Phosphates from detergents and eutrophication of surface water ecosystem in India

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Eutrophication, regarded as the most immediate environmental consequence of extensive phosphorus usage in contemporary societies, has received wide attention. If the current level of human-induced global environmental impacts continues, there is a chance of occurrence of nearly 2.4–2.7-fold increase in nitrogen and phosphorus driven eutrophication of terrestrial, freshwater and marine ecosystems in the near future. The main sources of phosphate in aquatic environment, is through household sewage water containing detergents and cleaning preparations, agricultural run-off containing fertilizers, as well as, industrial effluents from fertilizer, detergent and soap industries. The consumption of synthetic detergents is increasing year-by-year due to increasing urbanization and most of them contain phosphate as a 'builder', which increases phosphate loading rates in water bodies. The estimated annual consumption of phosphate-containing laundry detergents for the current population in India is about 2.88 million tonnes and the total outflow of P is estimated to be 146 thousand tonnes per year. Therefore, a major point of concern for checking eutrophication of water bodies, particularly in sensitive areas, is how to reduce P inputs to surface waters.

Keywords: Aquatic ecosystem, detergents, eutrophication, phosphate loading.

PHOSPHORUS (P) is one of the major nutrients needed to sustain all forms of life and cannot be substituted. Similar to fossil fuels, phosphorus (P) is a non-renewable resource and its availability for future generations is becoming obscure. Moreover, with increase in population growth, change towards meat-rich diets and growing demands for food production will further push an increasing demand for phosphate in the near future[1]. Globally, 148 million tonnes of phosphate rock is mined every year and approximately 90% is used for food production[2,3] primarily for the production of agricultural fertilizers (82%) and a smaller fraction for animal feed additions (7%) and food additives (1–2%). The remaining 9% goes to industrial uses such as detergents and metal treatment and other industrial applications[4]. Over a few decades, phosphate use in India which is dependent on imports[5,6] has increased largely and almost doubled in quantity between 2002 and 2009 (80% increase)[7]. It is clear that agriculture is the main user of mined phosphorus globally and only 20% of the phosphorus used in agriculture reaches the food we consume; most of the remaining phosphorus is lost in inefficient steps along the phosphorus cycle. Nearly, 33% of phosphorus entering the soil system is lost by erosion (both water and wind) and the livestock system loses about 45% of the phosphorus entering its system. On the other hand, only about one-tenth of the phosphorus entering the agriculture system is actually consumed by humans, but roughly 90% of the phosphorus entering human system is lost to water bodies through sewage discharge[8,9]. Domestic sources of phosphorus which include sewage, mostly from human and household waste and detergents, is most important as it contributes much of the bioavailable phosphorus for algal bloom. This lost phosphorus from several sources finally ends up in water systems causing widespread pollution in lakes, rivers and coastal areas, algal blooms, and dead zones in the oceans (together with nitrogen). Thus, ironically, phosphorus represents both a scarce non-renewable resource and a pollutant for living systems.

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Environmental impact of phosphorus uses

There exists a broad range of phosphorus-related environmental issues. In socioeconomic context, the major environmental issues are mineral reserve conservation, soil erosion and degradation of soil fertility, animal waste management, sewage and detergent use, and eutrophication. For many years, eutrophication has been regarded as the direct environmental consequence of widespread usage of phosphorus; and problems associated with it have received greater attention, particularly with respect to its effects on freshwater ecosystems[10,11]. Recent report of the United Nation Environmental Programme (UNEP) indicates that globally around 30–40% of the lakes and reservoirs have been affected by eutrophication[12]. Further, a national survey also reveals that more than half of the 40 lakes in China suffer from greater nitrogen and phosphorus enrichment[13].

