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Temporal variability of micro-organic contaminants in lowland chalk catchments: New insights into contaminant sources and hydrological processes

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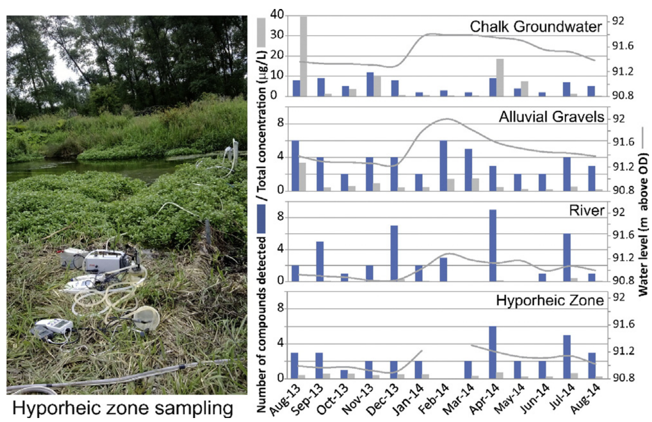
HIGHLIGHTS

- Trace microorganics found in all hydrological compartments in a chalk lowland catchment
- Greatest number of compounds found in Chalk groundwater compared to surface water
- The hyporheic zone shown to be important for MO contaminant attenuation
- High temporal variability has implications for the design of monitoring programmes.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper explores the temporal variation of a broad suite of micro organic (MO) compounds within hydrologically linked compartments of a lowland Chalk catchment, the most important drinking water aquifer in the UK. It presents an assessment of results from relatively high frequency monitoring at a well-characterised site, including the type and concentrations of compounds detected and how they change under different hydrological conditions including exceptionally high groundwater levels and river flow conditions during 2014 and subsequent recovery. This study shows for the first time that within the Chalk groundwater there can be a greater diversity of the MOs compared to surface waters. Within the Chalk 26 different compounds were detected over the duration of the study compared to 17 in the surface water. Plasticisers (0.06–39 μg/L) were found to dominate in the Chalk groundwater on 5 visits (38.4%) accounting for 14.5% of detections but contributing highest concentrations whilst other compounds dominated in the surface water. Trichloroethene and atrazine were among the most frequently detected compounds. The limit for the total pesticide concentration detected did not exceed EU/UK prescribed concentration values for drinking water. Emerging organic compounds such as caffeine, which currently do not have water quality limits, were also detected. The low numbers of compounds found within the hyporheic zone highlight the role of this transient interface in the attenuation and breakdown of the MOs, and provision of an important ecosystem service.

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1. Introduction

In the last few decades there has been a growing interest in the occurrence of micro-organic (MO) contaminants in the terrestrial and aquatic environment, and in their environmental fate and potential toxicity (Halling-Sørensen et al., 1998; Kolpin et al., 2002; Kümmerer, 2009). The contamination of groundwater by MOs is a growing concern and relatively poorly understood compared to other freshwater resources. It is clear from recent studies that trace concentrations of a large range of compounds can be detected in groundwater (Focazio et al., 2008; Lapworth et al., 2012; Loos et al., 2010; Stuart et al., 2012). This continues to be a global problem (Brausch and Rand, 2011; Jurado et al., 2012; Luo et al., 2014). A limited but growing number of studies are using MOs as tracers to fingerprint contaminant sources in surface water and groundwater and understand the transient nature of MOs processing at the groundwater–surface water interface (GSI) (e.g.Buerge et al., 2003; Burke et al., 2011; Engelhardt et al., 2011; Sorensen et al., 2014; Stuart et al., 2014).

A number of recent studies looking at MOs in vulnerable Chalk karstic systems have been published (Hillebrand et al., 2014; Reh et al., 2013), as well as studies focussed on the attenuation of selected MOs by Hillebrand et al. (2015). Research in these karstic settings has highlighted the transient nature of MO contamination and shows the need for greater temporal resolution if effective monitoring is to be undertaken in comparable hydrologic settings. A recent regional analysis of MO occurrence has been carried out for the Chalk of England and France (Lapworth et al., 2015), but this did not have a temporal component and to date there have been very few studies focussed on characterising the temporal variation of both the types and concentrations of MOs, including emerging organic contaminants, present in groundwater and the GSI in lowland chalk settings. In addition, with the development of broad screening techniques (e.g. Lapworth et al., 2015; Wode et al., 2015) it is no longer necessary to be restricted to small numbers of target compounds as has often been the case in previous studies to date.

