boDEREC-CE
WORKPACKAGE T2

O.T2.8 PILOT ACTION AREA NEUFAHRN BEI FREISING

VERSION 1

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1. Introduction

As a substantial part of the boDEREC-CE project, the occurrence of Pharmaceuticals and Personal Care Products (PPCPs) was intensively investigated in the water sources of each pilot action area. The knowledge form the monitoring is fundamental to understand the compound and site specific behaviour of PPCPs in the environment and to develop a site specific action plan if needed. The monitoring procedure and its findings was the main objective of the second thematic work package (WP T2). In total, eight pilot action areas were investigated in central Europe within the boDEREC-CE project. Each area showed different hydrological and socio-economic conditions.

This report summarizes the monitoring activities in the pilot action area of “Neufahrn bei Freising” next to Munich, in the southeast of Germany. The monitoring concept was designed to characterize the site specific conditions of the ground- and surface water. As the groundwater is used as potable water, it was sampled as well as the surface water, which may impact the groundwater.

In order to increase the comparability among the findings of the pilot action areas in the boDEREC-CE project, the sampling, and the laboratory analysis were uniformed. The procedure is also described in this document. The data gained by monitoring is presented and discussed here. So, transport processes can be estimated in a first analysis. Further, for the more detailed assessing of the data, transport models were developed based on the monitoring data within the thematic work packages (WP T3).

2. Pilot site characteristics

The pilot action area in “Neufahrn bei Freising” is depicted in yellow in Figure 1 and with an enlarged view in Figure 2. It is located 15 km north of Munich’s city centre in the southeast of Germany. The 20 km^2 area is bounded by the settlements “Dietersheim” in the south, “Neufahrn bei Freising” in the north, “Eching” in the west and by the river Isar in the east.

In accordance to the INVEKOS database, the pilot area is a rural area and is mainly used for agricultural purposes. Next to the river, in the middle of the pilot area, the wastewater treatment plant (WWTP) “Gut Marienhof” (built in 1998) treats a substantial part of Munich’s wastewater (Wünsch und Plail 2013). The wastewater to be treated is guided via a sewer channel through the southern part of the study area to the treatment plant.

In the northern part of the pilot action area, the water supplier “Zweckverband Wasserversorgungsgruppe Freising Süd” extracts shallow and deep groundwater, in order to provide the surrounding communities and industries with process and drinking water. The extraction wells (red dots nr. 3 and 4) can be seen in Figure 2.

