Relation between pharmaceutical consumption, environmental pharmaceutical burdens and current treatment technologies

Project MORPHEUS 2017 - 2019
Deliverable 4.2

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Key facts of the MORPHEUS project

MORPHEUS (Model Areas for Removal of Pharmaceutical Substances in the South Baltic) is a project financed by the European Union Interreg South Baltic Programme. The project duration is January 2017 – December 2019, with a total budget of EUR 1.6 million with a contribution from the European Regional Development Fund of EUR 1.3 million. The project has a total of 7 partners from four countries; Sweden, Germany, Poland and Lithuania: Kristianstad University (Lead Partner) – Sweden, EUCC – The Coastal Union Germany – Germany, University of Rostock – Germany, Gdansk Water Foundation – Poland, Gdansk University of Technology – Poland, Environmental Protection Agency – Lithuania and Klaipeda University – Lithuania. The project also has a total of 11 associated partners from these countries. For additional information on the project and activities please visit the MORPHEUS homepage at: www.morpheus-project.eu

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1 Introduction

The aim of this report called “Relation between pharmaceutical consumption, environmental pharmaceutical burdens and current treatment technologies” is to identify key linkages between pharmaceutical consumption data (WP3), monitoring data (WP4) and available treatment technologies (WP5). Based on previous findings and additional research, these key linkages will combine two different approaches of analysing the release of pharmaceuticals to the environment. On the one hand, a top-down approach, evaluating the emissions of pharmaceuticals starting at the source by collecting region-specific consumption data of selected pharmaceuticals and available human excretion data of these pharmaceuticals. On the other hand, a bottom-up approach, taking wastewater samples at the inlet of the 15 selected WWTPs and determining the actual burden of pharmaceuticals reaching the WWTP by performing chemical analysis of the samples. Additionally, taking wastewater samples at the outlet of the WWTPs made it possible to calculate the pharmaceutical specific removal efficiencies at the WWTPs in all four model areas in Lithuania, Germany, Poland and Sweden. Finally, the detailed inventory of existing treatment technologies gave information on similarities and differences in treatment characteristics between the WWTPs and allowed a first glance at potential correlations between technologies and removal efficiencies.

In WP3, a comprehensive data research (for the year 2015) was performed and prioritized. Region-specific pharmaceutical consumption loads have been calculated for the model areas in Lithuania, Poland, Sweden and Germany (results available in Deliverable 3.1: “Report on Pharmaceutical consumption patterns in four coastal regions of the South Baltic Sea, Germany, Sweden, Poland and Lithuania”). In order to predict the actual pharmaceutical load reaching the inflow of a specific WWTP, the excretion and the size of the WWTP have to be included. The excretion rate describes the share of a consumed pharmaceutical dose, which is not metabolized, but released unchanged by a human body via urine and/or faeces. Despite some average rate suggestions in literature, it is well known that excretion is substance specific. The size of a WWTP is normally identified by Personal Equivalents (PE) describing the wastewater inflow but is not suitable here since other wastewater sources than domestic ones are included, too. Therefore, the connected inhabitants of each WWTP were queried in the inventory of WP5. Moreover, all detailed information regarding WWTP technology applied in the model areas are summarised in Deliverable 5.1: “Inventory of existing treatment technologies in wastewater treatment plants, Case studies in four coastal regions of the South Baltic Sea Poland, Sweden, Lithuania and Germany”. By including all this information, it was possible to calculate the **Predicted Incoming Load – PIL** to individual WWTPs.

In WP4, sampling and further analysis of pharmaceutical concentrations have been conducted in receiving water bodies and in the inflow and outflow of model WWTPs in the summer of 2017 and winter of 2018 (results available in Deliverable 4.1: “Report on Determination of the Regional Pharmaceutical Burden in 15 Selected WWTPs and Associated Water Bodies using Chemical Analysis, Status in four coastal regions of the South Baltic Sea; Germany, Sweden, Poland and Lithuania”).

Using available analytical chemical data on wastewater inlet concentrations (the average of a summer and a winter sample) in combination with the WWTP inventory of WP5 it was possible to calculate the **Measured Incoming Load – MIL** for individual WWTPs.

By comparing the PIL value with the MIL value it was possible to study whether regional consumption data could be used to predict the actual incoming load to the selected WWTPs. Additionally, the obtained removal efficiencies (based on chemical analysis of inlet and outlet wastewater at the WWTPs in WP4) were directly compared to WWTP characteristics such as sludge age and applied technological solutions to investigate any possible relation between these characteristics and removal efficiency.

In this report, four pharmaceuticals have been selected according to the results of Deliverable 3.1 and 4.1. They represent different therapeutic classes and were either high in regional consumption or revealed the highest loads in WWTPs effluents in the studied model areas:

- **Azithromycin** (J - Antiinfectives for systemic use)
- **Carbamazepine** (N - Nervous system)
- **Diclofenac** (M - Muscolo-skeleton system)
- **Metoprolol** (C - Cardiovascular system)

2 Schematic overview and theoretical background

Combining top-down and bottom-up approaches of the WPs can help reveal the key linkages between pharmaceutical consumption data and analytical monitoring data under consideration of treatment techniques of the investigated WWTPs. Figure 1 shows the schematic overview of this report.

