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The contribution of household chemicals to environmental discharges via effluents: Combining chemical and behavioural data

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ABSTRACT

Increased concentrations and loads of soluble, bioavailable forms of phosphorus (P) are a major cause of eutrophication in streams, rivers and lakes in many countries around the world. To implement P control measures, it is essential to identify P sources and their relative load contributions. A proportion of P loading generated from household wastewaters is derived from detergents yet the P compositions of the range of domestic detergents and their usage is poorly understood. To quantify P loads from household detergents, we analysed a large range of detergents and cleaning products commonly available in the UK and Europe, comparing regular and eco-labelled products. Chemical data were coupled with survey results on typical household detergents preferences and usage (n = 95 households). We also determined whether the major and trace element signatures of these household detergents could potentially be used as anthropogenic tracers in watercourses. The greatest P concentrations were found for regular dishwasher detergents (43–131 mg P/g detergent) whilst the range of P in eco-labelled dishwasher detergents was much lower (0.7–9.1 mg P/g detergent). Other household cleaning groups contained relatively smaller P concentrations. Considering the survey results, detergents' total P loading generated from one household using either regular or eco labelled products, was 0.414 and 0.021 kg P/year, respectively. Given a household occupancy of 2.7, the P load from all detergent use combined was 0.154 kg P/person/year of which the dishwasher contribution was 0.147 kg P/person/year. In terms of elemental signatures, (DWD) dishwasher detergents were significantly (*P*-value <0.001) different from other household cleaning products in their As, Na, TP, Si, Sr, SRP, Ti, Zn and Zr signatures. Na, P and B were all positively correlated with each other, indicating their potential use as a tracer suite for septic tank effluent in combination with other indices. We conclude that forthcoming legislation for reducing P contents in domestic laundry detergents will not address the dominant environmental P load from DWD and studies such as this are important in promoting and allowing scenarios of benefits from future legislation for DWD.

1. Introduction

Phosphorus (P) is a key nutrient causing eutrophication in surface waters around the world (Smith et al., 1999; Wind, 2007; Withers and Jarvie, 2008). An increase in P concentration under certain conditions of ambient temperature, dissolved organic carbon and nitrogen concentrations in watercourses with long retention time, causes blooms of algae and harmful bacteria (cyanobacteria), (Hilton et al., 2006). This leads to oxygen depletion in fresh waters, discolouration and the unpleasant odour with subsequent threats to fish and aquatic biodiversity (Correll, 1989). Besides agricultural diffuse pollution, P can enter watercourses from municipal Waste Water Treatment Works (WWTW) and septic tanks (ST) that are used as onsite wastewater treatment systems. Since ST are highly variable in P removal and are not regulated for P discharges (unlike WWTW, which may include P stripping technologies), they constitute potential routes for delivery of P from household sources to surface waters (Zurawsky

Abbreviations: DWD, Dishwasher detergents; EDWD, Eco dishwasher detergents; ELD, Eco laundry detergents; FS, Fabric softeners; GCP, General cleaning products; HS, Hand soaps; LD, Laundry detergents; RDWD, Regular dishwasher detergents; RLD, Regular laundry detergents; ST, Septic Tanks; WUL, Washing up liquids; WWTW, Waste water treatment works.

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et al., 2004; Withers et al., 2011) adversely affecting water quality (Ahmed et al., 2005). Jarvie et al. (2006) indicated that point sources such as septic tank effluents represent more significant risk to river eutrophication than agricultural diffuse pollution particularly during low flow periods of spring and summer. Despite recent improvements in water quality following P reductions at major WWTW (Neal et al., 2010), many watercourses are still subject to pollution from municipal wastewater and agricultural runoff (Rothwell et al., 2010; EA, 2012; Withers et al., 2014).

Septic tanks specifically are not designed to remove phosphorus from wastewater. Apart from the initial sludge settling process which removes particulate P, most soluble P in ST effluent are discharged to the environment without treatment. Failing tanks pose even greater risk as all fractions of P including particulate P are released to the environment causing a threat to human health and environmental ecological impacts (Jarvie et al., 2006; Withers et al., 2012).

Household detergents are a source of bioavailable P (phosphate) present in municipal wastewater that if not removed, may eventually reach surface and ground waters (Pattusamy et al., 2013). Phosphates and sodium tripolyphosphates are important components of modern synthetic detergents which consume ~5% of total mined phosphate rock, (Gilbert and DeJong 1997; Prud'homme, 2010). Approximately, 80% of phosphate rock is mined for manufacturing fertilizers and the remaining 20% are used for detergents, animal feeds and other industries. However, the world reserves of the non-renewable resource of phosphate rock are declining in quality and economic-viability, leading to P being added to a European list of critical materials in May 2014 (EC, 2014). Although reserve estimates for phosphate rock are contested as based only on commercial data sources it remains clear that this fundamental resource vital to food production and nutrition is safeguarded for future generations, especially considering global projected population rise (Cordell and White, 2011; Edixhoven et al., 2013).