Eutrophication is the natural ageing of streams or lakes by nutrient enrichment and ultimately promotes aquatic vegetation or algae and phytoplankton causing several problems such as lack of oxygen needed for fish and higher mortality rate; thus, disrupting normal functioning of the aquatic ecosystem. Although eutrophication is a natural process, human activities can greatly hasten the process by increasing the rate at which nutrients and organic substances enter the aquatic ecosystems from the surrounding areas. If the human-induced global environmental impacts continue, nearly 2.4–2.7-fold increase in nitrogen and phosphorus-driven eutrophication will occur in terrestrial, freshwater and marine ecosystems[14]. Primary nutrient, such as carbon, nitrogen, phosphorus and in certain cases, silicon contribute to eutrophication. In fresh water ecosystem, phosphorus is the primary agent for eutrophication, as many algae are able to obtain N from the atmosphere[15]; whereas in estuaries and other marine ecosystems, nitrogen is mainly a limiting factor[16,17]. Moreover, if one element is limiting in excessive presence of other elements, carbon, nitrogen and phosphorus can generate its weight by 12, 71 and 500 times, respectively in algae[18]. Further, in many countries, a huge amount of untreated municipal sewage effluent is discharged directly into freshwater. Therefore, checking eutrophication of water bodies particularly in sensitive areas requires reducing P inputs to surface waters, despite the fact that P is an essential nutrient for crop and animal production.

Sources of P in the aquatic ecosystem

The main sources of phosphate in aquatic environment are household sewage water containing residues of detergents and cleaning preparations, agricultural runoff containing fertilizers as well as industrial effluents from fertilizer, detergent and soap industries[19]. Normally, point sources refer to the discharges from industry and urban wastewater. Diffuse sources (non-point sources) include background losses, losses from agriculture, losses from scattered dwellings, and atmospheric deposition on water bodies. The relative contribution from different sources may vary on regional and local scales depending upon the degree of urbanization, sewage management and intensity of agricultural practices. While industrial sources are locally important, the two major widespread sources of P into surface water are diffuse agriculture inputs and municipal wastewater. Around 16% of the phosphorus entering surface water of the European Union (EU) is derived directly from the application of fertilizers, whereas 34% is derived indirectly from livestock. The domestic sources of phosphorus include sewage mostly carrying human and household waste (24%) and detergents (10%), accounting for 34% of the phosphorus that enters surface waters in the EU (Figure 1)[20]. This source is particularly important, as it appears to represent much of the bioavailable phosphorus entering fresh waters[21]. Therefore, in areas around lakes without intensive agriculture (Lake Geneva and Lake Endine) where municipal wastewater is the primary source of phosphorus, wastewater treatment has been successful for controlling eutrophication. In contrast, agricultural inputs of phosphorus from intensive agriculture are dominant in Lake Sempach, Switzerland, where fertilizer phosphorus represents the main source of eutrophication and therefore an amalgamation of measures including best land management practices and improved wastewater treatment need to be employed.

Majority of P leaked into the environment, enters natural water in non-bio-available form, bound to particulate matter (from runoff and erosion), while only 5–10% occurs in soluble form. Phosphorus fractions in the pre-monsoon stage water samples of Upper Lake, Bhopal revealed that 68.94% of the total P (0.39 mg/l) was in particulate form (more than 450 nm) and remaining 31.06% was in ionic and dissolved form (less than 450 nm; Table 1)[22]. Basically, water samples were filtered with 450 nm membrane filter[23], to separate the particulate form from the dissolved P. In contrast, the soluble P may account for up to 90% of the total P content[24] as in case of municipal sewage water where P-containing detergents and human excreta are the main sources of phosphorus in the aquatic ecosystem. A case study of Lake Dianchi in southern China showed that the total net phosphorus load was 1693 tonnes in 2000, of which mining, phosphate industry, farming, livestock, urban and rural population subsystems contributed to 0.1%, 1.7%, 12.9%, 23.2%, 33.6% and 28.5% respectively[25]. Apparently, the human source, which accounted for 1052.1 tonnes phosphorus and agricultural activities, including both farming and livestock breeding responsible for 610.9 tonnes phosphorus load, dominated the sources for influencing the phosphorous-related water environmental problems in the Lake Dianchi[25].

Constituents of detergents

The term ‘detergent’ is now being applied to all synthetic washing substances. Initially, detergents were used mainly for hand and dish washing, and fine fabric laundering. Later on, with the advent of technologies, all-purpose laundry detergents were first introduced in the US in 1946. Detergents are generally referred to as

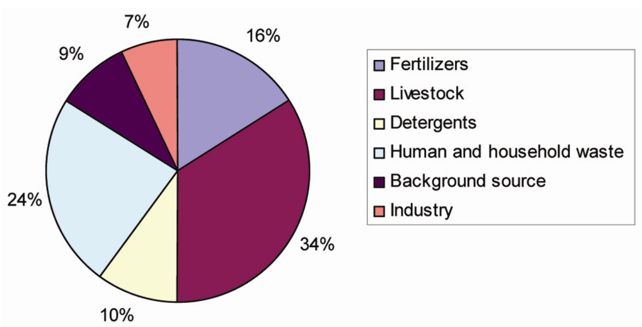

Figure 1. Different P sources entering surface waters of the European Union[20].