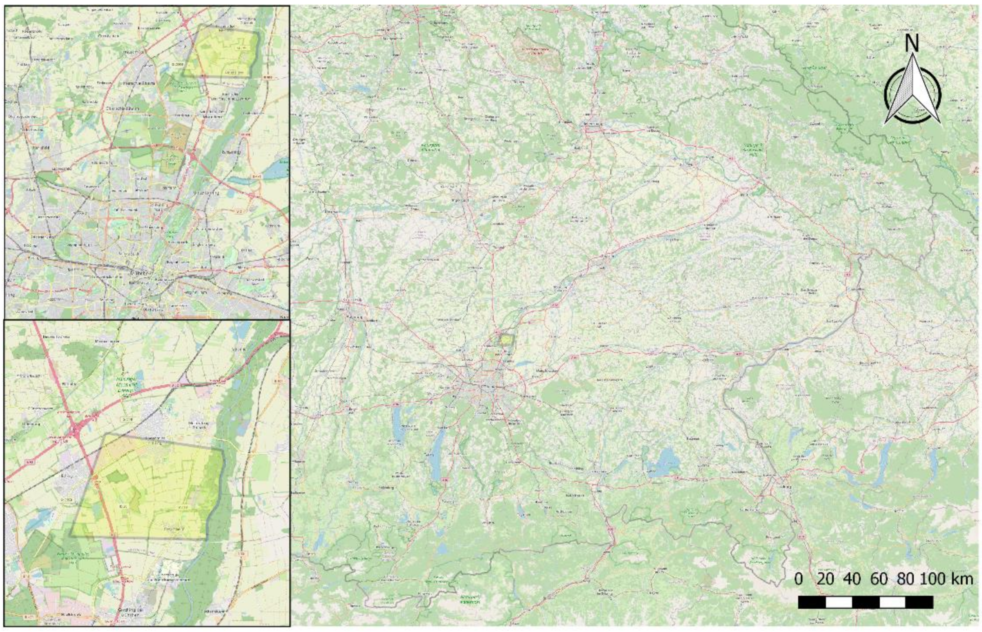

Figure 1 The location of the pilot action area (yellow) of Neufahrn bei Freising in Germany, created with © OpenStreetMap.

For the pilot action area in Neufahrn bei Freising, several potential PPCPs sources, causing a PPCP concentration in the aquatic environment (ground- and surface water), can be expected.

- **Surface water**: The WWTP effluent is discharged in the river Isar within the pilot area. Although, the WWTP compiles the legal requirements, it is not upgraded with an advanced water treatment (Wünsch und Plail 2013). Thus, PPCPs can be expected in the effluent and consequently in the river. If the surface water infiltrates in the groundwater, the PPCPs may be detected in the aquifer.
- **Landfills filled with municipal waste**: Although, it is not allowed to dispose untreated municipal waste to landfills in Germany (BayAbfG 2015), older leaking landfills in the study area may contaminate the groundwater with PPCPs.
- **Leaking sewer systems**: In urban water engineering, leaking sewer systems is a well-known issue (Ellis et al. 2009). For instance, Osenbrück et al. (2007) reported that the connection points to private sewer pipes might be the main source of exfiltrating wastewater. Perhaps, in this pilot action area the sewer itself or the connections to private households may be a PPCP source.
- **Agriculture**: The study area is mainly used for agricultural purposes. Formerly until 1995, sewer sludge was applied on fields south of the pilot action area but not in the area. A recently application of wastewater sludge is not known. Beside this, the application of PPCP containing pesticides may be another potential source (Lacorte et al. 2002).

2.1. Geographical and hydrological conditions

The pilot area lays in the northern part of the Munich gravel plain (Bayrisches Landesamt für Umwelt 2021). Three different kinds of glaciofluvial gravel can be found: in the west glacial gravel, in the middle alluvial gravel and in the east several floodplain deposits. Two porous groundwater aquifers were reported for this area by Schuler (1992): an unconfined quaternary and a confined tertiary aquifer, separated by a clay layer. Due to the low permeability of the clay layer, an exchange of both aquifers can be assumed to be negligible. However, due to geological formation, an aquifer connection cannot be ruled out completely. The upper quaternary aquifer shows an average depth of about 17 m below surface (Schuler 1992). Its water table is around 4 m below surface.

As the differences in the surface elevation between the boundaries (northwards and eastwards) are less than 10 m, the pilot action area can be assumed as flat.

The most important surface water in the pilot area is the river Isar. The river gives the eastern boundary for the pilot area. During a previous Interreg project (PROLINE-CE), the correlation between the river water level and the discharge was studied. This gained information could also be used for this project.

In the pilot area, no precipitation data is available. Yet, between 1993 and 2016 a mean annual precipitation sum of 762 mm and a mean annual air temperature of 9.1°C could be observed at the weather station at Munich's airport, which is located 10 km north-east. According to the German Meteorological Office (DWD), an actual evapotranspiration rate of 522 mm/a and a potential evapotranspiration rate of 656 mm/a was calculated for the same time span. (DWD 2017)

For the region of Munich a groundwater recharge of 187 mm/a was reported (Bayrisches Landesamt für Umwelt 2020) . The values were determined by the GWN-BW model, described by Gudera and Morhard (2015).

