The current state of understanding of Emerging Organic Compounds in European groundwaters

GeoERA HOVER WP8

BRITISH GEOLOGICAL SURVEY

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The current state of understanding of Emerging Organic Compounds in European groundwaters

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Summary

In Europe groundwater pollution by emerging organic compounds (EOCs) is still a growing research area. Prioritisation for monitoring EOCs was recently formalised (2019) in Europe by the development of the first voluntary groundwater watch list (GWWL) for emerging substances. Despite this, groundwater occurrence data in the peer reviewed literature for Europe has not been formally reviewed to date. In this study a total of 39 studies were identified for review based on specific selection criteria (geography, publication date, sample size >10, inclusion of EOC data). Data on specific compounds detected in each study, and associated meta-data are compiled and reviewed. The top 2 detected EOC's, Carbamazepine and Caffeine are analysed in further detail. Carbamazepine was detected in 22 studies, in concentrations of up to 2325 ng/L, and Caffeine in 15 studies at concentrations of up to 14.77 µg/L.

The categories used for grouping compounds within the studies are first identified, and the most frequently used are noted and discussed. Eight usage categories are selected for use in this review, building upon widely used categorisation methods and groupings. Significant discrepancies have existed between the categories that are used, their scale and the compounds which are contained within each. The most frequently reported category of compounds were 'Pharmaceuticals', a highly studied group with 135 compounds identified with 31 out of 39 studies (79%) reporting one or more detection of a compound within this category. The scale and purpose of each study are identified and reviewed, as these offer insight into the type of research currently being undertaken in this expanding field of science. A number of studies aim to answer a specific questions about pollution from a given source e.g. WWTP, whereas others aim to identify the baseline EOC's within a given geographical area. Four national scale, 23 regional scale and 12 targeted studies are reported. No links are observed between the year of publication and the scale of study, and no statistical significance between the number of compounds screened for and the number detected in groundwater; the number of compounds screened for and the number of groundwater sites used in the study; or the number of compounds detected in groundwater and the number of groundwater sites used.

Developments in a range of analytical techniques are expanding the number of EOC's that can be detected. Twenty-one, often slightly differing methods are represented, but are principally variants of the LC-MS/GC-MS methodology. Developments in extraction techniques also increases the likelihood that a compound could be detected. Increased number of studies alongside advanced extraction and analytical techniques mean that a large number of compounds can now be detected routinely. However, the use of these methods is not uniform across Europe, and this inevitably influences the current assessment of EOCs in groundwater across Europe.

1 Introduction

The term Emerging Organic Contaminants (EOC's) (Stuart, 2012) is used to describe organic contaminants that are not yet regulated, but may be of current or future concern. Although defined as emerging, they may not be new contaminants, but recently detected using improved sampling and analytical methods (Daughton, 2004). The term 'emerging' is therefore used in this review in the context of compounds of emerging concern. The number of EOC's is expected to increase as these methods develop, and new compounds continue to be released into the environment.

The European Commission's Groundwater Directive (2006/118/EC) sets out to 'prevent and limit input to groundwater of many contaminants of emerging concern. However, whilst there are no formal regulations to control these contaminants, there is currently no requirement to monitor or report on these contaminants.

A growing number of contaminants become regulated every year, it is therefore important to understand their occurrence, movement, fate and impacts. Furthermore, there is a need to understand what data/studies are available in order to progress the understanding of EOC's in groundwater. Questions surrounding their effect, toxicity, movement in the subsurface and unsaturated zone make the process of regulating their use more difficult.

In 2014, an amendment to Annex II of Directive 2006/118/EC stated that a lack of information meant that new groundwater quality standards could not be set for any pollutants. The review highlighted the need to 'obtain new information on other substances posing a potential risk' and this should be implemented by means of a 'Groundwater watch list (GWWL)' which was first implemented earlier this year through the European working group groundwater (CIS) (CIRCABC, 2019).

The impacts on human health and the environment is still relatively unknown for the majority of EOC's. Some studies (Juliano and Magrini, 2017) have reviewed the impact on health, a growing concern due to the relatively unknown impacts of some EOC's. However, the prioritization of compounds is still under review, with a number of prioritisation techniques discussed by Gaston et al., 2019. The authors conclude that a dynamic list of pollutants, where their use, properties and hazards are considered within a prioritisation framework would be the best solution.

EOC's are traditionally categorised by their use, rather than occurrence, transport or impact on the environment. A review of the most common categories used in recent European studies has been completed in section 3.2. Research studies often focus on one of the major usage groups, screening for selected compounds within the identified category. Analysis of EOC's is often costly and time consuming, so a targeted approach to EOC screening is a more practical approach for researchers. However, there is sometimes significant difficulty in categorising compounds into one of these groups, especially when they may belong to more than one grouping. In this review, a categorisation of the detected compounds is suggested based upon an assessment of categories presented in the selected studies and an element of expert opinion by the authors. This is not necessarily a final categorisation, but offers a basis from which to analyse the frequency of detection of different compounds.

Compared to surface water, studies of EOC's in groundwater are relatively novel, with few large studies focusing on the subsurface environment (Lapworth et al., 2012). However, there are an increasing number of national reviews into the state of research into a range EOC's (Banzhaf et al., 2017, van der Aa et al., 2013, Petrie et al., 2015, Cunha et al., 2017, Juliano and Magrini, 2017, Tiedeken et al., 2017, Wilkinson et al., 2017); yet no European scale study to understand the state of the science on a larger scale.

Lapworth et al. (2018) highlight the need for a coordinated approach in the EU to assess contaminants of emerging concern, especially in the groundwater environment due to a significant gap in policy. Collaborating to collect evidence about EOC's will help to inform policy and develop a regulatory framework for groundwater throughout the EU (Lapworth, 2018).

This paper compiles evidence from the latest studies (since 2012) on EOC's in groundwater in Europe. The constant monitoring of the state of knowledge surrounding EOC's is important to identify gaps in knowledge and understand areas for further research. Using studies from across Europe we can understand EOC's in greater detail and aim to develop a unified strategy for the classification, priorities monitoring of EOC's and better understand the balance between small scale vs European wide EOC contamination in groundwater. This review uses 39 European studies where compounds are known to be detected in groundwater.

The aims of this review are to (1) understand the current state of knowledge on EOC's in Europe and the developments in recent years, (2) Understand the different methods for sampling and analysing EOC's in Europe, and (3) highlight ongoing research and further areas for research necessary to develop a picture of EOC's in Europe.

2 Review methodology

The studies included in this critical review were selected based on a number of criteria. The study must be based in Europe; be published in a peer reviewed journals in English on or after 2012; building on the first global review by Lapworth et al (2012); and must identify organic substances that are not currently regulated in groundwater. This includes studies where the work was carried out before 2012, but not published until post-2012. The number of studies were further constrained by selecting studies with ten or more groundwater samples. Studies with fewer samples would considerably increase the number of small targeted studies reviewed, and may not be considered representative of the research carried out in a particular region. These criteria were developed to identify a range of studies that will provide a general understanding of the current state of knowledge and study being undertaken in the field of EOC's in Europe.

The studies were initially identified using a series of web searches using the search engines 'Google Scholar' and 'Web of Science'. The criteria was set for 2012 up to May 2019 and keywords 'Groundwater' or 'aquifer' used to help define the parameters of the search. Wildcards (*) were used to identify papers with selected keywords, increasing the potential for finding relevant studies. The search criteria used:

1. Emerging contaminant* and Europe
2. Groups of contaminants and Europe – pharmaceutical*, antibiotic*, sweetener*, antimicrobial*, caffeine*, polyfluorinated*
3. Specific examples of contaminants and Europe – sucralose, triclosan, DEET, carbamazepine, PFOS
4. Groups of contaminants and European member states

Secondly, a series of research partners with an interest in EOC's in Europe were emailed with the list of studies identified. They were asked to provide any additional published studies they knew about that fit the above criteria. This search was not exhaustive, and further studies likely exist in this topic area. However, the studies identified are adequate to give a good understanding to meet the review aims.

The review aims to look only at natural groundwater, and therefore does not include studies of artificial recharge, treated groundwater and re-injected treated wastewater (Candela et al., 2016). Where studies look at treated/un-natural groundwater, they often also observe the natural system (Cabeza et al., 2012). Only the results for natural groundwater are included in this review. Furthermore, review studies are not included, but where possible the original studies were found and included in this review.

Using the above criteria, a total of 39 studies from 16 European countries were selected for this review (

Figure ***1***).

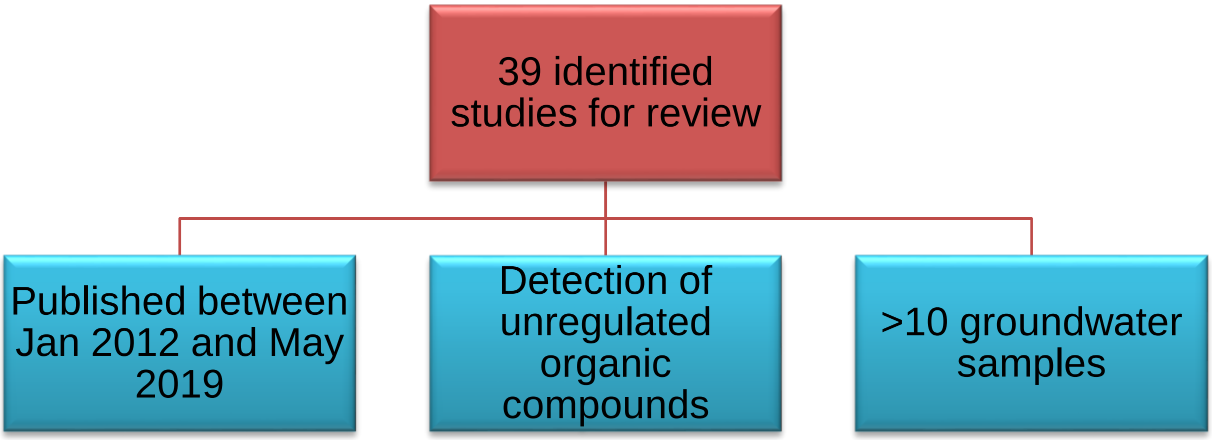


Figure 1: Schematic of the criteria used to identify the studies used in this review

There are a number of limitations to this type of review. The difference in reporting styles between European countries means that there is rarely the same level of detail provided in each study. Furthermore, the scale of a number of studies, means that it is not practical to report on the hydrological conditions, well type and depth, and other important information for a more detailed analysis. In some countries, more than one review has been analysed and reported, which may result in the duplication of some sites.

3 Review

3.1 CURRENT STATE OF KNOWLEDGE

Since the first major global review in 2012 (Lapworth, 2012) there have been developments in the field of EOC's in groundwater. For example, Balderacchi et al. (2014) report on the GENESIS project, which incorporated making suggestions of amendments to the Groundwater Directive. They highlight an increasing concern about emerging contaminants and the need for monitoring for the formulation of conceptual models and the eventual improvement of legislation. Furthermore, after the implementation of threshold values across EU member states, they suggest a consistent monitoring protocol.

One major step towards a unified understanding of the potential threat of EOC's was through the Groundwater Watch List (GWWL) (CIRCABC 2019), developed in response to the 2014 European Commission call for increased monitoring (Lapworth, 2018). The 2014 amendment to Annex II of Directive 2006/118/EC, encouraged an increase in research into organic contaminants, with the purpose of implementing management levels/concentrations for currently unregulated compounds in groundwater. The GWWL broadly mirrors the surface water watchlist (SWWL) in its aims and structure, acting to identify and monitor currently un-regulated contaminants in European groundwaters. In order to introduce regulation on some of the anthropogenic pollutants in EU groundwaters, it is recognised that significant evidence about the compounds is essential. Previous efforts have been made to prioritise emerging compounds in surface waters including Von der Ohe (2011), and a list of hazardous or non-hazardous pollutants in groundwater published by JAGDAG (2012) outlining the determination of these substances, using toxicity, persistence and potential to bioaccumulate. However, the groundwater watch list collates European monitoring data on EOC's that pose a threat to health or the environment, producing a list of substances ordered by their occurrence, mobility and toxicity (Lapworth, 2018). The process was documented so the list can be updated as studies further the knowledge about these attributes for different EOC's.

A diversity of studies is necessary in order to increase the available data in a particular field of science. Large-scale studies usually report on the presence of compounds across national or continental scale. Regional and local scale monitoring is also important to understand the spatial and temporal variations in the occurrence of EOC's. Loos et al. (2013) reported on a pan-European study of 164 groundwater samples from 23 countries completed in 2010, with particular attention to persistent organic pollutants. Since then, a number of countries have developed national monitoring of EOC's (Manamsa et al., 2016a, Lopez et al., 2015, Bono-Blay et al., 2012).

3.2 REVIEW OF COMPOUND CATEGORISATION

In this section, we identify the most common categories used to classify emerging compounds in the reviewed studies. There is no current standard for the classification of compounds, making it difficult to identify which areas need further study. As previously mentioned, categorisation is commonly by use, and can be categorised into different groups depending on the scale of the study and the area of research the study comes from. Primarily, sub-categories exist if a study is only focused on one dominant use category. These can help to build a picture of the anthropogenic uses of the contaminants, and often their source; offering more description than the larger scale groupings. It is important to understand what categories have been most commonly used, so these can be adapted and used to develop a more uniform classification for EOC's.

Not only does the categorisation of compounds need to be ascertained, but the terminology and size of classification group. For example drugs of abuse are reported by Jurado et al. (2012) but may also be termed illicit drugs, as reported by Castiglioni et al. (2018). Eschauzier et al. (2013) report perfluorinated alkylated acids (PFAAs) as a category and Castiglioni et al. (2018) report perfluorinated compounds.

