boDEREC-CE
WORKPACKAGE T1

O.T1.1 STATE-OF-THE-ART OF CURRENT PRACTICES IN RELATION TO EMERGING CONTAMINANTS IN THE WATER ENVIRONMENT

LEARNING TOOL

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1. Introduction

Emerging contaminants (ECs) have been in recent years recognised as significant pollutant. They are widely present and distributed in aquatic environment, and can have an impact on human and wildlife health, especially endocrine system. The main goal of work package T1 (WP T1) Discovering emerging contaminants in the water environment –State-of-the-art, is a review of the state-of-the-art (SOA) of handling, monitoring and treatment of ECs in water resources management, including different views and approaches: from political, legislation, monitoring and engineering point.

All 12 partners were involved and worked on this WP. Within this WP project partners (PPs) collected existing information about emerging contaminants through the existing transnational and national sources. In the first steps of the WP PPs identified emerging contaminants and collected relevant information about related problems and their appearance in the water environment which represents sources of drinking water supply. In the following steps, PPs did gather information about monitoring strategies and approaches and reviewed current analytical techniques. At the end of the work package, PPs also reviewed transnational and national findings on current attenuation strategies.

The SOA preparation process for the boDEREC- CE project can be understood as an illustration of the data collection approach related to ECs or similar substances. From this point of view, we consider the present report as a learning tool that is important for achieving the final objectives of the project and for the wider scientific and professional public at large.

2. Approach to State-of-the-art synthesis

The problems relating to the ECs represent a very broad field. In the scientific community and in various other socio-economic fields, researchers and managers are currently dealing with a wealth of information which has not yet been synthesised in a readable way at the level where it is clear and can be directly translated into practise and daily life. Such syntheses are urgently needed, and the efforts and objectives of the PPs in the boDEREC- CE project should contribute to these efforts, among others.

The preparation of the SOA with regard to the ECs was an identification and learning process, which started from the basics defined in the "project description", with the PPs already following a pre-development approach during the project proposal. Once the project was launched, it was necessary to fully develop procedures for the collection of available data and information. The boDEREC- CE procedure for data collection and SOA preparation can be applied as a step-by-step learning process. Following boDEREC- CE the preparation of a SOA can be seen as a learning tool for those who are working in a similar project or who are interested in the topic of ECs. Information for SOA has been collected based on the block diagram shown in Figure 1.

Emerging contaminants are an important topic in current science and professional life. At the same time, the term "emerging contaminant" is well accepted from a terminological point of view. However, the discussion among the partners and the review of the literature has shown that it is far from common agreement what is meant by the term and which substances are included. With this in mind, it was decided that emerging contaminants must be identified more precisely in the first step of the SOA. All information collected in connexion with the conceptual identification of the EC is summarised in the chapter "Overview of identified contaminants". From the definition and identification of contaminants follows the identification of problems. Certain compounds give rise to specific problems and, from this point of view, monitoring strategies must be related to them. Various aspects of ECs have been identified in the chapter "Monitoring strategies". Data were collected both at national level and from international sources, from the Internet to scientific papers. ECs are chemical substances that are present in the aquatic environment in very low concentrations. Such properties require specific chemical analysis and their application is closely linked to monitoring strategies. All relevant information has been summarised in the chapter "Analytical techniques. Monitoring provides results on which decisions must be based. The results provide a starting point for the decision process that follows the attenuation strategies. Information on them is very scarce and not much practical experience is available. Existing information has been summarised in the chapter "Attenuation strategies".

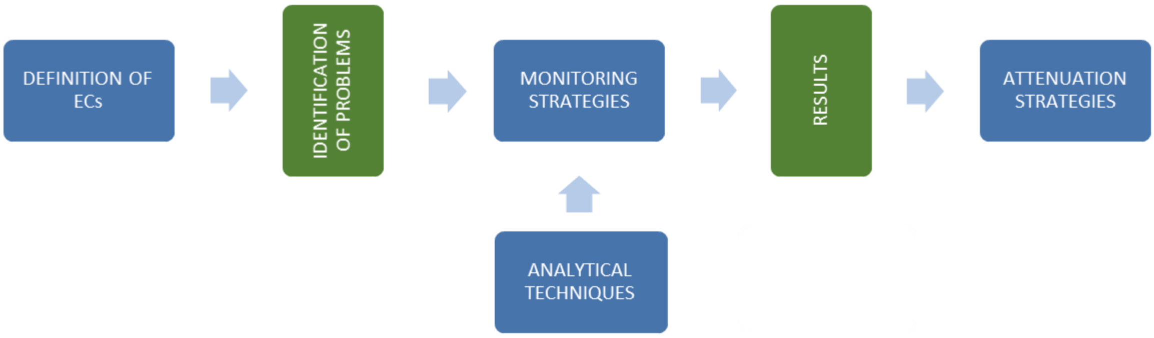

Figure 1 Collected information for state-of-the-art of emerging contaminants in water enivronment

A prepared SOA can also be understood as a learning tool within the framework of the boDEREC- CE project. The preparation of the SOA was a fruitful process for the PPs. They have synthesized national data on the ECs and, based on this, identified their own knowledge gaps. The process of preparing the national and transnational reports was important as an exchange process in which the PPs learn from each other and share experiences.

3. Overview of the indentified contaminants

In this chapter an overview and synthesise information on the identified contaminants is given based on the transnational report "Transnational synthesis report identifying problems related to emerging contaminants" which was prepared on the basis of the similarities and differences found in national reports and provides conclusions on the following main topics:

- Emerging contaminant (EC) terminology,
- EC sources,
- Release mechanisms of ECs in the environment,
- Problems regarding ECs.

3.1. Emerging contaminants definition

An exact, concise and coordinated lexical definition of ECs is needed for the continuing work on the project. As of now, based on the data included in the national reports and additional reviewed literature, there is no globally uniform definition of ECs. None of the PPs reported an official definition of ECs being given in their country. The cited definitions in the national reports where based either on different literature and web sources or included the project partner's (PP's) own synthesis. Definitions of ECs appear in certain scientific articles, project reports and websites. Some of the sources give a variety of definitions and some debate the commonly used terms and definitions used in the field. These observations all show that the scientific and non-scientific community has an interest in the issue of ECs, which is being actively studied and is still in the beginning phases of research.

The existing definitions mainly describe the most common general characteristics of EC. Figure 1 lists common definitions of ECs as they appear in all national reports of the project.

In current definitions, it has been recognised that ECs do not refer to only one type but in fact encapsulate a myriad of different groups (as the plural term itself indicates) which are most commonly referred to as chemicals or chemical compounds, substances, or simply, pollutants. Some of the definitions include microorganisms.

Since it has been established that ECs include not only anthropogenic substances but also naturally occurring ones and can also be microorganisms which are themselves composed of different substances, specific terms such as chemicals, chemical compounds and substances should not be used to unequivocally describe ECs but rather serve as examples of various different, albeit most common groups of ECs.

For a unique and firm definition, we can quite simply start with the fact that ECs represent a certain subclass of contaminants. Contaminants are defined as substances present above the natural background which have the potential to become pollutants, whereas pollutants, in turn, are defined as substances that have adverse biological effects (Chapman, 2007). The term pollutant is consequently not synonymous with contaminant and should not be equated with it, but it does represent a subclass of contaminants. Not defining contaminants as pollutants themselves is therefore important because they can be introduced into the environment where they are more or less bioavailable depending on many chemical and environmental factors as will be explained in the following chapters.

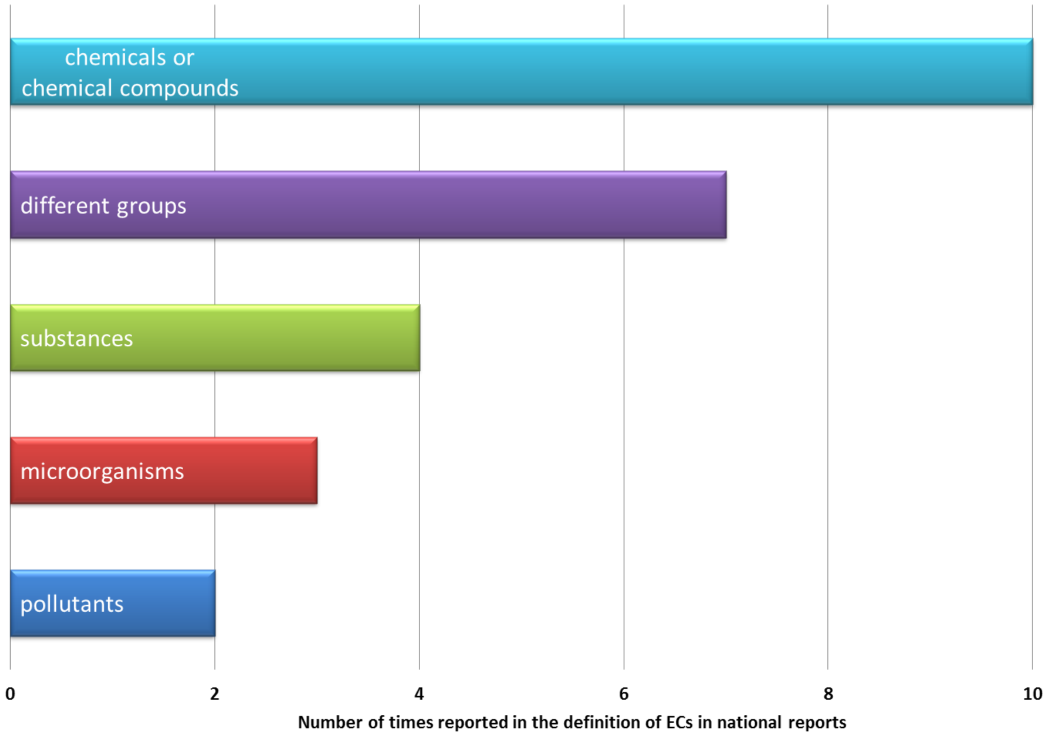

Figure 1 Different definitions of ECs in national reports

Defining ECs as a **subclass of potential pollutants** provides a group term for substances, compounds, mixtures and microorganisms and thus eliminates the problem encountered earlier.

Some existing definitions of ECs also consider this distinction, such as the one provided by the NORMAN network, a Network of reference laboratories, research centres and related organisations for monitoring of emerging environmental substances which was established in 2005 within a framework of a European project. The NORMAN project definition includes emerging substances and emerging pollutants separately with the following description (NORMAN Network Website, 2020):

- *"Emerging substances can be defined as substances that have been detected in the environment, but which are currently not included in routine monitoring programmes at EU level and whose fate, behaviour and (eco)toxicological effects are not well understood."*
- *"Emerging pollutants can be defined as pollutants that are currently not included in routine monitoring programmes at the European level and which may be candidates for future regulation, depending on research on their (eco)toxicity, potential health effects and public perception and on monitoring data regarding their occurrence in the various environmental compartments."*

Next, the defining characteristics of this subclass of potential pollutants need to be narrowed down and detailed further. ECs' descriptions most often include their characteristic sources, levels, occurrence, and behaviour, interaction with the environment, our knowledge and consideration of them. In the following paragraphs, these characteristics are described in more detail, covering the chemical, environmental and legislative frames of their occurrence. They are also a basis for several classification schemes of ECs described in the next chapter. Figure 2 shows most commonly described characteristics of ECs in national reports with relative sizes corresponding to the number of times they were mentioned in all of the reports.

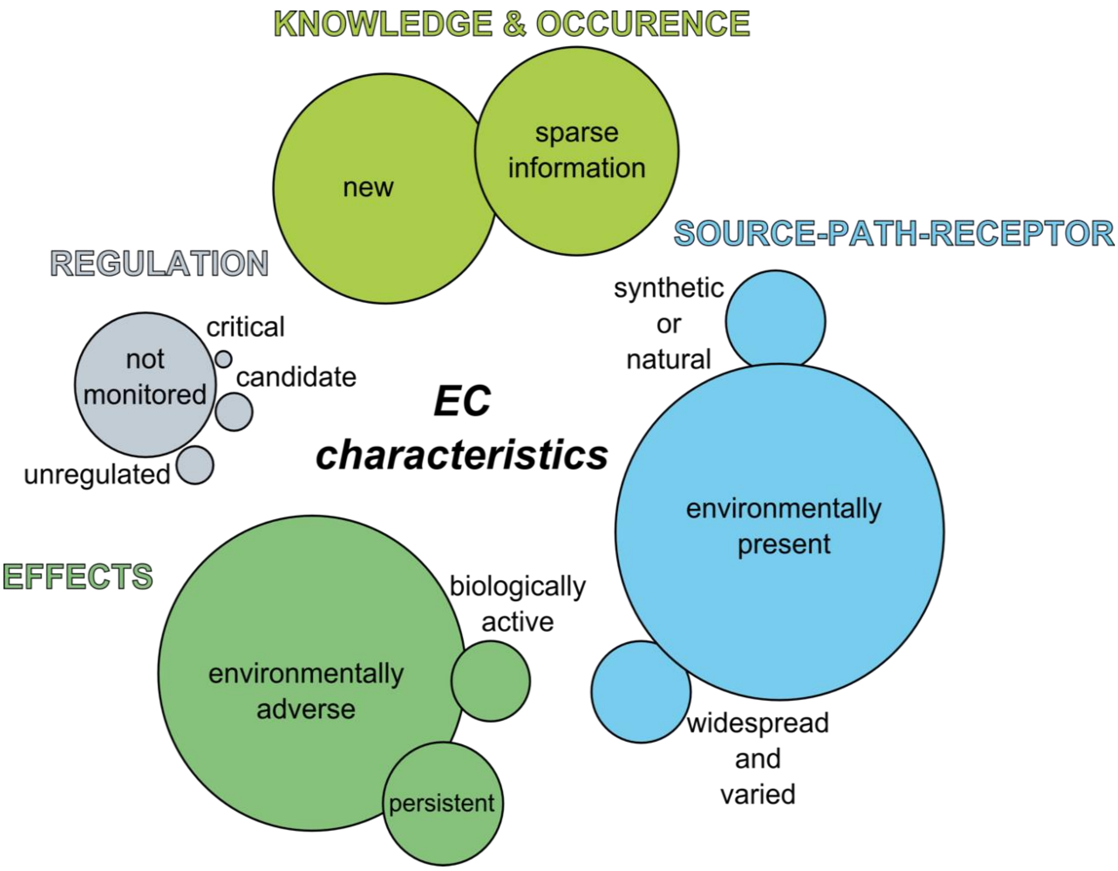

Figure 2 Most common characteristics of ECs in national reports

Sources

Sources of EC are most often anthropogenic although they can also be natural. An example of anthropogenic ECs would be pharmaceuticals, an example of natural ones are hormones or microorganisms. The sources of ECs are also described as being varied and as being continuously used in everyday life and other activities (industry, agriculture). In this way, they have the potential and in many cases often do enter the environment, which is a descriptive characteristic of ECs that often appears in their characterization. The sources of ECs can thus be described as various, widespread, regularly or even constantly present and in many cases, they are not simple but complex, composed or consisting of many different individual ECs.

Levels

The concentrations of those ECs that are most often studied are usually low, but they can range from ng/l to mg/l (Salimi, 2017). Even though their concentrations can be as low as trace elements, they are still relatively high considering that they were, not so long ago, expected to be non-existent in the natural environment. On the contrary, it would seem that some ECs are ubiquitous, as they are constantly being found globally in different parts of the environment, especially in different aquatic environments. This is particularly true of those ECs that have high mobility and high persistency.

ECs can also be referred to as micropollutants, pointing at the concentration levels at which they occur. It does however need to be pointed out that while they are found in some parts of the ecosystem in a certain levels of concentration, they can vary in others. For example, there can be a difference in levels of ECs in WWTP influents, effluents, and in their receiving surface waterbodies due to different water treatment methods and natural biotic or abiotic processes that influence the retention of contaminants from their entrance into and transport through the environment. The difference is even more pronounced when comparing different types of aquatic solution concentrations with their concentration in biosolids. Aga (2008) reports they can differ in several orders of magnitudes. Furthermore, some types of ECs can exhibit adverse effect to parts of the ecosystem even in small quantities (i.e. hormones).

As most ECs are artificially created, they can prove to be good tracers despite their low quantity as they indicate the presence of anthropogenic activity. Examples of newly studied potential tracers of urban wastewater in groundwater include carbamazepine, artificial sweeteners, and certain pesticides (McCance et al., 2018).

Behaviour and interaction

A defining characteristic of ECs is their potential to induce direct or indirect adverse effects in biota. Many of them are bioactive and affect different processes in organisms. They could be bioaccumulative and therefore biomagnify and/or they could be persistent in the environment, meaning they have a potential for long-range transport from their entry points into the natural environment. For example, ECs can circle the aquatic environment, entering surface water or groundwater by wastewaters and reappearing in drinking water originating from those sources. Not all ECs are persistent however – some are (bio)degradable and have metabolites and/or transformation products form from the parent compounds. Acesulfame (artificial sweetener) is for example very persistent (non-biodegradable), on the other hand butylparaben (PCP, preservative) is readily biodegradable (Margot, 2015).

Knowledge and occurrence

ECs are generally considered to be new, but that can refer to any number of different characteristics. For example, they can be new in the sense of production, detection, identification, etc.

It can be shown that different researchers might be talking about different types of ECs, among which our knowledge varies. The umbrella term ECs thus takes on a different meaning (of a subgroup of ECs) in each case. It is therefore not recommended to use the umbrella term ECs only to represent pharmaceuticals, for example, but to recognize that it can mean many different subgroups of natural or anthropogenic potential pollutants which have different characteristics, histories and documented information.

A review by Sauvé and Desrosiers (2014) highlights the relativity of the term “emerging” and states that the distinction in this case needs to be three-fold:

- contaminants that were not known in the past, meaning they were not yet produced (did not exist) or were not yet detected or identified,
- contaminants that existed, have been detected and identified but for which concerns about their adverse effects have only just been raised, and
- contaminants with already identified adverse effects the knowledge on which is currently being updated with newly conducted studies.

Only the first category of contaminants is truly “emerging”, while for the rest, the term CECs (contaminants of emerging concern) would be more appropriate, even though both are nowadays being used interchangeably.

Theoretically, this means that ECs might encompass those substances whose pollutant effects are poorly known or documented but merely suspected and would later be disproven. By not having any negative effects at all would in turn lead to its removal from the category. In this way, ECs are a fluid group, based on historical context and in a state of constant evaluation.

Regulation

Most of the currently recognized ECs are not included in routine environmental monitoring programmes and are not regularly monitored. This is however not true for all types of ECs, for example certain pesticides may already be included in routine national monitoring (i.e. atrazine).

This means that most ECs are currently unregulated but they do represent potential candidates for future legislative measures which could include regulatory monitoring and determination or criteria limiting their occurrence in the environment. You could say that this is intrinsic to the nature of ECs – constant emergence of new types of ECs or emergence of new interest in existing ECs cannot produce immediate regulatory changes. On the contrary, they will always be lagging due to the ongoing research which can take some time to confirm the urgency of regulation and also due to the complexities of the processes of preparation and adoption of new regulatory measures. As such, a constant stream of new knowledge and information on the potential pollutants and their ecotoxicology forces regulatory agencies to frequently re-evaluate their norms and guidelines. An example of this are the Priority and Watch Lists including ECs that were put forward by the EU legislators.

3.1.1. Proposed definition of ECs

ECs include a variety of substances and mixtures that originate from either natural or anthropogenic sources occurring in trace or higher concentration levels in various environmental compartments. They can already be regulated or they can be candidates for future legislative measures. They are either newly created, detected or our knowledge or interest in them has increased only recently. They are suspected or confirmed to have a potential to cause adverse effects in the environment if they are present in certain quantities and as such present a risk to the natural environment, including humans.

Based on the review of national reports and additional literature, we propose the following **definition** of ECs within the framework of the boDEREC-CE project and future research:

Emerging contaminants represent a group of potential pollutants that are either newly created, newly identified, newly detected or newly researched.

3.2. Groups of ECs

There are several ways in which we can classify ECs. These classifications are used to ensure a systematic overview and categorization of what is otherwise a long and diverse list of potential pollutants. There are some well-established acronyms which are being used in the field of ECs. In this subchapter we intend to provide a comprehensive overview of different groups of ECs and their main properties. The presented groups and possible classifications are largely related to the characteristics of ECs given in the previous chapter.

ECs groups that were listed in the national reports most often classify them based on intended use of their source. This highlights the required consideration of sources of these contaminants in the environment. Furthermore, they are grouped according to their chemical properties, their effect and the levels at which they occur in the environment (quantitative classification). Figure 3 shows the basis for several classification schemes presented in the following chapters.