The Chalk is the most important aquifer system in the UK, and in parts of England is the dominant source of drinking water (Defra, 2015). In the south east of England it can provide up to 80% of the drinking water supply, this area being classified as one of the regions globally with the lowest water availability due to high population density and relatively low rainfall (RGS, 2012). More widely, the Chalk aquifers of north-west Europe form a hugely important natural resource, providing drinking water and sustaining river flows across a large part of southern England and northern France as well as parts of Belgium, Germany, the Netherlands and Denmark. Discharge from the Chalk aquifer sustains river flow and groundwater dependent wetlands and groundwater dependent terrestrial ecosystem which require assessment under the Water Framework Directive (Directive 2000/60/EC). As such it is important to understand the nature, types and concentrations of the MOs found within Chalk groundwater (Lapworth et al., 2015), and in particular at the groundwater/surface water interface, to understand how these change over the hydrological year as well as in response to under the high flow conditions of early 2014. Recent research undertaken at bank infiltration sites in the UK (Ascott et al., 2015) has shown that following extreme high flow conditions it can take up to six months for water quality to return back to baseline conditions.

Both point and diffuse sources of MOs, including emerging compounds, contribute to contamination of groundwater. Factors controlling their entry to groundwater include land use and climate as well as the management of liquid and solid waste and treatment by-products (Bloomfield et al., 2006; Kümmerer, 2009; Schwarzbauer et al., 2002). The fate and concentrations of MOs in groundwater depend on a number of physicochemical processes, including sorption and degradation, as well as the hydrogeological setting including amount and distribution of groundwater recharge, residence times and pathways (Lapworth et al., 2012). The fate of MOs in the hyporheic zone (HZ, the zone of interaction between surface water and groundwater) is an important area of ongoing research (e.g. Lewandowski et al., 2011; Lu et al., 2015a, 2015b; Ward et al., 2015), this is due to its transient flow dynamics; relatively high microbiological activity and reduced phototransformation potential (compared to surface waters). This zone has a capacity to attenuate organic pollution within the river-groundwater continuum (Freitas et al., 2015), and providing an important ecosystem service (Griebler and Avramov, 2015).

This study was carried out at a well-characterised lowland Chalk research observatory in Southern England (Allen et al., 2010). The aims of this study were to determine i) how MO number and concentrations vary through the hydrogeological year ii) in particular whether there is a relationship between any variation observed and groundwater level fluctuations including during hydrological extreme conditions iii) the seasonal variability of the role of the hyporheic zone (HZ) in MO attenuation.

2. Study site

The study was conducted at the Boxford research site in Berkshire, UK. The site was instrumented with piezometers as part of the Lowland Catchment Research Programme (LOCAR) (Wheater and Peach, 2004), in order to characterise the interaction between groundwater, surface water and hyporheic zone in a lowland Chalk catchment (Allen et al., 2010). The site is rural, to the north of it lies a farm which undertakes both arable and dairy activities, while to the south lies the River Lambourn and an associated wetland, both designated as a Site of Special Scientific Interest (SSSI).

2.1. Hydrogeological setting

The geology of the underlying strata is shown in Fig. 1a and comprises Cretaceous Newhaven Chalk Formation overlain by superficial deposits of variable fractions of gravel and sand. The Newhaven Chalk is a soft microporous limestone with fracture and matrix flow both playing a role in movement of groundwater, which can be complex. The superficial deposits are of heterogeneous nature, alluvium with sand and gravel layers with discontinuous peat lenses present beneath the site. Head deposits, a term used to describe a diamicton of poorly-sorted chalk gravel, sand and silt, which are deposited due to periglacial mass flow movement, are found along the sides of and base of valleys in the chalk downs of southern England, see Newell et al. (2015) and references therein for further details on the geology of this area.

Interactions between the surface water and groundwater have been established by Allen et al. (2010), with hydraulic connectivity between the River Lambourn and the superficial gravels as well as the Chalk, these are schematically represented in Fig. 1b including the regional groundwater flow and stage dependant lateral and hyporheic exchange. There is however poor connectivity in the vicinity of the riverbank between the gravels and the Chalk. This is a result of lower permeability due to reworked chalk at the chalk–gravel interface. The overall groundwater flow direction is to the south and within the site is towards the river. A chalk mound present in the vicinity of the Y piezometer (Fig. 1b).

The established behaviour of these interactions during high and low water level is summarised below. During low water level conditions, there is notable input to the river from the Chalk as well as the gravels and alluvium from both southerly and northerly directions. During high water levels the proportion to the River from the Chalk is smaller and there is movement from the river to the gravels and alluvium to the south. The southerly flow of groundwater within the Chalk under the riverbed provides significant groundwater contributions within the wetland adjacent to the River Lambourn (Chambers et al., 2014).

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