We expect there to be discrepancies in the classification of compounds throughout Europe. An example of a compound that has been categorised differently in studies is caffeine. It is commonly categorised as a lifestyle compound, but occasionally considered a pharmaceutical (Pinasseau, 2019), due to the high levels found in a number of pharmaceutical drugs. Similarly, the compound nicotine may be categorised as a pharmaceutical by Estévez et al. (2012), but otherwise as a lifestyle compound (Manamsa et al., 2016a, Postigo and Barceló, 2015) or anthropogenic marker (Castiglioni et al., 2018) which is a very broad category. In this review both have been ascribed the category 'Lifestyle'. Furthermore, although most studies state the categories detected, occasionally it is unclear which compound has been included in which category.

Other compounds may be very difficult to categorise due to their large range of uses e.g. Alpha-pinene, described by (Mali, 2017) is used as an insecticide, cosmetic, solvent, plasticizers. Similarly, they describe benzaldehyde as a chemical intermediates, solvent, and a bee repellent. In this study we have categorised both alpha-pinene and benzaldehyde as personal care products (PCP's). A study by Cabeza et al. (2012) classifies the compound BHT, a food additive as a PCP, this may otherwise be categorised as a lifestyle compound as with other food additives such as BHA and Acesulfame. For this study we have categorised it as lifestyle to match the categorisation of other food additives. Similarly (Mali, 2017) describe Tris(2-chloroethyl) phosphate as a flame retardant, and then place it in the category of Plasticisers and additives. Whereas BDE 154, BDE 60, BDE 100 and BDE 47, all flame retardants, are classified by (Brueller et al., 2018) industrial compounds. For this review, flame retardants will be placed in the category industrial. EOCs can be classified regarding their use but also their chemical family. Mestranol is an example for which two classifications; both classification hormones and phenol, have been reported in the literature. However, the chemical classification is commonly used to assess transport and fate of EOCs in environment. Issues occur when both classification schemes are mixed. A third classification is based on the health effect, such as endocrine disruptors. In this review we have classified natural hormones such as estriol and estrone as Other EOC's, and synthetic hormones such as diethylstilbestrol and mestranol as Pharmaceuticals. Table 1 highlights some of these compounds and the categories in which they have been placed for this review.

Table 1: Compounds with different categorisation

Compound name	**Categories identified in studies**	**Category we used**
Caffeine	Lifestyle, Domestic and Personal, Anthropogenic marker, Pharmaceuticals active compounds (PhACs), Anthropogenic contaminant, Pharmaceuticals, Stimulant, pharmaceuticals and personal care products, Not categorised	Lifestyle
Alpha-pinene	Domestic and personal	PCP
Benzaldehyde	Domestic and personal	PCP
BHT	Food additive	Lifestyle
Tris(2-chloroethyl) phosphate	Plasticisers and additives	Industrial
Mestranol	EDCs	Pharmaceuticals
Nicotine	Lifestyle, Anthropogenic marker	Lifestyle

From a total of 39 studies considered, 36 categorise the compounds that are detected and 3 do not. The three studies that do not categorise the compounds detected often explain the compounds and their uses, but do not explicitly assign a grouping to them in the published paper or accompanying literature. This may be due to ambiguity in their categorisation, or that the authors did not feel they needed to be categorised. The five most reported categories are reported in Table 2 with the frequency they were reported in the 39 target studies.

Table 2: The top five categories used

Category	Number of studies
Pharmaceuticals	16
Pesticides	15
Industrial	9
Personal care products (PCPs)	8
Solvents	4

3.2.1 Pharmaceuticals

The mega-group 'Pharmaceuticals' is the most frequently reported category with 16 studies which explicitly look for Pharmaceuticals, alongside an additional 5 studies look for pharmaceuticals grouped with other compounds. These include 'pharmaceutically active compounds', 'pharmaceuticals and hormones', 'pharmaceuticals and personal care products' and 'pharmaceuticals and X-ray contrast agents'. Sub-categories within the Pharmaceuticals are commonly reported, reflecting the diversity of compounds included. Pinasseau (2019) report on antibiotics, antiepileptics, antihypertensive and nonsteroidal anti-inflammatory drugs as well as their metabolites. Pharmaceuticals may incorporate substances such as the sulphonamides reported in (Spielmeyer et al., 2017) which may be also known as veterinary drugs. As most of these compounds are also used in human treatment, in this review, these have been placed under the broad categorisation 'Pharmaceuticals'.

3.2.2 Pesticides

15 studies identify pesticides or pesticides and transformation products as a category for the categorisation of organic compounds. A further three studied use herbicides and one more uses fungicides as the primary categories. Therefore, a total of 18 studies categorise as pesticides, herbicides or fungicides, the latter two are sub-groups of pesticides. We have not included this as a group for emerging contaminants. Most pesticides are covered by the groundwater directive (REF). Others are managed at a regional or national scale and cannot therefore be compared between European studies. It is worth noting that this is a popular category that it still widely screened for. 53 different pesticides and their Transformation Products (TP's) were detected.. According to Annex 1 (Groundwater Quality Standards) pesticides, including their relevant metabolites, degradation and reaction products must not exceed 0.1 ug/l or 0.5ug/l total; where total refers to the sum of all pesticides detected in the monitoring procedure.

The regulation of specific pesticide compounds is devolved to each European country, and therefore their responsibility to identify pesticides and transformation products which are relevant locally and to decide which ones to monitor. A study by Kiefer et al. (2019) screened for more than 300 pesticides and 1100 pesticide transformation products, in 31 groundwater samples from Switzerland. The reported pesticide transformation product concentrations were greater than parent pesticides, and highlighted the need for greater screening for transformation products in European groundwater bodies.

3.2.3 Personal Care Products (PCPs)

As well as nine studies that use the category Personal Care Products, or PCPs, additional studies look for 'pharmaceuticals and personal care products'. The compounds included in this category typically included UV protectors such as sunscreen, insect repellent and some cosmetics. A number of compounds in this category cross over with those compounds included in the 'Lifestyle' category. For this reason some studies such as Lapworth et al. (2016) use the larger mega-group Personal Care Products and Lifestyle (PCPL).

3.2.4 Other categorisation

Where compounds are not categorised in the literature, the study tends to look for individual target compounds. E.g. (Hillebrand et al., 2012) target 4 compounds; Caffeine, Paraxanthine (metabolite), Theobromine (metabolite) and Carbamazepine. This may reflect the nature of the study, the analytical methods that are available to the researchers, or follow an existing scoping study that highlighted compounds of concern at the site of interest.

Apart from usage, other categorisation includes the potential hazards of the compounds. Three studies look at Endocrine disrupting compounds (Pignotti et al., 2017, Carvalho et al., 2015, Corada-Fernández et al., 2017), and Endocrine Disrupting Chemicals in (Brueller et al., 2018), although none are found in groundwater in (Pignotti et al., 2017). This category includes sub-groups such as PFAA's, synthetic hormones (e.g. estrone, estradiol, 17α-ethinylestradiol) and Phenols (e.g. Bisphenol A, Octylphenol, Mestranol and Nonylphenol).

Another reported category was anthropogenic markers and anthropogenic contaminants (Castiglioni et al., 2018). These are primarily compounds such as Caffeine and Nicotine, otherwise known as lifestyle compounds that are found in high concentrations in and around densely populated or urban areas.

3.3 SUMMARY STATISTICS OF REVIEW STUDIES

In this section we assess EOC results using a range of metadata, including the country of origin and date of publication, alongside other data including the number of EOC's screened for and detected and analytical methods used.

Thirty-nine studies have been reviewed and the summary statistics complied understand how EOC's have been studied in Europe, and the extent to which this has occurred. This review identifies all compounds recorded in the reviewed studies where EOC's are detected in groundwater. Regulated compounds, as listed in Annex 2 of the WFD (2000/60/EC) were removed to create a list of all EOC's detected within the review studies. For the purpose of this study, where possible, we have included compounds below the Limit of Quantification (LOQ), but above the Limit of Detection (LOD), as well as tentative detections. CAS numbers were assigned by cross-referencing the compounds with established lists e.g. NORMAN list of emerging contaminants. The categorisation used in the studies and its usage were used to establish a categorisation for each of the compounds detected. This is not a definitive list, but enables a greater understanding of what groups of compounds have been detected in the European studies. As discussed earlier, significant discrepancies exist where use and categorisation are not synchronised throughout Europe.

Section 3.3.1 looks at the overall statistics compiled from the selected studies, the following section 3.3.2 looks more specifically at a few categories in more detail.

3.3.1 Categories used

In total 7 categories were used (Table 3), where the categories used are primarily based upon the frequency of usage within the reviewed studies. Table 3 also shows the number of compounds categorised into each of these, and the total number of studies in which these compounds ascribed these categories were detected. Where the group contained less than 10 compounds, these were added to the Other EOC's category to prevent the over-representation of small categories, and the use of too many categories for comparison.

Assumptions must be made before categorisation of compounds can take place. In this review, we have adapted some of the categorisation so that similar uses were categorised together. Some of the uses described by each category are listed below.

Table 3: The chosen categories, sub-categories that are included and the number of compounds in each category that are reported in one or more of the reviewed studies

Category	**Sub-categories included**	**Compounds in category**
Pharmaceuticals	Synthetic Hormones, Psychiatric drugs, Antihypertensive, Cardiovascular, anti-epileptic drugs, antibiotics, antidepressants, lipid regulator, synthetic hormones, contrast agent, tranquilizers, anti-inflammatory	123
Industrial	Flame retardants, PFAS, Chemical intermediates, Dye intermediates	54
PCP's	UV filters, insect repellents, fragrance	22
Lifestyle	illicit drugs and stimulants, food additives, fragrances, sweeteners and caffeine	23
Solvents and THMs	chlorinated and non-chlorinated solvents, petroleum products, halogenated and non-halogenated solvents, CFCs, THMs	28
Plasticisers	Plasticisers, Plasticiser metabolites	12
Other EOC's	Sterols, natural compounds (including natural hormones)	13

Problems are encountered when identifying the compounds detected from within the reviewed studies, meaning not all compounds in the 39 studies are included in further analysis. Cerar and Mali (2016) identify 161 organic compounds in the first round of sampling and 166 in the second round. However, these compounds are not provided in the paper or supplementary material. Instead, 12 of the most commonly detected are reported in detail, which have been included in the study. Mali et al. (2017) similarly report 103 detections in the first sampling campaign and 144 in the second. 53 compounds are selected for further analysis, however there is no information to whether these are the compounds detected in both, or highest frequency or concentration of detection. Furthermore Ahkola et al. (2017) highlight the problem of Limit of Quantification (LOQ) vs Limit of Detection (LOD). We have used their notation <LOQ differently to n.d. (no detects), and assume in this case that compounds <LOQ are detected and those with n.d. are below

the LOD. These studies highlight the problem of differences in reporting between European countries, making an analysis of data across Europe difficult.

Table 4: List of categories used, including the number of compounds in each category and the number of studies that mention/related at least one compound in this category

Category	No of reported compounds	Number of studies that mention at least 1 compound in this category
Pharmaceuticals	123	31
Industrial	54	19
Lifestyle	23	19
Other EOC's	13	12
Solvents and THMs	28	9
Plasticisers	12	9

3.3.2 Distribution of studies

The distribution of studies (39) published since 2012 throughout Europe helps to understand the scale of the study area, and how this is developing spatially Table 5.

Table 5: Reviewed studies, including the number of groundwater sites, samples and the categories of compounds detected

Ref	*Year*	*Country*	*Scale of study*	*Number of groundwater sites*	*Number of (groundwater) samples*	*Our use categories of compounds detected*
Brueller et al.	2018	Austria	National	22	22	Plasticisers, Industrial
van Driezum et al.	2019	Austria	Targeted	7	22	Pharmaceuticals, Industrial
Hrkal et al.	2018	Czech Republic	Targeted	6	6	Pharmaceuticals, Lifestyle, Other EOC’s
Lapworth et al.	2015	England/ France	Regional	345	345	PCPs, Pharmaceuticals, Solvents and THMs, Plasticisers, Industrial, Lifestyle
Ahkola et al.	2017	Finland	Regional	6	Unknown	Pharmaceuticals
Lopez et al.	2015	France	National	494	988	PCPs, Pharmaceuticals, Solvents and THMs, Plasticisers, Industrial, Lifestyle, Other EOC’s
Pinasseau et al.	2019	France	Regional	5	10	Pharmaceuticals, PCP’s, Lifestyle
Hass et al.	2012	Germany	Targeted	9	36	Pharmaceuticals
Hillebrand et al.	2012	Germany	Targeted	1	157 (Spring)	Pharmaceuticals, Lifestyle
Müller et al.	2012	Germany	Regional	21	46	Pharmaceuticals
Hass et al.	2012	Germany	Regional	123	369	Pharmaceuticals
Reh et al.	2013	Germany	Regional	44	163	Pharmaceuticals, Industrial, Lifestyle