2.2. Water treatment techology

In the northern part of the study area, three (since 2020 four) shallow and six deep extraction wells are maintained by the water supplier "Zweckverband Wasserversorgungsgruppe Freising Süd". The extracted water of the shallow wells is used as process water, for instance, at the research campus of the Technical University of Munich in Garching. The water utility also supplies the local population in and next to Neufahrn with drinking water, extracted from the deep tertiary aquifer. In comparison to surface water, deep groundwater is not treated in multistage processes (apart from softening or the removal of iron and manganese ions at some sites).For the extracted groundwater in Neufahrn bei Freising, there is no need for any treatment before the supply. (Zweckverband Wasserversorgungsgruppe Freising-Süd 2022)

2.3. Socio – economic conditions and main end users

The main end users of the drinking water in this pilot action area are private households (14,000 households and circa 66,000 inhabitants) in the community of Neufahrn and in the vicinity (Zweckverband Wasserversorgungsgruppe Freising-Süd 2022). The private households are supplied only with drinking water extracted from the deep wells to ensure a high quality of the water. Whereas the main end user of the process water, extracted from the shallow aquifer, are industrial manufactures and cooling systems (e.g., the research campus Garching). Several potential pressures may result from socio-economic conditions, which are related to:

- generally increasing land use pressure resulting from land use conflicts (e.g. construction of a further runway at the international airport of Munich might cause a further need for more infrastructure, like roads, accommodations, utility services etc.) causing more possible sources of point pollution;
- increasing land use pressure as evidenced by increasing settlement spaces causes increasing sources of point pollution (e.g. leaky sewage systems);
- damaged private sewers can cause a deterioration of the groundwater quality through leakage of wastewater contaminants;
- old industrial locations and sector-specific residuals of possible contaminants pose a risk for the drinking water quality.

Although, the water supplier in this pilot action area complies all thresholds of the German drinking water directive, the utility is interested in providing a high water quality beyond the legal standard.

3. Monitoring methodology and available data

To the best of the authors' knowledge, there is no previous data about PPCP measurements of the aquifers and the surface water in the pilot area. In contrast, data on the flow of the Isar river, meteorological data (temperature, precipitation, and solar radiation and pressure), hydraulic head data of piezometers in and in the vicinity of the pilot area, data of mandatory hydrochemical analysis and operational data (extraction rates of all wells) from the waterwork is available. Additionally, the PPCP monitoring campaign of the boDEREC-CE project completed these data.

3.1. Sampling and laboratory analysis

The boDerec-CE monitoring on all project pilot sites was conducted according to common methodology. The analyses of the collected samples of surface and groundwater were carried out according to valid procedures and EPA method 1694 in the Vltava River Basin Authority laboratory.

Samples were collected in 60 mL amber glass vials (filled only halfway). The samples were stored in a freezer (in an inclined position). They were defrosted at a maximum temperature of 30 °C on the day of analysis. It was necessary to conduct the analysis immediately after defrosting.

One method was developed for the analysis of PPCPs (LC-MS/MS with combinated ESI+ and ESI- mode). The samples of water were centrifuged in headspace vials for 10 min at about 3500 rpm. Subsequently 1.50 g of each sample were weighed in a 2 mL vial on an analytical balance. Then 1.5 µL of acetic acid was added to each sample. An isotope dilution was performed in the next step. Deuterated internal standards of d10-carbamazepine, d6-sulfamethoxazole, d3-iopromide, d3-iopamidol, 13C2-erythromycin, d3-ibuprofen, d4-diclofenac, d3-naproxen, d5-chloramphenicol and others were used.

PPCPs were separated and detected by LC–MS/MS methods based on direct injection of the sample into a chromatograph. A 1290 ultra-high-performance liquid chromatograph (UHPLC) coupled with an Agilent 6495B Triple Quad Mass Spectrometer (MS/MS) of Agilent Technologies, Inc. (Santa Clara, CA, USA) were used.

Method; the separation was carried out on a Waters Xbridge C18 analytical column (100 mm x 4.6 mm, 3.5 µm particle size). The mobile phase consisted of methanol and water with 0.02 % acetic acid and 0.5 mM ammonium fluoride as the mobile phase additives. The flow rate was 0.5 mL/min. The injection volume was 0.050 mL.