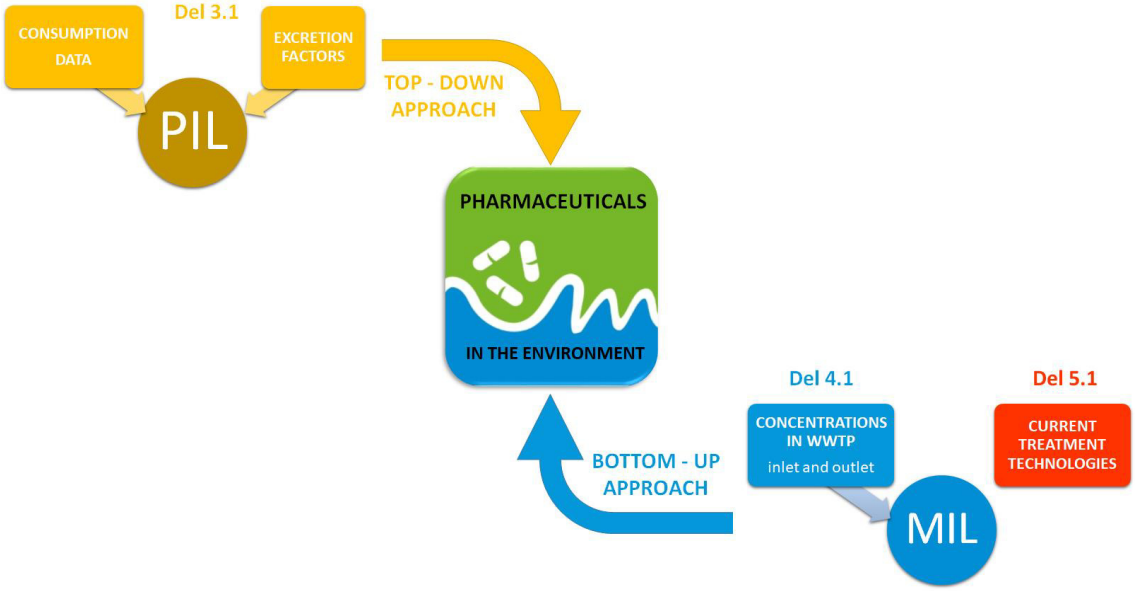

Figure 1. Schematic overview of the content of this report.

The predicted inflow loads (expressed as Predicted Incoming Load - PIL) and measured inflow loads (expressed as Measured Incoming Load - MIL) are the two parameters that were compared after calculation. PIL and MIL can be calculated according to Equation 1 and Equation 2, respectively:

- **PIL [kg/a]** = intake [g/inh./a] * excretion rate [%] * connected inhabitants of WWTP[-] /1000 Eq. 1
- **MIL [kg/a]** = c (inflow) [ng/L] * Qww [m³/a] *10^{-9} Eq. 2

In WP4, the inflow concentrations (ng/L) were already determined and were used to calculate MIL values, while the calculation of PIL required a literature review on excretion rates for selected pharmaceuticals. The comparison of MIL and PIL based on inlet loads has been conducted for the four selected pharmaceuticals Azithromycin, Carbamazepine, Diclofenac and Metoprolol, and completed for the 15 model WWTPs.

Additionally, the information available on both inlet and outlet concentrations provided the possibility to calculate the removal efficiency of the WWTPs as performed in Del. 4.1 according to Equation 3:

- **Removal efficiency** = ((Inlet conc.- Outlet conc.) / Inlet conc.) * 100% Eq. 3

By comparing these data with information about existing WWTPs technologies in the model areas (provided in Del. 5.1) potential correlations between removal efficiencies and technologies can be identified. Wastewater treatment technologies in the model areas are mostly based on the activated sludge system. There is still lack of information regarding pharmaceutical removal in the context of applied technology and technology operation parameters. For comparison of pharmaceutical removal efficiencies at different WWTPs the following characteristics were applied: number of connected inhabitants, average wastewater daily flow, sludge age, sludge digestion and technologies applied at the WWTPs. Detailed description of model WWTPs technologies in Lithuania, Germany, Poland and Sweden is available in Del 5.1.

3 Data Collection

For the identification of key linkages between WP3, WP4 and WP5, the following data sets from the model areas have been applied:

- regional consumption of pharmaceuticals per inhabitant in 2015 (Deliverable 3.1);
- excretion rates of pharmaceuticals (data from literature);
- pharmaceutical inflow loads of model WWTPs (Deliverable 4.1);
- pharmaceutical removal efficiencies of model WWTPs (Deliverable 4.1);
- model WWTP characteristics (Deliverable 5.1)

Summarized information of used data from all the four model areas in Lithuania (LT), Germany (GER), Poland (PL) and Sweden (SE) are presented below in Table 1 to Table 5.

Table 1. Regional consumption per inhabitant in 2015 (mg/inh./a). Data from Deliverable 3.1.

Pharmaceutical	Azithromycin	Carbamazepine	Diclofenac	Metoprolol
LT	7.6	204	258	619
GER	82	893	616	1 796
PL	73	716	210	82
SE	9.8	527	334	1 441

Table 2. Excretion rates (%) and corresponding sources. Data from literature.