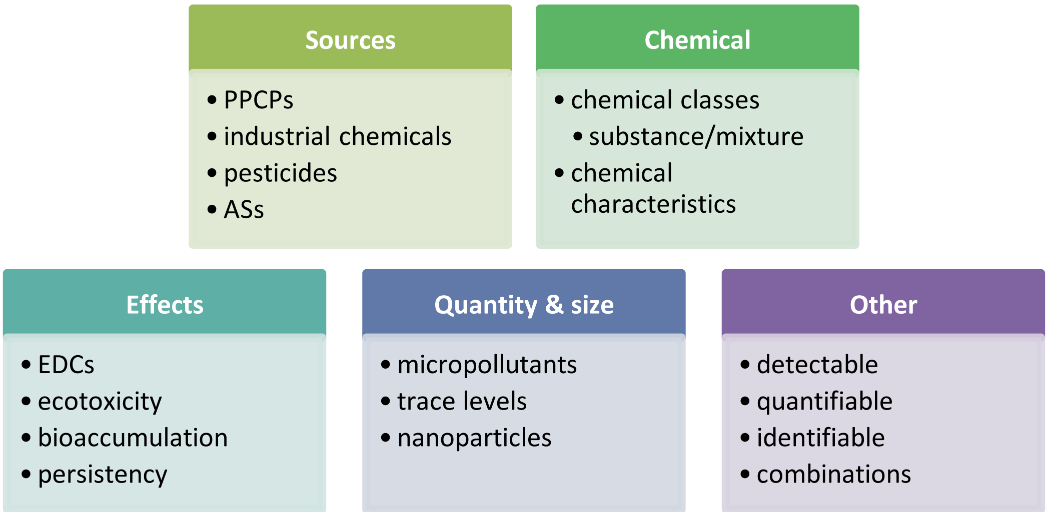

Figure 3 Different types of EC classifications

Classification based on sources

A human component is central to EC studies since the sources of ECs are most often anthropogenic, meaning they originate from human activity. ECs are also associated with anthropogenic matter and materials, also termed artificial or man-made, however, some ECs are naturally produced with no human contribution at all. In fact, there are certain ECs that can be both anthropogenic or naturally occurring.

ECs and their sources are further classified according to their purpose of use during different human activities. The main classification includes:

- industrial chemicals
- pharmaceuticals
- personal care products (PCPs)
- pesticides
- artificial sweeteners (ASs)

Main groups of ECs which are classified based on their source have their own subgroups which are classified according to their chemical class or again by their use. Each group is described in detail in the following paragraphs.

Industrial chemicals include flame retardants - FRs, perfluorinated chemicals - PFCs, surfactants, gasoline additives, intermediates of chemical factory industry, detergents, chlorinated solvents, perfluoroalkylated compounds, plasticizers, disinfectants, AOX, ethylene derivatives, polycyclic aromatic hydrocarbons, corrosion inhibitors.

Pharmaceuticals are a wide group intended for treatment of various humans and animal diseases and conditions. The acronyms PhACs (pharmaceutically active compounds) and APIs (active pharmaceutical ingredients) are sometimes used instead. Pharmaceutical environmental risk assessment is similarly shortened to PERA. Pharmaceuticals can be additionally classified into subgroups according to their type of use: antimicrobial agents (antiseptics, antibiotics), anti-inflammatory (corticosteroids), antihypertensives, lipid regulators, beta blockers, psychiatric drugs, analgesics, diuretics, hormones, radiographic contrast agents, stimulants, antidepressants, antiepiletics, antiulcerosum, hypolipidemics, fibrates, antiarhythmikum. We can also include illicit drugs (heroin, cocaine, amphetamine, MDMA, methamphetamine, cannabis) into this group of ECs.

Personal care products are a group of widely used chemicals in different every day products. They are classified according to their use into: UV filters (sunscreen agents), biocides (antimicrobials, insect repellents), siloxanes, preservatives (parabens), fragrances (musks), and detergents (Díaz-Cruz & Barcélo, 2015). Pharmaceuticals and personal care products are sometimes considered as a single group with the acronym PPCPs.

Pesticides are chemicals or biological substances used to kill or control pests (Waxman, 1998). They are used either for agricultural or non-agricultural purposes and can be classified according to the group of pest they target, for example into: acaricide, avicide, bactericide, nematicide, piscicide, predacide, fungicides, herbicides, insecticides, algicides, molluscicides, miticides, rodenticides, slimicides, etc. Biocides are pesticides intended to kill or inhibit growth of living organisms in industrial or consumer products (Karsa & Ashworth, 2002) and antimicrobials refer to either active compounds or preservatives used to control the presence of microorganisms in order to prevent or treat microbial disease or ensure the protection of microbiological safety and keeping quality of foods for consumption (Dhanasekaran et al., 2016; Taylor, 2015). Pesticides can also be classified according to their chemical class, for example organophosphate pesticides, organochlorine pesticides. Some pesticide groups are also considered to be PPCP and industrial chemicals.

Artificial sweeteners (ASs or ASWs) are sugar substitutes present in human nutrition. They are rarely metabolized and most remain unchanged as they find their way into the environment by human discharge and the absence of available WWTP removal technology (Salimi, 2017). They include the following: saccharin, cyclamate, acesulfame, aspartame, neotame, alitame, sucralose, neohesperidin dihydrochalcone. Since this group was introduced into human life and the environment only recently, it can prove to be a useful tracer of landfill or wastewater contamination, enabling age dating as well (Van Stempvoort et al, 2011; Roy et al., 2014). While the occurrence and persistency of ASs has been documented, the environmental and toxicological data is still poorly known (Praveena et al., 2019).

Chemical classification

Chemical classifications of ECs follow the basic chemical classifications of matter, starting with the division into elements or compounds and into pure substances or mixtures. An additional chemical classification distinguishes organic from inorganic matter, the former referring to carbon compounds and the latter to compounds without carbon. Chemical classifications also group matter with similar characteristics, which can be extended to ECs as well, for example, they can be classified according to whether they are polar, soluble, mobile, reactive, stable, conservative, degradable…

Some groups that belong to a specific class of chemical compounds are also chemically classified, such as polybrominated diphenyl ethers (PBDEs) or per- and polyfluoroalkyl substances (PFASs).

Some metabolites – products of biochemical reactions during metabolism and some degradation products are considered ECs as well.

Classification based on effects

Endocrine disrupting compounds (EDCs) are a group of ECs that are defined by the effect they have on the environment, specifically on living organisms. They interfere with the organism's endocrine system responsible for production, storage and secretion of hormones.

In the same way, ECs could be classified into those that have a pronounced impact on a certain group of organisms, for example aquatic, human…

There are a couple of terms that are used in relation to the effect of ECs on the environment, such as xenobiotics, bioaccumulation, biomagnification and ecotoxicity. Xenobiotics are all chemicals not naturally occurring in an organism. Bioaccumulation has different possible definitions and can mean a total uptake from the environment, the accumulation over time or the total retention, which can be difficult to determine based on many unknowns, like poor or scarce data (Díaz-Cruz & Barcélo, 2015). Biomagnification refers to a process of increase in concentration along the food chain, which is associated with bioaccumulation and other factors, such as longevity and organism size (Díaz-Cruz & Barcélo, 2015).

The acronym PBT is used for those contaminants that have the effect of being persistent, bioaccumulative and toxic.

Substance of very high concern (SVHC) is a term used by the EU regulation Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH, 2020) for chemical substances that have the following negative effects: they are carcinogenic, mutagenic, toxic for reproduction, persistent, bioaccumulative and toxic (PBT), very persistent and very bioaccumulative (vPvB) or if there is "scientific evidence of probable serious effects to human health or the environment which give rise to an equivalent level of concern".

Quantitative classification and classification based on size

ECs are sometimes referred to as micropollutants. Escher & Leusch (2012) describe micropollutants as man-made organic chemicals (pesticides, industrial chemicals, consumer products, pharmaceuticals) and natural compounds (hormones). As the name implies, micropollutants occur in the sub µg/l range, while macropollutants can be found in the mg/l to µg/l range. Macropollutants include toxicants, for example salts and metals (Escher & Leusch, 2012). A similar quantitative classification divides matter into trace and non-trace, the first occurring in concentrations below and the second above µg/l or ppm. With improvements in analytic technics and methods, matter can now be detected even at the sub ng/l levels or parts per quadrillion levels (Brooks & Haggett, 2012). Nanomaterials or nanoparticles are terms referring to the size, specifically to dimensions of up to 500 nm.

Other classifications

Some commonly used groups of ECs combine different types of classifications, for example trace organic contaminants (TOrCs) which includes two classifications; quantitative (trace) and chemical (organic). Another is anthropogenic trace compounds (ATC) which combines a quantitative classification (trace) and a classification based on the source (anthropogenic). An example of a combined classification is also the group of persistent organic pollutants (POPs) combining the classification based on effects (persistent) and a chemical classification (organic).

Finally, we could classify ECs based on our knowledge and regulatory consideration of them.

According to our current knowledge, there are certain ECs that can be identified with analytical methods and others that cannot. Similarly, we could make a distinction between ECs that can be quantified and those for which quantification is not yet possible.

There are some ECs that currently have a set or recommended or limit values, ones that have neither but have known ecotoxicological effects and some that have only suspected ecotoxicological effects. During our elaboration on the definition of ECs we concluded that ECs must always represent not only confirmed pollutants but also those that are only suspects and have not (yet) been confirmed as having any adverse effects on the environment.

3.3. ECs list

National reviews provided lists of varying number of ECs. Since there is no official comprehensive list in any of the partner countries, they were either summarizing the priority list and watch list put forward by the EU legislature in the Water Framework Directive or/and adding some additional ones based on the currently regulated pollutants within their individual legislature that could be considered ECs or based on available literature which was critically examined. The EU priority list includes priority substances in the field of water policy, while the watch list is frequently reviewed and updated. They are both detailed in a separate chapter of this report (Chapter 3.6 Priority lists of ECs). Some national reports cite the NORMAN network listing over 1000 different ECs.

3.4. Sources of ECs

In the beginning chapters, we have already mentioned a large spectre of possible sources of ECs and their varied nature. They are widespread and continuously used in modern day-to-day human activities.

Following the common contaminant characterization of sources that consequently determines the way in which we study and analyse them, sources fall into point or non-point categories, the latter also known as diffuse. They are anthropogenic in most cases, originating from one or more of the main human spheres; infrastructure, agriculture, industry or urban life (Figure 5).

Major EC sources include:

- pharmaceutical industries, industrial additives and agents, abandoned industrial sites, personal care products, factories, plants, mining activities,
- sewage, septic tanks, waste water treatment plants (effluent and sludge), landfills (leachate),
- households, hospitals, urban structures, stormwater runoff, roads and transport, cemeteries,
- agricultural land, farms, parks, gardens and horticulture.

One of the principle discharge sources of ECs is wastewater, containing non-point and point sources of industries, urban runoff, wastewater from households and water treatment facilities (Gogoi et al., 2018). Depending on the land use and industry type, wastewater has varying composition. In the vicinity of hospitals, for example, there can be an increase of ECs in WWTPs originating from medicinal activity and use (pharmaceuticals, diagnostic agents). Increased criminal activity contributes to higher level of narcotics in WWTPs. ECs enter the environment mostly through waste water treatment plants (WWTPs) having different levels of treatment (primary, secondary or tertiary) and different technologies of contaminant removal.

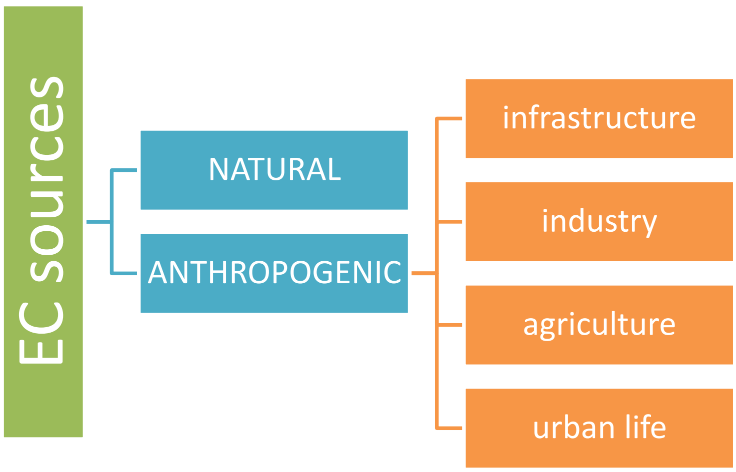

Figure 4 Sources of emerging contaminants

3.5. Release mechanisms and pathways of ECs in the water environment

ECs are being introduced into the environment through point or non-point (diffuse) sources, directly or indirectly, intentionally or unintentionally. They enter the environment from the source through application, discharges, leaks, and emissions. Media that enables transport of ECs to the aquatic environment is air, water (surface water, groundwater) and various porous media (soil, sediment, rock, Figure 6).

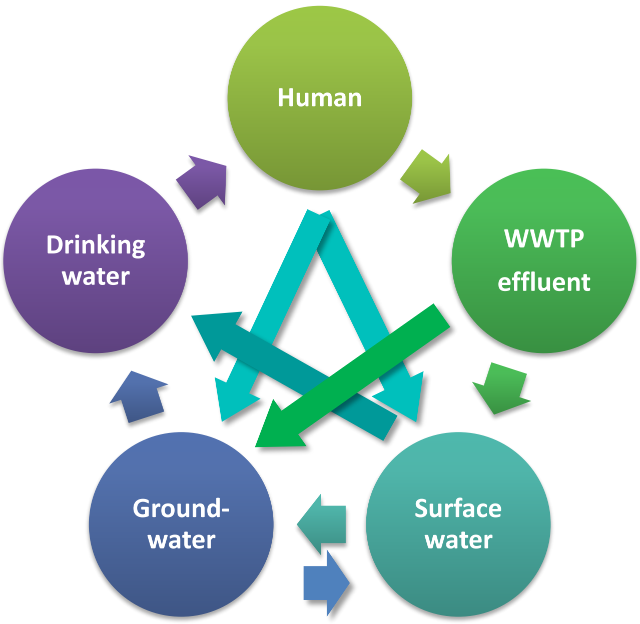

Figure 5 Possible pathways of wastewater EC transport through the water cycle

Currently, there are many unknowns regarding the pathways of some ECs in the environment. They depend on the environmental conditions and on the physicochemical properties of individual emerging contaminant along with its concentration levels, stability, hydrophobicity, reactivity, volatility and the type of removal technologies in place. Introduction of ECs to the environment is possible at the points of different water cycle components: directly into surface waters, directly into groundwater, onto soil surface which leads to infiltration through the soil zone and unsaturated zone into groundwater. Some ECs are able to travel through air transport and enter the water cycle after settling. The pathway of ECs through the environment from the source point onwards can be illustrated by the water cycle components that are connected to each other since the transport media is water. Along their transport pathway, processes of retention can influence their transport velocity. If groundwater or surface water is used for drinking water, the source of contamination becomes the receptor (human) and the cycle is completed and restarted.

3.6. Priority lists of ECs

Historically, several priority lists were specified that included contaminants with potential risks to the aquatic environment. For example, in the document Communication from the commission to the council on dangerous substances which might be included in List I of Council Directive 76/464/EEC, a priority list of 129 substances and groups was put forward with varying stages of research progress. Today, EU has put in place two main mechanisms - priority substances list and watch list which are described in the following chapters.

3.6.1. Priority substances list

Under the Water Framework Directive – WFD (2000/60/EC), a European strategy against the chemical pollution of surface waters was delineated, requiring the establishment of a list of priority substances (PS) and a subset list of priority hazardous substances (PHS) along with some management proposals. These priority lists include some emerging contaminants.

Decision 2455/2001/EC established the first priority list which included 33 substances or groups of major concern and 11 within this list as priority hazardous for which special management of inputs was needed (Table 1). Further 14 substances were identified as being subject to later review. Where groups of substances have been selected, typical individual representatives are listed as indicative parameters or the indicative parameter must be defined through the analytical method. These groups of substances normally include a considerable number of individual compounds. For some groups, appropriate indicative parameters cannot be given at present.

This first list was replaced by the second priority list in Annex II of the Directive on Environmental Quality Standards - EQSD (Directive 2008/105/EC), also known as the Priority Substances Directive. The second list reviewed the 14 proposed substances in the first list and added 8 other pollutants to the 33 priority pollutants.

Directive 2009/90/EC provided technical specifications for chemical analysis and monitoring of water status.

With the Proposal for a Directive amending the WFD and EQSD (COM(2011)876), 15 additional priority substances were added to the priority list, 6 of them are considered priority hazardous. This proposal also established a watch-list mechanism designed to allow targeted EU-wide monitoring of substances of possible concern to support the prioritisation process in future reviews of the priority substances list. According to the Proposal COM(2011)876, there are currently 48 priority substances ((Table 1, Table 2).

The priority substances and other pollutants complying with the limits are basis for reaching good chemical status of water bodies. On a national level, river basin specific pollutants are also identified and regulated.

Table 1 European Union Priority lists

1ST PRIORITY LIST *Decision 2455/2001/EC*	2ND PRIORITY LIST *Directive 2008/105/EC*	REVISED 2ND PRIORITY LIST *Proposal COM(2011)876*
33 priority substances/groups	33 priority substances/groups	48 priority substances/groups
11 priority hazardous	20 priority hazardous	28 priority hazardous
14 pollutants for later review		

Table 2 Current list of priority substances according to Proposal COM(2011)876

Number	CAS number	EU number	Name of priority substance	Identified as priority hazardous substance
1	15972-60-8	240-110-8	Alachlor	
2	120-12-7	204-371-1	Anthracene	X
3	1912-24-9	217-617-8	Atrazine	
4	71-43-2	200-753-7	Benzene	
5	not applicable	not applicable	Brominated diphenylethers	X
6	7440-43-9	231-152-8	Cadmium and its compounds	X
7	85535-84-8	287-476-5	Chloroalkanes, C_{10-13}	X
8	470-90-6	207-432-0	Chlorfenvinphos	
9	2921-88-2	220-864-4	Chlorpyrifos (Chlorpyrifos-ethyl)	
10	107-06-2	203-458-1	1,2-dichloroethane	
11	75-09-2	200-838-9	Dichloromethane	
12	117-81-7	204-211-0	Di(2-ethylhexyl)phthalate (DEHP)	
13	330-54-1	206-354-4	Diuron	
14	115-29-7	204-079-4	Endosulfan	X
15	206-44-0	205-912-4	Fluoranthene	
16	118-74-1	204-273-9	Hexachlorobenzene	X
17	87-68-3	201-765-5	Hexachlorobutadiene	X
18	608-73-1	210-158-9	Hexachlorocyclohexane	X
19	34123-59-6	251-835-4	Isoproturon	
20	7439-92-1	231-100-4	Lead and its compounds	
21	7439-97-6	231-106-7	Mercury and its compounds	X
22	91-20-3	202-049-5	Naphthalene	

Number	CAS number	EU number	Name of priority substance	Identified as priority hazardous substance
23	7440-02-0	231-111-14	Nickel and its compounds	
24	25154-52-3	246-672-0	Nonylphenols	X
25	1806-26-4	217-302-5	Octylphenols	
26	608-93-5	210-172-5	Pentachlorobenzene	X
27	87-86-5	231-152-8	Pentachlorophenol	
28	not applicable	not applicable	Polyaromatic hydrocarbons (PAH)	X
29	122-34-9	204-535-2	Simazine	
30	not applicable	not applicable	Tributyltin compounds	X
31	12002-48-1	234-413-4	Trichlorobenzenes	
32	67-66-3	200-663-8	Trichloromethane (chloroform)	
33	1582-09-8	216-428-8	Trifluralin	X
34	115-32-2	204-082-0	Dicofol	X
35	1763-23-1	217-179-8	Perfluorooctane sulfonic acid and its derivatives (PFOS)	X
36	124495-18-7	not applicable	Quinoxyfen	X
37	not applicable	not applicable	Dioxins and dioxin-like compounds	X
38	74070-46-5	277-704-1	Aclonifen	
39	42576-02-3	255-894-7	Bifenox	
40	28159-98-0	248-872-3	Cybutryne	
41	52315-07-8	257-842-9	Cypermethrin	
42	62-73-7	200-547-7	Dichlorvos	
43	not applicable	not applicable	Hexabromocyclododecanes (HBCDD)	X
44	76-44-8 / 1024-57-3	200-962-3 / 213-831-0	Heptachlor and heptachlor epoxide	X

Number	CAS number	EU number	Name of priority substance	Identified as priority hazardous substance
45	886-50-0	212-950-5	Terbutryn	
46	57-63-6	200-342-2	17alpha-ethinylestradiol	
47	50-28-2	200-023-8	17beta-estradiol	
48	15307-79-6	239-346-4	Diclofenac	

3.6.2. Watch list

The watch list (WL) mechanism of WFD determines that:

- WL substances and groups are to be monitored across the EU for up to 4 years,
- a maximum number of 10 substances or groups of substances shall be included in the first watch list,
- WL increases by one at each update,
- WL increases up to a maximum of 14 substances or groups,
- WL should not contain more than 25 substances or groups at any given time and
- WL should be updated every 2 years.

The 1st watch list was published in Commission Implementing Decision (EU) 2015/495 and included 10 substances: diclofenac, 17-Beta-estradiol (E2), Alpha-ethinylestradiol (EE2), Oxadiazon, Methiocarb, 2,6-ditert-butyl-4-methylphenol, Tri-allate, Imidacloprid, Thiacloprid, Thiamethoxam, Clothianidin, Acetamiprid, Erythromycin Clarithromycin, Azithromycin, 2-Ethylhexyl 4-methoxycinnamate.

After the review, a 2nd watch list was proposed with a JRC technical report in April 2018. In the report, it is proposed that 5 substances are removed and 3 new substances are added (Table 3, 4). The five proposed substances for removal were: diclofenac, oxadiazon, tri-allate, 2-ethylhexyl-4-methoxycinnamate and 2,6-di-tert-butyl-4-methylphenol. The three new substances to be included were: metaflumizone, amoxicillin and ciprofloxacin. The new list was put forward with the COMMISSION IMPLEMENTING DECISION (EU) 2018/840 of 5 June 2018 establishing a watch list of substances for Union-wide monitoring in the field of water policy pursuant to Directive 2008/105/EC of the European Parliament and of the Council and repealing Commission Implementing Decision (EU) 2015/495.