Spielmeyer et al.	2017	Germany	Targeted	4	88	Pharmaceuticals
Estevez et al.	2016	Gran Canaria	Targeted	7	37	Industrial, Solvents and THMs, Pharmaceuticals, Other EOC’s
Nagy-Kovács et al.	2018	Hungary	Targeted	2	30	Industrial, Pharmaceuticals, Lifestyle
Pignotti et al.	2017	Italy	Regional	Unknown	17	None detected
Castiglioni et al.	2018	Italy	Regional	53	53	Pharmaceuticals, PCP’s Lifestyle, Industrial
Banzhaf et al.	2012	Luxembourg	Targeted	5	47	Pharmaceuticals, Lifestyle
Kapelewska et al.	2016	Poland	Targeted	2	16	PCP’s, Lifestyle, Other EOC’s
Kapelewska et al.	2018	Poland	Targeted	8	23	Pharmaceuticals, PCP’s, Lifestyle, Other EOC’s
Carvalho et al.	2015	Portugal	Regional	13	13	Pharmaceuticals, Industrial, Other EOC’s
Paíga, and Delerue-Matos	2016	Portugal	Targeted	5	10	Pharmaceuticals
Koroša et al.	2016	Slovenia	Regional	14	56	Pharmaceuticals, Industrial, Lifestyle
Mali et al.	2017	Slovenia	Regional	15	28	Pharmaceuticals, Solvents and THMs, Lifestyle Plasticisers, Industrial, Other EOCs
Bono-Blay et al.	2012	Spain	National	131 (or 91)	131 - 40 springs and 91 boreholes	Industrial, Plasticisers
Jurado et al.	2012	Spain	Regional	36	36	Lifestyle, Pharmaceuticals
Estévez et al.	2012	Spain	Regional	4	14	Pharmaceuticals, Lifestyle, Industrial, Solvents and THMs, Other EOC’s
López-Serna et al.	2013	Spain	Regional	31	31	Pharmaceuticals
Jurado et al.	2014	Spain	Regional	31	31	PCP’s
Jurado et al.	2014	Spain	Regional	26	26	Pharmaceuticals
Luque-Espinar et al.	2015	Spain	Regional	12	85	Pharmaceuticals, Lifestyle
Corada-Fernández et al.	2017	Spain	Regional	29	57	PCP’s, Pharmaceuticals, Lifestyle, Other EOC’s
Filipovic et al.	2015	Sweden	Targeted	16	16	Industrial
Eschauzier et al.	2013	The Netherlands	Regional	7	15	Industrial
Kivits et al.	2018	The Netherlands	Regional	10	46	Pharmaceuticals
Stuart et al.	2014	UK	Regional	19	54	PCPs, Pharmaceuticals, Solvents and THMs, Plasticisers, Industrial, Lifestyle
White et al.	2016	UK	Regional	3	37	Solvents and THMs, PCP’s, Plasticisers, Industrial, Other EOC’s
Manamsa et al.	2016	UK	Regional	6	78	Plasticisers, PCPs, Pharmaceuticals, Solvents

						and THM's, Industrial, Lifestyle
Manamsa et al.	2016	UK	National	2650	2650	PCPs, Pharmaceuticals, Solvents and THMs, Plasticisers, Industrial, Lifestyle, Other EOC's

Figure ***2***2 highlights the distribution of the studies included in this review on a European scale. The largest number of studies were located in Spain (8), followed by the Germany (6).

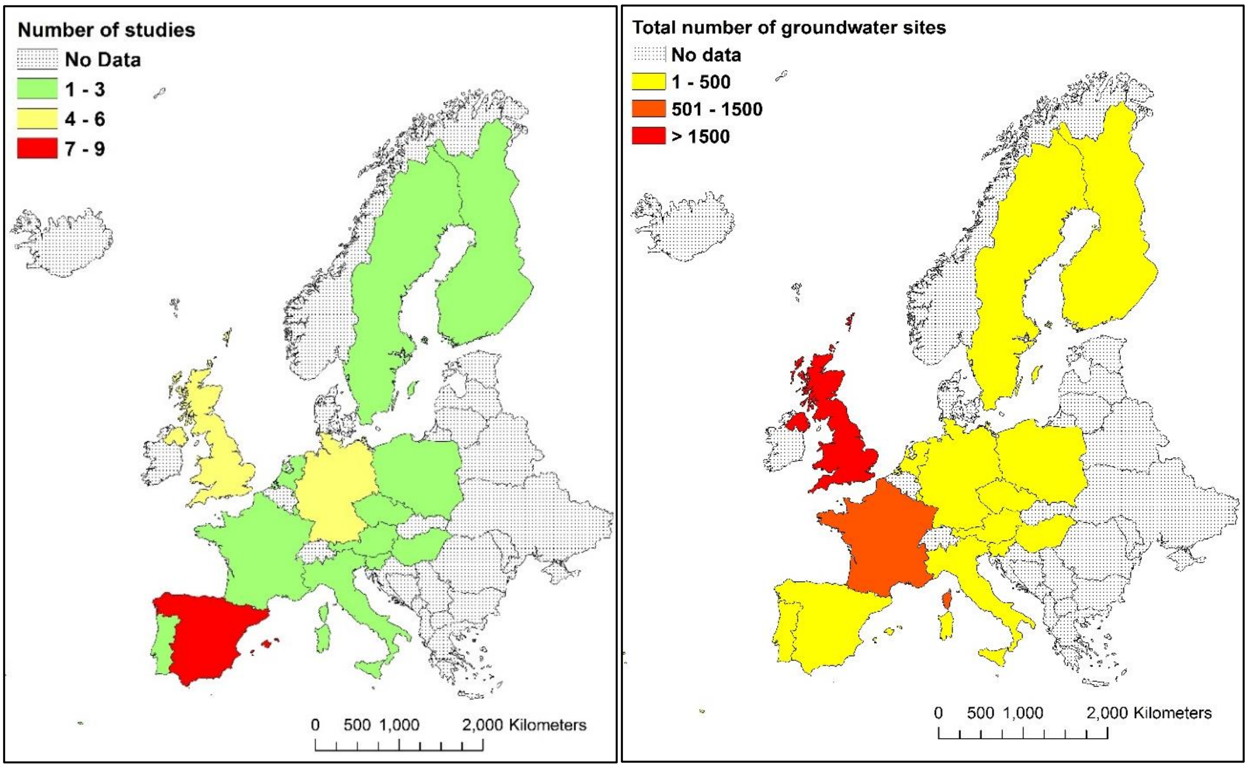


Figure 2: (a) The number of studies used in this review from each country. (b) The total number of groundwater sites from the selected review studies

Figure ***2*** also shows the total number of groundwater sites, using a summation of the number of sites used in each study within a given country. It must be noted that this does not represent the actual number of sites, as some are likely to be replicated in a number of studies. A lack of information about the sites means it is not possible to determine the actual number of discrete sites used. Some sites record at numerous well depths and take samples throughout the year. Groundwater sites is used here to reflect only the number of individual boreholes or wells sampled. In total 4222 groundwater sites were reported, with 5395 groundwater samples taken from those sites.

3.3.3 Sampling methods

It is important to consider the impact of different sampling methods when comparing pan-European studies, as different approaches make studies difficult to compare. Balderacchi et al.

(2014) suggest that a unified monitoring approach may need to be implemented after the unification of threshold values.

From reviewing the literature regarding the 39 selected studies, samples are primarily taken as grab samples from existing monitoring boreholes. However, other approaches such as passive sampling (PS) can be used to determine the presence of certain EOC's (Pinasseau, 2019, Ahkola et al., 2017, Mali et al., 2017, Cerar and Mali, 2016). These time-integrated methods are helpful for gathering reconnaissance data on the occurrence of EOCs in groundwater, particularly where these may be more temporally dynamic in terms of contaminant occurrence. Most of the studies used POCIS (polar organic compounds integrative samplers) tools or solid disk based PS (Ahkola 2017, Pinasseau, 2019), since they are dedicated to polar to mid-polar compounds. Other PS for a larger range of compounds have been developed (Mali et al 2017), however, there are difficulties in comparing data from PS and grab sampling approaches, for example, there are in-built assumptions required for translating PS data to equivalent concentration data and there may be site-specific considerations/calibration of PS required. Furthermore, low groundwater levels may limit contact time and can affect accumulation capabilities of the PS. In light of these factors, the main use of PS in groundwater is as a screening tool, rather than for quantitative assessments.

Regulatory monitoring typically follows a grab sampling protocol and it would be likely that this would be the case for EOCs in groundwater, at least for some time, particularly as in general residence times for groundwater are long, in the order of years to decades in most settings (Moreau et al 2019).

Peer review literature often reveals little information about the sampling regime undertaken. A number of studies complete sampling rounds at regular intervals throughout the year, some with high frequency (Hillebrand et al., 2012) and others just a single sample at multiple sites (Bono-Blay et al., 2012). Often a campaign during the summer and winter seasons are taken to reflect different groundwater states (for example (Jurado et al., 2014a, Lopez et al., 2015)), during which different groundwater levels may affect the type and concentration of compounds detected.

3.3.4 Analytical methods

In this section, we identify the analytical methods employed to quantify EOCs occurrence in groundwater. We first investigate the sample preparation techniques, then current analytical methods are reviewed before identifying the methods used in the reviewed studies.

3.3.4.1 Preparation/Extraction

Before the analytical method is implemented, there generally needs to be sample preparation by means of extraction. In the reviewed studies, primarily Solid-Phase extraction (SPE) was used, but in some cases, other methods were employed. SPE offers the benefit of extracting compounds with a wide range of properties (Martin-Pozo et al., 2018). Other methods of extraction include pressurized liquid extraction (PLE) and Liquid-liquid extraction (LLE) (Estévez et al., 2012, Lopez et al., 2015, Manamsa et al., 2016b) but also some more novel approaches such as ultrasound-assisted emulsification microextraction (USAEME) (Kapelewska et al., 2016, Kapelewska et al., 2018). Where passive sampling techniques are used, the extraction method is necessary is different, and is based on SPE.

3.3.4.2 Review of analytical methods

The principal analytical method for emerging contaminants is Liquid chromatography (LC) and gas chromatography (GC) coupled to mass spectrometry (Martin-Pozo et al., 2018).

Some substances require more work to analyse than others, for example, certain PFAS compounds have been particularly difficult to analyse, often owing to their range of chain lengths and characteristics. Recent developments in analytical methods have may screening a large number of compounds more cost effective (Richardson and Ternes, 2017).

Petrie et al. (2015) highlight the problem with targeted screening and low resolution mass spectrometry, meaning that some metabolites are often missed, whose impacts are often on the same level as the parent compound. A number of methods have been since developed, often for a specific group of compounds. Due to the large numbers of compounds detected, multiple methods are often employed within the same study.

High resolution mass spectrometry analysis allows conventional quantitative analysis (Brueller et al., 2018), but above the development of large compounds qualitative screening, (Pinasseau et al, 2019) without initial targeting of compounds to monitor. By this way new compounds of interest, such as EOCs transformation products can be identified in GW.

3.3.4.3 Methods used

A number of methods were used in the 39 selected review studies. 21 methods are cited in the studies, and listed in Table 6. The most popular methods are LCMS and GCMS methods, which both suit a wide range of compounds. The analytical method used depends on the type of EOC that has been screened for. Samples may screened for a few specific EOC's of interest e.g. Müller et al. (2012) and Hass et al. (2012b) or a full suite of over 1000 different compounds and metabolites e.g. Manamsa et al. (2016b), White et al. (2016).

Table 6: Analytical methods used by the reviewed studies.

Methods	**Reference**
Gas chromatography with mass spectrometry or tandem mass spectrometry (GC-MS) or (GC-MS/MS)	(Bono-Blay et al., 2012, Estévez, 2012, Jurado et al., 2014a, Stuart et al., 2014, Lapworth et al., 2015, Lopez et al., 2015, Cerar and Mali, 2016, Estévez, 2016, Kapelewska et al., 2016, Koroša et al., 2016, Manamsa et al., 2016a, Manamsa et al., 2016b, White et al., 2016, Corada-Fernández et al., 2017, Mali et al., 2017, Brueller et al., 2018, Hrkal et al., 2018, Kapelewska et al., 2018)
Liquid chromatography with mass spectrometry or tandem mass spectrometry (LC-MS) or (LC-MS/MS)	(Hass et al., 2012a, Jurado et al., 2012, Müller et al., 2012, Reh et al., 2012, Eschauzier et al., 2013, López-Serna et al., 2013, Jurado et al., 2014b, Carvalho et al., 2015, Castiglioni et al., 2015, Lapworth et al., 2015, Lopez et al., 2015, Antonio Luque-Espinar et al., 2015, Filipovic et al., 2015, Paíga and Delerue-Matos, 2016, Ahkola et al., 2017, Spielmeyer et al., 2017, Corada-Fernández et al., 2017, Pignotti et al., 2017, Castiglioni et al., 2018, Kivits et al., 2018, van Driezum et al., 2019)
Liquid chromatography High resolution mass spectrometry (LC-TOFMS)	(Estévez et al., 2012, Brueller et al., 2018, Pinasseau, 2019)
Gas chromatography–high resolution mass spectrometry GC/HRMS	(Lopez et al., 2015)
Continuous Flow Analysis	(Lopez et al., 2015)
semi-prep LC system with a diode-array detector (LC/DAD)	(Lopez et al., 2015)

Chemical Ionization Mass Spectrometry (CI-MS/MS)	(Lopez et al., 2015)
Ion chromatography	(Lopez et al., 2015, Spielmeyer et al., 2017)

3.3.5 Screening for EOC's

The number of compounds screened for is an important indicator of the type of studies reviewed. The number of compounds screened for does not necessarily represent the scale of the study, but often the budget and aims of the study. This reflects the nature of the study, whether it is targeted towards a few compounds, or a scoping study with a much larger number of compounds. In the reviewed studies, the average number of compounds screened for was 170, the largest being 1000 (White et al., 2016, Manamsa et al., 2016b, Stuart et al., 2014) and the smallest 4 (Hillebrand et al., 2012, Filipovic et al., 2015).

Figure ***3*** shows the cumulative distribution of the number of compounds screened for in the 39 reviewed studies. The largest category is the 10-100 range, representing intermediate studies where a category of compounds may be investigated or known existing EOC's are targeted. The log scale shows linearity in the data until around 100 screened compounds, highlighting a normal distribution of data.