Pharmaceutical	Azithromycin	Carbamazepine	Diclofenac	Metoprolol
Excretion rate [%]	50	14	15	10
Literature source	Besse et al. (2008)	Lienert et al. (2007) Björlenius et al. (2018)	Lienert et al. (2007) Ternes (1998)	Lienert et al. (2007) Ternes (1998)

Table 3. Measured incoming loads (MIL) of model WWTPs (kg/a) calculated using Eq. 2 above. The concentrations were determined by results from the chemical analysis performed in Deliverable 4.1, while the flow of wastewater was known from Deliverable 5.1 and are shown in Table 5 below.

MIL – Measured Incoming Load [kg/a]		Azithromycin	Carbamazepine	Diclofenac	Metoprolol
LT WWTPs	Klaipeda	4.2	6.5	41.0	20.7
	Palanga	0.4	0.8	5.5	3.3
	Kretinga	0.6	0.2	6.2	2.9
	Nida	<0.01	0.1	0.8	0.2
GER WWTPs	Rostock	28.3	13.9	50.4	43.8
	Laage	0.8	0.4	1.2	1.0
	Krakow	1.8	0.7	1.0	1.6
	Satow	0.4	0.1	0.3	0.3
PL WWTPs	Gdansk-Wschod	265	67.7	105	37.6
	Gdynia-Debogorze	248	37.4	76.2	22.0
	Swarzewo	15.4	4.06	13.0	1.69
	Jastrzębia-Góra	4.09	0.66	1.65	0.46
SE WWTPs	Kristianstad	1.51	5.24	5.2	7.33
	Tollarp	<0.01	0.08	0.14	0.32
	Degeberga	0.01	0.41	0.14	0.27

Table 4. Pharmaceutical removal efficiencies in model WWTPs (%). These were calculated in Deliverable 4.1 using Eq. 3 above. The concentrations were determined by chemical analysis of incoming and outgoing wastewater at each WWTP.

Removal efficiencies [%]		Azithromycin	Carbamazepine	Diclofenac	Metoprolol
LT WWTPs	Klaipeda	71.0	-8.3	20.1	22.7
	Palanga	74.2	-31.7	0.7	19.2
	Kretinga	95.6	2.6	32.9	67.5
	Nida	-73.3	-73.6	-20.0	-3.7
GER WWTPs	Rostock	95.1	-19.4	39.5	51.1
	Laage	95.6	21.1	66.1	83.5
	Krakow	81.7	6.3	39.0	66.0
	Satow	98.6	37.8	50.9	64.5
PL WWTPs	Gdansk-Wschod	33.9	41.4	13.7	17.6
	Gdynia-Debogorze	-102.7	44.4	14.4	14.2
	Swarzewo	75.9	51.9	62.2	32.6
	Jastrzębia-Góra	51.1	-24.3	42.95	44.1
SE WWTPs	Kristianstad	73.8	12.0	7.4	22.7
	Tollarp	30.0	-18.1	-68.1	-4.1
	Degeberga	96.2	15.4	16.3	93.8

*Table 5. Model WWTP characteristics for PIL calculations and correlation with removal efficiencies (%). Data from Deliverable 5.1. Abbreviations: AO - anaerobic/oxic, A2/O - anaerobic/anoxic/oxic system, DN-denitrification, N-nitrification, UTC - University of Cape Town concept, P - phosphorus, SBR- sequencing batch reactors; *summer season, **out of the season.*

WWTP characteristics		Connected inhabitants	Average flow in 2015 Q [m^3 /day]	WWTP technology	Sludge age [days]	Digestion of sludge	Removal efficiencies [%]			
							AZI*	CAR*	DIC*	MET*
LT WWTPs	**Klaipėda**	170 000	41013	UCT	22	YES	71	-8.3	20	23
	Palanga	13 000	7552	A2O chemical P	32	NO	74	-32	0.7	19
	Kretinga	18 127	3576	AO	22	NO	96	2.6	33	68
	Nida	1 700	620	AO	27	NO	-73	-74	-20	-3.7
GER WWTPs	**Rostock**	235 645	42314	UCT + BIOFOR-N/DN	11	YES	95	-19	40	51
	Laage	4 516	880	Convention. N/DN	43	NO	96	21	66	84
	Krakow	3 964	630	Convention. N/DN	35	NO	82	6.3	39	66
	Satow	1 303	218	SBR with a downstream clarification pond	no data	NO	99	38	51	65
PL WWTPs	**Gdansk-Wschod**	571 350	92958	A2O	14 - 28	YES	34	41	14	18
	Gdynia-Debogorze	360 000	55294	Bardenpho with Carussel system (simultaneous DN)	29	YES	-103	44	15	14
	Swarzewo	35 668	6164	SBR N/DN	63	YES	76	52	62	33
	Jastrzebia-Gora	10 000	1678	modified Bardenpho; UV disinfection of final effluent	9** 24 ***	NO NO	51	-24	43	44
SE WWTPs	**Kristianstad**	52 000	22427	N/DN chemical P	13	YES	74	12	7.5	23
	Tollarp	3 400	989	N/DN chemical P	no data	NO	30	-18	-68	-4
	Degeberga	880	216	Convent. N/DN chemical P	8	NO	96	16	16	94

4 Calculation of PIL and comparison with MIL

In order to compare the MIL values (see Table 3) with PIL values, the PIL values had to be calculated according to Eq. 1 above. The required data include intake [g/inh./a], excretion rate [%] and connected inhabitants of WWTP[-] /1000 which are available in Table 1, Table 2 and Table 5, respectively. MIL values (from Table 3) are listed along with the calculated PIL values in Table 6 below.