The two groups of substances on the watch list are macrolide antibiotics and neonicotinoids, which are specified in detail. The group of macrolide antibiotics includes: Erythromycin (CAS number 114-07-8, EU number 204-040-1), Clarithromycin (CAS number 81103-11-9), Azithromycin (CAS number 83905-01-5, EU number 617-500-5). The neonicotinoids group includes: Imidacloprid (CAS number 105827-78-9/ 138261-41-3, EU number 428-040-8), Thiacloprid (CAS number 111988-49-9), Thiamethoxam (CAS number 153719-23-4, EU number 428-650-4), Clothianidin (CAS number 210880-92-5, EU number 433-460-1), Acetamiprid (CAS number 135410-20-7/160430-64-8).

Table 3 European Union Watch lists

1ST WATCH LIST *Decision 2015/495*	2ND WATCH LIST *Decision 2018/840*
10 substances/groups	8 substances/groups

Table 4 Current European Union watch list according to Decision 2018/840

Number	Name of substance/ group	CAS number	EU number	Indicative analytical method	Maximum acceptable method detection limit (ng/l)
1	17-Alpha-ethinylestradiol (EE2)	57-63-6	200-342-2	Large-volume SPE - LCMS-MS	0,035
2	17-Beta-estradiol (E2), Estrone (E1)	50-28-2, 53-16-7	200-023-8	SPE - LC-MS-MS	0,4
3	Macrolide antibiotics			SPE - LC-MS-MS	19
4	Methiocarb	2032-65-7	217-991-2	SPE - LC-MS-MS or GC-MS	2
5	Neonicotinoids			SPE - LC-MS-MS	8,3
6	Metaflumizone	139968-49-3	604-167-6	LLE - LC-MS-MS or SPE – LC-MS-MS	65
7	Amoxicillin	26787-78-0	248-003-8	SPE - LC-MS-MS	78
8	Ciprofloxacin	85721-33-1	617-751-0	SPE - LC-MS-MS	89

On 11 March 2019, a strategic approach to pharmaceuticals in the environment was adopted covering all lifecycle phases of pharmaceuticals (design, production, use and disposal).

As stated in the 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, a watch list for pollutants of groundwater should be established under the Common Implementation Strategy for Directive 2000/60/EC to increase the availability of monitoring data on substances posing a risk or potential risk to bodies of groundwater, and thereby facilitate the identification of substances, including emerging pollutants, for which groundwater quality standards or threshold values should be set. Groundwater Watch List (GWWL) will include new or emerging substances that Member States should include into monitoring programmes. The Commission Implementing Strategy

(CIS) Working Group on Groundwater (WG GW) was mandated to elaborate a concept for the establishment of this list. The methodology for GWWL is based on the occurrence of substances in groundwater (based on monitoring data) and the theoretical leaching potential of substances (based on the substance properties). The combined outcome of these two assessments, called "Combined groundwater leaching potential score", is linked with the hazard potential of these substances to form a ranked list of "Integrated groundwater score". This list serves as a basis for the determination of substances either to be selected for the GWWL, or to be listed to facilitate the Annexes I and II review process of the GWD.

3.7. ECs identification in water bodies

First analytical detections of pharmaceuticals and their metabolites were reported in the 1970s in sewage effluent. The following decades saw a worldwide increase in publications on pharmaceutically active substances (PASs) in the environments worldwide. Refinement of analytical instruments and methods contributed to widening the scope of detection and identification of ECs, which was followed by environmental risk assessments and guidelines given to and recognized by regulators (Brooks & Huggett, 2012).

Today, systematic monitoring on a national scale or scientific research studies funded through national or international programmes contribute to the identification and quantification of relevant ECs in water bodies. Some monitoring of ECs is performed in the scope of internal monitoring of drinking water providers or waterworks. In most cases, the research is targeted, meaning it focuses either on specific ECs or on specific groups or environments. Among the EU funded projects that investigated ECs in the past are: NORMAN, Pharmaqua, and LIFE PharmDegrade.

An overview of wide and varied EC research projects and programmes in partner countries of the boDEREC-CE project is given below.

In **Austria**, ECs were studied in the scope of 8 main national programmes and projects:

- Wassergüte project performed from 2014-2016 consisted of a comprehensive and continuous periodic monitoring of surface and groundwater analysing physical-chemical parameters and some of the priority substances delineated in the WFD, such as pesticides and their metabolites.
- Pesticides and their metabolites were also measured by a one-campaign programme Sondermessprogramm für Pestizide und Metabolen in selected groundwater and river monitoring sites, focusing on new active ingredients as well.
- A list of parameters including pharmaceuticals was investigated in selected monitoring points in a study Monitoringprogramm von Pharmazeutika und Abwasserindikatoren in Grund- und Trinkwasser.
- Some threshold values of selected drug metabolites not regulated under the drinking water legislation were aimed to be established by the report Abgeleitete Toleranzwerte für Ausgewählte Arzneimittelwirkstoffe in Trinkwasser.
- Certain PPCPs were measured in selected WWTP in the frame of the project SCHTURM.
- Some pharmaceuticals of selected WWTPs were studied within the project Pharmaqua.
- 33 priority substances and other 47 specified in the surface water legislation were analysed in municipal wastewaters within the scope of the project "Emissions of organic and inorganic substances from municipal wastewater treatment plants". 94 priority and other substances were measured in 8 sewage treatment plants as a follow up project "Emissions of selected priority and other substances from municipal wastewater treatment plants"

Croatian research projects have focused on:

- atrazine and pharmaceuticals in groundwater
- antimicrobials in municipal wastewater
- selected pharmaceuticals in surface waters
- antibiotic-contaminated effluents from pharmaceutical industries
- ECs in wastewaters and receiving rivers
- macrolide antibiotics in receiving river sediments
- spatiotemporal distribution of selected illicit drugs and therapeutic opioids (codeine, methadone) in surface water.

Several studies of occurrence of PPCPs in the **Czech Republic** were focused on treated wastewater effluents from hospitals. Occurrence of pharmaceuticals was studied in wastewater along with their interaction with shallow aquifers. PPCPs were also monitored in drinking water supply systems.

German Federal Environmental Agency (UBA) currently lists 339 pharmaceuticals that are regularly being assessed at different sites even though they are not subject to any legal specifications yet. Special attention is given to water-endangering compounds presenting potential negative effects on the quality of the surrounding water bodies. PPCPs classified as water-endangering are e.g. Atorvastatin, Bezafibrate, Bisfenol, Butylparaben, Caffeine, Carbamazepine, Climbazole, Cyclamate, Cyclophosphamide, Ethylparaben, Iopamidol, Ketoprofen, Methylparaben, Metoprolol, Telmisartan and Triclosan.

In **Italy**, research and monitoring has been most intensively performed on pesticides and pharmaceuticals. Research on pesticide pollution in surface and groundwater is conducted by research institutes, national environmental Institutes and Regional Offices and River Basin District Authorities:

- Since 1997, 13 italian research works have investigated the occurrence of industrials in both surface and ground water.
- To date, 15 studies investigating the occurrence of 67 pharmaceuticals in surface water and ground water have been published between 2000 and 2013.
- Since 80'-90', in the Po river basin endocrine disruptors have been discovered and analyzed through fishes sampling and investigation.
- In 2013, PFAS have been detected in drinking water so monitoring of several PFAS in groundwater and surface water has been performed.
- Several research and public sector surveys have focused on substances such as PBDE, PPCP and pharmaceutical.

Polish EC research and monitoring is conducted in the framework of State Environmental Monitoring (SEM) where 60 organic compounds are being assessed in groundwater bodies. More scientific research work regarding ECs was performed in surface water than in sea and groundwater. The following types of ECs were present in different environments:

- pharmaceuticals, pesticides, polycyclic aromatic hydrocarbons, adsorbable organic halides and polychlorinated biphenyls in rivers and lakes,

- pharmaceuticals, pesticides, trichloroethylene and tetrachloroethylene in groundwater,
- antibiotics in sea water

According to the **Slovenian** national monitoring the most common substances yearly between 2014 and 2019 were caffeine, carbamazepine and sulfamethoxazole. Active and passive sampling with active carbon is performed in the country. Qualitative passive sampling method for the detection of a wide range of organic compounds was used in groundwater by gas chromatography mass spectrometry in a couple of slovenian aquifers identifying 1,3-dimethyl-2,4,5-trioxoimidazolidine, atrazine and tetrachloroethene as well as typical benzotriazoles. A parallel analysis of passive samples and quantitative chemical analysis in a Slovenian aquifer detected 382 organic compounds (unknown, urban, pesticides, solvents and other) and determined certain transport properties of selected pollutants (propyphenazone, caffeine and carbamazepine). Other EC research performed in Slovenia included the following:

- anticancer drug residues cyclophosphamide and ifosfamide were analysed in wastewaters by grab and time proportional sampling,
- spatio-temporal variability of 48 EC compounds was analysed in WW and SW including caffeine, carbamazepine, UV-filters, preservatives, diclofenac,
- CECs were analysed in effluents of WWTPs as well as surface water with analyses made with SPE procedure with caffeine being the most abundant in both matrices and several bisphenols (B and E in WW and AP, CL2, P and Z in SW) were quantified for the first time in Europe.

A review of analytical EC identification in 2019 (Galindo-Miranda et al. 2019) showed that the techniques mostly used for extraction seem to be SPME and SPE, while out of chromatographic techniques, LC and GC seem to be most used. UHPLC has also recently showed to be highly efficient. For identification, mass spectrometry techniques are most sensitive but photodiode array detector (PAD) has also been used. In the EU watch list (Decision 2018/840), the following extraction and analytical methods are provided that differ according to the substance or group: LLE — liquid liquid extraction, SPE — solid-phase extraction, GC-MS — Gas chromatography-mass spectrometry, LC-MS-MS — Liquid chromatography (tandem) triple quadrupole mass spectrometry. Passive sampling monitoring targets time-average samples and is less expensive, but a validation procedure including sampling uncertainty evaluation still presents a challenge. The passive sampling method has proved to be an elegant solution for the monitoring of complex aquifers with different influences on the groundwater quality (Auersperger et al., 2016).

Finally, the reports on current state-of-the art of ECs are presented in various conferences and journals all around the world. For example, every 4 years, Polytechnic of Milan hosts a conference on ECs. Other meetings and conferences specifically on the subject of ECs are: Emerging Contaminants Summit, Emerging Contaminants in the Environments Conference, but ECs are among the subject topics in many other conferences covering several environmental sciences. Similarly, ECs appear as topics of special issue journals while new journals dealing specifically with ECs have been established as well, such as the journal Emerging Contaminants, which started in 2015.

4. Monitoring strategies and approaches

The main objective in this chapter is to synthesize information, identify similarities and differences on analytical techniques and monitoring strategies and approaches provided from all participating countries in the boDEREC-CE project. The overview is gathered in transnational report "*Transnational review on state-of-the-art in monitoring*" which was prepared from seven national reports. National reports ware created by using the questionnaire as a guideline for describing the main topics and they were encouraged to use both national and transnational reference sources. The data reported in all of the national reports cites a lot of different literature sources, including applicable legal acts, national and international standards, monitoring reports as well as scientific papers, online sources, published by researches and institutions and other entities dealing with environmental issues. Additional information was also collected during meetings with stakeholders, representatives of regional or local authorities, institutions and research entities.

4.1. Official state monitoring of ECs present in countries

In the project partner countries, more or less systematically monitor the quality of surface and groundwater at national level. It varies from country to country, depending on the different aquatic environments and the range of ECs controlled parameters. The monitoring program follows the requirements of the EU Water Directive. Since 2015, a watch list has been established at European Union level.

In **Austria** among the substances to be analyzed and monitored under the monitoring program "Water quality of Austria" ("Wassergüte in Österreich", BMLRT), the emerging contaminants are not specifically included, however priority substances listed in the Water framework directive and many pesticides are included. During the past decades, some monitoring or research studies concerning the presence of pharmaceuticals and pesticides in groundwater mainly and in drinking and waste- waters were performed by governmental institutions, such as the Federal Environmental agency (Umweltbudesamt), Federal ministry of social affairs, health, care and consumer protection (Bundesministerium für Soziales, Gesundheit, Pflege und Konsumentenschutz) and by the Federal Ministery of Agriculture, Regions and Tourisms (Bundesministerium für Öandwirtschaft, Regionen und Tourismus). However, they do not represent any official state of the art monitoring for emerging contaminants within the Austrian environment. Within the operational monitoring, two measurements per year are planned according to the legislation in the selected monitoring points in Austria, and four times per year measurements are performed at the non-good status groundwater bodies.

In **Croatia** the national reports on monitoring of the surface and groundwater are prepared each year (Musić et al., 2011); however, they are not publicly available and the extent of ECs monitoring is not clear. According to the Monitoring Compliance Program (Program usklađenja monitoringa, Croatian Waters, 2016) monitoring of the ECs substances from the Watch list (EU Decision 495/2015) was planned for 2016. According to the recent paper published by the experts from Croatian waters, monitoring of the parameters from the Decision 840/2018 is going on 2019 in Croatia (Bujas et al., 2019).

In the **Czech Republic**, the Czech Hydrometeorological Institute (CHI) carries out a long-term, systematic monitoring of surface and groundwater quality on a nationwide scale of the territory of the Czech Republic. Since 2015 also substances from the so called Watch list are gradually implemented to the national monitoring program. The Monitoring program framework does not specify exactly which EC has to be monitored and where. However, in general, surface water which is affected by EC should be monitored. ECs in groundwater are not monitored systematically.

In **Germany** does not exist an official state monitoring of ECs due to the fact that monitoring falls in the responsibility of every particular federal state of Germany (BMJV 2019a). Every state, however, has established its own monitoring. Although not being legally required (except for any ECs enlisted on the

Watch List, priority substances, water body specific substances etc.), all states perform monitoring of ECs and have contributed to national studies (Bergmann, Fohrmann and Weber 2011). However, state monitoring varies in many important aspects (how often measurements are taken, what compounds are addressed and measured, from what matrices samples are taken etc.). Other ECs, e.g. microplastics, are targeted in research projects carried out by federal environmental agencies as well as private institutions and universities which usually comprise entire regions and are then evaluated on a national level (Heß 2018).

In **Italy** according to the definition of ECs taken from the NORMAN network for surface water and groundwater the ECs are not monitored except for the following PFAS: PFPeA, PFHxA, PFBS, PFOA, PFOS. About drinking groundwater, subject to a specific site monitoring, the operators also monitor some ECs. In Italy, monitoring of Watch List have been started in 2015. The monitoring plan has been set within the National technical group involving all the Environmental Agencies, coordinated by ISPRA (National Institute for Environmental protection and Research, 2017).

In **Poland** the occurrence of selected groups of ECs in water environment is systematically observed as part of surface water monitoring (inland waters, transitional and coastal waters), groundwater quality monitoring, and Baltic Sea monitoring. Among ECs groups that are investigated in water environment in Poland there is no regular monitoring of PPCPs. The scope of the ECs determination varies for individual aquatic environments and is subject to changes during its conduct, adapting to Polish and European strategic documents relating to the environment.

In **Slovenia**, the Environmental Agency of the Republic of Slovenia is responsible for the monitoring and monitoring of some ECs within the framework of the Monitoring program for the chemical and ecological status of water.

4.2. Objectives of the ECs monitoring

The aim of ECs monitoring is to carry out investigative monitoring, which in most countries also involves the surveillance of ECs pollutants and operational management based on established facts (Figure 7).

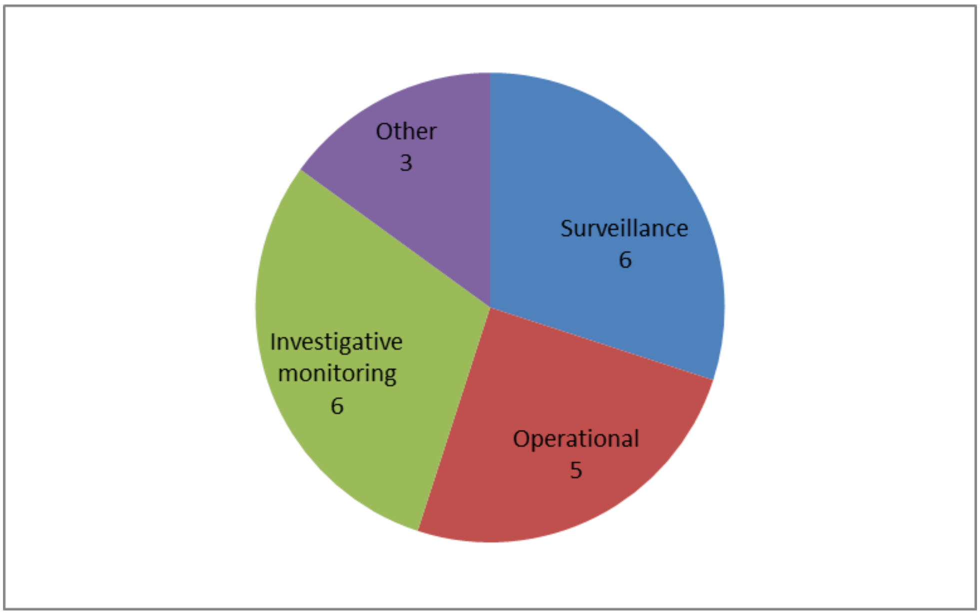

Figure 7: Pie chart of ECs monitoring objectives

In **Austria**, surveillance and operational management are used for river basin management, and operational management is primarily performed for water bodies which are at risk of missing the good quality status. Both are covered by the monitoring program "Water quality in Austria" (Federal Ministry of Agriculture, Regions and Tourisms). In **Slovenia**, investigative monitoring is conducted only on surface waters. In **Germany**, ECs monitoring is also aimed at conducting research, and in Poland monitoring of protected areas (eutrophication endangered areas, recreation areas, drinking water supply areas, Natura 2000 sites). The purpose of the research is to acquire the knowledge necessary to take action to improve the state and protect water against pollution.

4.3. Monitoring of ECs performed for scientific purpose

Monitoring of ECs, which is carried out for scientific purposes and operates in different projects or studies, is ongoing in all project partner countries.

In **Austria** in the past decade, several monitoring and research studies were performed in order to obtain information concerning the presence of pharmaceuticals and pesticides in the water bodies. Some of the research studies are SCHTURM, "Monitoring of pharmaceuticals and wastewater indicators in ground and drinking water", Pharmaqua and published in the paper from Zoboli et al. 2019. Within the project SCHTURM ("Spurenstoffemissionen aus Siedlungsgebieten und von Verkehrsflächen", 2014; BMLFUW) the presence of selected micro pollutants in different emission pathways was investigated. The project "Monitoring of pharmaceuticals and wastewater indicators in ground and drinking water" consisted of the monitoring of selected ground- and drinking water measuring points through analyzing selected pharmaceuticals (in particular antibiotics) and wastewater indicators. The research project presented by Zoboli et al. aims to quantify the presence of selected micropollutants in water bodies in Austria, on one hand in order to create a database for future monitor and management strategies, and on the other hand to have a better insight concerning the behavior of micropollutants in the aquatic environment. The project Pharmaqua sampled and analyzed wastewater influents, not with a monitoring purpose, but only to collect data for the further development of treatment possibilities of micro pollutants with combination of membrane and activated carbon.

In **Croatia** several scientific projects were related to the ECs:

1. Assessment of Hazardous Chemical Contamination in the Sava River Basin, 2007-2010, IRB;

2. "Reduction of Environmental Risks, posed by emerging Contaminants, through advanced treatment of Municipal and industrial Wastes – EMCO", 2004-2007, IRB;

3. Innovative biodiagnosis meets chemical structure elucidation-Novel tools in the effect-directed analysis to support the identification and monitoring of emerging toxicants on the European scale EDA-EMERGE, 2011-2015, IRB ;

4. Comprehensive assessment of the environmental behavior and fate of pharmaceutically active contaminants: macrolide antibiotics and opioid analgesics – COMPASS, 2015-2019, IRB;

5. Environmental Implications of the Application of Nanomaterials in Water Purification Technologies – NANOWAP, 2014-2017, FKIT;

6. The fate of the environmental pharmacists during the advanced water treatment processes - PharmaFate, 2015-2019, FKIT,

7. Study of the effect of the pharmaceutical industries wastewater on the composition and the profile of antibiotic resistance of exposed mycobacterial communities in freshwater sediments, 2014-2017, IRB,

8. Recently, there are several more projects dealing with the technology issues: "Modelling of Environmental Aspects of Advanced Water Treatment for Degradation of Priority Pollutants (MEAoWT), FKIT, 2015-2019.

In the **Czech Republic**, 5 projects are described. In the first they studied pharmaceuticals in drinking water (Kožíšek et al., 2013), in the second they monitored organic micropollutants, including pesticides and drugs (Maršík et al., 2016), in the third they contained PPCP substances in two water reservoirs (Hrkal et al., 2018), in the fourth and fifth they dealt with the occurrence of pharmaceuticals in groundwater (Hrkal et al., 2018; Rozman et al., 2017).

Also in **Germany**, sampling and monitoring of ECs in the aquatic environment have been carried out for scientific purposes. Research projects concerning ECs addressed varying issues. In Bavaria, for example, research projects addressed the removal efficiencies and elimination processes of pharmaceuticals for which waste water treatment in- and effluents were monitored.