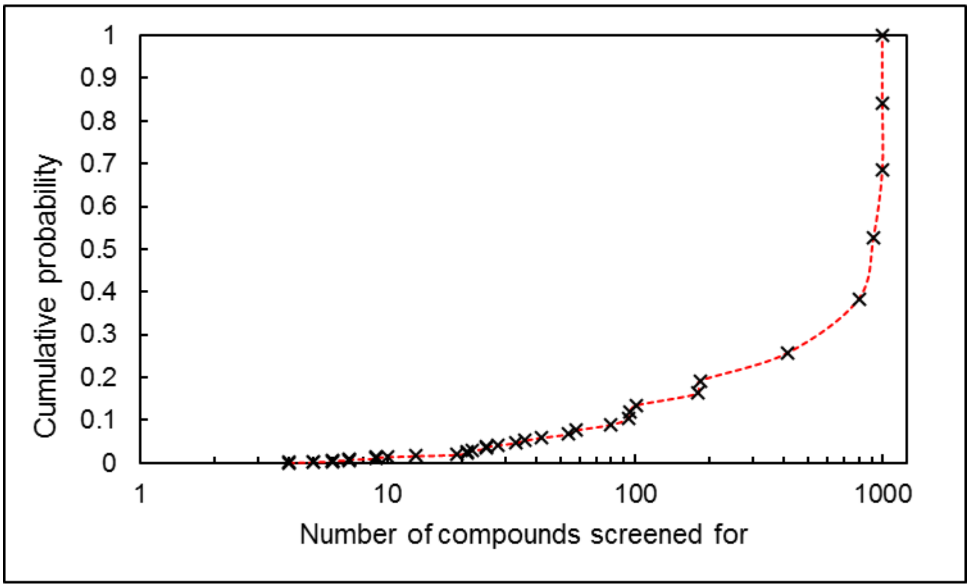


Figure 3: Cumulative probability plot of number of compounds screened for

We are interested in how the study of EOC's has changed through recent years. Figure 4 shows no strong relationship between year and the number of compounds screened for. Large-scale national studies that fit the review specifications were primarily completed in the years 2014 to 2017. More recently, the studies show smaller number of compounds are screened for, which may suggest a more targeted approach following earlier scoping studies, or the desire to characterise a few targeted compounds in more detail. These results suggest that there is an array of research taking place, both large scoping studies, and smaller, more targeted ones.

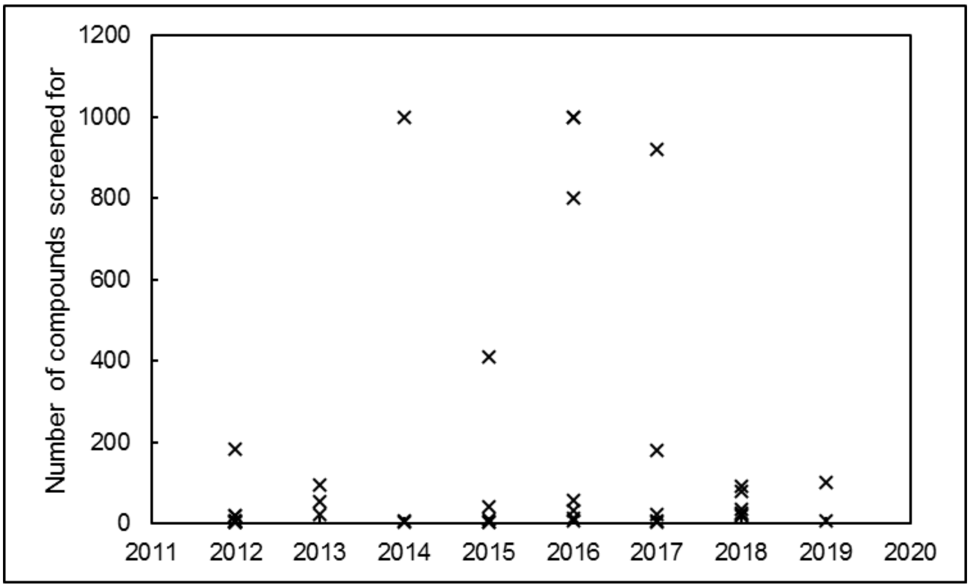


Figure 4: The number of compounds screened for within studies in each year, where each study is represented by one marker

Figure ***5*** shows there is a generally increasing trend between increasing the number of compounds screened for, the number of groundwater sites, and the number detected in groundwater. However, the number of compounds detected in large scale screening does not increase linearly. In Figure 5a the number of compounds detected continues to increase as the number of sites in the study increases, up until around 1000 groundwater sites. Figure 5b shows a similar effect, whereby the number of compounds screened for continues to increase until around 1000 compounds are screened for. This is likely due to the targeted nature of the smaller scale studies, where a scoping procedure or known problem means that there is a higher hit rate of EOC's in groundwater. This highlights the need for a prioritisation approach, showing that simply increasing the number of sites and compounds screened for will not always increase the number of detects. Pearsons correlation is used to describe a correlation between two variables, where r = 1 is a strong positive correlation and r = -1 is a strong negative correlation, where $-1 \leq r \geq 1$. No significant correlation is observed between the number of compounds screened for and detected in groundwater; the data giving a Pearsons correlation of r = 0.032, meaning no significant correlation is observed at p=0.1 significant level. Annex B shows the distribution of compounds screened for and the total number of groundwater sites. Generally, a larger number of studies have screened for a large number of samples. An apparent exponential relationship between the number of compounds detected and the number screened for until after 100 compounds screened for. However, there are still very few large scale studies with only one study with 500+ groundwater sites (Manamsa et al., 2016a).

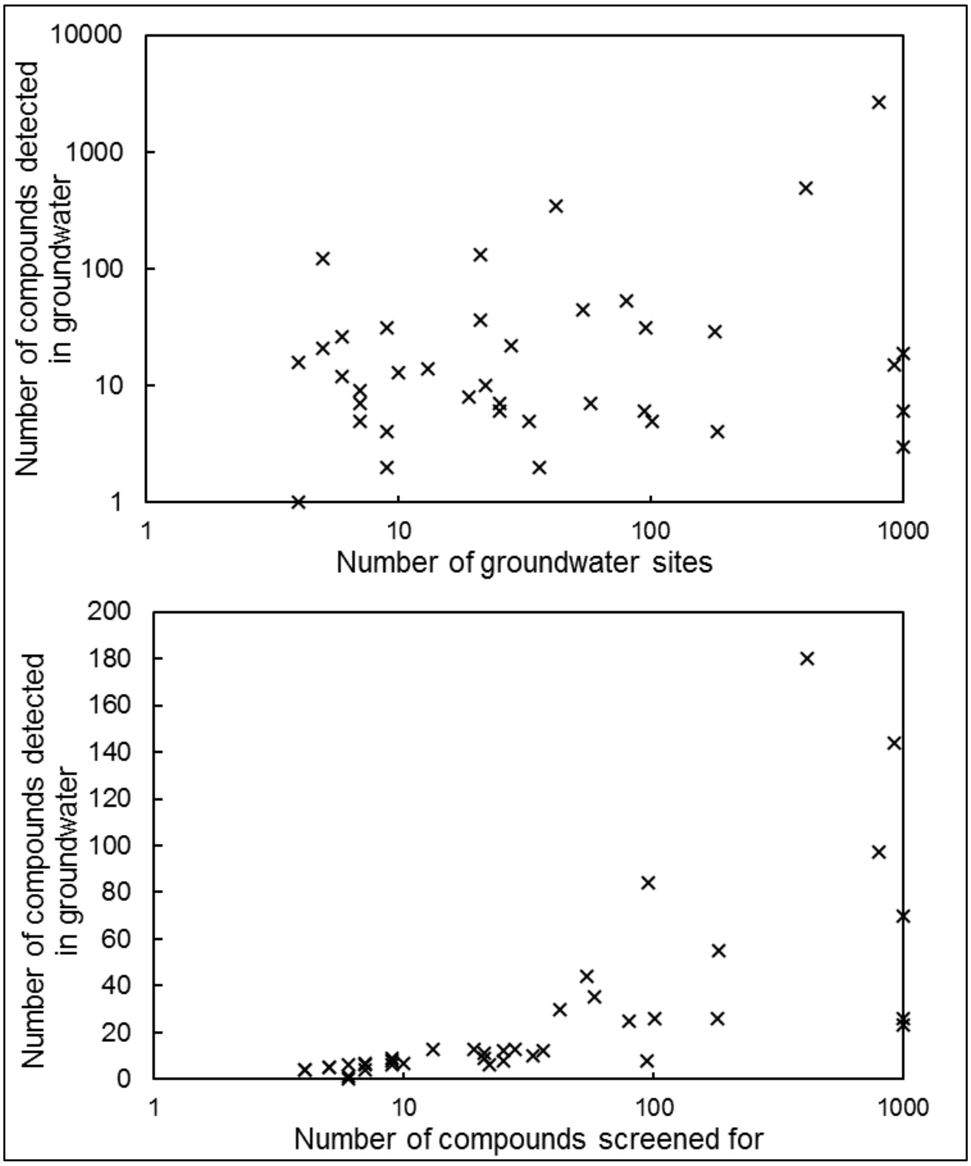


Figure 5: a) The number of groundwater sites sampled vs the total number of compounds detected in groundwater, and b) The number of compounds screened for vs the number of compounds detected in groundwater.

It appears that the number of sites does not influence the scale of the study (Appendix B). This includes both the number of compounds screened for and the number of compounds detected in groundwater. Furthermore, there is no strong correlation between the number of compounds screened for and the number of groundwater sites used in the study (r = 0.050) or the number of compounds detected in groundwater and the number of groundwater sites used (r = 0.002). This reflects the diversity in size and scale of the studies.

Furthermore, there is no statistical correlation between the number of groundwater samples and the number of groundwater sites in the 39 studies considered, where r = 0.001, and is still not significant at the p = 0.1 significance level (Appendix C). This is likely due to the range in scale of the studies. We might expect more targeted studies to have a smaller number of sites and therefore smaller number of samples. However, targeted studies are often part of longer-term monitoring programmes (e.g. Hillebrand et al., 2015), whereas national scale GW EOC studies often only take sample from each site once or twice (e.g. Lopez et al., 2015).

Most of the reviewed studies do not report on their LOD and LOQ values, however large discrepancies are likely to exist. Limited cross-country reporting means that different countries and laboratories are likely to report on different values. Improved analytical techniques allow for greater lab performance, and increased detections of compounds existing in small concentrations. However, constraints such as funding and lack of facilities, mean that the same techniques have not been universally incorporated by all European countries.

3.3.6 EOC's detected

7 shows the top 10 compounds detected in all groundwater samples in the selected 39 studies). Six of the top 10 were classified as Pharmaceuticals, 1 as lifestyle, 1 as a plasticiser and 1 as a pesticide and 1 as Industrial (

7).

The compounds detected are compared to those recently added to the GWWL (CIRCABC, 2019), however, here we do not consider the toxicity or mobility in the environment as it is done for the GWWL assessment, solely the occurrence of their detection in reviewed studies. The top ten compounds presented in

are in broad agreement with the results of the GWWL. Both Carbamazepine and Sulfamethoxazole were reported in enough studies that they were removed from the initial GWWL, with enough evidence of potential groundwater contamination for a standard to be designed. Caffeine is widely reported, but due to its low toxicity, is not ranked highly on the watch list. Diclofenac is highly ranked in the GWWL methodology, ranking 21st in the list of pharmaceuticals considered for the watch list. Ibuprofen was also highly ranked, and is one of the nine pharmaceuticals suggested to be added to the GWWL.

Table 7: The top 10 compounds detected, their occurrence in number of studies in which they are detected, their use and proposed categorisation.

CAS	Compound	Number of studies reporting one or more detection	Use	Category
298464	Carbamazepine	22	Anti-epileptic drug and other	Pharmaceuticals

			pharmaceutical applications	
58082	Caffeine	15	Lifestyle	Lifestyle
723466	Sulfamethoxazole	13	Antibiotics	Pharmaceuticals
80057	Bisphenol A	13	Resins for food packaging	Plasticisers
15687271	Ibuprofen	12	Anti-inflammatory agent with analgesic properties	Pharmaceuticals
103902	Acetaminophen	9	Non-Prescription Drugs	Pharmaceuticals
134623	N,N-diethyl-m-toluamide	8	Insect repellant	PCP's
15307865	Diclofenac	8	Anti-inflammatory agent	Pharmaceuticals
108907	Chlorobenzene	8	Chlorinated solvent	Solvents and THMs
41859670	Bezafibrate	7	Lipid regulator	Pharmaceuticals

number of studies that report a detection of one or more of the compounds in this category.

Figure 6 shows the number of discrete compounds that have been categorised into each of the 7 categories. The figure shows the number of individual compounds detected which have been assigned each category, and the number of studies that report a detection of one or more of the compounds in this category.