The range of analysis and detection limit for each analyte is shown in the table below (**Fehler! Verweisquelle konnte nicht gefunden werden.**).

Each series of samples were verified by calibration control and by maintaining a clean environment, equipment, and agents. The performance of the analytical system was ensured by blank and spiked samples. The chemicals used for the preparation of calibration solutions had a certified purity of 99%. Calibration solutions were prepared from neat analytes or from solutions with certified concentration. Each fifth sample in a series was processed by the method of standard addition, which was 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 were under regular control, and measuring vessels were metrologically tested.

The chemicals used were supplied from renowned manufacturers in the EU and USA: Dr. Ehrenstorfer GmbH (Augsburg, Germany), LGC Ltd. (Teddington, Middlesex, UK), Honeywell International Inc. (Morris Plains, NJ, USA), HPC Standards GmbH (Cunnersdorf, Germany), Absolute Standards Inc. (Hamden, CT, USA), CIL Inc. (Tewksbury, MA, USA), Analytika spol s.r.o. (Prague, Czech Republic).

Table 1. Analysed PPCPs within the monitoring campaign of the pilot action area in Neufahrn bei Freising.

Pharmaceutical and Personal Care Products (PPCP)	Limit of Detection (LOD) [ng/l]
17a-ethynilestradiol	<2.0
17-alpha-estradiol	<1.0
17-beta-estradiol	<1.0
1-H-Benzotriazole	<20.0
1-methyl-1-H-Benzotriazole	<50.0
4-formylaminoantipyrine	<10.0
5-methyl-1-H-Benzotriazole	<20.0
Acebutolol	<10.0
Acesulfame	<50.0
Alfuzosin	<10.0
Amitriptyline	<10.0
Atenolol	<10.0
Atorvastatin	<10.0
Azithromycin	<10.0
Bezafibrate	<10.0
Bisphenol B	<50.0
Bisphenol S	<50.0
Bisoprolol	<10.0
Bisphenol A	<50.0
Butylparaben	<10.0
Caffein	<100
Carbamazepine	<10.0
Carbamazepine 10,11-dihydro-10-hydroxy	<10.0
Carbamazepine 10,11-dihydroxy	<10.0
Carbamazepine 10,11-epoxid	<10.0
Carbamazepine-2-hydroxy	<10.0
Celiprolol	<10.0
Chloramphenicol	<20.0
Ciprofloxacin	<20.0
Citalopram	<20.0
Clarithromycin	<10.0
Climbazole	<10.0
Clindamycin	<10.0
Clofibric acid	<10.0
Cotinine	<20.0

Cyclamate	<100
Cyclophosphamide	<10.0
DEET - diethyltoluamide	<10.0
Diatrizoate	<50.0
Diclofenac	<20.0
Diclofenac-4'-hydroxy	<20.0
Diltiazem	<10.0
Disopyramide	<10.0
Doxycycline	<50.0
Enoxacin	<20.0
Enrofloxacin	<20.0
Eprosartan	<10.0
Erythromycin	<10.0
Estriol	<10.0
Estron	<1.0
Ethylparaben	<10.0
Fexofenadine	<10.0
Fluconazole	<10.0
Fluoxetine	<10.0
Furosemide	<50.0
Gabapentin	<10.0
Gemfibrozil	<10.0
Hydrochlorothiazide	<50.0
Ibuprofen	<20.0
Ibuprofen-2-hydroxy	<30.0
Ibuprofen-carboxy	<20.0
Iohexol	<50.0
Iomeprol	<50.0
Iopamidol	<50.0
Iopromide	<50.0
Irbesartan	<10.0
Ivermectin	<50.0
Ketoprofen	<10.0
Lamotrigine	<10.0
lincomycin	<10.0
Losartan	<10.0
Lovastatin	<10.0
Memantine	<20.0
Metformin	<20.0
Methylparaben	<30.0
Metoprolol	<10.0
Mirtazapine	<10.0
Naproxen	<50.0

