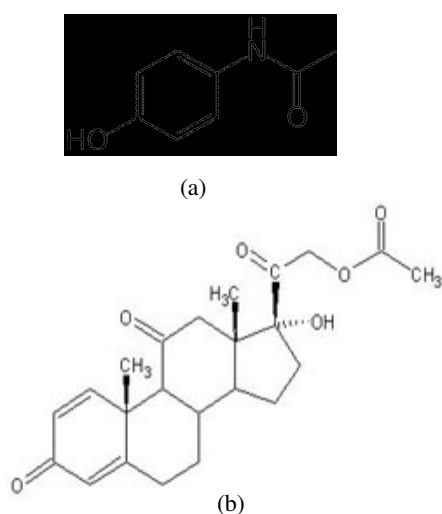
Fig. 1. Molecular structure of paracetamol ($C_8H_9NO_2$) (a) and prednisone acetate ($C_{12}H_{22}O_{11}$) (b).
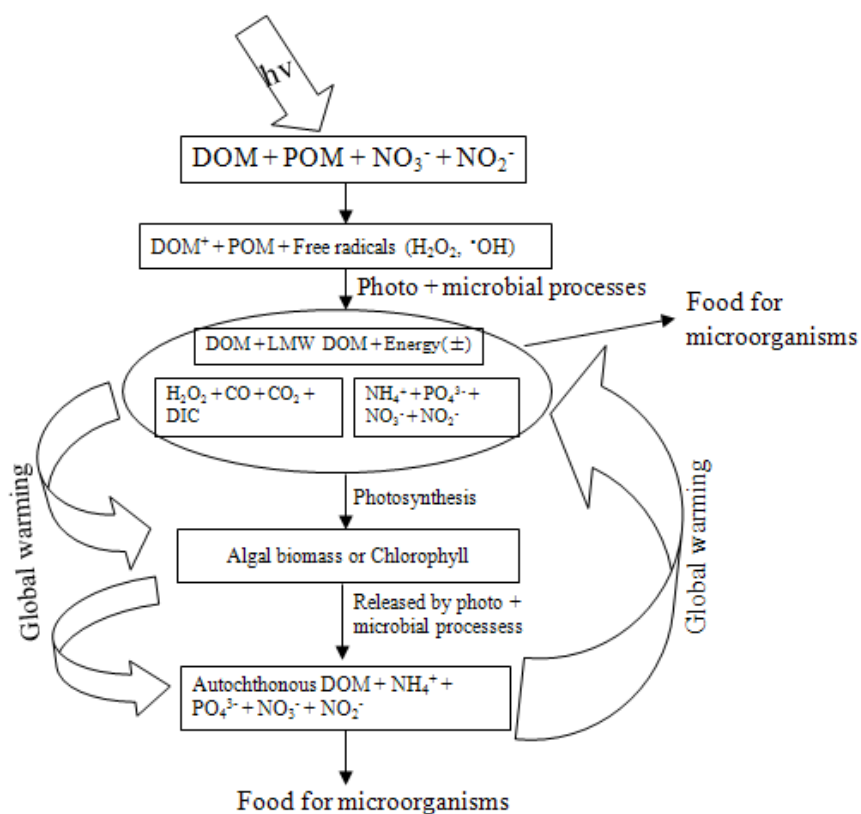

Fig. 2. A conceptual schematic diagram about the response to global warming effects of photoinduced and microbial processes of DOM and POM photoproducts, as well as their possible effects on key biogeochemical processes in natural waters (Data source: Mostofa et al. 2012d).