Table 1. Dissolved and particulate P content in water sample of Upper Lake of Bhopal[22]

Stages		% of Total P		
		Total P (ppm)	Dissolved P (<450 nm)	Particulate P (>450 nm)
Premonsoon stage	Range	0.28–0.47	20.51–43.24	56.75–79.48
	Mean STDEV	0.386 ± 0.045	31.06 ± 5.962	68.94 ± 5.962
Post-monsoon stage	Range	0.31–0.59	24.47–47.56	60.56–82.46
	Mean STDEV	0.424 ± 0.071	27.49 ± 3.20	72.51 ± 11.71

synthetic compounds that contain an active agent called surfactant. Surfactant has hydrophilic and lipophilic functional groups that emulsify the oily matter and keep the fabric wet and the soil in suspension. Apart from surfactants, another substance called builder is added to detergents to increase efficiency. Phosphate in the form of sodium tripoly phosphates (STPP) is the most commonly used builder which forms complexes with Ca^{++} and Mg^{++} ions creating a favourable condition for detergent action by reducing hardness of water. Soda ash, sodium chloride, sodium sulphate, sodium silicate and zeolite are also used as builders in the detergent. However, STPP as a builder in detergent, is preferred largely due to its greater ability to clean. In addition to builders, detergents have additives such as anti-re-deposition, whitening, bluing and bleaching agents; foam regulators, certain enzymes, perfumes and substances that assure crispness of the material.

Common laundry detergent contains over 40% STPP ($Na_5P_3O_{10}$), although the global development is towards reducing this quantity, because it adversely affects the quality of the aquatic ecosystem and induces eutrophication (algal blooms, kills fishes and poor water quality). The P content in the STPP of detergent is 25.27%. The STPP content of the commonly used detergents in different countries[26] is provided in Table 2; and in India, it ranges from 8–35% (ref. 27). Detergent standards in India require minimum quantity of phosphates as an ingredient; 11% for grade 1 and 7% for grade 2 detergents. However, laboratory testing carried out in 2008 showed that almost all the detergents had much higher phosphorus[28]. No detergent brand available in the market has opted for the Indian eco-label (known as the Ecomark), which certifies environment-friendliness of a product. Further, the detergent P as STPP when released along with the laundry waste water is quickly hydrolysed to orthophosphate. Thus, where STPP is used as builder in laundry detergents, it is estimated to contribute 25–50% of soluble (bio-available) P in untreated municipal water[29].

Scenario of P emissions from detergent use in India

Synthetic detergent, being a mass consumption item, has shown dramatic growth since its inception in 1957. During the last five years, the demand for detergents has been on the rise at an annual growth rate of 10–11% (Figure 2). In India, because of increasing urbanization, the consumption of synthetic detergents is increasing year-by-year, but is less, in comparison to the other Asian countries. In India, the per capita consumption of detergent is approximately 2.8 kg per year (ref. 30), whereas, in countries such as Philippines and Malaysia, the per capita consumption is 3.7 kg, and in USA it is around 10 kg. P contributions from the use of phosphates in detergents are largely dependent on use patterns and marketing conditions. It is a matter of concern that not much information is available about the current production and consumption of phosphate-containing detergent types in India. Due to lack of published data, some assessment about the scenario of P-emissions from detergents in India can be made in the following two ways:

(1) As per the data compiled by Indiastat from Central Statistical Organisation (CSO), about 817,933 tonnes of synthetic detergents were produced during 2009–2010 (Figure 2). Almost all the laundry detergents in India contain STPP, ranging from 8–35% and the STPP content to the tune of 20% can be considered as average value[27]. Thus, total amount of STPP use in detergents is estimated to be 0.16 million tonnes. Since, STPP contains 25.27% P, the estimated outflow of P from detergents is around 41,000 tonnes.

(2) In 1994, per capita consumption of detergents in India was 2.8 kg/annum[30] and it is expected to rise to over 4 kg/annum. Assuming that 60% of the total detergents are consumed as laundry detergents, the estimated consumption of phosphate-containing laundry detergents for the current population is about 2.88 million tonnes and the total outflow of P is estimated to be 145,555 tonnes/annum.