Table 6. Comparison of MIL (Table 3) and calculated PIL using Eq. 1 and data from Table 1, Table 2 and Table 5.

Average inflow loads [kg/a]		**Azithromycin**		**Carbamazepine**		**Diclofenac**		**Metoprolol**	
	WWTP	**MIL**	**PIL**	**MIL**	**PIL**	**MIL**	**PIL**	**MIL**	**PIL**
LT	Klaipeda	4.19	0.65	6.50	4.86	40.99	6.55	20.71	10.51
	Palanga	0.43	0.05	0.75	0.37	5.50	0.50	3.32	0.80
	Kretinga	0.55	0.07	0.23	0.52	6.16	0.70	2.86	1.12
	Nida	<0.01	0.01	0.07	0.05	0.81	0.07	0.16	0.11
DE	Rostock	28.32	9.66	13.91	29.45	50.42	21.78	43.77	42.32
	Laage	0.78	0.19	0.42	0.56	1.19	0.42	0.97	0.81
	Krakow	1.81	0.16	0.72	0.50	1.03	0.37	1.58	0.71
	Satow	0.39	0.05	0.07	0.16	0.26	0.12	0.30	0.23
PL	Gdansk-Wschod	265.31	20.86	67.69	57.27	104.69	18.00	37.60	4.69
	Gdansk-Debogorze	247.72	13.14	37.43	36.09	76.24	11.34	21.98	2.95
	Swarzewo	15.35	1.30	4.06	3.58	12.97	1.12	1.69	0.29
	Jastrzebia-Gora	4.09	0.37	0.66	1.00	1.65	0.32	0.46	0.08
SE	Kristianstad	1.51	0.25	5.24	3.83	5.20	2.61	7.33	7.49
	Tollarp	<0.01	0.02	0.08	0.25	0.14	0.17	0.32	0.49
	Degeberga	0.01	<0.01	0.41	0.06	0.14	0.04	0.27	0.13

A better visualisation of MIL and PIL values for each pharmaceutical are presented in the figures below. Before proceeding, the WWTPs were sorted according to size in terms of number of connected inhabitants from lowest to highest as shown in Figure 2 (note the logarithmic scale). This was assumed to also given an increase in both MIL and PIL values from left to right in Figure 3.

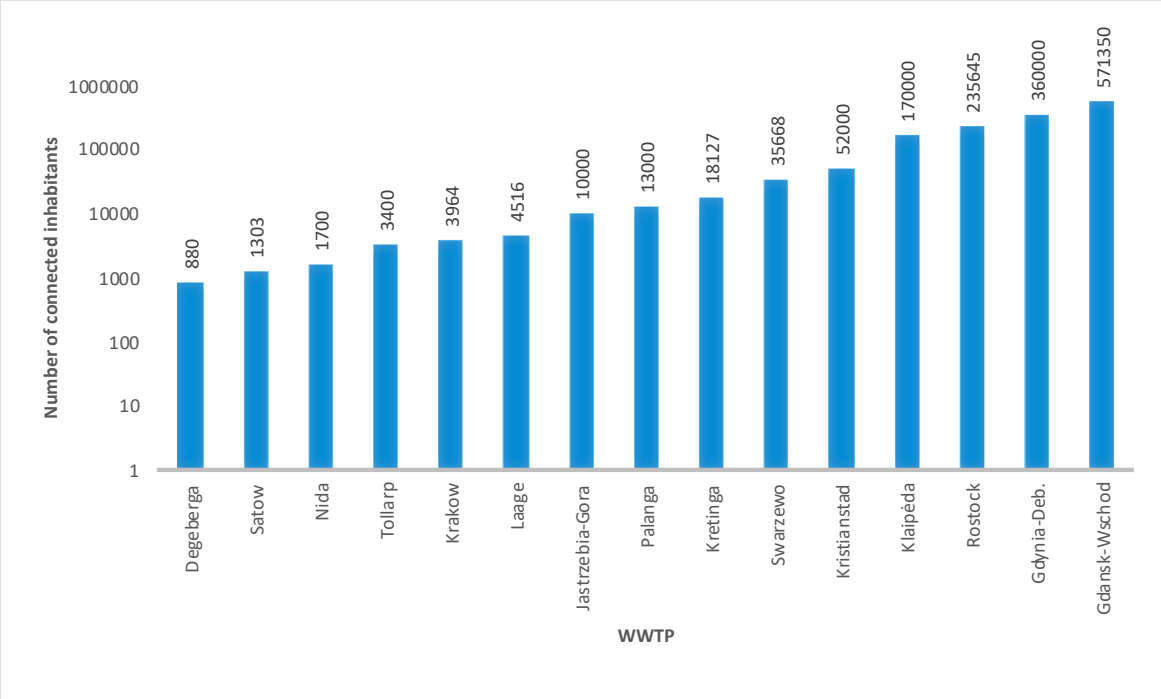

Figure 2. WWTPs sorted according to size in terms of number of connected inhabitants from lowest to highest (note the logarithmic scale).