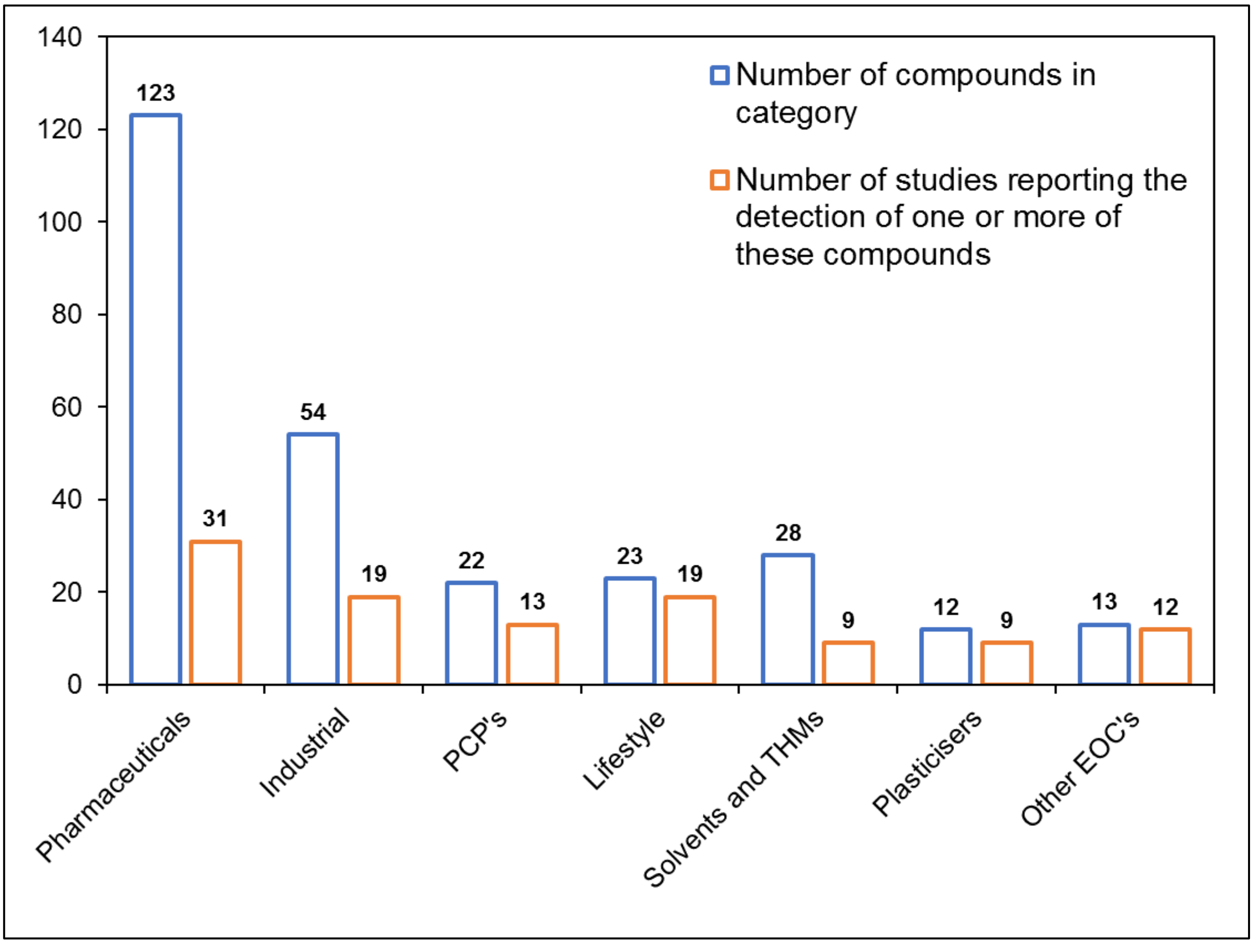


Figure 6: The number of compounds detected in each of the 8 selected use categories and the number of studies that report a detection of one or more of the compounds in this category

3.3.6.1 Pharmaceuticals

Pharmaceuticals is the most widely observed category in this study, with 123 individual compounds being detected in one or more study. Thirty-one of the 39 studies reported the detection of one or more compound classified as a Pharmaceutical. The frequency of detection of pharmaceuticals is likely to be much greater, as each study is recorded here as one detection. In reality, this number does not reflect the number of individual positive sample detects encountered within each study which may be numerous on large national scale studies. The top 5 most commonly detected pharmaceuticals are the anti-elliptical drug Carbemazepine, the antibiotic Sulfamethoxazole, the anti-inflammatories Diclofenac and Ibuprofen and the Lipid regulator Bezafibrate. As well as 12 occurrences of Sulfamethoxazole detections, a further 2 detections are made in the form of Sulfamethoxazole metabolites 4-Nitro-SMX and Desamino-SMX (Reh et al., 2012).

Pharmaceuticals are commonly used as groundwater tracers, Müller et al. (2012) use a selected 5 pharmaceuticals to indicate the presence of sewage in groundwater at 21 sites in Germany. Banzhaf et al. (2012) use 7 EOC’s to trace the interaction between surface and groundwater in riverbank deposits. They are of particular concern due to their potential effects on wildlife and humans. The detection of pharmaceuticals after water treatment is not regularly reported, but a number of reviews show that the process may be insufficient for the adequate removal of a number of EOC’s (Yang et al., 2015). Pharmaceuticals have been widely screened for and detected in studies throughout Europe, with the data being used to assess methods of removal from drinking and aquatic water (Rodriguez-Narvaez et al., 2017; Yang et al., 2017; Wang and Chu, 2016).

3.3.6.2 Personal Care Products (PCP's)

A total of 22 PCP compounds were detected at least once in 13 of the 39 studies, where there is likely to be more than one different compound in this group within the same study. The top 5 most commonly detected PCPs were the compounds Benzophenone, N,N-diethyl-m-toluamide (DEET), Triclosan, Benzophenone-3 and Propylparaben.

3.3.6.3 Endocrine Disrupting compounds (EDC's)

EDC's are studied by Carvalho et al. (2015) who study 10 different EDC's in 13 groundwater samples from within a water supply system. 7 compounds were detected in from the 13 groundwater sites sampled. All compounds were detected at concentrations of less than 0.1 ug/L, the proposed values for some unregulated compounds such as pesticides and Polycyclic aromatic hydrocarbons (Water Framework Directive, 2013). Pignotti et al. (2017) screened for six EDC's, but found that no compounds were detected in concentrations above the MQL in groundwater (ranging from 0.21-2.02 ng/l). They conclude that dilution by rainfall makes the compounds undetectable, natural attenuation processes and distance from vulnerable recharge zones are also discussed. Brueller et al. (2018) screened for 28 compounds known or suspected of having endocrine disrupting properties. Phthalates were detected in 11 groundwater samples. 8 samples contained Perfluoroalkyl substances, 4-nonylphenol monoethoxylate was found in 2 groundwater samples and Bisphenol A in 1 further sample. However, 576 (93.5%) out of 616 measurements in groundwater detected no compounds above the Limit of Quanitification (LOQ). Corada-Fernández et al. (2017) screened for 8 EDC's but only detected one compound (Triclosan) at a concentration of 83+- 20 ng/l.

The category EDC's is solely reported in papers where this is the only category used, as it cannot be compared to the use categories. For this reason, although a popular classification, may not be suitable for a large-scale review, and therefore not used as a category within this study. Personal care products often contain endocrine disrupting compounds that are shown to have negative impacts on human health and the environment in which they are detected (Kabir et al., 2015).

3.4 CASE STUDIES

The two most detected EOC's in the review were chosen as case studies have been selected to extend our understanding of how EOC's have been studied in Europe.

In this section we look at the case studies, Carbamazepine and Caffeine, the two most detected compounds within the review studies. We look at the distribution of studies and detections, as well as maximum recorded concentrations. The large number of detection of these compounds means they are commonly screened for, and therefore known to be present and likely to have an associated risk or hazards. However, it is the less detected compounds which potentially need further work.

The concentrations given are the maximum concentrations in any one sample in a given location. The concentration is therefore not necessarily representative of the whole country, but indicates the maximum concentration that has been recorded in that country.

3.4.1 Carbamazepine

Carbamazepine is a widely applied anticonvulsant used to treat epilepsy, bipolar disorder, and trigeminal neuralgia (Banzhaf et al., 2012). Carbamazepine has been shown to threaten aquatic organisms (Oetken et al., 2005).

Carbamazepine was detected in 22 of the 39 studies. The maximum reported concentration was 2325 ng/l (Müller et al., 2012), recorded in the vicinity of a waste water treatment plant (WWTP) where the groundwater is thought to be influenced by recent sewage water. In this study of pharmaceuticals as indictors of sewage-influenced groundwater, Carbamazepine was reported in

20 of the 46 groundwater samples (43.5%). Hillebrand et al. (2012) reported Carbamazepine was detected in 57.3% of the 157 spring water samples taken, but was not quantified in any sample. The average recovery in the 21 groundwater studies that reported their recovery was 60.1%.

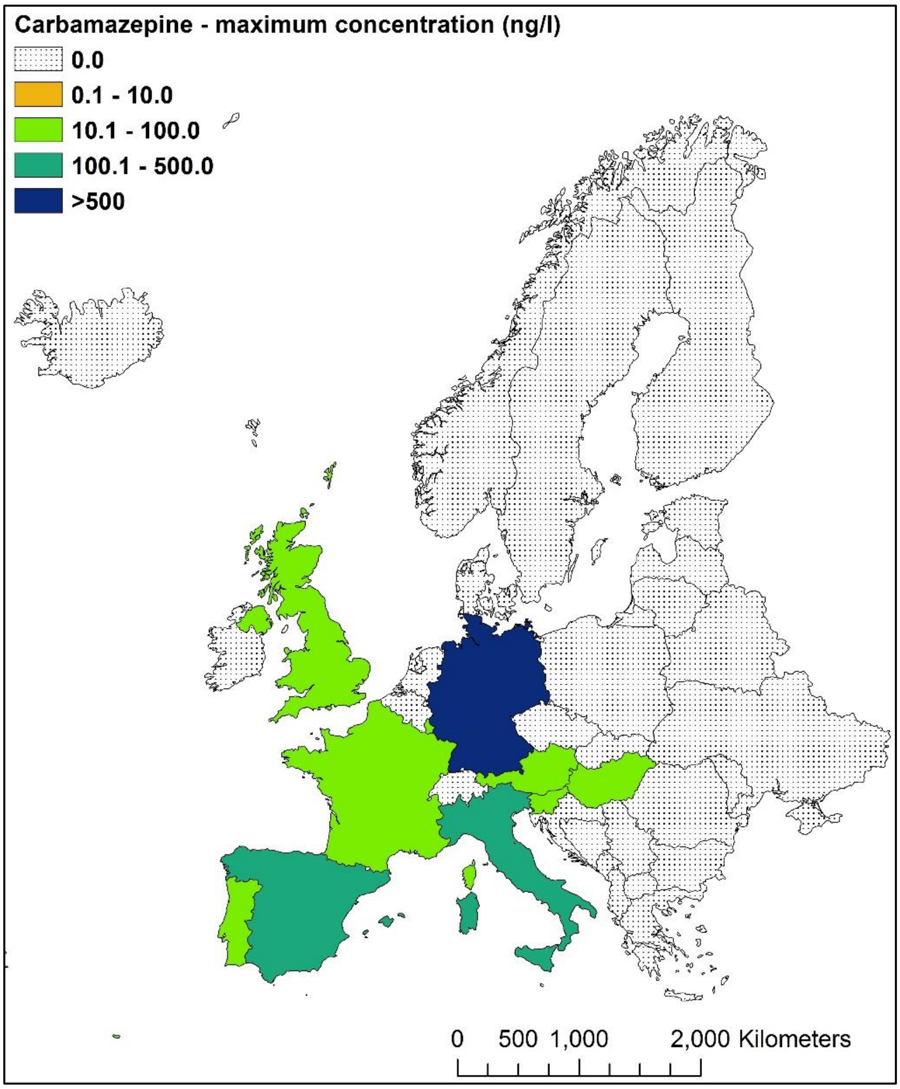


Figure 7: Max concentration of Carbamazepine in ng/L for each European country reporting detections of Carbamazepine

3.4.2 Caffeine

Caffeine can fall into a number of EOC categories, but in this study has been classified as a lifestyle compound. Caffeine was detected in 15 studies , where the maximum concentration was reported in a groundwater sample from southern Spain (Luque-Espinar et al., 2015) were a concentration of 14.77 µg/L was detected in the vicinity of a wastewater treatment plant. The reported percentage of positive detections ranged between 3.1 (Manamsa et al., 2016a) and 100 % (Pinasseau, 2019) of groundwater samples.

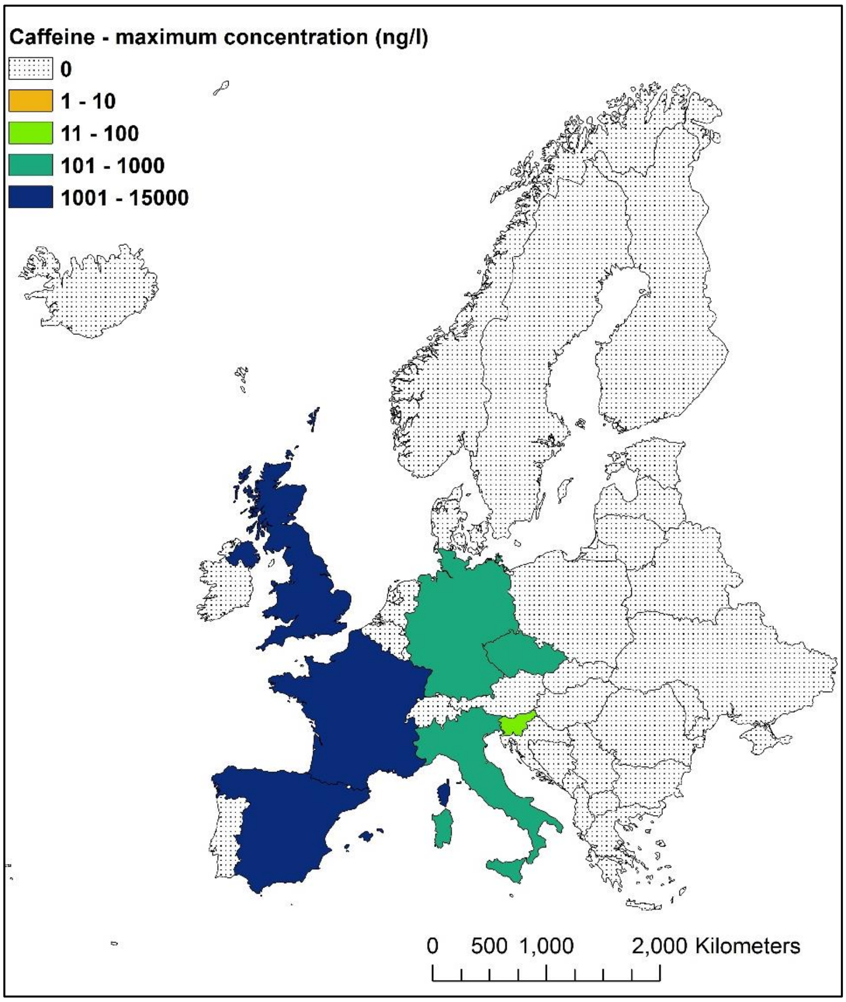


Figure 8: Maximum concentration of caffeine in groundwater only detections in Europe

3.5 PURPOSE OF STUDY

Current understanding of EOC's in groundwater varies considerably between European countries. The scope and scale of the study depends on funding, interest, capability, and perceived threat.