In **Italy**, the most important issues in the ECs are science issues such as: compounds to monitor; sampling strategies; measure accuracy; uncertainty of expected concentration compared to consumptions; prioritization of compounds; removal efficiency of treatment plants; promising technologies for removing key pollutants; management of wastewaters from hospitals; environmental risk assessment of pharmaceuticals residues; antibiotic resistant bacteria and effectiveness of measures for reducing them. In Italy, various professions and institutions are linked to these issues. They are also involved in some European projects: "MAT4TREAT" (which considers the management aspect and has produced low-cost innovative solutions for the removal of organic ECs), "MOTREM" (which aims to provide new technologies for water treatment and improve existing ones through the development of integrated processes for monitoring and treating EC in the current waterline of municipal wastewater treatment plants), WP JPI (the aim is to be able to reuse the treated water, after having reduced to a minimum, with new assessment and management methods, the risks deriving from ECs biological pollutants.)

In **Poland** the monitoring of ECs performed for scientific purpose was carried out by Polish Geological Institute – National Research Institute, in the framework of chemical status monitoring and as a part of State Environmental Monitoring. In the years 2009-2014 determination of pesticides was conducted for groundwater samples from 708 measurement points in Poland. Pharmaceuticals were a subject of the monitoring program performed in 2017.

In **Slovenia** in the past years, several scientific and research projects were carried out in order to obtain information about the occurrences of ECs in water environment. Some examples: samples from several boreholes and piezometers in the area of Ljubljansko polje and Ljubljansko barje were collected to present the distribution of caffeine, carbamazepine and propyphenazone (Jamnik et al, 2009); wastewater samples from five Slovene hospitals were samples with additional samples collected from wastewater treatment plants influents and effluents where connection is possible (Česen et al., 2015); passive samplers were built in 19 piezometers in Drava field and were left there four times for six months to detect organic pollutants. Besides qualitative results from passive sampler, active sampling for certain compounds was performed for quantitative analyses (Koroša et al., 2017); several samples were collected from nine Slovene wastewater treatment plants and receiving surface waters (rivers) to analyse seasonal and spatial variations in the occurrence, mass loadings and removal of compounds of emerging concerns (Česen et al., 2018); samples from three Slovenian and three Croatian wastewater treatment plants were collected as 24 h time-proportional samples, as well as seven samples from River Sava prior or after the wastewater discharges as grab samples (Česen et al., 2019); a study was published a study, where they sampled and analysed waste water on 10 wastewater treatment plants, five surface water samples and 15 samples of drinking water from tap water (Trontelj et al., 2018).

4.4. Water bodies to be monitored

In general, groundwater and surface water are monitored, sampled and analysed, especially those intended for drinking water supply. The manner, scope and institution that conducts the monitoring varies from country to country. Countries have a network of monitoring stations in place, and at the same time they carry out monitoring as part of individual research projects. The monitoring network covers those areas with higher environmental risk (landfills, wastewater treatment plants, high population density, tourism, industry, etc.).

4.5. Sampling locations of ECs

Most project partners sample ECs most commonly in flowing water, followed by groundwater (observation boreholes and water supply wells) and standing water bodies (natural and artificial). Sampling of ECs at springs is only carried out in Italy and Slovenia (Figure 8).

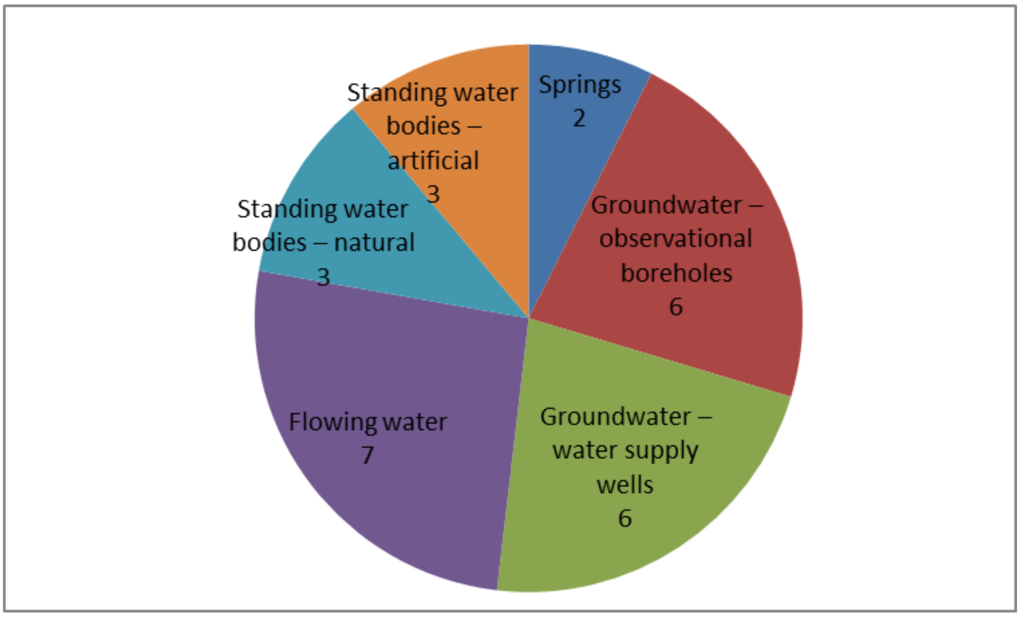

Figure 8: Pie chart of the most common sampling locations of ECs

4.6. General natural conditions for water body were monitoring is performed

In **Austria**, water bodies were divided into groundwater and surface water bodies. The total groundwater bodies monitored were 138, including 63 near-surface isolated pores groundwater bodies, 66 near surface groundwater bodies, and 9 deep groundwater bodies. There are 2000 measuring points on groundwater bodies. Nationally, a quarter of the measuring points are located in populated areas and a fifth in forested or natural areas. Some measuring points are also located in karstic areas (central and northern alpine limestone areas), and in agriculture areas (which percentage varies in each federal state). Surface water bodies are divided into streaming and standing water bodies (lakes). The total number of measuring points on streaming water bodies is 100. 31 of these representing significant cross-border surface water bodies with catchment area > 2500 km2, 9 of these points are referential and are located in water bodies which are only slightly influenced by anthropogenic activities and 60 measuring points are located in tributaries to large rivers and in regions characterized by intensive agriculture use and tourism. There are 33 measuring points on 28 natural standing bodies of water.

In the **Czech Republic**, they have groundwater monitoring points in porous, fissured rocks and combined types of aquifer. Scientific monitoring is generally performed in shallow aquifers and state monitoring network includes also deep monitoring boreholes. Generaly forests, agriculture, industry are located in the recharge area. The surface water body type are rivers, which are monitored across the country.

Germany has described 9 water bodies: Ampere close to Inkhofen, Franconian Rezat at the "Spiegelmühle", Ebrach close to Ebersberg, Würm close to Dachau, Isar bei Plattling / Deggendorf, Danube close to Bad Abbach, Main close to Marktheidenfeld, Loisach close to Burgau, Dresden-Hosterwitz. The largest of these is Dresden-Hosterwitz. Its recharge area covers 50,000 km2. Land use is primarily forestry, agriculture and urban settlements. The surface water body in this area is represented by the Elba River with a medium flow of 331 m3 / s. The type of aquifer is porous, the depth to groundwater is 6-9 m, the thickness of the aquifer is 15 m.

In **Italy**, they focused on the water body of the Po river where the case study of Pontelagoscuro is located. Yearly mean discharge is 1500 m3 / s and recharge area is 74,000 km2. In the recharge area there are 45% forest and grassland, 46% agriculture, 7% urban and industrial area, 2% wetland and water bodies.

Slovenian pilot area Ljubljanska kotlina encompasses a large part of central **Slovenia**. It is also the most populated area in Slovenia, with the capital of the Republic of Slovenia and many other cities. The largest share of more than half of the entire Ljubljana basin is occupied by agricultural areas. They are followed by forest and semi-natural areas and artificial surfaces, where each class occupies a fifth of the pilot area. According to the guidelines following the Water framework directive, Slovenia is divided into 21 groundwater bodies. The pilot area is positioned in the same area as the groundwater body Savska kotlina – Ljubljansko barje (SIVTPODV1001 – Sava basin and Ljubljana Marsh). Inside of the groundwater body several aquifers are defined, which are predominantly intergranular. In the northern and central part of groundwater body, Quaternary intergranular aquifer consisting of sandy gravel deposits of the Sava River and its surface tributaries predominates. Aquifer is extensive with medium to high yield. Medium aquifer thickness is 55 m (Priloga 2 - Ur.l. RS, 63/05). Second important aquifer is positioned in Mesozoic carbonate rocks as a karstic-fissured aquifer. It is positioned mainly on the rims of the groundwater body and in some parts extends bellow the Quaternary aquifer. It is extensive, however locally bounded with faults and other hydrogeological barriers. Consequently, it has varying yield, from low to high. Medium aquifer thickness is > 60 m (Priloga 2 - Ur.l. RS, 63/05). The discharge characteristics of the Sava River in the upper part (north of Radovljica) of the pilot area indicate an Alpine nival-pluvial regime. In the Ljubljana basin, the Sava River and its tributaries' discharge regime changes to Alpine pluvial-nival and remains the same throughout the rest of the flow in Slovenia. The average discharge is 40 m3/s in the upstream part of PA (Radovljica gauging station) and 85 m3/s in the downstream part. Ljubljansko polje is the most important unconfined intergranular aquifer in the basin and a drinking water source for the capital of Slovenia and its surroundings. The phreatic groundwater recharges from rainfall, from the Sava River and via groundwater inflow from Ljubljansko barje multi aquifer system from the south.

4.7. Performing ECs monitoring on the recharge zones of drinking water resources and drinking water protection zones

In most project partner countries, monitoring is carried out on the recharge zones of drinking zones and / or drinking water protection zones as well as outside these areas. In Austria, some measuring points are located in groundwater wells used for water supply.

4.8. Expectations of the monitoring program

As ECs monitoring is not yet fully regulated, project partners expect the program to provide comprehensive, ongoing monitoring of water status and quality. Most want to create a database that will allow them to know the most important substances found in the waters, learn about the sources, routes and behaviour of detected ECs, control pollution levels and act on pollution, improve water treatment methods, identify seasonal and long-term changes in ECs concentrations and drinking water safety.

4.9. Analysed ECs substances (groups)

In **Austria** concerning the monitoring program “Water quality in Austria” the scope of the monitoring is divided among ground and surface water bodies.

Concerning groundwater, a total of 129 parameters are included in the monitoring program (GZÜV), divided into groups of types of parameters:

Block 1: sampling and on-site parameters, chemical parameters

Block 2: dissolved metals, volatile halogenated carbons, pesticides

Concerning the streaming water bodies, the parameters included in the monitoring plan are the following: physico-chemical parameters, non-synthetic and synthetic pollutants (priority substances), biological parameters (pythobenthos, macrozoobenthos, fish, macrophytes, plankton), hydromorphological parameters (flow, morphology).

Concerning the standing water bodies, the parameters included in the monitoring plan are the following: physico-chemical parameters, non-synthetic and synthetic pollutants (priority substances) for sediment, biological parameters (pythoplancton, fish, macrophytes), hydromorphological parameters (water level, water supply, morphology).

Also under this program, an analysis of selected priority substances in sediments and biota in five selected measuring points along rivers in Austria was carried out. The substances monitored were selected form the updated list of priority substances (2013/39/EU).

Concerning the project “Monitoring program of pharmaceuticals and wastewater indicators in groundwater and drinking water” 18 parameters were analysed in the samples collected: tetracycline (doxycycline), sulphonamide (Acetyl-Sulfamethazin, Acetyl-Sulfamethoxazol, Sulfadimethoxin, Sulfamerazin, Sulfamethoxypyridazin, Sulfamonomethoxin, Sulfaquinoxalin, Sulfisoxazol), macrolide (erythromycin-anhydro, spiramycin, tylosin), quinolone/gyrase inhibitors (danofloxacin, flumequin, nalidixin acid, oxolin acid, sarafloxacin), lincosamide (lincomycin). In the framework of this project, waste water indicators were measured as well: acesulfam, sucralose, benzotriazol, tolyltriazol, carbamazepine, 10,11-dihydro-10,11-Dihydrocarbamazepine, Sotalol and metopropol.

Pharmaceuticals are analysed in **Croatia**: 15 sulfonamides, 2 macrolides and 2 aromatic antibiotics along 20 rivers in Croatia in Danube and Adriatic catchment area.

Czech Republic analyses pesticides and PPCPs, which are further divided on pharmaceuticals and their metabolites, hormones and antibiotics.

In **Germany** several different groups of ECs are targeted in monitoring programs and research projects by federal environmental agencies. A particularly well-researched group is pharmaceuticals. Antibiotics and natural and synthetic hormones are of concern. Anti-inflammatory drugs and beta-blockers are also being explored (Lfu 2015). The next group is biocides and pesticides as well as microplastics (Lfu 2019b).

In **Italy** the ECs groups analysed are: pfas, hormones (estrone (e1); β-estradiol (e2); estriol (e3); 17-α-ethynylestradiol (ee2), bisphenol a (bpa); 4-n-nonylphenol (np); 4-octylphenol (op); 4-t-octylphenol (t-op), phenols (bisphenol a (bpa); 4-n-nonylphenol (np); 4-octylphenol (op); 4-t-octylphenol (t-op)), atrazine, drugs (carbamazepine, cefazolin, diclofenac, paracetamol, erythromycin, clarithromycin, clofibric acid, diatrizoid acid, primidon, plant protection products: desetil-desisopropil-atrazina (dact), simazina, atrazine, propazine, terbutylazine, desetil-atrazine, desetil-terbutylazine, linuron, diuron, fludioxonil, metalaxyl, boscalid, dimethomorph, bentazone. In particular for the monitoring programme of the Watch

List (Ispra, 2017) and for the National Programme of the monitoring of the “Extremely worrying” substances (SNPA, 2018) ECs may be grouped according to their use: nanomaterials, pesticides, industrials additives and by-products, etc.

In **Poland**, the monitoring of the ecological and chemical status of surface water bodies mainly analyzes pesticides, insecticides, fungicides, chemical parameters, etc., such as: alachlor, anthracene, atrazine, benzene, chlorfenvinphos, chlorpyriphos, diuron, endosulfan, hexachlorobenzene (HCB), hexachlorocyclohexane (HCH), isoproturon, pentachlorobenzene, pentachlorophenol, polycyclic and hydrocarbons, aromatic sumazine, tributyltin compounds, trichlorobenzenes (TCB), trichloromethane, trifluralin, dicofol, quinoxyfen, aklonifen, bifenox, cybutryne, cypermethrin, dichlorphos, heptachlor and heptachlor epoxide, terbuthrin, dezinine as well as aldrine, para,para-DDT and total DDT. Some substances should be determined monthly or at least once a year.

In **Slovenia** priority compounds in surface water are: metals, phytopharmaceuticals, biocides, industrial chemicals and solvents. Some other compounds were also added to monitoring programs e.g. steroid hormones, some newer insecticides, antibiotics, other pharmaceuticals, cosmetic ingredients and herbicide. In groundwater monitoring program included: industrial chemicals and solvents, pharmaceuticals.

4.10. Institutions responsible for the interpretation of the ECs monitoring results

Certain institutions with their professional staff are responsible for interpreting the ECs monitoring results in all project partner countries. Quality status is mostly taken care of by water management institutions. Ecotoxicological issues are addressed by water management or environmental institutions, public health institutes and the Ministry of the Environment. Human health issues are in charge of public health institutes and sanitary inspections.

4.11. Criteria for sampling locations

4.11.1. Guidelines/regulations used for ECs sample collection

There are no precise regulations for the sampling of ECs in surface and groundwater. Thus, international or national sampling guidelines / regulations are applied in each project partner country. Some countries follow the same international regulations, some apply their national guidelines.

Austria uses a national guideline to sample. For the sampling of groundwater and drinking water, they take into account the “Ordinance for monitoring of the water bodies status” (GZÜV). ÖNORM M guidelines are followed for sampling systems of wastewater related samples. **Croatia**, the **Czech Republic** and partly **Germany** and **Italy** use international standards ISO 17025 and / or ISO 5667. In Germany, monitoring is based mainly on the national standard that generally governs the sampling and analysis method, DIN 38407-47 (LfU 2015). More details on regulations concerning the exact procedures are given in other regulations and guidelines, the DIN 38407-47 refers to. For example, information about the extraction of samples from water bodies is given in DIN 38402, for groundwater aquifers e.g. DIN 38402-13. When sampling standing surface water bodies, the national standard DIN 38402-12 is applied. Italy also uses Ispra / SNPA guidelines / regulation for sample collection. Moreover, it is important to consider UNICHIM Manual 157 and Document EPA/540/S-95/504 April 1996. In **Poland** there is currently no standardized practice or protocol for the sampling and analytical determination of pharmaceuticals in water or any other environmental media that ensures the comparability and quality of the data generated. Several ISO standard from series 5667 Water quality – Sampling are also most commonly used for sample collection in **Slovenia**. Samples of watercourses should be taken at a depth of 0.5 m. For waters shallower than 1 m, samples should be taken at half depth. Groundwater is sampled from wells approximately 1 m below groundwater level or where filters are and in the case of shallower wells, on the half of the water column. If the height of the water column is less than 0.5 m, groundwater sampling shall not be carried out.

4.11.2. Official national standards on the EC sampling

There are no official national standards for sampling ECs, but national guidelines may nevertheless be followed. The exception is Italy, which has the SNPA Guidelines n. 7/2018 and n. 13/2018 which contains indications and the National standards on ECs sampling.

In **Austria**, sampling guidelines for wastewater, for surface water bodies for water matrix and for groundwater bodies from the Ordinance water methodology are used (Methodenverordnung Wasser, MVW, 2019). These sampling guidelines are:

- for wastewater:
  - DIN 19559-1 1983-07-01
  - DIN 38402-11 (DEV A 11) 2009-02-01
  - DIN 38402-30 (DEV A 30) a) 1998-07-01
  - ÖNORM EN 16479 2014-10-01
  - ÖNORM EN ISO 5667-1 2007-04-01
  - ÖNORM EN ISO 5667-16 2018-02-01
  - ÖNORM EN ISO 5667-3 2013-04-15
  - ÖNORM EN ISO 6817 1996-04-01
  - ÖNORM M 5880 1998-02-01
  - ÖNORM M 6258 1992-01-01
  - VDI/VDE 2642 1996-12-01
- for surface water bodies for water matrix:
  - ÖNORM EN ISO 5667-1 2007-04-01
  - ÖNORM EN ISO 5667-14 2016-11-15
  - ÖNORM EN ISO 5667-3 2013-04-15
  - ÖNORM EN ISO 5667-6 2017-01-15
  - ÖNORM ISO 5667-4 2015-01-01
- for groundwater bodies:
  - ISO 5667-11 2009-04-15
  - ÖNORM EN ISO 5667-1 2007-04-01
  - ÖNORM EN ISO 5667-14 2016-11-15
  - ÖNORM EN ISO 5667-3 2013-04-15

Croatia applies the guidelines HRN ISO 5667-11, HRN ISO 5667-6, HRN ISO 5667-10 for groundwater sampling, for sampling on rivers / streams and for waste water sampling. In the **Czech Republic**, sampling is officially standardized only by ČSN EN ISO / IEC 17025. In **Germany**, DIN 38407 applies to the examination of water, waste water and sludge. Part 47 of the DIN deals with the determination of selected active pharmaceutical ingredients and other organic substances in water and waste water. The sampling of non-liquid matrices is accounted for in other national standards e.g. to DIN 19747. In **Slovenia** the only sampling ISO standards are for passive sampling - SIST ISO 5667-23: 2011.

4.11.3. Discharges with potential ECs in the vicinity of the sampling locations

6 project partners confirm the presence of discharges containing potential ECs in the water near the sampling points. The exception is **Poland**, which has not provided an answer, as there are no official requirements for monitoring them.

In **Austria**, according to the monitoring program “Water quality in Austria”, about half of groundwater measuring points are located in agricultural areas, but this value varies within each different federal state. In the **Czech Republic**, healthcare and wastewater treatment plants represent a potential source of ECs and in Italy industrial and agricultural production. **Germany** described Bavaria, where wastewater treatment plants are those that could affect the state of eight rivers by releasing ECs (Lfu 2015). In **Slovenia**, discharges with potential ECs from hospitals, laundries, industry, wastewater treatment plants and landfills are present in the vicinity of some groundwater and surface water quality measuring sites.

4.12. Sampling type and frequency

4.12.1. Monitoring of ECs: who implements it and how often

Monitoring of ECs in project partner countries is carried out by different institutions at different time intervals.

In **Austria** the sampling is taken care by the Federal Ministry of Agriculture, Regions and Tourisms. The measuring frequency is based on a six-year observation monitoring for both ground and surface water bodies. The frequency of monitoring in groundwater, concerning the priority pollutants, is increased. At the end of the six-year period, an evaluation of the observed data is performed, and it is considered as basis for the next following six-year cycle. During the six-year period, the first year is dedicated to initial observation monitoring, and the rest of the five years are dedicated to repetition observation monitoring. The results of the 5-year monitoring will be the basis for the new 6-year cycle. The groundwater bodies measuring points are monitored from one to four times per year. The monitoring frequency for streaming surface water bodies is usually twelve times per year, analysing basic chemical and physical parameters and if necessary additional pollutants. The monitoring of chemical, physical and of phytoplankton in standing water bodies (lakes) is performed usually four times per year. The biological and hydromorphological parameters in streaming and standing water bodies are usually analysed once per year.