Detergent phosphates are used as builders to decrease the hardness of water by binding to calcium and magnesium ions present in hard waters which otherwise reduce the effectiveness of the surfactant. This then allows a reduced amount of surfactant to be used. They act by loosening mud, fats, oils and greases from fabrics and surfaces, breaking them into small particles, keeping them suspended in solution and preventing their re-attachment to the cleaned surface (Duthie, 1972).

Many studies have used Boron (B) as a tracer for wastewater (Colin et al., 1998; Jarvie et al., 2006). Boron (B) in the form of borate is also used in modern detergents as a bleaching agent and its use as tracer for municipal wastewater pollution is due to its conservative behaviour in natural waters (Neal et al., 2010). Borate is best suited to hot water washing and since there is a shift in public preference to low temperature washing in the recent years, many detergents manufacturers have substituted oxygen-based bleaching agents instead of borate. The effect of this shift was highlighted by Metzner et al. (1999) who reported a decrease in B concentration in municipal wastewater as a result of replacing B with other bleaching agents. A reduction in the sale of sodium perborates in Europe and UK from 1997 has been reported (RPA, 2008; RIS, 2006) that is expected to lead to lower boron concentrations in surface water. While Bundschuh et al. (1993) stated that elevated B concentrations are indicative of anthropogenic effect, Verstraeten et al. (2005) recommended not to rely solely on B alone in tracing sources of catchment pollution.

Attempts to reduce the environmental impact of detergents have been made by the production of compact powders and tablets. Some alternative builders have been used but were found to have low performance, be less effective or too costly (Kohler, 2006). An alternative builder to sodium tripolyphosphates is Zeolite A which is an artificial zeolite derived from aluminium oxide. It is a relatively inert substance, insoluble in laundry detergents and hence separates from laundry waters. It has a reasonable performance but has limitations as a builder, requiring a co-builder (polycoarboxylic acids) to fully substitute for phosphates (Morse et al., 1995; Kohler, 2006).

Many countries in Western Europe, US, Canada and Japan regulate the use of detergent products containing sodium tripolyphosphate as a measure to control eutrophication through reduction of P loading to WWTW and subsequent discharge to streams and rivers. In the UK, The Detergents Regulations 2010 (SI, 2010 740) restricts the amount of phosphates in domestic laundry detergents from January 2015, stating: “It is an offence to place on the market a detergent intended for use in domestic laundry if the weight of phosphorus as inorganic phosphate contained in the detergent is greater than 0.4% of the weight of the detergent”. This is derived from the 2013 amendment to the EU detergents regulation (684/2004), (EU Parliament Regulation, 2004). It is being discussed that similar restrictions will be placed on dishwater detergents from 2017 and so studies are needed as evidence for the current situation with P in detergents and allow for benefits scenarios for upcoming policies.

In this study 80 laundry products and household detergents available on supermarket shelves, and widely used in the UK and Europe, were tested for P and trace elements concentrations. A survey was devised and distributed in 4 catchments to gain real information on the different types of detergents and household cleaning products that are normally used and the frequency of their use. Detergent products were in a variety of forms (condensed tablets, powders, gels and liquids) of different classes and uses: Laundry detergents (LD), dishwasher detergents (DWD), fabric softeners (FS), washing up liquids (WUL), general cleaning products (GCP) and hand soaps (HS). The aims were: 1) to examine P concentrations in these household chemicals across different usage categories including those determined as ‘eco-products’ compared to regular products; 2) to combine concentration data with survey data on household product usage and cleaning habits to produce household P loading scenarios for regular and eco-product use, and 3) to investigate whether the major and trace element signatures of these household chemicals could be used as an indicator for anthropogenic wastewater input to watercourses.

2. Materials and methods

2.1. Questionnaire on household detergents usage

To gain an informed knowledge of typical household detergent preferences and their usage, we surveyed household detergent use in four catchments in the North East of Scotland as representatives of many catchments in Europe. The catchments varied from intensive agricultural (Rivers Dee and Lunan) to peri-urban (Rivers Don and Ythan), (Fig. 1) with no demographic discrimination, having been selected on the basis of estate tenanted farms, rented accommodation, catchment focus groups and local house to house distribution. The purpose of the survey was to gather real information on detergents use and habits, product preferences and the frequency of their use. The survey was intended to gain information relevant to assessing environmental impacts of septic tanks (ST) but also included those discharging effluent to WWTW. All respondents were questioned whether eco or low P products were considered as viable options as a substitute to regular products and, for those who use eco-products, to comment on their performance, price and availability. Additional questions focussed on ST system condition and management. The response rates to the survey were 58%, yielding 95 responses using ST as wastewater disposal systems and 24 using WWTW.