The purpose of each study is usually well defined and specific to the investigation work to be undertaken. A large majority of reviewed studies principally aim to investigate the occurrence; transport and fate of a group or key EOC's that have been identified in a defined catchment, area or geological unit (e.g. regional aquifer system).

This may be in relation to the threat upon a particular resource e.g. (Hass et al., 2012b, Ahkola et al., 2017) in relation to drinking water.

Occasionally EOC's have been used as a tracer to develop a greater understanding of the hydrogeology of the region being studied (Stuart et al., 2014, White et al., 2016, Pinasseau, 2019). Banzhaf et al. (2012) specifically use EOC's as tracers of surface-groundwater interactions.

3.6 SCALE OF STUDY

Various scales of study are required to understand aspects of EOC's occurrence. Large-scale studies offer an insight into spatial occurrence and trends in EOCs, and allow researchers to understand how widespread or diffuse a particular EOC is in the groundwater system, this aspect will be an important consideration for regulating EOCs in the future. Smaller scale studies are primarily used to understand temporal variability and specific hot-spots where EOC contamination may be more likely to occur. Although the majority of studies are still focused on point sources, in areas where EOC's have previously been detected or known to have been released. There are an increasing number of regional and national studies (Lopez et al., 2015, Manamsa et al., 2016a, Brueller et al., 2018).

In this review, each study was classified to a scale to gain a greater understanding of the studies previously undertaken. Although a procedure was used, some studies may be classified differently. Where a large scale campaign was undertaken across the country as a whole, the study was classified as 'National'. Where a range of sites around a given city/aquifer/region were studied, the study was classified as 'Regional'. If the study focused on a specific stretch of river, WWTW or study site the study was classified as 'Targeted'.

Out of 39 studies used in this analysis, 4 were national scale, 23 were regional and 12 were targeted studies, highlighting a consistent focus on regional and targeted studies. There are no obvious trends in the scale of studies published in each year (Figure 9), however the number of studies included may not be large enough for any trends to be apparent.

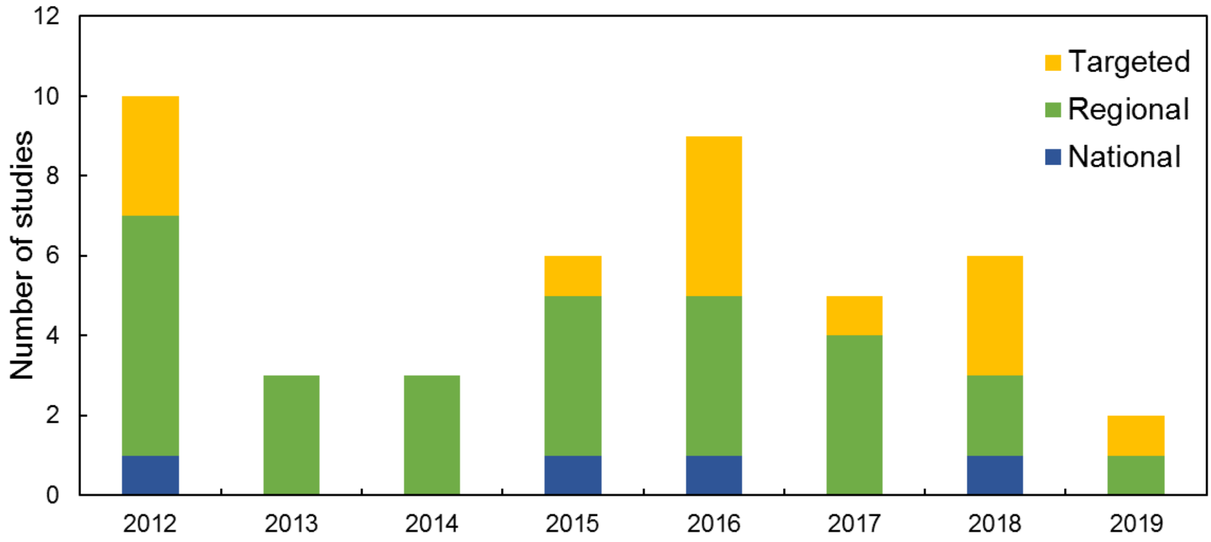


Figure 9: The scale of studies reported in each year considered in this review

Both the number of groundwater sites and the number of groundwater samples are useful statistics to understand the scale of studies in Europe, and how these two variables compare. The total number of groundwater sites, where the statistic was published, totalled 4350. The total number of recorded groundwater only samples was 5696. Medium scale studies were most popular, with 19 studies recording in the order of 10-100 groundwater samples (Figure 10).

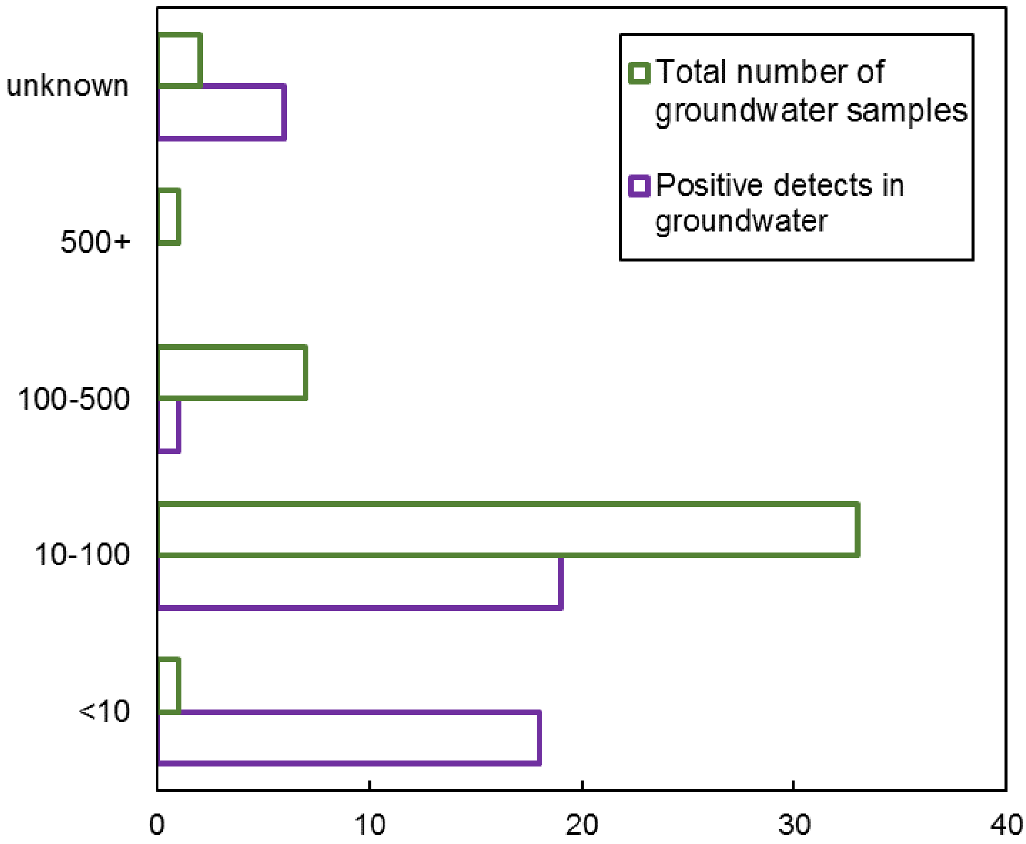


Figure 10: Number of groundwater samples in each research study reviewed

3.6.1 National scale

Four of the 39 selected studies are at a national scale. National scale studies principally develop the scientific understanding of EOC's in a country and act as a baseline for further studies. National scoping studies highlight areas for concern and further study, whether that is geographically or linked to the geology, land use or environmental setting. These types of study are important to gather evidence about EOC's in groundwater across Europe, and will help to define threshold values and a regulatory framework. But these types of study require a large organisational element, and organisation, are difficult to implement and expensive. Due to the large amount of data for analysis, the national scale studies presented here are primarily semi-quantitative (Stuart et al., 2014, Manamsa et al., 2016a, White et al., 2016).

3.6.2 Regional Scale

In this study, we have defined regional scale as studies that investigate the groundwater across a large geographic area. Examples include large cities or a specific geological area or aquifer system. 23 of the 39 selected studies have been classified as regional studies.

Regional scales primarily study a single aquifer across a larger region (Reh et al., 2012, Jurado et al., 2014b), but sometimes incorporate more than one aquifer (Jurado et al., 2012, Antonio Luque-Espinar et al., 2015, Koroša et al., 2016, Corada-Fernández et al., 2017, Pignotti et al., 2017, Castiglioni et al., 2018). Aquifers that provide a public water supply are primarily studied, as EOC's present may pose a threat to public health.

3.6.3 Targeted studies

Twelve of the 39 selected studies have been classified as targeted studies. The majority of targeted studies focus on a particular area, often where there is a known problem or presence of EOC's. Practically, this helps to direct the study and keeps costs down. Targeted studies are often employed where EOC's have been detected in the past, due to the presence of a current or previous structure or environmental setting. The study may then screen for a larger range of compounds to determine the scale of the contamination of groundwater in this area.

Wastewater treatment plants are commonly studied (Pitarch et al., 2016, Hass et al., 2012b), alongside current and disused landfill sites (Kapelewska et al., 2016) and industrial areas (Castiglioni et al., 2018).

Urban areas are often prevalent in studies on drugs (Cunha et al., 2017, Jurado et al., 2012) and pharmaceuticals (López-Serna et al., 2013, Hass et al., 2012a, Müller et al., 2012, Banzhaf et al., 2012, Rozman et al., 2015, Paíga and Delerue-Matos, 2016, Ahkola et al., 2017) in groundwater.

4 Conclusions and future outlook

The aims of this review was to; understand the current state of groundwater sampling of EOC's in Europe and the developments in recent years, understand the different methods for sampling and analysing EOC's in Europe, and highlight ongoing research and further areas for research necessary to develop a picture of EOC's in Europe. In this section we review these aims in with the analysis of the reviewed studies.

4.1 ONGOING RESEARCH

Analytical and extraction methods continue to improve. Zhong et al. (2019) describe the development of an automated system for the extraction and analysis of 87 emerging contaminants, including those previously considered difficult to extract such as PFAS. The process uses an online solid phase extraction liquid chromatography tandem mass spectrometry method, requiring just 30 minutes and reporting 82% of analytes with a recovery of between 70% and 130%.

Alongside EOC's, their transformation products can often be found in equal or greater concentrations (Stuart and Lapworth, 2014) and can have just as detrimental impacts. Stuart and Lapworth (2014) highlight the relatively few studies conducted in the area of emerging contaminant transformation products. Specific groups such as Pesticides, Disinfection By-products, Alkyl phenols and other endocrine disruptors and caffeine and nicotine are highlighted as some groups with transformation products of concern.

The recent publication of the GWWL (CIRCABC, 2019), establishes a ranking of compounds of current concern. Eleven compounds classified as either pharmaceuticals or PFAS are proposed for the GWWL, and a further 4 PFAS selected as candidates for the list (CIRCABC, 2019). This will likely help focus effort on the top ranked compounds of concern in GW until sufficient detail is collected for a regulatory level to be set. It is anticipated that this will be a dynamic process as compounds are studied, become regulated, and are replaced by the next highest ranking compound or a different group of compounds.

The detection of compounds continues to be a large part of the research process so that the presence of compounds can be assessed. However, the accurate quantification of compound across a large number of geologic environments, and using a range of techniques is equally imperative. The quantification of EOC concentrations is a key component of developing standards and threshold concentrations which may later be implemented into groundwater regulations

The lack of knowledge in the field of EOC's means that the majority of the studies are still at an investigative level. Aims of the studies, stated in the associated papers are commonly to understand the occurrence, transport and fate of EOC's within a given environmental setting. A lack of knowledge on every aspect of the EOC alongside limited monitoring data mean that threshold values have not been set (Lapworth et al., 2012), and therefore remain unregulated.

4.2 AREAS FOR FUTURE STUDY

There is still limited work published on the current state of EOC's in groundwater at a national level. In some countries, national reviews may be undertaken; but as they aren't published in English in peer reviewed journals, they are not included in this review. Although interest in the topic has increased in the past years, studies still tend to focus on small pilot study areas where all aspects of the occurrence transport and impacts of certain EOC's are analysed. The number of large-scale studies, and those with a large number of analytes (>500) are still relatively low, owing to the high cost of screening and the logistical complexity of screening for large numbers of compounds. Currently there are a number of small-scale studies where a specific problem compound has been identified. This allows specific compounds, like those identified on the GWWL to receive a greater level of study than others that may not pose such a site-scale threat, or may be less mobile in the environment. The quantification of these compounds allows threshold values and standards to be developed for a range of geological environments throughout Europe. However, there may be many more compounds present that have not been screened for, skewing our understanding of groundwater quality at that site to reflect the targeted compounds. It is therefore important that both targeted studies are conducted to quantify compounds of highlighted concern, whilst national and regional scale studies report the presence of other EOC's that can be studied further to determine their potential impacts.

The majority of studies included in this review include pharmaceutical compounds, an area that has been heavily studied in previous years. The data presented therefore shows these compounds as frequently screened for and detected, which may distract from other compounds which are screened less regularly. The GWWL aims to report selected compounds of particular concern, once their toxicity and mobility has also been considered, leading to a more targeted approach to screening for EOC's in groundwater.

For the development of EU regulation on EOC's, there needs to be a greater emphasis on understanding the occurrence of EOC's in groundwater throughout Europe. Meanwhile studies on the possible impacts of the compounds must also start to develop a better understanding of the effect of the compound(s) on the aquatic environment and groundwater dependant ecosystems. The additional impact of synergistic affects, whereby an impact is compounded by the presence of more than one type of compound, must also be considered. Currently each compound is primarily assessed independently, but future studies must also assess the impact of mixtures of compounds, considering a potential cocktail effect.