In **Croatia** monitoring of the parameters from the Watch list is performed by the accredited laboratories of Croatian Waters or Public health institute Andrija Stampar from Zagreb. According to Ivešić et al. (2017) monitoring of the pharmaceuticals in 20 Croatian rivers was performed once per year in 2012 and 2013 and four or three times per year in 2014.

In the **Czech Republic**, 4 different institutions carry out monitoring frequently. WRI (T.G. Masaryk Water Research Institute) irregularely/monthly, CHMI (Czech Hydrometeorological Institute) quarterly or yearly, NIPH (National Institute of Public Health) two times per year and RBAs monthly.

In **Germany**, official monitoring of any substances including ECs falls within the responsibility of the federal states. Monitoring is carried out by the according environmental agencies which define their monitoring programs, hence, monitoring sites, substances addressed, and the frequency of monitoring. A generalization on how often the monitoring is carried out can therefore not be made. Considering Bavaria, where the pilot action is located, monitoring is performed by the “Bayerische Landesamt für Umwelt”. As part of the strategic monitoring in Bavaria, WTTP effluent samples were taken three to four times a year, the same frequency applies to surface (flowing stream) water bodies (LfU 2015).

In **Italy**, monitoring is taken care of by the regional environmental agencies involved in SNPA and ISPRA. Sampling periods have been defined according to the period of maximum use of the different monitored substances (winter for antibiotics and anti-inflammatory, spring-summer for pesticides) and in order to synergize with the current monitoring of water bodies (at least one time per year). The monitoring plan works on a period of at least four years. According to monitoring plan of extremely worrying substances set by SNPA (SNPA, 2018) sampling frequency will be monthly for surface waters while for ground water a single sampling will be enough (as foreseen by the WFD). The monitoring period will be totally six months.

In **Poland** regional Inspectorate for Environmental Protection is responsible for monitoring of surface water, and in justified cases – the Chief Inspectorate for Environmental Protection. Diagnostic monitoring of surface water bodies is carried out in annual cycles, with a frequency of not less than every 6 years. As part of the monitoring, the frequency of analyses of a particular indicator throughout the year varies depending on the type of water investigated (rivers, lakes, transitional waters, coastal waters) and ranges from 1 to 12 times a year. Monitoring of substances particularly harmful to the aquatic environment included in the watch list is carried out for a period of at least 12 months for at least 15 representative control points. The Chief Inspectorate for Environmental Protection and the Polish Geological Institute - National Research Institute - State Hydrogeology Survey are responsible for groundwater monitoring. Groundwater samplings under operational monitoring are carried out at least once a year (2 samplings), excluding the year in which diagnostic monitoring of the chemical status of groundwater bodies is carried out.

In **Slovenia** the monitoring program is performed by Slovenian Environment Agency. For surface waters, the sampling frequency is 12 times per year for chemical parameters. For the priority and priority hazardous substances for surveillance monitoring the sampling frequency is 12 times per year and for the priority and priority hazardous substances for the operational monitoring it is 4 to 12 times per year. The frequency for groundwater sampling is 1-2 times per year and is determined based on legal regulations and recommendations, on the results of risk analysis, analysis of monitoring results at individual sites for a 5-year period, also regarding on the data in groundwater protection zones and data on aquifer load and vulnerability.

4.12.2. Spatial representation for any type of monitoring sites

In **Austria**, 54 measuring points for sampling and further analysis have been identified as part of the project "Monitoring of Pharmaceuticals and Wastewater Indicators in Groundwater and Drinking Water". These measuring points were selected to have near a drinking water station, allowing the comparison of ground and drinking waters, in order to observe any influence. The measuring points are spread throughout the Austrian territory.

In **Croatia**, sample sites are selected downstream from the industrial and hospital wastewater discharges. Samples were collected at 23 sampling points (Danube and Adriatic basins) to determine the need for monitoring. 5 locations were selected to monitor the parameters from the Watch list.

In the **Czech Republic**, monitoring studies conducted by CHI, RBAs and NIPH cover the whole territory.

Originally, **Germany** had a large number of monitoring sites in Bavaria for pharmaceuticals including waste water treatment plant effluents, flowing streams as well as groundwater aquifers (Lfu 2002). Because most groundwater aquifers did not contain any residues from pharmaceuticals, they reduced the number of monitoring sites to eight flowing streams and four wastewater treatment plant outlets (Lfu 2015).

There are 25 stations in the whole of **Italy**, which are already included in the current water quality monitoring. According to the monitoring plan set by the SNPA for extremely worrying substances, the position of the measuring stations will be such as to maximize the representativeness and significance of the data collected (SNPA, 2018).

ECs other than PPCPs are regularly monitored in surface and groundwater bodies in **Poland**. The national monitoring network of rivers and dam reservoirs is evenly distributed throughout the country. The same goes for monitoring groundwater bodies.

Within the Program for monitoring the chemical and ecological status of water in **Slovenia** there are 204 monitoring sites for surface waters, of which there are 162 sites in the Danube river basin district and 42 sites in the Adriatic Sea basin district. Surveillance monitoring is implemented on 31 monitoring sites. For groundwater the density of the measuring grid is higher on water bodies with higher anthropogenic pressures, which enables the determination of the chemical state with the highest degree of reliability. In the program 205 measuring locations were determined on all groundwater bodies, but nevertheless, new measuring locations can be added, especially on water bodies, where there are currently fewer measurement sites.

4.12.3. The type of active sampling applied

According to the monitored environmental medium, 5 project partners - Croatia, Czech Republic, Italy, Poland and Slovenia use in situ sampling (water samples in field) for active sampling. However, Austria and Germany collect water samples and then analyse them in the laboratory (Figure 9). Laboratory batch sediment extraction is not conducted in any country.

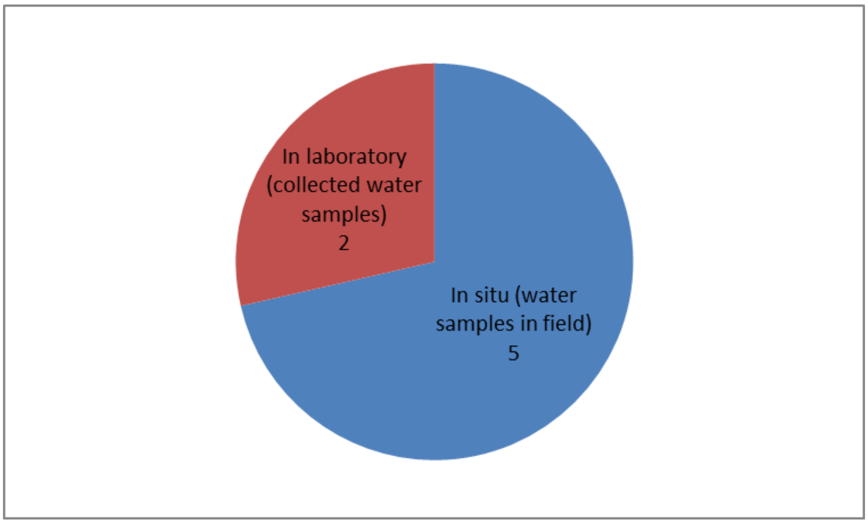

Figure 9: Pie chart of the most common types of active sampling

4.12.4. Time frequency for active sampling

The time frequencies for active sampling vary from country to country. The sampling frequency, for example, depends on the institution conducting the monitoring, on the legislation in each state or on the type of monitoring program.

In **Austria**, the selected groundwater measuring points are monitored from one to four times per year. The streaming surface waters are monitored twelve times per year, and the standing surface waters four times per year ("Water quality in Austria"). According to the Ordinance of wastewater emissions (AAEV 1996) the frequency and sampling intervals depends on the discharge of the wastewater constituents.

Croatia performs active sampling once to 4 times per year.

In the **Czech Republic**, active sampling is also performed at different time frequencies (monthly, quarterly / yearly, 2x per year), depending on the monitoring institution.

In **Germany**, the frequency of active sampling varies between federal states. In Bavaria, three to four times a year was sampled during strategic monitoring of pharmaceutical products. Different frequencies between years occur because sampling was performed in parallel with other regular monitoring programs (Lfu 2015).

In **Italy**, the frequency of active sampling varies from a minimum of one to a maximum of two per year. According to the monitoring plan set by SNPA for extremely worrying substances, time frequency is one per month for surface waters and for ground water one sampling per year (SNPA, 2018).

In **Poland**, the time frequency depends on the type of monitoring program. In diagnostic monitoring of surface water bodies most ECs are sampled every month during one year measuring cycle, performed once per six or three years. In operational monitoring of surface water bodies ECs should be sampled every month during one year measuring cycle, performed every year. In monitoring of protected areas, sampling frequency depends on the number of people supplied with water from the area of interest (4 times per year - number of supplied people is less than 10 000, 12 times per year - number of supplied people exceeds 30 000. In case of monitoring of chemical status of groundwater bodies, priority substances and other ECs are not obligatory for sampling.

In **Slovenia** within the Program for monitoring the chemical and ecological status of water the frequency for active sampling is determined for surface waters (12 times per year for chemical parameters and priority/priority hazardous substances – surveillance monitoring, and for the priority/priority hazardous substances – operational monitoring it is 4 to 12 times per year) and groundwaters (1 to 2 times per year).

4.12.5. Sampling for active sampling

Sampling for active sampling in all countries is done manually by grab sampling or snapshots. In addition, mixed samples are used in Poland and Slovenia and automatic sampling in Austria (Figure 10).

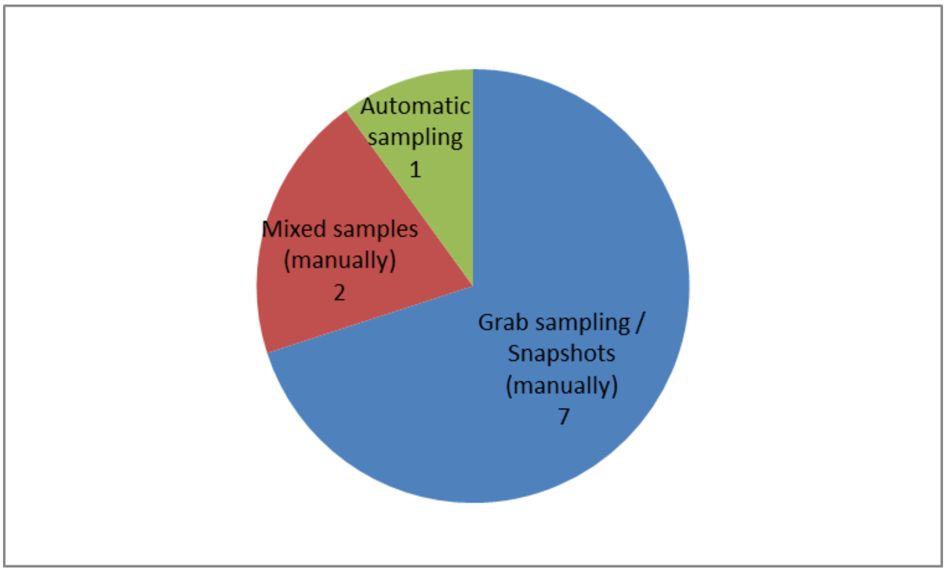

Figure 10: Pie chart of sampling modes for active sampling

4.12.6. Monitoring with passive samplers

Monitoring with passive samplers is only performed in Slovenia and not in other countries (Figure 11). ISO standards and ASTM D7929-14: A standard guide for the selection of passive sampling techniques for groundwater monitoring wells, are used for sampling. In passive sampling, diffusion-based (liquid sampling) passive sampling with active carbon fibers is used for water samples. For the receiving phase of the sampler is used porous adsorbent material – active carbon.

5. Analytical techniques

The chapter analytical techniques is divided four sub-chapters; sample containers and sampling, sample transportation and storage, sample preservation and sample analyses.

5.1. Sample containers and sampling

5.1.1. Type of sample vessels used

All project partners use glass bottles for sampling. In addition, plastic containers are used in the Czech Republic, Italy and Poland, and aluminium bottles are used in Austria (Figure 11).

In Austria sample container for organic trace substances should be made of glass and only in particular cases of plastic, depending on the substances to be analysed. The closuring cap should be of glass or PTFE. For specific substances like endocrine disruptors, metallic vessels (Aluminium) are recommended.

In Croatia, samples are collected in 2.5 L amber glass bottles, in the Czech Republic 60 ml amber glass vials with HDPE screw cup are used and for sediment plastic containers of approximately 500 ml capacity. In Germany, samples are stored in 1 L glass bottles, in Italy in dark glass bottles. Slovenia uses glass bottles with PTFE stop box caps. In Poland for most ECs normal or specific glass bottles are recommended. Usually dark glass is preferred, especially for TCE, PCE, PAHs, BTEX or pesticides. Ground glass stopper is recommended for many ECs. Plastic vessels, however, are acceptable for some ECs like phenols or surfactants. PE and PTFE materials are recommended.

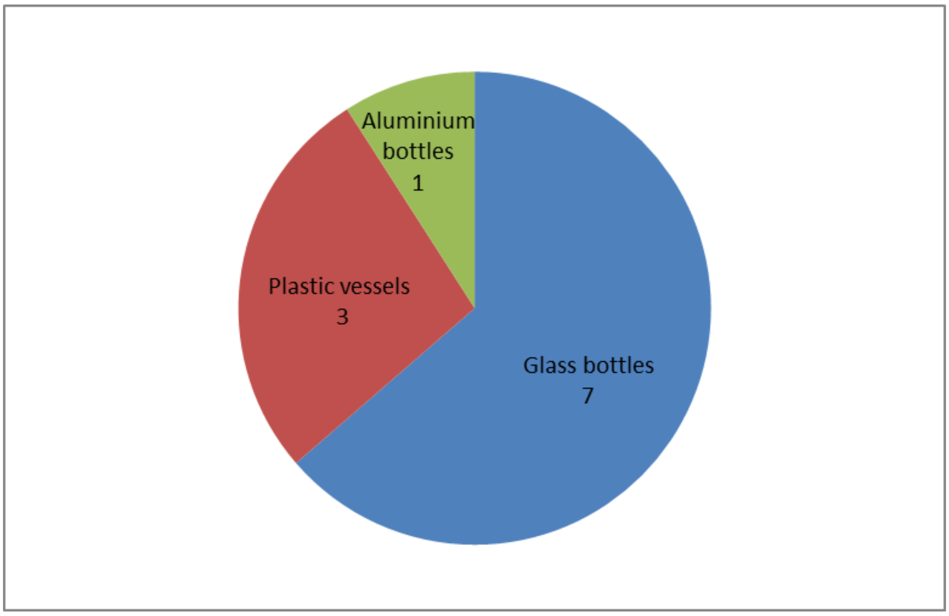

Figure 11: Pie chart of the most common types of sample containers used

5.1.2. Guidelines / regulations used for choosing the right sample containers

Countries follow different guidelines / regulations or recommendations for the selection of sample containers.

In Austria for special sampling the guideline to follow to choose the right material of the sampling vessel are in:

- DIN EN ISO 5667-1 (A4), Preparation of sampling program and sampling techniques (general principles)
- DIN EN ISO 5667-2 (A3), Instructions for sampling techniques (with the aim to obtain water quality data)
- DIN EN ISO 5667-3 (A2), preservation and handling of water samples.

More in specific the guidelines used for the different types of water bodies/water pathways are:

- DIN 38402-11 (A11), Sampling of wastewater (program and techniques for sampling of all wastewaters, raw sewage, industrial, cooling waters for industries, municipal)
- DIN 38402-12 (A12), Sampling of stagnant waters
- DIN 38402-13 (A13), Sampling of aquifers
- DIN ISO 5667-5 (A 14), Sampling of drinking water from treatment plants and canalisation
- DIN 38402-15 (A 15), Sampling of streaming waters
- DIN 38402-17 (A 17), Sampling of precipitations (fluid state)

Croatia uses ISO 5667-3 to select containers. According to the "Monitoring Program for the Chemical and Ecological Status of Water for the Period 2016 to 2021", ISO 5667-3 is also taken into account by Slovenia, in addition to ISO 5667-11 (Groundwater Sampling Guideline) and ISO 5667-6 (River and streams Sampling Guideline). The Czech Republic follows the recommendations of EPA method 1694 and Germany selects containers according to national standard DIN 38407-47. In Italy, the selection of sample containers is in accordance with ISPRA Guideline 173/2018 and SNPA Act n. 25/2018. In Poland, the right sample containers are recommended by laboratories. In addition, they use a manual entitled “Catalog of selected physical and chemical pollution indicators in groundwater and analytical methods” (Witczak et al., 2013), which contains all the detailed information and recommendations for sampling.

5.1.3. Preparation of sample containers

In Croatia and Germany, the sampling container is prepared by washing it with ultrapure / distilled / deionized water, or rinsing with sample water or using an organic solvent. In Poland, the vessel is washed with ultrapure / distilled / deionized water or with sample water, and in the Czech Republic only with ultrapure / distilled / deionized water. In Slovenia, the sampling container is prepared by rinsing with sample water, using organic solvents or by drying at 300 degrees Celsius. In Austria and Italy, other approaches are used to prepare sampling vessels. Austria follows the guideline of DIN EN ISO 5667-1, which suggests that it is necessary to check for possible scratches, signs of wear, and damages of the vessel. It is also necessary to prepare the bottles correctly according to the substance to be analysed, to check their cleanliness. In Italy, acetone, hydrochloric acid (4% or 25%) and nitric acid 10% are generally used to prepare sample vessels (Figure 12).

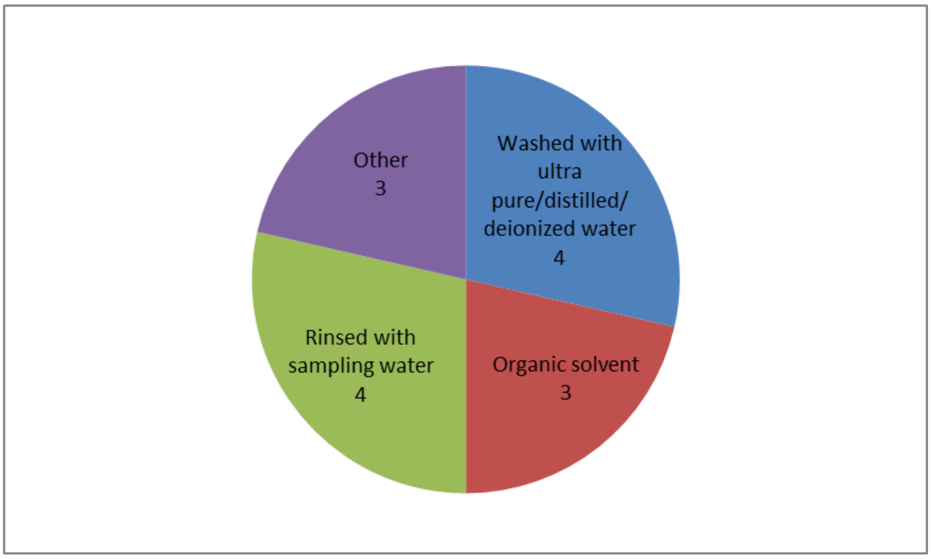

Figure 12: Which kind of preparation of sample containers is performed?

5.1.4. Optimum sample volume needed for analysis

In some countries, the optimum sample volume for analysis is determined in advance, and in some countries the volume varies and depends on the purpose and type of analysis, the method of sample preparation and the method used.

In Croatia, the samples are collected in 2.5 L bottles, in Slovenia the optimum sample volume is 1 L. In the Czech Republic, the ideal sample volume is 2x30 ml. In Germany, according to DIN 38407-47, the optimum sample volume is 250 ml. In Austria, Italy and Poland, the optimal amount required for analysis is not always the same (SNPA, 2018; SNPA, 2019, ONORM). In Austria it depends on the purpose of analysis and on the method of sample preparation. The partial sample volume can be determined by knowing the minimum volume for the analysis, (if possible to predict) the maximum outflow of a precipitation event and therefore the expected outflow volume to be sampled, the duration of a partial sampling. By defining the total volume and the partial volume of the sampling phase, the minimum and maximum number of partial samples is estimated. In Poland, the volume of the sample container is determined by the laboratory after selection of the analysis method. For the determination of most ECs, 1L vessels are usually used and 0.25 L bottles are used for TCE and PCE.

5.1.5. Requirements for filling the containers

In Germany, Italy and Poland, turbulence and air contact in container are avoided when filling the containers. Austria and the Czech Republic have their own filling requirements (Figure 13). In Austria, the following standard sampling procedures are followed. When the freezing (-20°C) of the collected sample is required, the vessels must not be completely filled up, to allow the ice expansion. Otherwise, the containers should be completely filled. The volume of the sample should be as high as possible, in order to avoid higher contamination from the vessel itself. The time of contact between the sample and the (automatic) sampler device should be as short as possible, to avoid contamination.

According to the guidelines DIN EN ISO 5667-1, the following procedure has to be followed in order to avoid contamination:

- Rigidly protect sample bottles from contamination
- Avoid disturbances at the sampling location
- Rinse equipment thoroughly
- Clean the funnel inside and outside after the sampling
- Clean syringe barrels and filter media before use
- Carefully store the plugs and caps
- Do not touch specimens (neither with hands nor with gloves, especially for microbiological sampling)
- Clean all equipment after the sampling
- Place all exhausted gas-producing devices (aggregates) on the side facing away from the wind.
- Inspect each sample for large particles (leaves) and if necessary, take new ones.