The results from comparison of MIL and PIL values are shown in Figure 3.

The first general observation to be made is that in most cases the predicted values of analysed pharmaceuticals are lower in comparison to the measured values, since the graph clearly shows that MIL values nearly always exceeds PIL values.

The second general observation to be made is that of the four investigated pharmaceuticals Carbamazepine and Metoprolol showed the best overall correspondence between consumption and occurrence data in most WWTPs. For Azithromycin and Diclofenac, the differences between MIL and PIL values were much larger. It is worth noting that both Carbamazepine and Metoprolol are characterized by the lowest excretion rates, even though Diclofenac also has a low value (Table 2). To easier see the actual differences between the MIL and the PIL values the MIL/PIL ratio was calculated. In Figure 4 the results are shown and the WWTPs are once again listed from smallest to largest to study any possible relation between size of WWTP and MIL/PIL ratio.

<20 000 inh. >30 000 inh.

Figure 3. Comparison of MIL and PIL values for Azithromycin, Carbamazepine, Diclofenac and Metoprolol for the 15 WWTPs. The WWTPs are listed from lowest to highest number of connected inhabitants as shown in Figure 2. The graphs are split in two parts due to large differences in inflow loads.

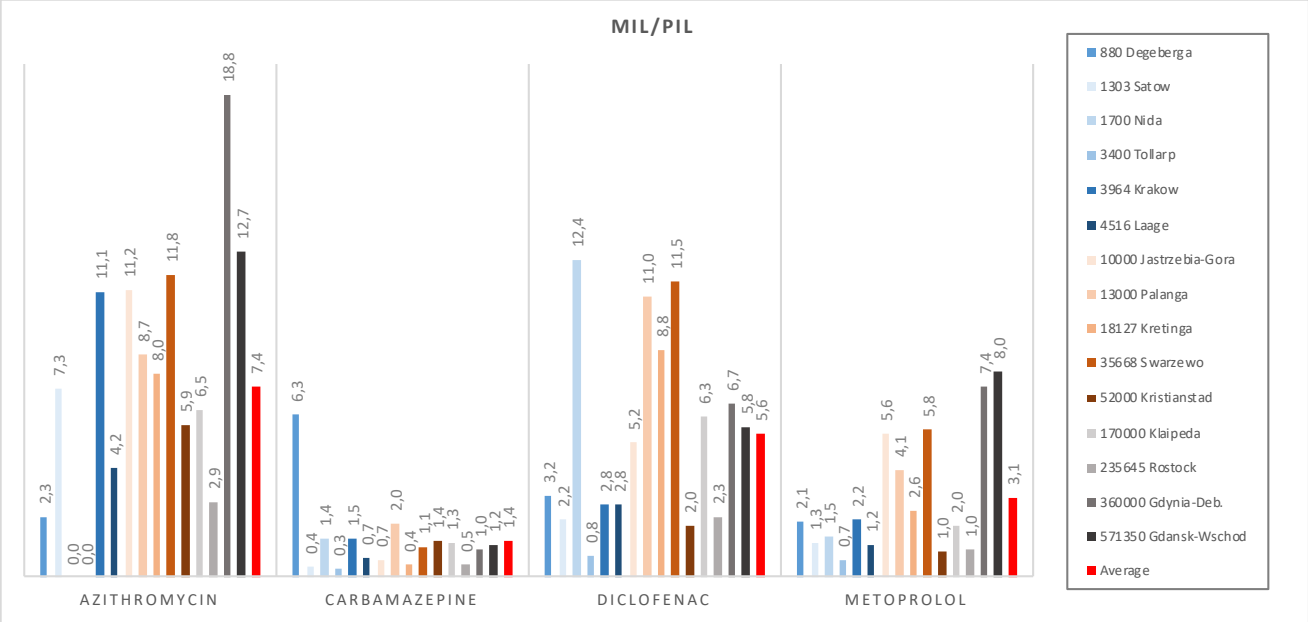

Figure 4. Calculated MIL/PIL ratios for Azithromycin, Carbamazepine, Diclofenac and Metoprolol. The WWTPs are listed from lowest to highest number of connected inhabitants.

Azithromycin showed the best MIL/PIL ratio for Degeberga 2.3 (880 inh.) and Rostock 2.9 (235 645 inh.), while the highest MIL/PIL ratio was observed for Gdynia-Deb. 18,8 (360 000) followed by Gdansk-Wschod 12.7 (571 350 inh.). The average MIL/PIL ratio was 7.4, but with a wide span. This was also the highest average ratio of all pharmaceuticals. There was no clear trend that the size of the WWTP had any influence on the MIL/PIL ratio. The linear relation between MIL and PIL values is shown in Figure 5.