We also need to improve the knowledge of relationships that link anthropogenic land uses and activities with the potential impact on groundwater quality taking into account pathways and fate of molecules that interact with physico-chemical contexts of soils and underground. This knowledge is crucial for measures to be taken on the right targets (industries, WWTP, etc..), and applied at the right scales.

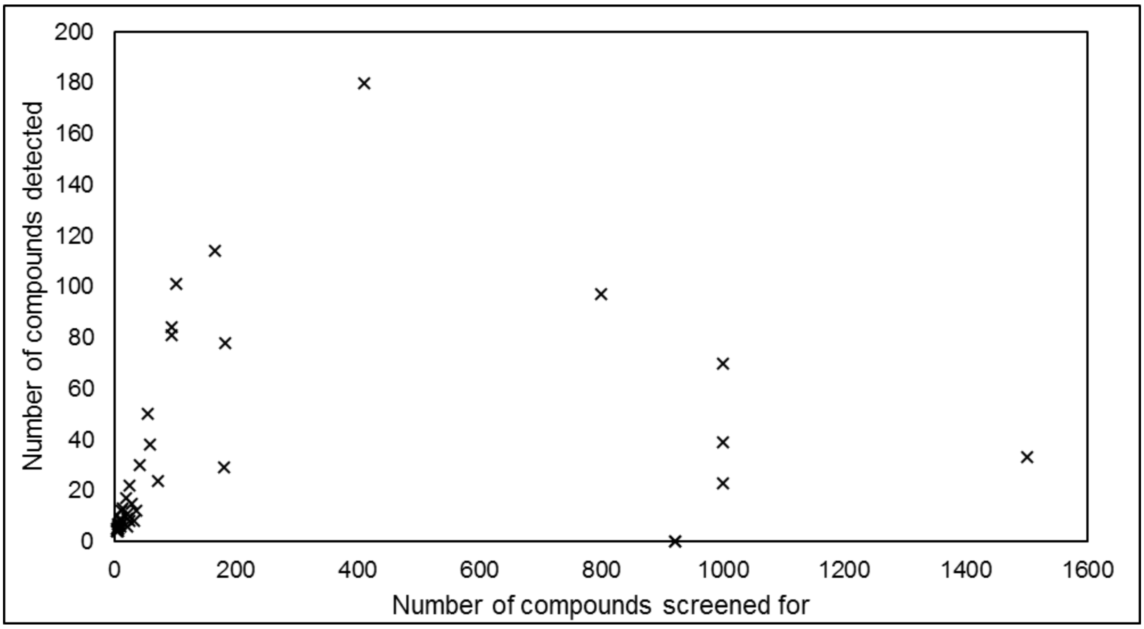
Future studies should define the source of their detections, as in a number of published studies it is uncertain as to the source of a positive detection, and cannot therefore be included in this study. It would also be useful to follow a standardised approach to reporting. Although positive detections are usually defined as above the LOQ or LOD, studies report these intermittently. Furthermore, it would be useful to report on the maximum concentrations and % hits for compounds of greater concern. This would enable a more detailed review to be conducted.

As shown in Figure 5, an apparent increase in number of compounds assessed for stops increasing the number of detected compound at around assessed 100 compounds. This may be the limit of our current analytical scope and the number of compounds known to have the potential to reach groundwater. An increase in groundwater sites however, appears to continue to increase the number of detected compounds. This highlights the need for prioritisation of the compounds known to exist in groundwater. Furthermore, if we wish to identify a greater number of compounds, the data suggests that we should increase the number of sites at which we search for compounds, rather than just increasing the number of compounds analysed.

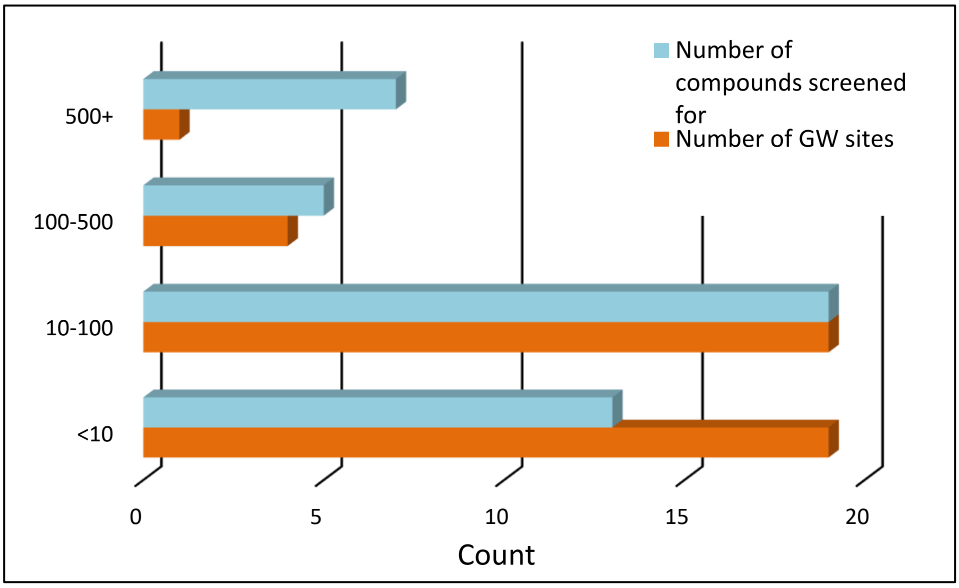
5 Recommendations

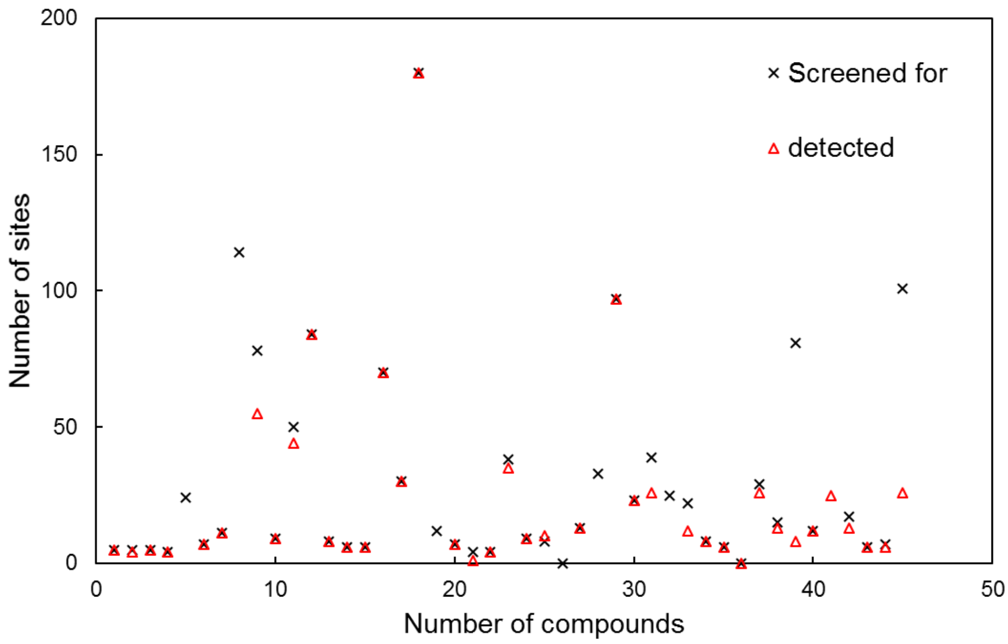
- There exists a high frequency of detections of a number of EOC's throughout Europe, a number of which are also detected at high concentrations. Although this helps to develop the distribution of EOC's, it doesn't include toxicity/hazard information and is heavily biased towards a small number of compound groups that have been more frequently investigated.
- It is important to continue advancing extraction and analytical methods which allow new EOC's to be detected. Continue to develop greater understanding of those already detectable, so suitable limits can be set and implemented.
- The GWWL is likely to be implemented throughout Europe, to help prioritise which compounds to look for. This is a relatively small list for pragmatic reasons, however, this should not detract from the need to continue to screen for a wider number of compounds that are not on the GWWL.
- Increasingly, EOC's are used as tracers for surface water/groundwater interactions or interaction with infrastructure e.g. sewer networks, treatment plants (Hillebrand et al., 2012, Wolf et al., 2012). Although these studies have a different goals, they further our understanding of the occurrence, transport and impact of EOC's.
- Studies focussed on artificial recharge and re-injected water are not considered in this study, but offer further insight into EOC's that may not be removed during treatment being injected into groundwater.
- Increased quantification of EOC's in groundwater is needed to aid the development of threshold values. The GWWL highlights compounds of particular concern, giving researchers a direction for future studies to be focused. However, it is important to continue large scale scoping studies which are invaluable for assessing the occurrence of EOC's in groundwater bodies across a range of environmental settings.
- Due to the wide range of reporting styles between European countries, it is difficult to conduct a more detailed study. A template of reporting would be necessary in order for studies to report on the same statistics, in a way that would be comparable between countries, the GWWL process may go some way to help overcome this obstacle.

Appendix A

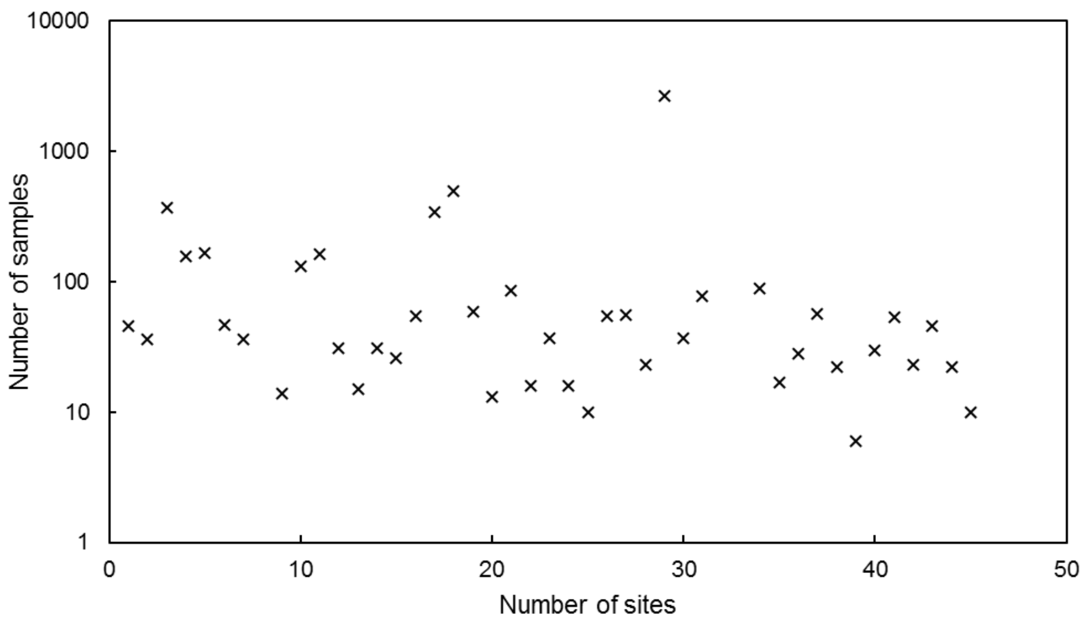


Appendix B





Appendix C



Appendix D

CAS number	Compound name/commonly known as	Number of papers with at least one positive detection	Use	Our categorisation
298464	Carbamazepine	22	anti-epileptic drug and other pharmaceutical applications	Pharmaceuticals
58082	Caffeine	15	Lifestyle	Lifestyle
723466	Sulfamethoxazole	13	Antibiotics	Pharmaceuticals

80057	Bisphenol A (BPA)	13	resins for food packaging	Plasticisers
15687271	Ibuprofen	12	Anti-inflammatory agent with analgesic properties	Pharmaceuticals
103902	Acetaminophen (Paracetamol)	9	Non-Prescription Drugs	Pharmaceuticals
134623	N,N-diethyl-m-toluamide (DEET)	8	insect repellant	PCP's
15307865	Diclofenac	8	Anti-inflammatory agent	Pharmaceuticals
108907	Chlorobenzene	8	Chlorinated solvent	Solvents and THMs
41859670	Bezafibrate	7	Lipid regulator	Pharmaceuticals
95147	Benzotriazole	6		Industrial
119619	Benzophenone	6	fragrance fixitive/sunscreen agent	PCP's
94133	Propylparaben	6	PCP	PCP's
29122687	Atenolol	6	Cardioselective beta-adrenergic blocker	Pharmaceuticals
218019	Chrysene	5	PAH	Industrial
129000	Pyrene	5	PAH	Industrial
120478	Ethylparaben	5	food additive	Lifestyle
53167	Estrone	5	Steroids and Hormones	Other EOC's
3380345	Triclosan	5		PCP's
99763	Methylparaben	5	PCP	PCP's
81103119	Clarithromycin	5	Antibiotics	Pharmaceuticals
114078	Erythromycin	5	Antibacterial agent	Pharmaceuticals
25812300	Gemfibrozil	5	Lipid-regulating agent	Pharmaceuticals
22071154	Ketoprofen	5	Analgesics/anti inflammatories and TPs	Pharmaceuticals
60800	Phenazone	5	Analgesic/anti-inflammatory	Pharmaceuticals
68152921	Nonylphenol	4		Industrial
126738	Tributyl phosphate	4		Industrial
85018	Phenanthrene	4	PAH	Industrial
486566	Cotinine	4	Nicotine metabolite	Lifestyle
519095	Benzoylecgonine	4	Cocaine metabolite	Lifestyle
50282	Estradiol	4	natural hormone	Other EOC's
496117	Indane	4	Petrochemical compound	Other EOC's
882097	Clofibric acid	4	Lipid regulator	Pharmaceuticals
54910893	Fluoxetine	4	Antidepressant	Pharmaceuticals