In the Czech Republic, containers are filled only to half, due to later freezing in the laboratory. In Slovenia, they keep as little air as possible in the container. In Croatia, the requirements for filling containers are unknown.

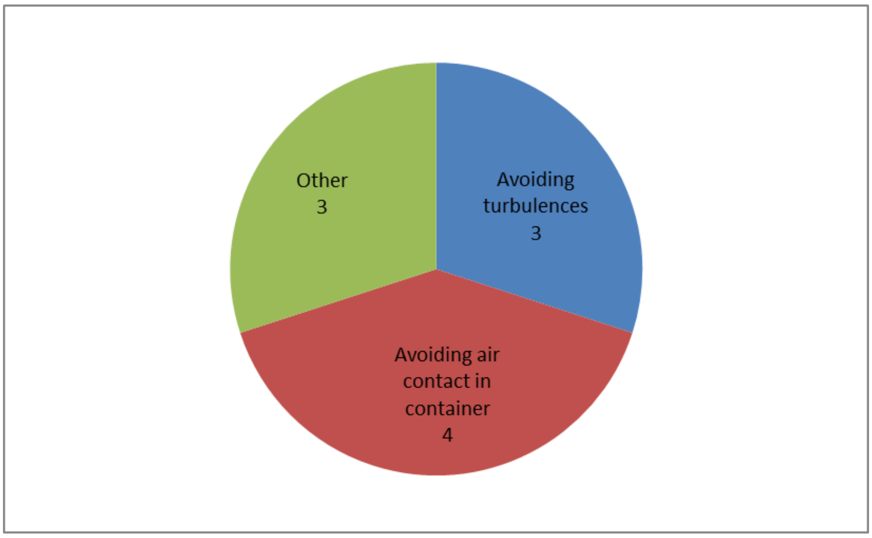

Figure 13: Pie chart of container filling requirements

5.2. Sample transportation and storage

5.2.1. Performing additional sample stabilization / preservation

In all countries the sample is protected from light (Figure 14). In addition, chemicals are added to the samples in Italy and Poland. Austria also adds chemicals to the samples or stabilizes / preserves the samples in the manner agreed with the laboratory performing the analysis. Slovenia also cools the samples at 5 °C ± 3 °C and uses 1 ml of IS solution internal standard.

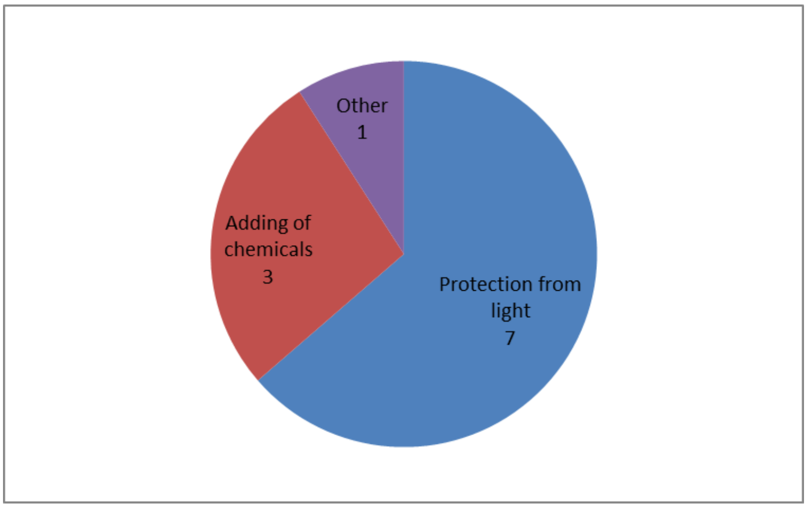

Figure 14: Pie chart of additional measures to stabilize / preserve samples

5.2.2. Conditions for transport of samples to the laboratory

In most countries, the condition for the transport of samples to the laboratory is a reduced temperature, which can reach from 3-8 °C or freezing of samples at -20 °C (Figure 15). The following is the condition of the required time for sample delivery. Typically, samples should be delivered to the laboratory as soon as possible - the same day. Otherwise, the requirements may also change depending on the agreement with the laboratory performing the analysis.

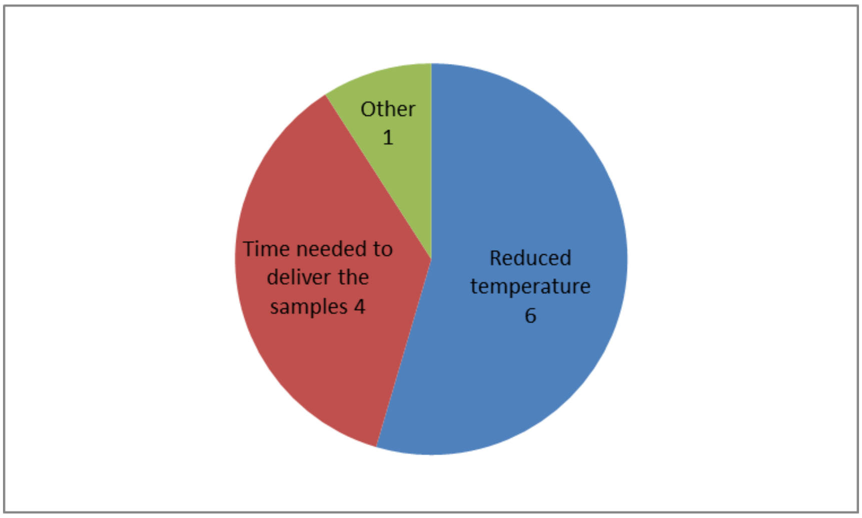

Figure 6: Pie chart of additional conditions for transportation of samples to the laboratory

5.2.3. Steps to be performed before transportation

Additional steps taken before transporting the samples are carried out in Austria, Germany, Italy, Poland and Slovenia (Figure 16). These pilot partners perform filtration with a pore width between 0.40 and 0.45 µm. Filtration is an important stabilization and preservation step for water samples. This is mainly to remove water micro-organisms, which can contribute on quality changes in water during the storage period or in order to minimize any chemical reactions, altering the concentrations of ECs (ISO 5667-3). Sometimes it is required to filter the sample "in situ, such as in the case of determination of the dissolved fraction of metals in ground water.

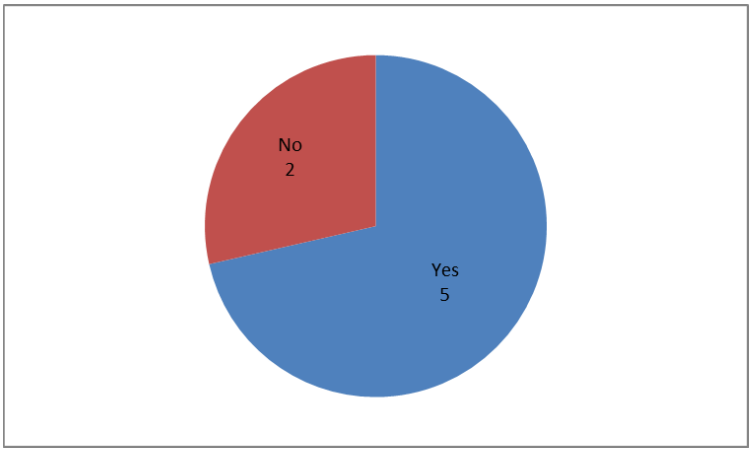

Figure 7: Are there any steps that should be performed before transportation?

5.3. Sample preservation (in the laboratory)

5.3.1. Additional sample preservation steps performed in the lab

The most common additional sample preservation step performed in the laboratory is light protection. Often, samples are also filtered, pH changed or chemicals are added to the samples. Centrifugation is the least used as an additional sample preservation step. In some countries, e.g. in the Czech Republic samples are stored in the laboratory by freezing them, and in Austria they carry out, among other things, ionic strength adjustments (Figure 17).

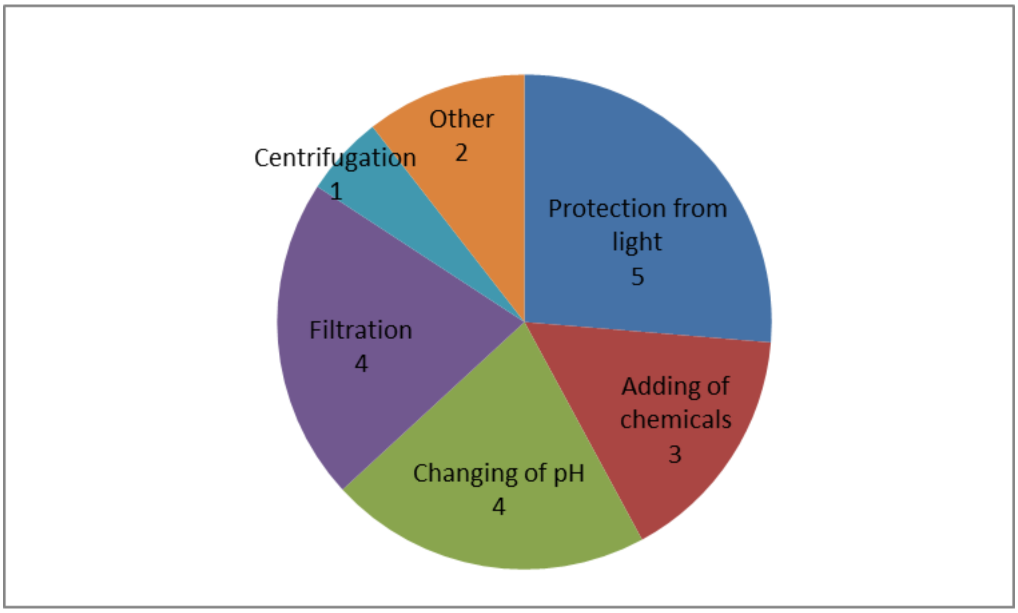

Figure 8: Pie chart of additional steps performed in the laboratory

5.3.2. Maximum allowed time between sampling/arriving of samples in the lab and analysis

Regarding the maximum time between collection and analysis, it is recommended to always perform the analysis on the samples, as soon as possible after collection. In some countries there are less, in some more strict, rules regarding the maximum allowed time between sampling / arriving of samples in the lab and analysis.

In Austria and the Czech Republic, maximum time limits are not set or specific rules do not apply. After sampling, the sample should be submitted to the laboratory and analysed as soon as possible – on the same day. In the case of storage at -20 °C, there is no time limit. The shortest possible time between sampling and analysis according to DIN 38407-47 is also required in Germany. Thus, the time between collection and analysis should not exceed three weeks. In Croatia, all samples were extracted within a week. In Italy, a sample submitted to the laboratory within 6 hours after collection is considered appropriate. If more than 6 hours have elapsed since sampling, the temperature shall be checked first and, if less than 10 degrees, the sample is appropriate. Likewise, the sample can be frozen after stabilization (SNPA, 2018). In Poland and Slovenia, the maximum allowed time between sampling and analysis cannot exceed 24 hours. For some analyses, in Poland, longer time is acceptable, for example 48 hours in the case of surfactants or even 7 days in the case of some specific substances. In Slovenia, samples can be stored for up to 30 days if filtered and cooled to 4 °C or longer if frozen.

5.4. Sample analysis

5.4.1. Kind of sample preparation performed before analysis

Before analysis, project partners prepare samples with solid-phase extraction (SPE) or liquid-liquid extraction (LLE) or evaporation (Figure 18). In many countries, however, samples are also prepared by other, own procedures that vary depending on the instrument used or the method of analysis. These procedures are: centrifugation / ultracentrifugation, acidification, isotope dilution. Some countries also use / test different procedures in different projects or research. These procedures are: divinylbenzene (DVB) extraction disks (Kotowska & Jasińska, 2011), ultrasound-assisted emulsification microextraction (USAEME) (Kapelewska et al., 2018).

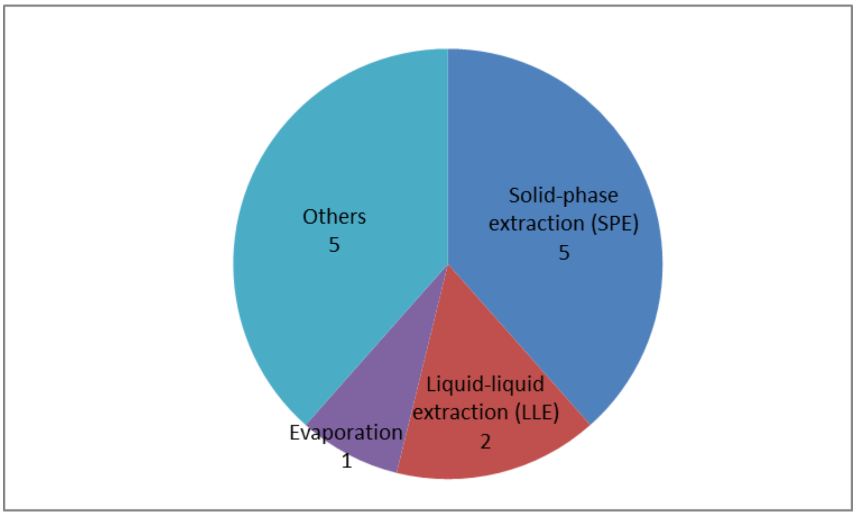

Figure 9: Pie chart of sample preparation types before analysis

5.4.2. Laboratory analytical standards and procedures: are they ISO/IEC 17025 accredited?

Analysis procedures follow national or European standards. In Austria and Germany, the standard method DIN 38407-47 (German standard methods for the examination of water, wastewater and sludge) is used for the analysis of pharmaceuticals. In the Czech Republic, analyses are carried out according to the applicable procedures and according to EPA method 1694. In Italy analytical methods have been fixed by the Legislative Decree n. 219/2010 implementing the Directive 2009/90/EC, which gives minimal efficiency criteria for analytical methods used in water status monitoring. SNPA Guideline n. 20/2019 contains specific procedures for priority substances and establishes internal analytical standards for laboratories. In Poland currently, there is no obligatory monitoring for pharmaceuticals or pesticides in the water environment, therefore scarce information exists about laboratory analytical standards and procedures.

Sampling and laboratory analyses for the determination of pollutants have been carried out in accordance with ISO / IES 17025 in all project partner countries.

5.4.3. Applied analytical approach

A targeted analytical approach is used in all project partner countries, including a non-targeted one in Poland and Slovenia (Figure 19).

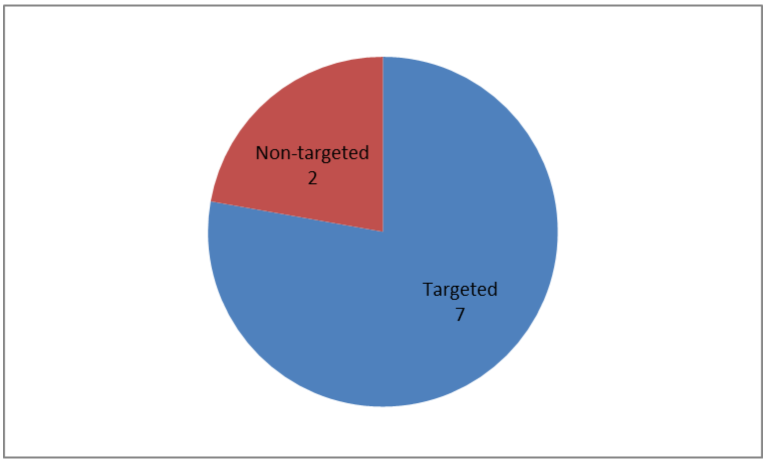

Figure 10: Pie chart of analytical approaches used

5.4.4. Applied method of analysis

All project partners use liquid chromatography (LC-MS) as an analytical method (Figure 20). The exceptions are Italy and Poland, which also use gas chromatography (GC-MS) and other methods, and Slovenia that uses only gas chromatography (GC-MS). Poland also uses Thin Layer Chromatography (TLC) in the case of scientific research, and Italy uses MS-MS, SPE-LC, LLE-SPE.

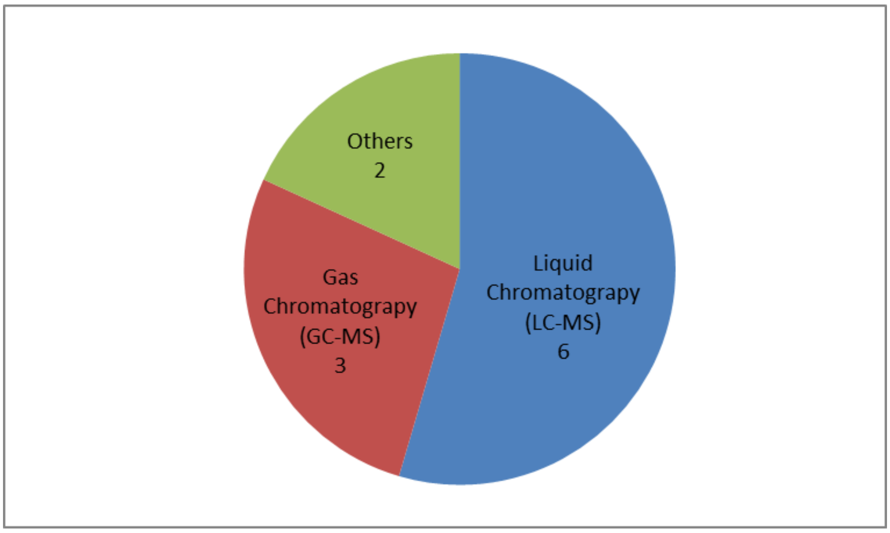

Figure 20 Pie chart of analytical methods used

5.4.5. Quality assurance performed for the analysis

Project partners have different views on quality assurance for analysis, or the quality assurance procedures are unknown – in **Croatia**.

Within the monitoring program "Water quality in **Austria**", the laboratories undergo through a quality assurance procedure to ensure the quality of the methodology employed and results obtained. This occurs by reviewing and inspecting the laboratory procedures. The performance of the analysis is externally evaluated, by internationally recognized audit expert institutions (e.g. EURACHEM, CITAC). The laboratories in charge for this analysis are reviewed by the official accreditation body "Accreditation Austria". One important step of quality assurance is the evaluation of the measurement procedures through external quality evaluation. For that, laboratories are participating on laboratory proficiency tests, organized from independent national or international companies. These tests may help the laboratories in assessment of performance characteristics of the method(s) used, according to. ÖNORM ISO/TS 13530 Wasserbeschaffenheit, Richtlinie zur analytischen Qualitätssicherung in der Wasseranalytik (Water quality, guideline for analytical quality assurance in water analysis). In Germany and Italy, quality is assured from sampling to sample analysis in accordance with the international standard ISO 5667-3. The following are important: preparation of the sampling car, selection of the sampling location, qualification of the sampler, proper labelling of the samples, selection of the appropriate sampling container and its preparation, etc.

Poland to assess accuracy or quality assurance uses basic and control samples. These samples are important for assessing the uncertainty of the results obtained, which must be included in the monitoring of surface and groundwater quality (PN-EN ISO / IES 17025: 2005).

In **Slovenia**, according to the "Monitoring Program for the Chemical and Ecological Status of Water in the Period 2016 to 2021", sampling for the analysis of physicochemical parameters is carried out in accordance with internationally valid standards. Validated test methods and calibrated equipment are used for analysis, and measurement uncertainties are specified. Procedures for quality assurance of results, traceability and inter-laboratory comparisons are implemented.

The **Czech Republic** verifies samples through calibration control and by maintaining a clean environment, equipment and agents. The performance of the analytical system is ensured by blind and spiked samples. Each fifth sample in a series is processed by the method of standard addition, which is used to control the effect of the matrix of the sample and to reset the actual recovery ratio of a specific analyte. The measuring instruments is under regular control, and measuring vessels are metrologically tested.

5.4.6. Performance characteristics of the methods used for analysis

The analysis shall take into account European rules on laboratory analyses. It is necessary to keep in mind, as described by **Italy**, that the new generation instrumentation is certainly better than the more dated (higher sensitivity, higher quality and precision), so the same analytical method used with two different instruments can give different results of the limit of quantification LOQ and of the limit of detection LOD. The development of the technique was also highlighted by Germany. In Germany, however, the maximum deviation that is tolerated between the standard and the sample is +/-20 %. The maximum deviation still tolerated for the retention time of the analytes between the calibrated solution and the sample is +/-2.5 % (LfU 2015).

Poland stated that LOQ and LOD values depend on the method used, the compounds to be determined and the different types of water samples. In the case of the monitoring performed by Cabalska et al., limits of quantification (LOQ) of the GC-ECD method were dependent on determined compounds and ranged between 0.001 and 0.020 ug/L. For LC-MS/MS (MRM) and GC-MS (SIM) methods applied in the monitoring performed by Kuczyńska LOQs were 1-5 ng/L and 5-30 ng/L. Precision was assessed and defined as relative standard deviation (RSD). During the study conducted by Kuczyńska, RSD values did not exceed 10%, which is considered as good accuracy in terms of determination of ECs concentration. It is noteworthy that other methods applied for scientific research are characterised by various values of LOQ and limits of detection (LOD). LOD and LOQ are often dependent on a particular EC. LOQ may also differ in various types of water samples, e.g. using HPLC method LOQ for non-steroidal anti-inflammatory drugs (NSAIDs) determination was 1-5 ng/L in surface and groundwater and 5-25 ng/L in waste water (Kasprzyk-Hordern et al., 2008). In another research (application of GC-MS/MS (SIM)), LOQ values were 0.38-498.78 ng/L (RSD 5.1-23.1%) for groundwater and 1.1-241.6 ng/L for wastewater (RSD 6.4-23.6%) (Kapelewska et al., 2018).