Naproxen-O-desmethyl	<20.0
Norfloxacin	<20.0
Norverapamil	<10.0
Octyl methoxycinnamate	<1000
Ofloxacin	<20.0
Oxcarbazepine	<10.0
Oxypurinol	<50.0
Paracetamol	<10.0
Paraxanthine	<100
Penicillin G	<10.0
PFOA	<10.0
PFOS	<5.0
Phenazone	<10.0
Primidone	<10.0
Progesterone	<0.5
Propranolol	<10.0
Propylparaben	<20.0
Propyphenazone	<10.0
Ranitidine	<10.0
Roxithromycin	<10.0
Saccharin	<50.0
Salbutamol	<10.0
Sertraline	<10.0
Simvastatin	<10.0
Sotalol	<10.0
Sucralose	<1000
Sulfamerazine	<10.0
Sulfamethazine	<10.0
Sulfamethoxazole	<10.0
Sulfanilamide	<50.0
Sulphapyridine	<10.0
Telmisartan	<20.0
Testosterone	<0.5
Tiamulin	<10.0
Tramadol	<10.0
Triclocarban	<10.0
Triclosan	<20.0
Trimethoprim	<10.0
Valsartan	<10.0
Valsartan acid	<10.0
Venlafaxine	<10.0
Venlafaxine O-desmethyl	<10.0
Verapamil	<10.0

Warfarin	<10.0

3.2. Objective of monitoring

The objective of the monitoring was to quantify the PPCPs occurrence in the shallow and the deep groundwater as well as in the river Isar. Moreover, the analysis of the different water samples aimed to investigate a potential interaction between the water resources.

The obtained PPCP concentrations can also be used, in a next step, for the development of transport models as well as their calibration and validation process. With these models, potential site specific sources may be identified or at least can be ruled out.

3.3. Sampling

In total, six different sampling points were selected to determine the water quality related to the PPCP concentrations in the pilot action area of “Neufahrn bei Freising”. The samples were withdrawn during the year 2020 and 2021. In Figure 2, the sampling points are depicted and are additionally described in the following.

For the groundwater sampling, the shallow groundwater (3) and the deep groundwater (4) was sampled separately. Additionally, tap water (5) of a random household in Neufahrn was sampled once. To investigate the water quality of the surface water, the river Isar was sampled at two points, at its inflow (1) to the pilot area next to Garching and circa 4 km downstream at the outflow (2) of the pilot area at the Isarbrücke. In addition, the effluent of the wastewater treatment plant (WWTP) (6), which is discharging in the river Isar in the middle of the study area was analysed regarding the PPCP concentrations.

