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Research article

National scale assessment of total trihalomethanes in Irish drinking water

Connie O'Driscoll [a, b, *], Jerome Sheahan [c], Florence Renou-Wilson [d], Peter Croot [e], Francesco Pilla [f], Bruce Misstear [a], Liwen Xiao [a, **]

[a] Department of Civil, Structural and Environmental Engineering, Trinity College Dublin, Dublin 2, Ireland
[b] Department of Civil Engineering, National University of Ireland, Galway, Ireland
[c] School of Mathematics, Statistics and Applied Mathematics, National University of Ireland, Galway, Ireland
[d] School of Biology and Environmental Science, Science Centre West, University College Dublin, Belfield, Dublin 4, Ireland
[e] Earth and Ocean Sciences, School of Natural Sciences, National University of Ireland Galway, Galway, Ireland
[f] Department of Planning and Environmental Policy, University College Dublin, Dublin, Ireland

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ABSTRACT

Ireland reported the highest non-compliance with respect to total trihalomethanes (TTHMs) in drinking water across the 27 European Union Member States for the year 2010. We carried out a GIS-based investigation of the links between geographical parameters and catchment land-uses with TTHMs concentrations in Irish drinking water. A high risk catchment map was created using peat presence, rainfall (>1400 mm) and slope (<5%) and overlain with a map comprising the national dataset of routinely monitored TTHM concentrations. It appeared evident from the map that the presence of peat, rainfall and slope could be used to identify catchments at high risk to TTHM exceedances. Furthermore, statistical analyses highlighted that the presence of peat soil with agricultural land was a significant driver of TTHM exceedances for all treatment types. PARAFAC analysis from three case studies identified a fluorophore indicative of reprocessed humic natural organic matter as the dominant component following treatment at the three sites. Case studies also indicated that (1) chloroform contributed to the majority of the TTHMs in the drinking water supplies and (2) the supply networks contributed to about 30 $\mu g\,L^{-1}$ of TTHMs.

1. Introduction

Trihalomethanes (THMs), the most prominent class of halogenated disinfection by-products (DBPs) (Krasner et al., 1989), are formed during raw water disinfection. Although many DBPs can be formed during disinfection only THMs and bromate are regulated by the EU (EU, 2014). THMs are formed when chlorine reacts with natural organic matter (NOM) that remains following drinking water pre-treatment. Dissolved organic carbon (DOC - used to quantify NOM) varies in quantity and quality depending on catchment characteristics. Greater quantities of peat soil cover, precipitation, coniferous forestry and lower catchment slope can increase DOC concentrations in water (Cool et al., 2014; Liu et al., 2014; Parry et al., 2015; Valdivia-Garcia et al., 2016). Land-use change can also influence DOC concentrations in water, i.e. damaged peatlands are a net source of aqueous carbon and forest harvesting (Freeman et al., 2001; Nieminen et al., 2015). DOC concentrations are reported to be increasing on an international scale and there has been much debate around the causes for this (De Wit et al., 2007; Evans et al., 2005, 2012; Freeman et al., 2001). In the absence of scientific consensus, misunderstanding about the importance of land-use in water treatment policies has developed (Evans et al., 2012). Increasing DOC concentrations will amplify the challenges for water companies in treating NOM and achieving total THM compliance (total THMs; 100 $\mu g\,L^{-1}$, EU, 2014; 80 $\mu g\,L^{-1}$, US EPA, 2006).

* Corresponding author. Department of Civil, Structural and Environmental Engineering, Trinity College Dublin, Dublin 2, Ireland.
** Corresponding author. Department of Civil, Structural and Environmental Engineering, Trinity College Dublin, Dublin 2, Ireland.
E-mail addresses: connieodriscoll@gmail.com (C. O'Driscoll), liwen.xiao@tcd.ie (L. Xiao).

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Pivotal to the success of NOM removal is an understanding of NOM quality and water managers are being encouraged to adopt an all-encompassing approach by including 'catchment' in drinking water safety plans (WHO, 2011) and understanding the role of 'catchment management' in affecting drinking water quality (Brooks et al., 2015). The use of fluorescence excitation-emission matrix (F-EEM) spectroscopy to differentiate humic- and protein-like fluorescent signals in drinking water is increasing (Baghoth et al., 2011). F-EEM spectroscopy together with PARAFAC (multi-way data analysis using parallel factor analysis, Stedmon et al., 2003) is a direct and economical approach to identify different sources of NOM in drinking waters (Kraus et al., 2010) and many studies have used this approach successfully to establish problematic NOM fractions remaining in drinking water following pre-treatment (Baghoth et al., 2011; Shutova et al., 2014).

In a comparison of the EU Member States, Ireland reported the highest non-compliance for THM exceedances, at least twice those in the next Member State with the next highest percentage non-compliances (EC, 2014). In Ireland, 20% of land area is classified as peat (Connolly and Holden, 2009), containing more than 75% of the national soil organic carbon (Renou-Wilson et al., 2011). Near-intact peatlands actively sequester carbon; however, only 15% of Irish peatlands are deemed as near-intact (Wilson et al., 2013) and following drainage, increases in DOC fluxes of up to 60% have been observed in temperate peat catchments (Evans et al., 2016). Forestry accounts for just above 10% of land area in Ireland compared to a European average of 43%, approximately half of which is located on peat soils and the majority of which are first rotation coniferous plantations. Agricultural area accounts for the greatest percentage of land-use in Ireland (67.36%) and with plans for agricultural expansion and intensification underway (EPA, 2016a,b) an investigation into land-use is warranted to ascertain whether any links exist between Ireland's land-use and the higher TTHM concentrations observed.