58935	Hydrochlorothiazide	4	Diuretic	Pharmaceuticals
37350586	Metoprolol	4	Antihypertensive	Pharmaceuticals
611596	Paraxanthine	4	Psychoactive central nervous system stimulant. Metabolite of caffeine	Pharmaceuticals
125337	Primidone	4	Anticonvulsant, sedative	Pharmaceuticals
68359	Sulfadiazine	4	Antibiotics	Pharmaceuticals
57681	Sulfamethazine	4	Antibiotics	Pharmaceuticals
82419361	Ofloxacin	4	Antibiotics	Pharmaceuticals
83670	Theobromine	4	Stimulant	Pharmaceuticals
738705	Trimethoprim	4	Antibiotic	Pharmaceuticals
86737	Fluorene	4	PAH	Pharmaceuticals
84662	Diethyl phthalate	4	Plasticiser	Plasticisers
3622842	N-Butylbenzenesulfonamide	4	Plasticizer, metabolite of chlorobenzenes	Plasticisers
123911	1,4-Dioxane	4	Solvent	Solvents and THMs
79016	Trichloroethene (TCE)	4	Chlorinated solvent	Solvents and THMs
1763231, 132324119	Perfluoro-n-octane sulfonate (PFOS)	3	PFAS	Industrial
13973143	Perfluorooctanoic acid (PFOA)	3	PFAS	Industrial
29385431	Tolyltriazole	3	Corrosion inhibitor	Industrial
95169, 128366289	Benzothiazole	3	Rubber additive, antimicrobial agents, flavors	Industrial
87412, 1135443467	1(3H)-isobenzofuranone (Phthalide)	3	Dye intermediate	Industrial
3320830, 51134033	2-Chlorophenyl isocyanate	3		Industrial
56553	Benz(a)anthracene	3	PAH	Industrial
50362	Cocaine	3	illicit drugs	Lifestyle
57272	Morphine	3	Opioids	Lifestyle
54115	Nicotine	3	Lifestyle	Lifestyle
25013165	Butylated hydroxyanisole (BHA)	3	Food additive	Lifestyle
128370	ButylatedHydroxytoluene (BHT)	3	Food additive	Lifestyle
131577	Benzophenone-3	3	PCP	PCP's
1222055, 80450664	Galaxolide	3	Musk fragrance	PCP's
85721331	Ciprofloxacin	3	Antibacterial agent	Pharmaceuticals
439145	Diazepam	3	Sedative, anxiety-relieving and muscle-relaxing (Valium)	Pharmaceuticals

443481	Metronidazole	3	Antibiotics	Pharmaceuticals
22204531	Naproxen	3	Anti-inflammatory agent with analgesic and antipyretic properties	Pharmaceuticals
54217	Salicylic acid	3	Analgesics/antiinflammatories and TPs	Pharmaceuticals
3930209	Sotalol	3	Antihypertensive	Pharmaceuticals
60548	Tetracycline	3	Antibiotics and TPs	Pharmaceuticals
76573	Codeine	3	Analgesic agent	Pharmaceuticals
154212	Lincomycin	3	Antibiotic	Pharmaceuticals
61687	Mefenamic acid	3	Analgesics/antiinflammatories and TPs	Pharmaceuticals
604751	Oxazepam	3	Other Prescription Drugs	Pharmaceuticals
525666	Propranolol	3	Cardiovascular drugs and TPs	Pharmaceuticals
479925	Propyphenazone	3	Analgesics/antiinflammatories and TPs	Pharmaceuticals
84742	Dibutyl phthalate (DIBP)	3	Plasticiser	Plasticisers
74953	Dibromomethane	3	Volatile Solvent	Solvents and THMs
120821, 63697187	1,2,4-Trichlorobenzene	3	Insecticide, degreasing, pigments, chemical intermediates	Solvents and THMs
95136	Indene	3	Solvent	Solvents and THMs
375224	Perfluorobutanoic acid (PFBA)	2	PFAS	Industrial
307551	Perfluorododecanoic acid (PFDoDA)	2	PFAS	Industrial
375859	Perfluoro-n-heptanoic acid (PFHpA)	2	PFC	Industrial
307244	Perfluoro-n-hexanoic acid (PFHxA)	2	PFC	Industrial
82382125	Sodium perfluoro-1-hexane sulfonate (L-PFHxS)	2	PFAS	Industrial
934349, 4464599, 60362052, 92353214	2(3H)-Benzothiazolone	2		Industrial
126863	2,4,7,9-Tetramethyl-5-Decyne-4,7-diol (TMDD)	2	Industrial	Industrial
105602	Caprolactam	2		Industrial
108941, 9003412, 11119770, 9075994	Cyclohexanone	2	precursor	Industrial

123637	Paraldehyde	2		Industrial
83329	Acenaphthene	2	PAH	Industrial, Pharmaceutical
134725	Ephedrine	2	Illicit drugs	Lifestyle
57885	Cholesterol	2	Steroid, manure, septic tanks, sewage intrusion	Other EOC's
53703	Dibenzo(a,h)anthracene	2		PAH
36861479	4-Methylbenzylidenecamphor (Enzacamene)	2	skincare	PCP's
94268	Buthylparaben	2		PCP's
6197304	Octocrylene	2	UV filter	PCP's
90437	O-Phenylphenol	2	Disinfectant	PCP's
88150429	Amlodipine	2	Cardiovascular	Pharmaceuticals
36507309	Carbamazepine epoxide	2	Other Prescription Drugs	Pharmaceuticals
56757	Chloramphenicol	2	Antibiotics and TPs	Pharmaceuticals
57625	Chlortetracycline	2	Antibiotics	Pharmaceuticals
59729338	Citalopram	2	Antidepressant	Pharmaceuticals
94088854	Doxycycline	2	Antibiotics	Pharmaceuticals
75847733	Enalapril	2	Cardiovascular drugs and TPs	Pharmaceuticals
93106606	Enrofloxacin	2	Antibiotics	Pharmaceuticals
42835256	Flumequine	2	Antibiotics	Pharmaceuticals
54319	Furosemide	2	Diuretic	Pharmaceuticals
78649419	Iomeprol	2	Iodinated contrast media	Pharmaceuticals
79794755	Loratadine	2	Antiallergic	Pharmaceuticals
846491	Lorazepam	2	Psychiatric drugs and TPs	Pharmaceuticals
114798264	Losartan	2	Other Prescription Drugs	Pharmaceuticals
57534	Meprobamate	2	tranquilizer	Pharmaceuticals
50066	Phenobarbital	2	Barbiturates	Pharmaceuticals
7206760, 80147406	Phenylethylmalonamide	2	metabolite of primidone	Pharmaceuticals
13523869	Pindolol	2	Cardiovascular drugs and TPs	Pharmaceuticals
80214831	Roxithromycin	2	Antibiotic	Pharmaceuticals
54965241	Tamoxifen	2	Cancer treatment	Pharmaceuticals
93413695	Venlafaxine	2	Antidepressant	Pharmaceuticals
51481619	Cimetidine	2	Antihistaminics and TP	Pharmaceuticals
112398080	Danofloxacin	2	Antibiotics and TPs	Pharmaceuticals
1095905	Methadone	2	Opioids	Pharmaceuticals
965526	Nifuroxazide	2	Antibiotics and TPs	Pharmaceuticals
76744	Pentobarbital	2	Barbiturates	Pharmaceuticals

144832	Sulfapyridine	2	Antibiotics	Pharmaceuticals
58559	Theophylline	2	Stimulant, metabolite	Pharmaceuticals
2440224	Drometrizole	2	UV screen for plastics	Plasticisers
124481	Chlorodibromomethane	2	THM	Solvents and THMs
100414	Ethylbenzene	2	Solvent	Solvents and THMs
128686033	M & p-xylene	2	Petroleum products, chemical intermediates	Solvents and THMs
95476, 68411847	O-xylene	2	Petroleum products, chemical intermediates	Solvents and THMs
127184	Tetrachloroethene (TCE)	2	Chlorinated solvent	Solvents and THMs
75252, 4471185	Bromoform	2	THM	Solvents and THMs
98828	Isopropylbenzene	2	Volatile solvent	Solvents and THMs
13401564	1,1-Dimethyl-3-chloropropanol	1		Industrial
207122154	2,2´,4,4´,5,6´-Hexabromodiphenylether (BDE 154)	1	flame retardant	Industrial
60348609	2,2´,4,4´,5-Pentabromodiphenylether (BDE 99)	1	flame retardant	Industrial
189084648	2,2´,4,4´,6-Pentabromodiphenylether (BDE 100)	1	flame retardant	Industrial
79755434	2,2´,4,4´-Tetrabromodiphenylether (BDE 47)	1	flame retardant	Industrial
96764	2,4-Di-tert-butylphenol (2,4-DTBP)	1	Industrial	Industrial
128392	2,6-Di-tert-butylphenol	1	UV stabaliser	Industrial
104767	2-Ethyl hexanol	1		Industrial
149304	2-Mercaptobenzothiazole	1		Industrial
91-57-6	2-Methylnaphthalene	1	PAH	Industrial
626437	3,5-Dichloroaniline	1	Chemical intermediates	Industrial
108601, 52438912	Bis(2-chloroisopropyl) ether	1		Industrial
106650	Dimethyl succinate (Butanedioic acid, dimethyl ester)	1	precursor	Industrial
104358, 26027383	Nonylphenol-mono-ethoxylate	1		Industrial
375735	Perfluorobutane sulfonate (PFBS)	1	PFAS	Industrial
335762	Perfluorodecanoic acid (PFDA)	1		Industrial
3871996	Perfluorohexane sulfonate (PFHxS)	1	PFAS	Industrial

375951	Perfluorononanoic acid (PFNA)	1	PFAS	Industrial
2706903	Perfluoropentanoic acid (PFPeA)	1	PFC	Industrial
2058948	Perfluoroundecanoic acid (PFUnDA)	1	PFAS	Industrial
1763231	Perofluorooctanesulfonic acid (PFOS)	1	PFAS	Industrial
115968	Tris(2-chloroethyl) phosphate	1	Flame retardant	Industrial
38998753, 67562394, 67652395	1,2,3,4,6,7,8-Heptachlorodibenzofuran (HTCBF)	1	automotive manufacture	Industrial
35822469, 37871004	1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin (HTCBD)	1	automotive manufacture	Industrial
39227286	1,2,3,4,7,8-Hexachlorodibenzo-p-dioxin (HTCBD)	1	automotive manufacture	Industrial
51207319	2,3,7,8-Tetrachlorodibenzofuran (TCBF)	1	automotive manufacture	Industrial
924163	N-nitrosodi-n-butylamine	1	automotive manufacture	Industrial
59892, 67587568	N-nitrosomorpholine	1	automotive manufacture	Industrial
36038536	1,1,4,4-Tetrachlorobutadiene	1	Chlorinated solvent	Industrial/Solvent
6136374	1-Methylxanthine	1	Stimulant, metabolite	Lifestyle
30223735	2-Ethylidene-1,5-dimethyl-3, 3-diphenylpyrrolidine	1	illicit drugs	Lifestyle
42542109	3,4-methylenedioxymethamphetamine (MDMA)	1	Amphetamines	Lifestyle
33665906	Acesulfame	1	Food additive	Lifestyle
28981977	Alprazolam	1	Illicit drugs	Lifestyle
529384	Cocaethylene	1	Cocaine metabolite	Lifestyle
149326	Erythritol	1	Artificial sweeteners	Lifestyle
125291	Hydrocodone	1	Opioids	Lifestyle
61507, 68677258	N,N-Dimethyltryptamine (DMT)	1	Illicit drugs	Lifestyle
60426417	Norbenzoylecgonine (BNE)	1	Cocaine metabolite	Lifestyle
76426	Oxycodone	1	Opioids	Lifestyle
81072	Saccharin	1	Sweetener	Lifestyle
21145777	Tonalide	1	Fragrance	Lifestyle
20189428	2-Ethyl-3-methylmaleimide	1	Natural compound	Other EOC's
108689	3,5-Dimethylphenol	1		Other EOC's
14035337	3,5-di-tert-butyl-4-hydroxyacetophenone	1		Other EOC's

2300063	6β-Methylpregn-4-ene-3,20-Dione	1	Steriod	Other EOC's
80091	Bisphenol S	1		Other EOC's
624920, 68920649	Dimethyl disulfide	1	Natural compound	Other EOC's
3658808	Dimethyl trisulfide	1	Natural compound	Other EOC's
50271	Estriol	1	natural hormone	Other EOC's
10544500	Sulphur S8	1	Petroleum products, decomposition of tyres, reduction of sulphate	Other EOC's
611994	4,4'-Dihydroxy benzophenone	1	UV filter	PCP's
1137424	4-Hydroxybenzophenone	1	UV filter	PCP's
25766181	Alpha-pinene	1	Insecticide, cosmetic, solvent, plasticizers	PCP's
100527	Benzaldehyde	1	Chemical intermediates, solvent, bee repellents	PCP's
131566	Benzophenone-1	1	UV protector	PCP's
4065456	Benzophenone-4	1	PCP	PCP's
83834597, 5466773	Ethylhexyl methoxycinnamate	1	Sunscreening agent	PCP's
1843056	octabenzone	1	UV absorber/screener	PCP's
68155668	Octahydrotetramethyl Acetophenone (OTNE)	1	fragrance	PCP's
27503817	Phenylbenzimidazole Sulfonic Acid (PBSA)	1	UV absolper	PCP's
4640011	Triclosan methyl	1	disinfactant, soap	PCP's

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British Geological Survey holds most of the references listed below, and copies may be obtained via the library service subject to copyright legislation (contact libuser@bgs.ac.uk for details). The library catalogue is available at: https://envirolib.apps.nerc.ac.uk/olibcgi.

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DIRECTIVE

COMMISSION DIRECTIVE 2014/80/EU of 20 June 2014 amending Annex II to Directive 2006/118/EC of the European Parliament and of the Council on the protection of groundwater against pollution and deterioration.