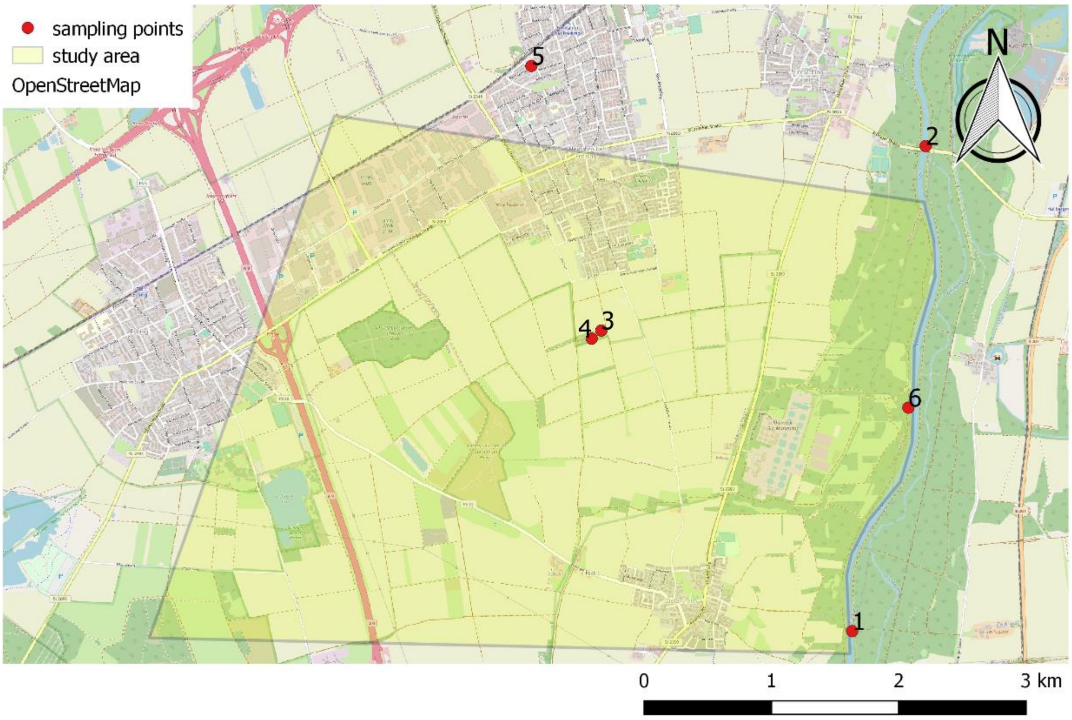

Figure 2 Location of the sampling points (displayed by red dots) in the pilot action area of Neufahrn bei Freising, created with © OpenStreetMaps.

Individual sampling points are characterised as follows:

Point 1: Isar - Garching	
Objective: obtain information on the quality of surface water from the Isar river, upstream at the inflow of the river to the pilot area.	
Method of sampling: sampling directly from surface water.	

Point 2: Isar - Isarbrücke	
Objective obtain information on the quality of surface water from the Isar river, downstream at the outflow of the river to the pilot area.	
Method of sampling: sampling directly from surface water.	

Point 3: shallow groundwater	
Objective: get information on groundwater quality in the upper quaternary aquifer, which is hydraulic connected to the river and used as process water.	
Method of sampling: sampling in one of the three shallow extraction wells in a dynamic state.	

Point 4: deep groundwater	
Objective: get information on groundwater quality in the deeper tertiary aquifer, which is used as drinking water.	
Method of sampling: the sample is taken from a deep well in a dynamic state.	

Point 5: tap water	
Objective: get information on the tap water in the supplied area, represented by one household.	
Method of sampling: the sample is taken from the water tap installed in the kitchen of a household in Neufahrn.	

Point 6: WWTP effluent	
Objective: get information on the WWTP effluent, discharging in the river Isar in the study area.	
Method of sampling: the sample is taken from the discharge after the last treatment step of the WWTP.	

4. Monitoring results and discussion

In this section, selected PPCP concentrations are presented and discussed. To interpret and analyse the sampling results, a mean value of all detections for one sampling point (between 2020 and 2021) was calculated, the extreme values (maximum and minimum) were determined and the detection frequency throughout the samplings was investigated. These values are given for the compounds 4-formylaminoantipyrene (4-FAA), 5-methylbenzotriazole benzotriazole, DEET, diatrizoate, diclofenac, gabapentin, iohexol, iomeprol, lamotrigine, metformin, oxypurinol, telmisartan, valsartan and valsartan acid in Table 2. Moreover, the mean values are visualized in Figure 3. Note that a logarithmic scale was used to display the observed range of 5 orders of magnitude. For the selection, it was aimed to obtain the compounds with a detection frequency of 100 % in the WWTP effluent and to investigate the compounds, which were detected in the shallow wells in more than one sample. All project related monitoring data are gathered in deliverable D.T2.3.1. Further, the raw data can be found in the monitoring database in D.T2.3.2.

Table 2 Mean value]ng/l], maximum and minimum detected concentration [ng/l] and the detection frequency [%] of selected PPCPs for the different sampling points.