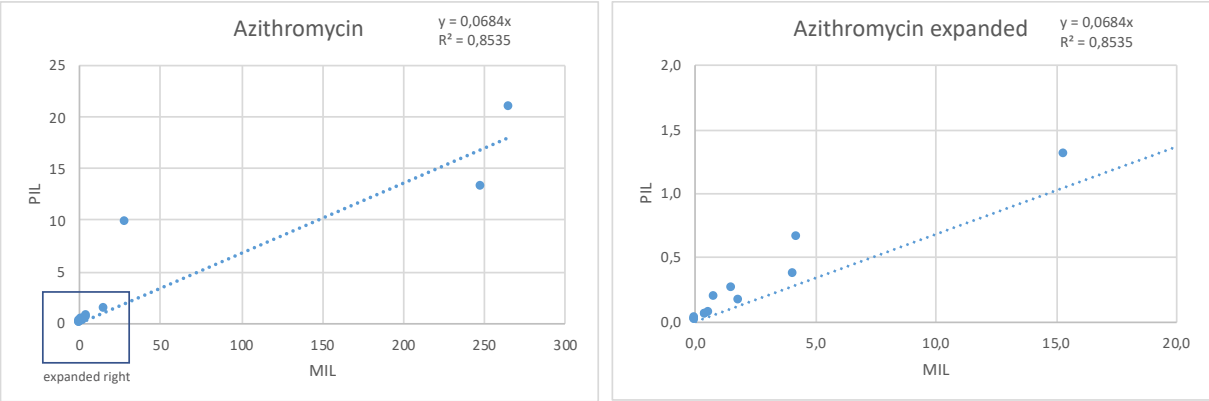

Figure 5. Linear relation between MIL and PIL for Azithromycin.

The Azithromycin relation is described by the function PIL=0.0684*MIL with an R^2 of 0.8535.

The MIL/PIL ratios of Carbamazepine were in general very close to 1.0. The only main exception was the smallest WWTP in Degeberga (880 inh.) with a high MIL/PIL value of 6.3. Here it can be noted, that high Carbamazepine concentrations have been observed in Degeberga at several occasions during chemical analysis, meaning that there possibly is a specific source in Degeberga, which will have a large effect on the incoming concentrations to this small WWTP. Excluding this WWTP, the best MIL/PIL ratios of Carbamazepine were identified in Gdynia-Deb. 1.0 (360 000 inh.) and Swarzewo 1.1 (35 668 inh.), while those with the least fit between MIL/PIL were in Tollarp 0.3 (3 400 inh.) and Palanga 2.0 (13 000 inh.). The average MIL/PIL ratio was 1.4 with a relatively narrow span. This was also the lowest average ratio of all pharmaceuticals. Once again, there was no obvious trend that the size of the WWTP had any influence on the MIL/PIL ratio. The linear relation between MIL and PIL values is shown in Figure 6.

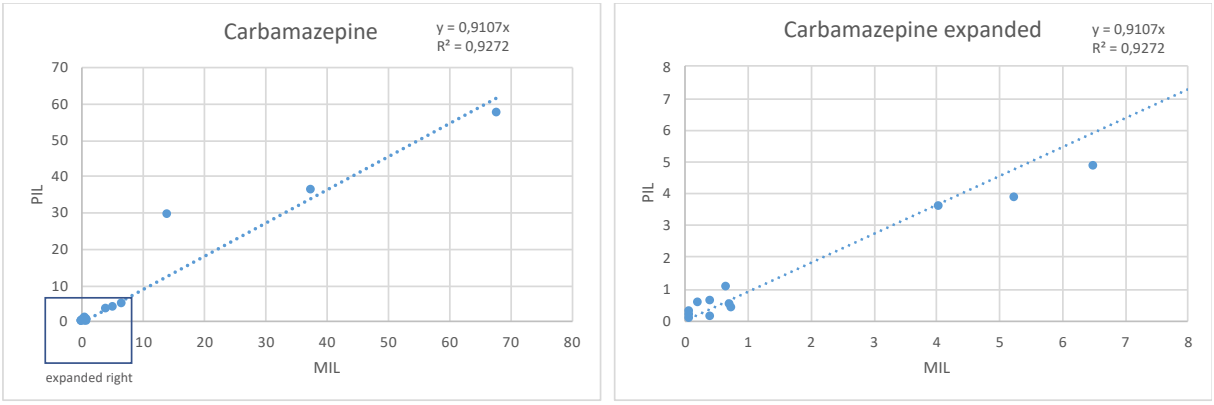

Figure 6. Linear relation between MIL and PIL for Carbamazepine.

The Carbamazepine relation is described by the function PIL=0.9107*MIL with an R^2 of 0.9272.

For Diclofenac the best MIL/PIL ratios were seen in Tollarp 0.8 (3 400 inh.) and Kristianstad 2.0 (52 000 inh.). The highest MIL/PIL ratio were observed in Nida 12.4 (1 700 inh.) and Swarzewo 11.5 (35 668 inh.). These ratios were almost as high as those observed for Azithromycin. The average MIL/PIL ratio was 5.6 and somewhat lower than that observed for Azithromycin, but the second highest of all four pharmaceuticals. The span was also relatively high, but somewhat smaller than for Azithromycin. Just as was the case for Azithromycin and Carbamazepine there was no clear trend that the size of the WWTP had any influence on the MIL/PIL ratio. One could possibly argue that, apart from the high value observed in Tollarp (12.4), the smaller WWTPs (<5000 inhabitants) showed MIL/PIL ratios that were somewhat more homogenous, but this is only vaguely indicated. The linear relation between MIL and PIL values is shown in Figure 7.