Thus, in India, total out flow of P from detergents to the wastewater is somewhere between 41,000 to 145,555 tonnes/annum.

Trophic status of surface water bodies: global and Indian perspective

On a global basis, researchers have demonstrated a strong correlation between total phosphorus inputs and algal biomass in lakes[31]. Since 1950, phosphorus inputs to the environment have been increasing as the use of phosphate-containing fertilizer, manure and laundry detergent has become more common[32]. Eutrophication is a serious threat to many European lakes[33], for example Lake

Pamvotis in Northwest Greece[34] is under eutrophic condition for the last 40 years. In South Africa, hypertrophic conditions are prevailing at all monitoring stations of Berg River; moreover, the phosphate levels have increased 10-fold due to anthropogenic inputs over the last 20 years[35]. In Zimbabwe, Lake Chivero was also reported to be hypertrophic[36]. According to an estimate in 1965, greater than 80 tonnes of phosphates from sewage, detergents and fertilizers were added daily in the Lake Erie[37]. In China, nutrient unloading from local industries and agricultural fields to the Lake Taihu leads to eutrophication, particularly at Meiliang Bay[38]. Similarly, coastal eutrophication has also received greater attention as a result of nutrient enrichment through runoff from agricultural land into the ocean via rivers and streams. Sewage has been discharged into the oceans all over the world, mostly from urban settlements. The annual global loss of ecosystem service including damage to fisheries from coastal N and P pollution-related hypoxia alone amounts to an estimated US$ 170 billion[39].

According to an estimate by Central Pollution Control Board (CPCB), nearly all the sewage (2.5 billion litres per day) generated along the banks of River Ganga goes directly into the river[40]. Out of the total waste dumped into River Ganga, municipal wastewater and industries contribute 80% and 15% by volume respectively. In the middle Ganga basin, between Allahabad to Ballia, domestic waste water drains are the major sources of pollutants[41]. Usharani *et al.*[42] observed that River Noyyal in Perur, Tamil Nadu, is used for washing purposes and the detergents had enhanced the P load of this river. Similarly at Lake Ranchi, the entry of nutrients particularly P, was due to the use of detergents and the influx of sewage water from surrounding areas[43]. In the 15 years from 1959 to 1974, phosphate–phosphorus concentration in the near-shore waters of Bombay increased from 0.82 to 1.13 μmol/l, i.e. by about 40% and it increased to 2 μmol/l in 1984 (refs 44, 45)

Table 2. STPP contents of some of the known brands of detergents[26,27,*]

Countries	Detergents with STPP builders having STPP content (%)
Denmark	20
Finland	10
France	50
Greece	50
Portugal	70
Spain	60
Sweden	15
UK	45
Hungary	70
Poland	85
India*	8–35

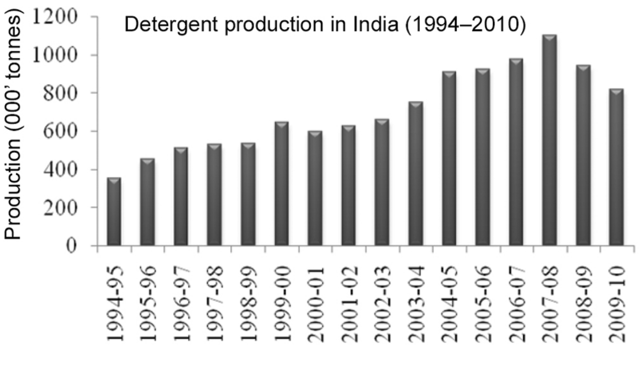

Figure 2. Industrial production of detergents in India (November 1994 to December 2010).