	Isar - Garching				Isar - Isarbrücke				WWTP effluent				Shallow well				Deep well			
	Mean	Max.	Min.	DF	Mean	Max.	Min.	DF	Mean	Max.	Min.	DF	Mean	Max.	Min.	DF	Mean	Max.	Min.	DF
4-FAA	27.1	40.8	17	100	128	220	70	100	2398	4460	1160	100	-	-	-	0	-	-	-	0
5-methylbenzotriazole	25.2	27.8	22	67	156	238	38	100	2432	4250	1130	100	-	-	-	0	-	-	-	0
Benzotriazole	60	82	37	100	320	490	70	100	5300	8420	1840	100	-	-	-	0	-	-	-	0
DEET	52	62	43	33	42	62	19	83	122	404	15	100	25	43	14	50	21	30	13	50
Diatrizoate	81	168	57	100	176	335	55	100	560	1010	263	100	73	95	56	50	-	-	-	0
Diclofenac	-	-	-	0	67	113	21	100	1250	1960	643	100	-	-	-	0	-	-	-	0
Gabapentin	21	31	17	100	102	163	32	100	1010	1470	602	100	-	-	-	0	-	-	-	0
Iohexol	72	90	52	100	205	323	88	100	2426	5970	516	100	-	-	-	0	-	-	-	0
Iomeprol	-	-	-	0	245	583	52	100	6506	16000	1800	100	-	-	-	0	-	-	-	0
Lamotrigine	13	16	11	50	52	66	14	100	800	960	620	100	-	-	-	0	-	-	-	0
Metformin	40	57	24	100	99	170	39	100	1221	2660	646	100	-	-	-	0	-	-	-	0
Oxypurinol	72	111	52	66	476	725	231	66	11184	15100	7340	100	71	89	52	100	-	-	-	0
Telmisartan	-	-	-	0	47	67	22	100	661	850	393	100	-	-	-	0	-	-	-	0
Valsartan	22	39	12	100	127	319	26	100	1775	3720	355	100	-	-	-	0	-	-	-	0
Valsartan acid	22	32	15	83	50	129	25	100	421	1240	127	100	-	-	-	0	-	-	-	0

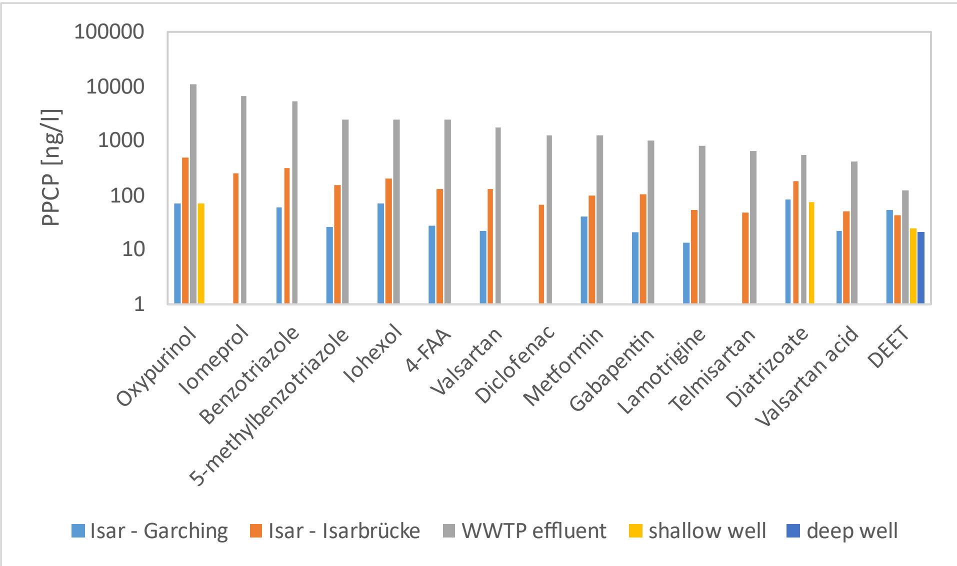

Figure 3. Mean concentrations [ng/l] of selected PPCPs in the river (blue and orange), the WWTP effluent (grey) and the wells (yellow and blue).