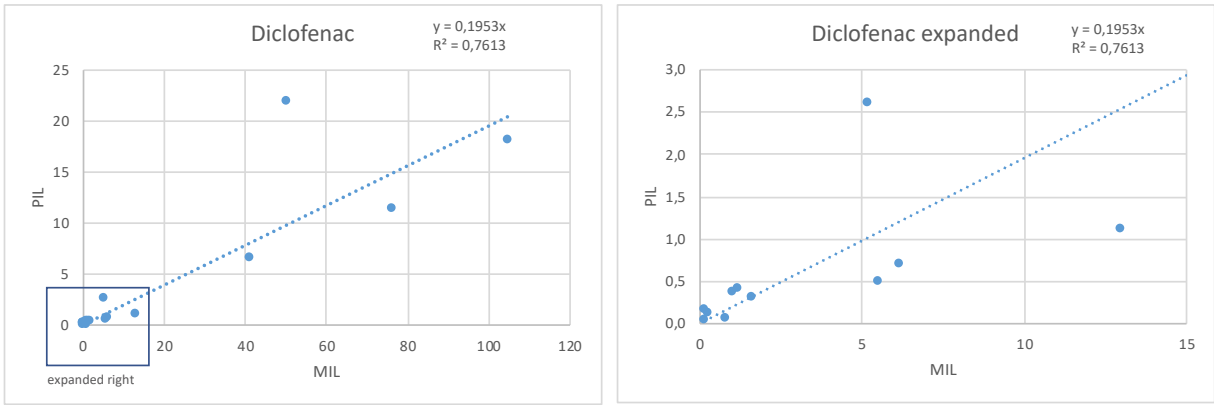

Figure 7. Linear relation between MIL and PIL for Diclofenac.

The Diclofenac relation is described by the function PIL=0.1953*MIL with an R^2 of 0.7613.

The MIL/PIL ratios of Metoprolol ranged between 0.7 to 3.1 for 10 of the WWTPs, while 5 WWTPs had MIL/PIL ratios between 4.1 to 8.0. The best MIL/PIL ratios of Metoprolol were observed in Kristianstad 1.0 (52 000 inh.) and Rostock 1.0 (235 645 inh.), while the highest MIL/PIL ratios were seen in Gdynia-Deb. 7.4 (360 000 inh.) and Gdansk-Wschod 8,0 (571 350 inh.). The average MIL/PIL ratio was 3.1 with a narrower span than those observed for Azithromycin and Diclofenac, but not as small as that observed for Carbamazepine. This was also the second lowest average ratio of all pharmaceuticals. For Metoprolol there was a weak trend that the predicted values were better in line with measured values for WWTPs with <5000 inhabitants. The linear relation between MIL and PIL values is shown in Figure 8.

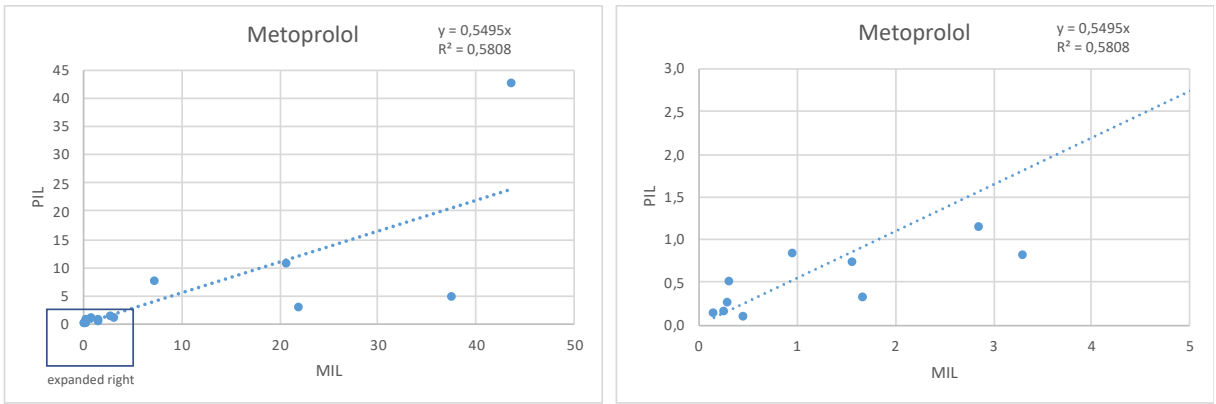

Figure 8. Linear relation between MIL and PIL for Metoprolol.

The Metoprolol relation is described by the function PIL=0.5495*MIL with an R^2 of 0.5808.

Based on the above results Carbamazepine is a good candidate to be used as predictor of expected chemical load to a WWTP using consumption data in a certain region.