Austria showed LOQ and LOD values for selected substances from the SCHTURM project and the “Monitoring of pharmaceuticals and wastewater indicators in ground and drinking water” project. The SCHTURM project analyzed: metals, xenohormones / Industrial chemicals, organotin compounds, perfluorinated surfactants, oestrogen steroid / hormones, phthalates, polybrominated diphenylethers, polycyclic aromatic carbons, pesticides and metabolites. LOQ and LOD values were the lowest in polybrominated diphenylethere (LOQ ranges from $7.8*10^{-6}$ µg/L to 0.18 µg/L ; LOD ranges from $8.5*10^{-8}$ µg/L to 6*10-4 µg/L) and the highest in the case of metals (LOQ ranges from 0.001 µg/L to 5 µg/L; LOD ranges from 0.0005 µg/L to 0.5 µg/L). The UHPLC-HRMS analytical method was used in the project “Monitoring of pharmaceuticals and wastewater indicators in ground and drinking water”. LOQ and LOD values were lowest (LOQ=1.0 ng/l; LOD=0.5 ng/l) in the case of acetyl-sulfamethoxazole, erythromycin, erythromycin-anhydrol, flumequinan, sulfadimethoxine, sulfamonomethoxine and trimethoprim, and highest (LOQ=5.0 ng/l; LOD=2.5 ng/l) in the case of chlortetracycline, marbofloxacin and Oxy-tetra cyra. In the same project, wastewater indicators were also analyzed using the LC-MS / MS method. LOQ and LOD values were lowest in the case of carbamazepine (LOQ=1.0 ng/l; LOD=0.5 ng/l) and highest (LOQ=10 ng/l; LOD=5.0 ng/l) in the case of sucralose, 1H-benzotriazole and tolyltriazole.

The Vltava RBA **Czech** Laboratory analyzes 114 substances, including medicines, hormones and antibiotics. Drug LODs range from 5 ng / l for PFOS (perfluorooctane sulfonic acid) to 1000 ng / l for octyl methoxycinnamate (OMC). Uncertainty estimates range from 30% to 45%. For hormones, LODs range from 0.5 ng / l for progesterone and testosterone to 10 ng / l for estriol. The hormones progesterone and testosterone have an uncertainty rating of 30% and the hormones estrone, estriol, 17-beta-estradiol, 17-alpha-estradiol and 17a-ethinylestradiol of 35%. Most antibiotics have a LOD of 20 ng / l, only the antibiotic doxycycline has a LOD of 50 ng / l. All antibiotics have an uncertainty rating of 35%.

Croatia provided data from Iveŝić et al. (2017). The lowest recoveries were obtained for SPY amounted to 62 % and the highest value was recorded for sulfamethoxazole, 115 %. The precision expressed as relative standard deviation (RSD) of recoveries was also satisfactory, in a range from 2 to 10%, and exceeded only for SMTX and SMM. The limits of detection LOD, based on the recovery and precision data, were arbitrary set at the lowest spiked level at which the following requirements could be easily achieved: recovery within the range of 50–130% and RSD ≤ 25% and were estimated for all analytes after measurement of six independent spiked samples. The same principle was applied in determining the LOQ provided that requirements were much stricter: recovery within the range of 70–120% and RSD ≤ 15% and also were estimated for all analyses after measurement of six independent spiked samples. The LOD values for all sulfonamides amounted to 0.05 μgL−1, for CAP was 0.01 μgL−1, and for FUM, TOR, and MCs were 0.1 μgL−1. The LOQ values for all sulfonamides amounted to 0.1 μgL−1, for CAP was 0.02 μgL−1, and for FUM, TOR, and MCs were 0.2 μgL−1.

5.4.7. Screening and fingerprinting methods

Screening or fingerprinting methods are used by project partners only in certain cases or may not be used at all.

There are no screening methods in Austria for the implementation of official ECs monitoring programs. With the exception of the monitoring program "Water Quality in Austria", pesticides were analysed using this method. In Italy, according to ISPRA Report 105/2011, screening of the chemical state is performed when information on the presence of emissions is not available. In Slovenia, screening and fingerprinting methods are applied and achieved with passive samplers only in the selective areas of Slovenia. In Poland, information on screening is only available in monitoring by Kuczyńska and Cabalska et al. In Germany and Czech Republic screening or fingerprinting methods are not used. Screening data for Croatia are unknown.

6. Attenuation strategies

Emerging contaminants (ECs) are widely present and distributed in aquatic environment. They have been in recent years recognized as significant pollutant and can have an impact on human and wildlife. There is a growing need to develop reliable methods of wastewater treatment, which enable the efficient removal of emerging contaminants at trace levels. Those pollutants can be to some extent removed with natural attenuation processes (e.g. physical, chemical or biological processes taking place in the environment to reduce the toxicity, volume, concentration of contaminants). An important role in removal of EC also has passive attenuation strategies (e.g. policy instruments, regulations and recommendations) and active attenuation strategies (e.g. treatment plants, nanofiltration, membrane filtration, biodegradation and photolysis).

The content of this report is divided into three subchapters. The first one is dedicated to active attenuation measures; the second to passive attenuation measures and the third to evaluation of effectiveness of the applied attenuation approaches.

The information concerning the active and passive attenuation strategies and their implementation reported in all the national reports have searched and cite a lot of different information sources, such as, international and national legislations, programmes and strategies, different research and scientific papers and presentations.

6.1. Active attenuation measures

6.1.1. Internationally available technology for the removal of ECs

All project partners are familiar with technologies for removing ECs. Countries have listed many different technologies, some of which are known in several countries.

The **Czech Republic** has stated that ECs can be removed in a water treatment process by ozonation, activated carbon filters or by reverse osmosis. Ozonation degrades organic molecules by oxidation, activated carbon filters remove contaminants by sorption, while reverse osmosis treats water by passing it through a semi permeable membrane, which is impermeable for dissolved salts and organic molecules (Romeyn et al., 2016; Justo et al., 2013; Sundaram et al., 2014). Another technology for mitigating EC contamination is artificial infiltration, which uses natural attenuation of contaminants in the aquifer.

As wastewater treatment plants are the main source of ECs entering the environment and conventional primary and secondary treatment do not remove ECs from wastewater, **Germany** cited several different advanced treatment technologies: ozonation and chlorination, UV-light disinfection, high- and low-pressure membranes, passive treatment technologies like lagoons, wetlands or soil aquifer treatment as well as the removal during riverbank filtration. Ozonation and high-pressure membranes have proven to highly sufficiently remove many of the environmentally relevant pharmaceuticals and personal care products (Rodriguez-Narvaez, 2017; Oulton, 2010).

Italy stated that in the case of drinking water, disinfection/oxidation is useful for the removal of ECs in the case of oxidation by ozone or advanced oxidation processes. However, activated carbon adsorption (granular activated carbon, GAC) is considered to be the best available ECs removal technology, potentially improved when used as biologically activated carbon. Other processes that can be used are pressure driven separation processes membrane filtration, nanofiltration, ultrafiltration and reverse osmosis (UN, 2017). In the case of wastewater, various factors affect the treatment applicability and efficiency. Conventional wastewater treatment plants include primary, secondary and tertiary treatment and disinfection (flocculation / precipitation, chemical oxidation, adsorption, membrane filtration and stripping) for phosphorus, residual suspended solids, microorganisms, Pharmaceuticals and Personal Care Products (Mezzanotte, 2018). Adsorption on GAC also produces the abatement of ECs depending on the kind of substance. For the removal of endocrine disruptors, drug residues, biocides compounds from Urban WasteWater, and pesticides from drinking water in over ten pilot plants in the entire world using a novel process combining PAC (Powder Activated Carbon) and ballasted clarification. The development of treatment inside wastewater and drinking water treatment plants is carried out through the construction of ozone diffusion systems and coupled ozone and inactivated carbon systems. Wetlands are also increasingly used in urban environments to mitigate the impact of polluted storm water runoff and wastewater. Both natural and constructed wetlands also biodegrade or immobilize a range of emerging pollutants, including some pharmaceuticals. For some chemicals, they may offer the only solution (UN, 2018). There are also limits to how NBS (nature based solutions) can perform. For example, NBS options for industrial wastewater treatment depend on the pollutant type and its loading. The efficiency of wetland operation is changed by operational settings (the increase of the water residence time, the growth of the macrophytes cover and the installation of the aeration system).

Poland is also familiar with several advanced sewage and drinking water treatment technologies that have been successfully adapted for the disposal of ECs (Yang et al., 2017). Widely applied techniques are membrane filtration, adsorption on active carbon, and a group of advanced oxidation processes (AOP). The most effective group of methods of ECs removal are AOP, such as ozonation, UV and photocatalysis and Fenton reaction, which are widely used in water treatment plants and, less often, in sewage treatment plants (Huber et al., 2003; Klavarioti et al., 2009; Gerrity et al., 2010).

Slovenia and **Austria** divided the technologies for removing ECs into 3 groups (Rodriguez-Narvaez et al., 2017):

- phase-changing technologies (adsorption using activated carbon, adsorption-using biochar, adsorption in carbon nanotubes, adsorption by clay minerals, other adsorbents materials, membrane technology-pressure driven membrane technologies)
- biological treatments (aerobic and anaerobic activated sludge, aerobic soil filtration, biological filtration)

- advanced oxidation processes (UV photolysis, ozonation, titanium oxide photo catalysis or combination of them)

In general, the best removal efficiencies were obtained with specific “substance-based” treatment technology and/or the combination of different technologies. Most of them showed a lack of scaling-up experiments (as in realistic scenarios) or more detailed studies.

6.1.2. Nationally available technology for the removal of ECs

In **Austria**, conventional treatment plants equipped with conventional activated and inactivated sludge system, biological degradation, adsorption to sewage sludge, membrane bioreactor system, (Clara et al. 2004a, 2004b), chemical oxidation (ozonation) are used to remove ECs (POSEIDON, 2004).

Croatia is conducting a number of scientific studies, but so far there is no such technology at any of the WWT plants.

In the **Czech Republic**, the AQURIUS project evaluated the efficacy of attenuation of pharmaceuticals in wastewater treatment plants, constructed wetlands, and in a shallow Quaternary aquifer (Rozman et al., 2017). They also conducted laboratory tests for the sorption of pharmaceuticals from wastewater onto an activated carbon filter and its sorption capacity.

Germany states that, although they know and have technologies for removing ECs, it is necessary to distinguish between the availability and the actual installation of these technologies. Although many advanced treatment technologies are available and could be used, this has barely been done. Advanced treatment technologies require high installation and operation and maintenance costs. This is the reason that most municipalities have not yet installed or even planned on doing so. Nevertheless, pilot experiments for the removal of ECs in wastewater are being encouraged in some federal states. The most commonly tested technologies applied in these experiments are activated carbon filters and ozonation (LfU, 2019c).

In **Italy**, different technologies for removing ECs are being studied with different projects and in different institutions. The PerFORM WATER 2030 project focuses on the removal of inorganic and organic pollutants (drugs and perfluorinated compounds) by adsorption on activated carbon, chemical oxidation processes and the use of microalgae and nanoparticles. The University of Milan Bicocca has assessed the priority and emerging contaminant levels and removal in conventional WWPTs, considering the removal mechanism and efficiencies in systems not designed for the removal of ECs. A similar study carried out on drinking water treatment plants was carried out by the Politecnico di Milano. Department of engineering of Ferrara has been studying ECs removal efficiency, especially with microbiological techniques. CNR-Water Research Institute has studied the degradation of emerging organic pollutants (polybrominateddiphenyl ethers) in sewage sludge by ozonation treatment. Aquality, an International and Academic-Industrial Multidisciplinary Network investigates the removal of pollutants emerging from the water system through sustainable hybrid treatments from an economic and environmental point of view which are the oxidation processes that use solar energy (sun driven), combined with membrane filtration systems (new materials, high flow ceramics, etc.

In **Poland**, water suppliers use attenuation technologies to remove ECs on a daily basis. There are no studies describing the effectiveness of technologies for removing ECs from drinking water. The project “Development of an innovative method of removing pharmaceuticals from wastewater” (Internet 1) focuses on the use of ozonation to remove PPCPs from sewage up to 80-90%. Additional aim of the project is preventing the formation of carcinogenic substances, as side products, and elimination of use additional chemicals in the ozonation process. The project represents the first attempt of taking up the PPCPs attenuation problem in wastewater at the national level.

Slovenia does not have national technologies for removal of ECs. However, some water utilities mix different sources of drinking water in case of excessive concentration of compounds in drinking water. This dilutes the excess concentration and thus achieves compliance with the standards.

6.1.3. Active attenuation measures present in the countries

Active attenuation measures for removal of ECs are present in all project partner countries, except **Croatia** and **Slovenia**. In both Slovenia and Croatia, wastewater treatment plants are mechanically and biologically treating the wastewater, which is controlled and in compliance with the regulations and is then returned to the natural environment.

The main active measure present on **Austrian** territory is wastewater treatment plants. They use an activated sludge system with biological degradation and sludge adsorption (Clara et al., 2004a, b, c, 2005). Beside this, pilot plants for research purposes may include ozonation, activated carbon and membrane bioreactor.

In the **Czech Republic**, several drinking water treatment plants have applied filters with activated carbon. Currently such filter stage is being built in the biggest drinking water treatment plant in the country.

In **Germany**, there are some municipalities that have set up active mitigation measures in the form of advanced technologies as part of pilot measures in wastewater treatment plants. Technologies that are tested and implied in these pilot experiments are (granulated) activated carbon filters as well as ozonation. In situ active attenuation measures exist particularly in highly-populated areas or areas with low precipitation where riverbank filtration is used in order to obtain drinking water. These remove contaminants, however, were not specifically designed for ECs. There is no national or regional approach to install a network of active attenuation measures to reduce occurrence of ECs in the aquatic environment (LfU 2019a, b).

In **Italy**, the removal of ECs is carried out by different waterwork groups in various projects. The perFORM 2030 project of the CAP group will explore and test technologies for the exploitation of sludge and the removal of emerging pollutants. The MM group is carrying out monitoring and removal tests for emerging contaminants in raw drinking water and treated water. SMAT group participates in the Horizon 2020 Aquality project, which includes the testing of innovative ECs removal techniques. Romagna Acque Spa and Milan Polytechnic study the applicability of Advanced Oxidation Processes for the removal of emerging micro-pollutants. HERA that manages pilot action Pontelagoscuro, at the drinking water treatment plant level, has implemented adsorption and/or filtration techniques through membranes of proven efficiency for the removal of PFASs in the production and distribution chain of drinking water.

In **Poland** the active methods of attenuation are widely used by water suppliers in Drinking Water Treatment Plants. Using of particular methods depends mainly on raw water quality and target quality of water, meeting the requirements of safe drinking water. Methods used by the Drinking Water Treatment Plants are:

- surface coagulation on contact filters and disinfection (SUW Czaniec);
- preozonation, coagulation, filtration, indirect ozonation, activated carbon filtration and disinfection (ZUW Goczałkowice);
- preozonation of raw water, contact coagulation in the fast and slow mixing chambers, rapid filtration on the anthracite-sand filers, indirect filtration through activated carbon deposits (SUW Kozłowa Góra);
- contact coagulation on fast filters with oxidant ($KMnO_4$) dosed periodically and correction of water pH, disinfection of water with sodium hypochlorite (SUW Maczki);

- disinfection with concentrated sodium hypochlorite (SUW Zawada) (as in boDEREC-CE report D.T2.1.1.).

6.1.4. Programs for removal or treatability of ECs in waste water

Most project partners are familiar with programs for the removal or treatment of ECs in wastewater both internationally and nationally. The exceptions are the **Czech Republic**, which knows neither international nor national programs, and **Slovenia**, which knows only international programs.

Croatia is internationally aware of the reverse osmosis/nanofiltration (RO/NF) membrane treatment processes which are widely used to remove emerging contaminants from the water. At the national level, Croatia conducts a number of studies on the removal of ECs. Dolar et al. (2012a) studied the removal of anthelmintic drugs and their photodegradation products from the water with RO/NF membranes. Dolar et al. (2009) also demonstrated that complete removal of the antibiotics sulphonamides, diaminopyrimidine, and fluoroquinolone is possible with typical RO/NF (XLE) membranes and by the tight nanofiltration membrane NF90. Membrane bioreactors (MBR) which combine activated sludge process and membrane separation are used for pharmaceutical removal from wastewater (Matošić et al., 2008; Dolar et al., 2012b). Cvetnić et al. (2018) tried to evaluate UV-C/H_2O_2 and UV-C/$S_2O_8^{2-}$ photooxidative processes for the treatment of 14 ECs. The kinetics of UV-C/$S_2O_8^{2-}$ processes were several times faster than in the UV-C/H_2O_2 cases. Cvetnić et al. (2019) used the QSPR methodology to predict the second-order degradation kinetics by $HO^{\cdot}$ and $SO_4^{\cdot -}$ for the selected ECs, more specifically pesticides and pharmaceuticals.

At international (European) level, Germany mentions EU-funded research projects aimed at removing micro-pollutants. **Germany** also participated in these projects. An example of such a project is the ATHENE project, which examines “new technical wastewater treatment solutions targeted for organic micropollutant biodegradation, by understanding enzymatic pathways and assessing detoxification” (ATHENE, 2019). In all parts of Germany, however, pilot experiments have been established on the removal efficiency of advanced treatment technologies. This represents a national strategy on the removal of trace substances from waste water which includes the orientational testing of different elimination technologies. In addition, Germany also has programs and strategies for individual groups of ECs, such as antibiotics. One such strategy for antibiotic-resistant microorganisms is “DART 2020”, which funds research projects on the most effective advanced treatment technologies, particularly ultrafiltration, since this technology has been established to be the most effective in the removal of resistant bacteria (DART, 2019).

Italy states that both international and national projects are involved in reducing ECs in wastewater. A study from Austria showed that in addition to longer treatment times of micro pollutants, ozonation will also be required. However, Abargues et al. (2018) dealt with the removal of endocrine disruptor compounds in wastewater using micro algae. Disposal of ECs on a national scale is discussed in the DEMEAU project, where different processes (oxidation processes, sand filtration or biologically activated carbon filtration) are studied and compared to advance elimination efficiency and to eliminate oxidation by-products and transformation products. DEMEAU's contribution to controlling these technologies at full-scale will improve the long-term stability and robustness of the processes, thereby facilitating the uptake of these technologies.

Poland cited EMAS (EcoManagement and Audit Scheme) as the only international program for the removal of ECs in sewage, which is a European eco-management and audit system. In order to reduce contaminants, especially micro-pollutants, from wastewater, the program recommends the introduction of a third stage of treatment (adsorption on activated carbon dust or oxidation with the use of non-chlorine-containing oxidants (mainly ozone)). At national level, too, Poland has approved the operation of the EMAS scheme. According to the decision of the EU Commission (Commision Decision (EU) 2019/61), it will be necessary to take appropriate measures in the field of sewage management, including the removing of ECs.

Slovenia, on an international scale, knows the review performed by the OECD on Advantages and disadvantages of advanced wastewater treatment options to remove pharmaceuticals (OECD, 2019 adapted from Rizzo et al., 2019). Advanced wastewater treatment processes are reverse osmosis, ozonation, activated carbon, membranes and advance oxidation and can achieve higher removal rates for pharmaceuticals in comparison to conventional secondary wastewater treatment (OECD, 2019). The effectiveness of these processes depends on the physico-chemical properties of active pharmaceutical ingredients and their metabolites. Activated carbon adsorption (with both powered activated carbon (PAC) and granular activated carbon (GAC)), ozonation and filtration by nanofiltration or reverse osmosis membranes, have been demonstrated to effectively remove most pharmaceuticals.

6.1.5. Pilot experiments in waste water treatment performed for the removal of ECs

Pilot tests of wastewater treatment for the removal of ECs are being carried out in all project partner countries.

In **Austria**, the KomOzAk project "Further purification of municipal wastewater with ozone and activated carbon for the removal of organic trace substances" from the Federal Ministry for land, forest, environment and water management ("Weitergehende Reinigung kommunaler Abwässer mit Ozon sowie Aktivkohle für die Entfernung organischer Spurenstoffe", Bundesministerium für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft, 2015) wanted to explore a combination of additional treatment technologies to purify municipal waters from organic trace substances: ozonation followed by activated carbon, or activated carbon as standalone. The experimental plant was situated in the vicinity of the main wastewater treatment plant and it was composed of two pilot plants disposed in parallel.

In **Croatia**, all technological projects include pilot experiments. Thus, wastewater treated with membrane bioreactor (MBR), nanofiltration (NF), and reverse osmosis (RO) for its reuse in the agricultural irrigation was assessed at the waste water treatment plant in Čakovec. They found that the treated wastewater meets the standards for irrigation in agriculture (Dolar et al., 2019).

In the **Czech Republic**, as part of the project "Technology using photochemical oxidation with sorption for the elimination of micropollutants nature of pharmaceutical substances from wastewater", a prototype of a treatment unit was developed and tested its efficiency as a tertiary stage of a conventional wastewater treatment plant. The device treated water by oxidation with H_2O_2 activated by UV light and sorption on different materials. The in situ experiments resulted in setting the optimal parameters of the process.