Increasing concerns over the effects of THMs on human health have amplified scientific interest in this subject. THMs have shown both genotoxic and carcinogenic effects particularly in the liver and kidney of laboratory animals in epidemiological studies (WHO, 2011). Recent studies report conflicting findings into the relationship between DBPs and reproductive health in human population based studies. Hwang and Jaakkola (2012) report that the risk of stillbirth is related to prenatal exposure to DBPs. Nieuwenhuijsen et al. (2008) found no apparent relationship between THM concentrations in the public drinking water supply and risk of congenital anomalies. Similarly, Iszatt et al. (2013) report that concentrations of THMs in public drinking water do not appear to be related to semen quality in England and Wales. However, Zeng et al. (2014) found that human exposure to DBPs may contribute to reduced semen quality in China.

With increasing attention on the presence of DBPs in drinking water and potential effects of THMs on human health, understanding the role of catchment processes and management in affecting THM concentrations in drinking water quality is vital. This study is the first to address the issue of Irelands THM exceedances at a national scale using geographic and operational factors as were employed by Valdivia-Garcia et al. (2016), but with the addition of catchment land-use factors. Agriculture in particular can cause nutrient enrichment, leading to increased autochthonous activity in receiving waters. The case study sites examined here in detail, using a suite of parameters, have allowed the findings of the statistical analysis to be teased out further. The main objective in this study was to evaluate the relative contribution of geographical parameters and catchment land-use to TTHM concentrations in Irish drinking water.

2. Materials and methods

2.1. High risk catchment maps

Routinely collected data on TTHMs concentrations were obtained from the Irish Environmental Protection Agency (hereafter referred to as the EPA) for the period 2006–2013 in the Republic of Ireland (hereafter referred to as Ireland). Geographical Information System (GIS) coordinates for the abstraction sources were obtained and further GIS datasets included: soil data from the National Soil Dataset, EPA; the Derived Irish Peat Map (DIPM) (Connolly and Holden, 2009); Long-Term Rainfall Averages (LTA) for Ireland, (Walsh, 2012); the Digital Elevation Map for Ireland, EPA; and Irish Rivers and Lakes Segments, EPA. Groundwater vulnerability mapping, derived by combining two GIS layers of subsoil thickness and permeability, plus karst features where present, was obtained from www.gsi.ie (Misstear et al., 2009; Hunter Williams et al., 2013).

While four types of THMs - chloroform, bromodichloromethane, dibrochloromethane and bromoform - are routinely measured in water supply zones, only total THM data (i.e. the sum of the concentrations of the four individual THMs) were available for this study. Water supply zones (hereafter referred to as schemes) differ in area, in the size of the population served and in the volume of water supplied. Not all schemes are accounted for in the final dataset owing to lack of abstraction information. Repeated measurements from each scheme were averaged between years and within years to eliminate seasonality and temporal variation. Only those schemes for which the water source did not change over the study period were included, i.e. 684 water schemes serving 1,062,700.91 m^3 of water daily to a population of 2,289,539. Of these, 270 schemes abstracted from surface water (serving 1,777,888) and 414 from groundwater (serving 511,651).

Maps showing high risk areas were created by integrating geo-located data from three layers, which detailed information about presence of peat, yearly average rainfall (>1400 mm) and slope (<5.25%). The national THM dataset was imported to ArcGIS. Inverse Distance Weighting (IDW) analysis was carried out with the THM values for which a location's coordinates could be attributed.

2.2. Statistical approach

Firstly, using ANOVA we studied the effect of vulnerability on TTHM while adjusting for treatment code, and we studied the effect of treatment code on TTHM while adjusting for vulnerability on the full dataset (684 observations) to determine significant differences between abstraction vulnerability (high and low vulnerability and surface water) with two levels of treatment code. For this, two levels of treatment code were adopted, treatment code level 1 – absence of NOM treatment/chlorination only (409 observations); and treatment code level 2 – treatment with some degree of NOM removal which included all other treatment types (coagulation, flocculation and clarification – 127; dissolved air flotation and filtration – 16; membrane filtration – 10; ozone and GAC treatment – 9; pressure filtration – 10; rapid gravity filtration – 22; simple filtration – 38; slow sand filtration – 43). Concerning the six combinations of treatment code and vulnerability we conducted both parametric and non-parametric procedures. For example, the Kruskal-Wallis test showed strong evidence that not all six population medians were equal. We then conducted pairwise comparisons to see which pairs, among the fifteen possible pairs differed.

Secondly, a General Linear Mixed Model (GLM) approach was used to study the effects on TTHMs of certain land-use variables on surface water abstractions (270 observations) at 7 levels of treatment (chlorination only, coagulation, flocculation and clarification;