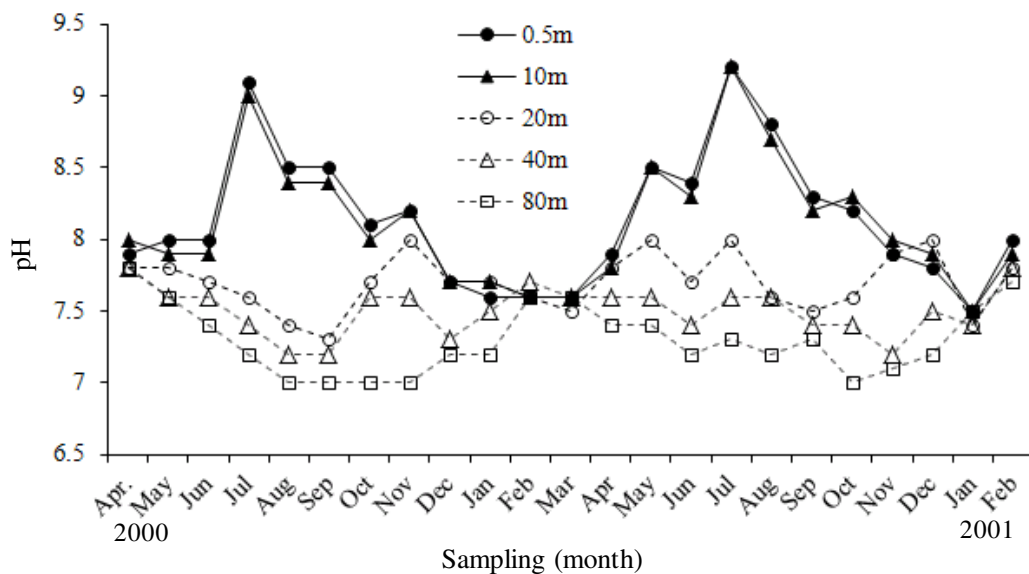
Fig. 3. Month by month variation of pH in the waters of Lake Biwa (Mostofa et al. unpublished data)
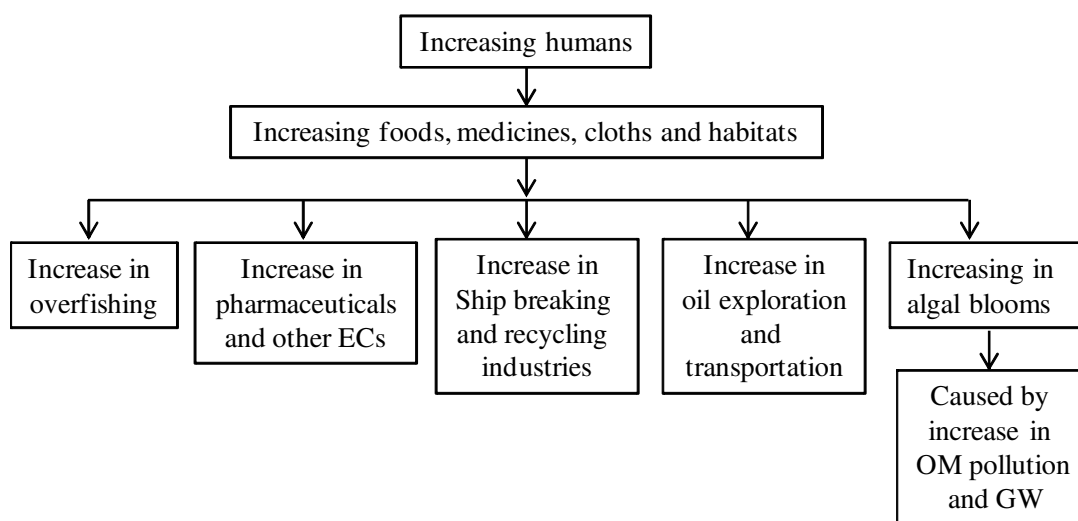

Fig. 4. Relationship between increasing human population and problems in marine ecosystems.
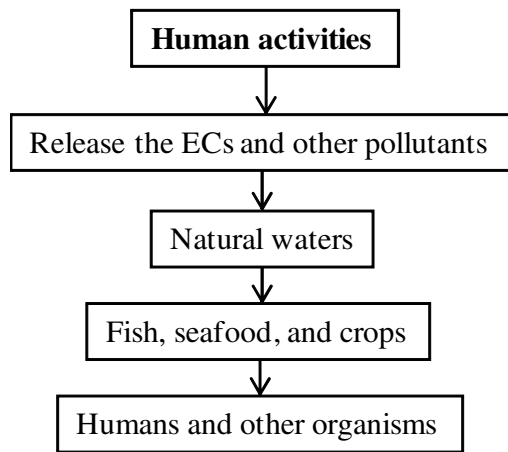
Fig. 5. Transmission of contaminants to humans and other organisms through food consumption.