There are also several treatment plants in **Germany** that have installed advanced treatment technologies as part of pilot experiments. In Bavaria, in the town of "Weißenburg", there is a pilot wastewater treatment plant that uses two different advanced treatment technologies in succession. The first one is ozonation (ozone generator and contact reactor), subsequently filters of two different materials were installed. Part of the water was filtered through sand, the other through granulated activated carbon in order to be able to compare both filter materials (Gruber, 2019; LfU, 2019c).

Italy mentions several major pilot projects for wastewater treatment, including Nosedo-Milan and pilot action Pontelagoscuro (Ferrara).

Poland provides data on two pilot wastewater treatment experiments. In 2018, the removal of diclofenac from treated sewage was tested in a Wastewater Treatment Plant located in the Silesian Voivodeship. For this purpose, ozone treatment was applied using a pilot station located on a truck. A 98 % reduction in diclofenac concentration was achieved (Kosiniak, 2019). The project "Development of an innovative method of removing pharmaceuticals from wastewater" is also being implemented. Its goal is to develop a pilot station which will use ozonation to reduce 80-90% pharmaceutical residues from sewage (Internet 2).

In **Slovenia**, two studies examined the removal of pharmaceutical residues in a pilot wastewater treatment plant. In the first study, five pharmaceuticals were removed by biodegradation and/or by adsorption: ibuprofen, naproxen, ketoprofen, diclofenac, and clofibric acid (Kosjek et al., 2006). Another study investigated the removal of six pharmaceuticals: ibuprofen, naproxen, ketoprofen, carbamazapine, diclofenac, clofibric acid with hydrodynamic cavitation and added hydrogen peroxide. The research showed pharmaceutical which are resistant to biological treatment, can be largely removed by introducing additional non-biological procedures in to wastewater treatment, in this case hydrodynamic cavitation and added hydrogen peroxide proved to be very sufficient (Petkovšek et al., 2015).

6.1.6. In situ treatments for ECs in surface water bodies

Project partners do not have data on in situ treatment for ECs and surface water bodies, with the exception of **Germany**. In **Austria**, however, they have studies on river bank filtration, but still at a research level, and not as an established treatment method. Germany also mentions riverbank filtration, which in some parts of the country is a common form of in situ surface water treatment used for drinking water. Riverbank filtration, however, is no in situ treatment specifically designed to efficiently remove emerging contaminants. Drinking water obtained from riverbank filtration has also been included in the projects of the Federal Environmental Institution. Parent compounds and metabolites of pharmaceuticals, cocaine, and the synthetic sweetener acesulfame were discussed. 25% of the probes contained residues of the anti-epileptic carbamazepine, in few sulfamethoxazole and primidone were additionally positively detected. This leads to the conclusion that riverbank filtration is not capable of completely removing ECs (LfU, 2019b).

6.1.7. In situ treatments for ECs in monitoring bodies

In 5 project partner countries (**Austria**, **Croatia**, **Czech Republic**, **Germany**, **Slovenia**) there are no in situ treatment for ECs in monitoring water bodies. The exceptions are **Italy** and **Poland**, where in situ treatment technologies for some groundwater bodies were applied. In Italy there were wells for potable use with high concentrations of PFAS. In addition to the closure of these wells, activated carbon filters have been adopted for the new wells that feed the public network, the only ones capable of retaining perfluoroalkyl substances. In Poland, however, in situ treatment technologies were used for groundwater contaminated with PAHs. In most cases a pump and treat method was used. Also impermeable barriers are implemented to reduce the spread of contamination.

6.1.8. Prescribed strategies for the chemical status improvement

In **Austria** no specific and explicit reference to pharmaceuticals, pesticides or personal care products (or ECs in general) is stated in the potential pressures and impacts (point source pollution or diffuse source pollution) to the river basins (Danube, Rhine, Elbe) (EC, 2012, River Basin Management Plans, Austria).

In **Croatia**, wastewater treatment plants should implement new advanced treatment technologies in order to preserve the good chemical status of river basins in the context of ECs presence in water.

The **Czech Republic** has no specific strategies focused on ECs. Dangerous substances, including ECs, are subject to the following measures:

- identify possible point sources of contamination;
- if the identified sources are regulated by discharge permission, contact the relevant authority and in cooperation with the polluter set a procedure, which will lead to elimination of the source;
- record the discharge to the national discharge registry;

- add the substance to the wastewater discharge regulation, so that regulation will include all the substances which are relevant for chemical status assessment.

In **Germany**, most water bodies do not achieve good chemical status. In the management plan (2016-2021) with preventive approaches focused on production / resources through technological measures, they want to reduce the risk of contaminants in the environment. The measures are adapted to the specific water bodies and substances to be addressed. To efficiently be able to reduce the entry, one has to differentiate between diffuse (run-off from urban and agricultural areas) and point sources. There are measures that specifically address waste water treatment plant effluents which is the main source for most ECs entering the aquatic environment. Among the suggested measures that can be undertaken in these waste water treatment plants are the adjustment or new construction of (old) municipal plants, the optimization of the mode of operation in order to obtain the required reduction in entry of nitrogen and phosphorous as well as “other substances” into the environment. These measures may already contribute to a reduced entry of ECs, however, the “LAWA” (“Bund- / Länder-Arbeitsgemeinschaft Wasser”) as well as the Bavarian State Ministry of the Environment (“Bayerisches Staatsministerium für Umwelt- und Verbraucherschutz”) have included other measures to reduce the amount of contaminants in waste water treatment plant effluents. The Bavarian measures include the installation of quaternary advanced treatment technologies which would efficiently improve the chemical status of water bodies concerning ECs (BStUV, 2015).

In the Po river basin management plan in **Italy**, measure KTM14-P1-a053 “Increased knowledge on endocrine disruptors (quantities and effects on biological communities) present in the surface waters of the Po River” was envisaged. It is a cognitive measure because the loads of ECs passing through the year are not known on the river Po.

In accordance with River Basin Management Plans (Internet 3), **Poland** assesses the chemical status of surface water bodies on the basis of the concentration of priority substances and other pollutants for which environmental quality standards are set. The assessment of the chemical status of surface water bodies is conducted in accordance with the requirements included in the Regulation of the Minister of the Environment (2016) and is based on a comparison of the results of measurements of water quality indicators with environmental quality standards. The classification of the surface water bodies’ chemical status is performed on the basis of at least 12 measurement results of priority substances and other pollutants. Good chemical status is determined if the average annual concentration of the measured indicators and the maximum concentrations do not exceed the environmental quality standards.

In **Slovenia** river basin management plan according to WFD are not considering ECs directly. The intentions are focused in general to achieve good status of water bodies.

6.1.9. Consideration of natural attenuation in relation to ECs and its harnessing

In all countries of the project partners the advantage of natural attenuation is taken. Different countries use more or less different procedures carried out in the framework of national plans or research projects.

In **Austria** natural attenuation methods for removing ECs are wetlands and riverbank filtration. Many research and data is also available on the development of constructed wetlands, but no data are available specifically concerning the removal of emerging contaminants. The natural attenuation method of riverbank filtration is often used for drinking water purposes (van Driezum, 2018; van Direzum, 2014; Gillefalk, 2018). It is a natural infiltration of surface water into nearby aquifers. The effects of removing organic micro-pollutants (including pharmaceuticals and personal care products) have been studied in various studies and found positive attenuation results (van Driezum, 2018).

In **Croatia** there are constructed wetlands (e.g. in Kaštelir) for wastewater treatment which enable the removal of organic contamination through absorption on biofilm and clay particles, as well as degradation with the help of bacteria in soil. However, until now they are not put in the context of ECs removal. Given the large number of smaller settlements in Croatia with the need for wastewater treatment, the use of constructed wetlands poses as promising option.

In **Czech** legislation natural attenuation is not considered in relation to ECs, however it has been studied in the AQUARIUS project and has been relatively effective. It turned out that only carbamazepine and its metabolites were detected in unchanged concentrations in a borehole 100 m away from the infiltration of polluted water. Concentrations of sulfamethoxazole, hydrochlorothiazide, gabapentin, tramadol, and sulfanilamide were low and apparently affected by natural attenuation in the soil and in the aquifer.

In **Germany**, the fate of different groups of ECs in the aquatic environment has been studied as part of various national and international studies. Natural attenuation depends on the physiochemical properties of the individual contaminant, so generalization is not possible for all ECs. ECs are mainly attenuated by dilution along with adsorption onto suspended solids / sediments, photolysis, and aerobic biodegradation (Pal, 2010). Most ECs are ubiquitous in surface water bodies and are also quite persistent there. This can be caused by microorganisms which are not able to biodegrade the compounds (ibuprofen, carbamazepine, acetaminophen mefenamic acid, and propranolol) in the river water environment or by a constant inflow of WWTP effluents counterbalances a measurable reduction. The phenomenon is called pseudo-persistence. Pseudo-persistent pollutants are different classes of antibiotics such as macrolides, sulfonamides, and beta-lactams (Li, 2014). The studies therefore considered natural attenuation and examined whether it could be used as an effective attenuation measure (e.g. removal of ECs in constructed wetlands) (Rühmland, 2015).

In **Italy** sludge adsorption, stripping, biodegradation and photo degradation are considered natural attenuation processes, especially for the molecules produced synthetically but which are also present in nature (hormones). For ECs that have only synthetic molecules (PFAS), these removal approaches are less effective.

Poland has identified two groups of natural processes that can contribute to the gradual reduction of the load and concentration of micro-pollutants in the aquatic environment:

- degradative processes – they degrade pollution permanently: biodegradation, biological and chemical transformations, photolysis,
- non-degradative processes – they do not degrade pollution, but reduce its concentration: sorption, immobilization, volatilisation, diffusion, dispersion, dilution as a result of mixing with clean water and rainwater.

These processes contribute to the attenuation of chlorinated solvents, polycyclic aromatic hydrocarbons and phenols (Malina, 2011). Due to these processes, ECs concentration, their toxicity and mobility may decrease and, as a consequence, self-attenuation may occur. The most important procedures for the natural attenuation of ECs are: photodegradation (photolysis), biodegradation, sorption processes (adsorption, desorption, ion exchange). The most important and the most effective process which leads to the degradation of ECs in the aquatic environment is biological decomposition influenced by microorganisms (biodegradation). Adsorption methods are often used to remove PPCPs from water and wastewater and they are highly effective (Czech, 2012).

In **Slovenia**, natural attenuation is used mainly in water utilities, where they provide adequate and safe drinking water through dilution and dispersion.

6.2. Passive attenuation measures

6.2.1. Passive attenuation measures present in the countries

Passive attenuation measures are present in all project partner countries except **Croatia** and the **Czech Republic**. In some countries they are more developed and used in some less.

In **Austria**, the passive attenuations are mainly the presence of legislation, which regulate some of the classified ECs. In the Austrian legal environment no ordinances/legislation are present concerning precisely and specifically emerging contaminants. Some of the substances considered as ECs are listed in the national water quality legislations. Accordingly, the environmental limits of synthetic and non-synthetic substances and pesticides are set to achieve good ecological and chemical status of water bodies.

In **Germany**, only a few passive attenuation measures concern ECs in the aquatic environment. At the national level, a strategy has been established for the elimination of trace substances in waste water treatment plants, which served the initial orientation on the effectiveness of various advanced treatment technologies. In addition, individual federal states have established their own programs / measures to eliminate the entry of trace substances into the aquatic environment. Through these programs, the installation of advanced treatment technologies is financially supported. Results might contribute to passive measures such as the obligement to install advanced treatment measures in waste water treatment plants (LfU, 2019c). Passive attenuation measures have also targeted various subgroups of ECs that are of particular concern to the environment and human health. These groups include endocrine disrupting compounds and antibiotics.

In **Italy**, according to the DPSIR, the focus is on passive attenuation measures influencing Drivers (human activities like industry agriculture) and less States (emission of pollutants). Among the policy instruments we can consider the environmental legislation on water protection. Among regulations and recommendations, it is possible to include:

- specific passive attenuation measures related to environmental monitoring, production process regulations and water protection zones regulations
- National and Regional Programmes for Rural development (PSRN, 2019)
- the National Plan for the Sustainable Use of Phytosanitary Products
- regional and national information campaigns for reducing the use of antibiotics;
- voluntary actions and sectoral initiatives for reducing pollution impacts from specific industrial productions

In **Poland**, the main passive attenuation measures for groundwater are protection zones of water intakes, protection zones of aquifers as well as all kinds of prohibitions and restrictions on land and water use. The important part of passive attenuation measures in terms of water protection is also groundwater monitoring, including the so-called defensive detection monitoring associated with early warning about threats for major sources of water supply.

In **Slovenia**, passive attenuation measures take the form of prohibited compounds/restricted chemicals to be used or applied because they pose a risk to human health or the environment. Groundwater protection areas are also a very important passive measure, where there are several prohibitions and restrictions on land use and compounds used. One of the strategies of passive attenuation is also the Decree on management with waste medicine (Official Gazette of the RS, No. 105/08 in 84/18 - ZIURKOE) in which the conditions for the collection and disposal of unusable medicines and residues are written.

6.3. Evaluation of effectivness of attenuation approach

6.3.1. Effectiveness of active attenuation measures and measurements of their effectiveness

5 project partner countries confirm the effectiveness of active attenuation measures, and 2 countries - **Croatia** and **Slovenia** do not have effective attenuation measures. In **Slovenia** the only measure in some very rare cases is mixing water from different natural sources to lower concentrations of some pollutants in drinking water.

In **Austria**, the effectiveness of active measures such as biodegradation and adsorption to sewage sludge has been reported in Clara et al. (2004). The measures are relatively effective in removing ECs from wastewater, as determined by observing and analysing samples of the influent and effluent of a wastewater treatment plant. In addition to wastewater, active measures in Italy are also effective in drinking water treatment processes. Also in the **Czech Republic** the effectiveness of attenuation is measured by comparing the concentrations in the inflow to the system and in the outflow from the system.

In **Germany**, the most effective active attenuation measures are ozonation and granulated activated carbon filters that are commonly used in pilot experiments concerning EC removal in wastewater treatment plants. These advanced treatment technologies in wastewater treatment plants are able to efficiently remove environmentally harmful micro-contaminants. The pilot experiment conducted in Bavaria for example aimed for an overall removal rate of 80 % in order to call the removal "effective". This was the case for both ozonation and subsequent sand filtration as well as subsequent activated carbon filtration. However, sand filtration was not able to improve the elimination rates, while the activated carbon filter was indeed able to do so. All in all, the ozonation achieved an overall removal rate of 83.3 %; the succession of ozonation and subsequent granulated activated carbon filter even reached 92 %. Note that pharmaceuticals (i.e. carbamazepine) recalcitrant to common wastewater treatment are efficiently removed through these processes (removal rate for carbamazepine >95 %) (LfU, 2019c).

In **Poland**, the effectiveness of active attenuation measures on different contaminants is often different. The removal efficiency (RE) is usually expressed as the percentage of reduction between the ECs concentration in raw water (wastewater) (C_{IW}) and the ECs concentration in the treated water (wastewater) (C_{EW}) (Kapelewska et al., 2018).

$$RE = ((C_{IW} - C_{EW}) : C_{IW}) \times 100\%$$

The removal efficiency (RE) may be also expressed as the ratio of concentration after treatment (C) to initial concentration of a given compound (C_0) (Budzik-Niemiec & Dudziak, 2015).

$$RE = C / C_0$$

The active attenuation measures applied in Poland for water and sewage treatment in removal of ECs are:

- activated sludge
- denitrification and nitrification chambers
- UV treatment (UV) and UV treatment coupled with ozonation (UV/O_3)
- membrane bioreactor (MBR)
- nanofiltration
- photocatalysis in the presence of TiO_2 and ZnO
- gravel filters
- constructed wetlands

- biological reactor
- activated carbon
- FK/UF/NF sequential system (photocatalysis/ultrafiltration/nanofiltration)
- reverse osmosis

For water treatment from micropollutants there are also enhanced natural attenuation methods (biostimulation, bioaugmentation, phytoremediation, agrotechnical treatment and composting) (Malina, 2011) which supports natural degradative and non-degradative (self-attenuation) processes that lead to reduction of the pollution load in the water environment.

6.3.2. Implementation of passive attenuation measures

Passive attenuation measures are implemented in all project partner countries except **Croatia** and the Czech Republic. In the **Czech Republic**, attenuation measures are not implemented because ECs do not have limit values and are not relevant for water chemical status assessment (except for pesticides).

In **Austria**, passive attenuation measures are implemented as national laws. This means that for instance the listed pesticides must be monitored and analysed for surface water bodies, within drinking waterworks according to the frequency and methodology requested in the legislation statement.

In **Germany** passive attenuation measures include recommendations and financial support for the installations of quaternary treatment technologies as well as the national strategy on antibiotic-resistance. Nevertheless, most passive attenuation measures consist of non-binding recommendations that can voluntarily be applied. German policies concerning ECs in the aquatic environment lack of effective and binding measures that are capable of efficiently and promptly reducing the amount of ECs entering/present in the aquatic environment (LfU, 2019c; DART, 2019).

In **Italy**, passive attenuation measures have been identified as effective actions for reducing ECs both in drinking water and waste water treatment processes. Regulations for prohibited and restricted compounds are also being implemented in Slovenia, but the question is if the regulations are being respected and to what extent.

In **Poland** currently, protection zones of water intakes are established according to the new regulations, in accordance with the new Act of Water Law (Water Law of 20 June 2017). The necessity of determination of protection zones is determined based on the results of risk assessment. It is estimated that approximately 20% of water intakes in Poland have established protection zones. Protection zones of aquifers are also being established. Within these protection zones, some prohibitions and restrictions on land and water use are in force in order to protect water resources from quality degradation.

7. Conclusion

Based on the State-of-the-art procedure developed at the beginning of the thematic work package, we have collected a large amount of information. It was organised according to the predefined steps, which were defined as a learning tool.

Currently, there is no globally uniform definition of ECs. The existing definitions mainly describe the most common general characteristics of EC. Based on the review of national reports and additional literature, we propose the following definition of ECs within the framework of the boDEREC-CE project:

Emerging contaminants represent a group of potential pollutants that are either newly created, newly identified, newly detected or newly researched.

There are several ways in which we can classify ECs. Most often, they are classified based on intended use of their source into industrial chemicals, pharmaceuticals, personal care products (PCPs), pesticides and artificial sweeteners (ASs). We also recognize other possible classifications such as chemical, quantitative, based on effects and several combined classifications.

ECs group is comprised of over more than 1000 different chemical substances or mixtures, most of them are currently unregulated.

Sources of ECs are mainly anthropogenic activities originating from one or more of the main human spheres such as infrastructure, agriculture, industry or urban life. Human waste, ending up in wastewater and landfills is recognized as a principle source of origin for ECs that provides entrance of ECs into the environment through various release mechanisms and at different compartments of the water cycle.

Although there is a common European framework guiding the national legislatures of all partner countries, there are considerable differences in research and monitoring stages of each individual country. European projects like boDEREC-CE therefore provide a useful platform to synthesize and share the current SOA knowledge among the PPs with the final goal to aid in the management of ECs on a transnational level.

The definitions and classifications gathered in this report will help in the systematization of further questions and challenges relating to ECs in the scope of state-of-the-art activities that follow and in the course of the entire boDEREC-CE project as well.

Current state-of-the-art reviews are essential for providing new information on any topic, especially for such field as ECs because it is increasingly popular leading to new research being conducted and new results constantly being provided. New advances in analytical extraction and measurement technologies and methods provide us with tools to detect, identify and quantify ever smaller and more reliable analyses of ECs levels in the environment. Additionally, investigating the topic of ECs is important for the society in general, as it deals with the possible pollution of the natural environment which can have a negative impact on the ecosystems and consequently human life. Only sufficient knowledge can enable implementation of correct and timely measures to avoid the adverse effects of ECs to our health and life.

The monitoring of emerging contaminants in project partner countries is not highly developed. The monitoring of surface and groundwater is performed regularly, however, few pollutants belonging to emerging contaminants is taken into account. Some ECs are monitored from the priority list according to the EU legislative requirements, some from the watch list, and some from different projects and studies. The results of these projects show the importance of the problem with ECs, as it has been found that waters are ubiquitously contaminated with ECs (pesticides, drugs, hormones…).

The transnational report provided specification regarding the sampling location, method and methodology of analysis in all project partner countries. As the problem of the occurrence of ECs in the aquatic environment is relatively new, no specific guidelines or regulations for sampling and analysis have been established. All the characteristics and procedures are for general standard sampling.

Most countries are interested to know more about the properties of these hazardous substances in water. They need to record sources, routes and behaviour of detected substances, have control over pollution levels and take action in the event of pollution, improve methods of drinking and wastewater treatment, and identify seasonal and long-term changes in ECs concentrations.

The project partner countries are aware of the issues related to ECs and are researching the occurrence and attenuation of ECs in surface and groundwater and wastewater. Effective procedures for removal of ECs have not yet been established internationally and, in most countries, also nationally. Because the rate of removal depends on the characteristics of the substance and the technology used, it is difficult to find an effective system for removing ECs. Countries are testing different treatment methods such as oxidation, activated carbon filtration, constructed wetlands, adsorption, river bank filtration, ozonation, ultrafiltration, reverse osmosis etc. Each method has advantages and disadvantages, and the rates of removal of ECs also differ.

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