Surface active agent known as 'surfactant' is the basic component of the detergent which has greater biodegradability and is moderately toxic to aquatic organisms, particularly fish. The threshold values which are detrimental for aquatic life are 3–12, 20 and 3–38 mg/l respectively for anionic, cationic and non-ionic detergents. A study revealed that surfactant values in the groundwater and surface waters in urban Kolkata were 0.29 and 0.148 mg/l, respectively; while the corresponding values were 0.025 and 0.90 mg/l in semi-urban districts of Hugli and Howrah[46]. It is clear that groundwater shows lower concentration of surfactants than surface water. At certain places of Kolkata Metropolitan District, the anionic surfactant values in groundwater (0.015 to 0.032 mg/l) and surface water (0.025 to 0.425 mg/l) exceed the critical level of 0.2 mg/l. However, for drinking purpose in India, they are within the permissible limit of 1.0 mg/l (ref. 47). In another study, STPP content in sewage water samples collected from sewage outlets was influenced by the different income groups of the city (Table 3)[48]. Despite the fact that the source apportionment in this article gives a generic picture about India, the information clearly shows the relative importance of detergent P released to our ecosystems.

Specific action plan to reduce P from detergents to aquatic ecosystem

Environmental impact of phosphates in detergents leads to introduction of different control measures and restriction on the use of phosphate-based detergent for household purposes in order to reduce P loading in the surface water bodies. During the last decades, several countries have taken action to reduce phosphorus loading in surface water bodies. Most studies, indicate that a phosphorus reduction by 70–90% is compulsory to trim down the

Table 3. STPP levels (mg/l) in sewage water in the domestic sewage outlets from different income group of four major cities of MP[48]

Cities	HIG	MIG	LIG
Bhopal	11.93	2.38	10.78
Indore	12.87	4.56	9.45
Jabalpur	9.54	2.47	7.84
Gwalior	10.23	3.21	9.48

Table 4. Total treatment efficiency (P-removal) in sewage treatment containing phosphorus[50]

Sewage treatment	P removal (%)
Primary treatment	5–15
Secondary treatment	10–40
Tertiary treatment	>90

eutrophication level and recover original trophic status of water bodies. A significant load reduction up to 40% on phosphorus that enters the surface water bodies can be achieved by imposing a ban on phosphate-based detergents; however, this alone cannot be sufficient for any sizeable improvement. Wastewater treatment facility improvements may result in 30% reduction of phosphorus loading in the aquatic ecosystem. In several developed countries, major improvements in surface water bodies (up to 70–90% reduction in phosphorus loading) were observed when enhanced wastewater treatment and reduced phosphate detergent usage measures were implemented together (Table 4)[49,50]. Apart from phosphate load reduction and sewage treatment, there has been extensive research to find alternatives to phosphates and STPP as a builder in detergents. Sodium citrate, ethylene-diamine-tetraacetic acid (EDTA), nitrilotriacetic acid (NTA) and Zeolite A – polycarboxylate have been extensively researched worldwide as an alternative to phosphate builder. As compared to STPP, sodium citrates are not effective in removing calcium and magnesium cations and also not cost-effective[51]. Similarly, EDTA and NTA builders are less effective in particle dispersing properties as compared to STPP[52]. Moreover, NTA binds with carcinogenic heavy metal ions in the sewage sludge and increases its mobility[52]. A better alternative for STPP has been Zeolite A, which is comparatively inert and derived from aluminium oxide[53]. Zeolite A is non-toxic to humans and aquatic fauna; and produces relatively less toxic by-products. Developed countries such as USA, Switzerland, many countries of the European Union including Germany and Italy have widely adopted use of Zeolite-A as a substitute for STPP[54].

In India, Class-1 cities and Class-II towns together generate 38,254 MLD of sewage, out of which only 30.8% (11,787 MLD) is being treated[55]. Thus in our country, sewage or domestic waste, is a major cause of aquatic pollution and would undoubtedly be a major threat in years to come.

In order to tackle problems related to phosphate-containing detergents, the following action plan is suggested.

- In India, none of the synthetic detergents is phosphate-free due to lack of mandatory legislations. Therefore, there is need for enacting strong mandatory legislation to regulate phosphate content in laundry detergents.
- There is need to create a national database on the total production and consumption of phosphate containing detergents in India and total out flow of P in rural and urban sectors.
- There is need to generate research-based data on the relative input of P into surface water from human excreta, detergents and agriculture for different regions of India.
- As urbanization in India shows an increasing trend, the per capita consumption is also likely to increase, as a result of which the total out flow of P from detergents to the sewage system will also increase remarkably in the near future. Therefore, there is need for mandatory legislation to set up sewage treatment plants in almost all cities so as to recover P not only from detergents, but also from human excreta.
- A partnership approach needs to be promoted in eutrophication management, at both local and national levels, since solutions are generally beyond a single regulatory body or party.

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