As it can be seen in Table 2 and in Figure 3, the deeper aquifer is almost free of any measured PPCPs. Only the insect repellent DEET can be found in 50 % of the samples at very low concentrations (< 30 ng/l)close to the detection limit (20 ng/l). Since the concentration of DEET in the lower ng/l range very unlikely exhibit an adverse effect on the ecology and on human health (Schriks et al. 2010), there is no need for any concern about the drinking water quality. In the analysed tap water, none of the investigated PPCPs were detected in Neufahrn. As the tap water origins from the deep aquifer, these results should be in agreement.

In the shallow aquifer, in contrast, low concentrations of some PPCP were observed. For example, oxypurinol was analysed in each sample. It is a metabolite of the pharmaceutical allopurinol (Funke et al. 2015). In the shallow well it showed a mean concentration of 71 ng/l. The insect repellent DEET and the X-ray contrast agent diatrizoate were present in 50 % of the samples at a mean concentration of 25 ng/l and 73 ng/l. Additionally, bisphenol A, benzotriazole, caffeine, PFOS and salbutamol were detected once. The one-time detection might occur due to contaminations during the sampling, the preparation or the analysis of the samples. For instance, the caffeine concentration could be related to an exhalating person, who had consumed coffee before. Besides this, bisphenol A as a plasticizer, could be present in the sampling, if any plastic material was used for the sampling or the analysis. As the detection limits are very low (ng/l), the analysis is very sensitive regarding any contamination. Moreover, the seasonal usage of PPCPs may lead to different concentrations over the year. As benzotriazole can also be used as a de-icing agent in winter, a higher release to the environment is expected.

In contrast to the very low concentrations in the extraction wells, the PPCP concentrations in the river were higher. The observed concentrations were below 100 ng/l. Although, they are in the same order of magnitude as the PPCP concentrations detected in the shallow aquifer, many more compounds can be found in the river water. For example, gabapentin, iohexol, metformin and valsartan were detected in each river sample in Garching, but not in the shallow groundwater.

At 4 km downstream (Isarbrücke), the measured PPCP concentrations were increased up to one order of magnitude, in comparison to the upstream located sampling point in Garching. Moreover, additional PPCPs can be found there. For example, the pharmaceuticals diclofenac, iomeprol and telmisartan were not present in the samples withdrawn in Garching. This observed difference can very likely be explained by the PPCP containing effluent of the WWTP. To further investigate this hypothesis, the effluent of the WWTP was analysed.

The effluent exhibited the highest measured PPCP concentrations of all sampling campaigns. The maximum PPCP concentration was observed for iomeprol (16 µg/l) and oxypurinol (15 µg/l). Compared to the downstream PPCP concentrations, these are one order of magnitude higher. As the WWTP is not upgraded with an advanced water treatment step (e.g.; ozonation or activated carbon), the high PPCP concentrations were to be expected.

Compared to other drinking water sources in the boDEREC-CE project, the PPCP concentrations in the shallow groundwater wells are negligible. However, the concentrations in the river downstream of the wastewater treatment plant is comparable to detected concentrations in other surface water sources in this project.

5. Conclusion

The monitoring data showed the occurrence of several PPCPs in the aquatic environment for the pilot action area in “Neufahrn bei Freising”. The concentration levels were the highest for the WWTP effluent, followed by the surface water (downstream > upstream) and the groundwater (shallow > deep aquifer). The tap water was completely free of any PPCPs. Thus, there is no need to remove PPCPs from the extracted water before supplying it as drinking water in this pilot action area.

The different fingerprints of the water bodies showed that transport processes have a substantial impact on the PPCPs’ behaviour in the study area. However, the monitoring dataset is too small to describe the PPCPs’ behaviour adequately. Thus, flow and transport models are required to further investigate the PPCPs’ behaviour. Especially, the occurrence of oxypurinol, DEET and diatrizoate in the shallow groundwater needs to be explained. Potential sources may be investigated by flow and transport models. The PPCP modelling for this case study is applied in the WP T3.

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