5 Relation between WWTP characteristics and removal efficiency

In the project a detailed inventory of existing treatment technologies in the 15 WWTPs were gathered and presented in Deliverable 5.1: "*Inventory of existing treatment technologies in wastewater treatment plants Case studies in four coastal regions of the South Baltic Sea Poland, Sweden, Lithuania and Germany*". A summary of these are presented in Table 5 above and include number of connected inhabitants, average flow Q [m^3/day) WWTP technology, sludge age and sludge digestion. This information can now be compared with knowledge about removal efficiencies presented in Table 4 above which comes from Deliverable 4.1: "*Report on Determination of the Regional Pharmaceutical Burden in 15 Selected WWTPs and Associated Water Bodies using Chemical Analysis, Status in four coastal regions of the South Baltic Sea; Germany, Sweden, Poland and Lithuania"*. The results from this comparison is discussed below to identify possible relations between these WWTP parameters and removal efficiency of the four pharmaceuticals. In Figure 9 the removal efficiency of the four pharmaceuticals at the 15 WWTPs is graphically shown. The data are sorted from highest to lowest removal efficiency based on Azithromycin data. Each country is colour coded; Germany light blue, Sweden light green, Poland dark green and Lithuania dark blue. The average removal efficiency for all WWTPs is shown in *italic* at the bottom of the graph.

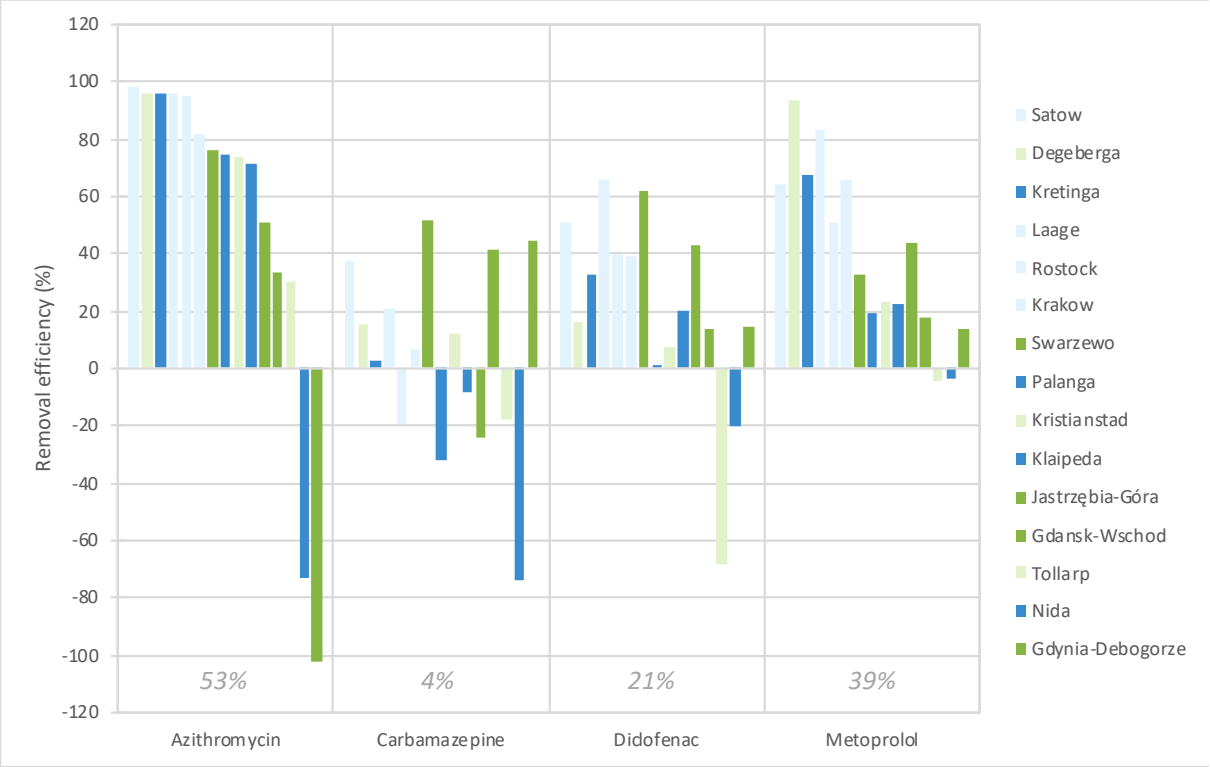

Figure 9. Removal efficiency (%) of the four pharmaceuticals at the 15 WWTPs. The data are sorted from highest to lowest removal efficiency based on Azithromycin data. Each country is colour coded; Germany light blue, Sweden light green, Poland dark green and Lithuania dark blue. The average removal efficiency for all WWTPs is shown in italic at the bottom of the graph.

The highest average removal efficiency was seen for Azithromycin with 53%, followed by Metoprolol 39%, Diclofenac 21% and lastly Carbamazepine 4%. However, there are large differences between WWTPs and the average value only gives an indication of the degree of removal. Yet, it is not surprising that Carbamazepine has a low removal efficiency since it is known from a number of previous studies to be persistent. Also, Diclofenac is relatively persistent.

5.1 Connected inhabitants, daily flow and removal efficiency

Two parameters that reflect the size of the WWTPs are number of connected inhabitants and daily flow. The relation between the number of connected inhabitants and removal efficiency is shown graphically in Figure 10.

The results in Figure 10 do not suggest that the number of connected inhabitants have any major effect on removal efficiency of the four pharmaceuticals. Azithromycin and Metoprolol show a very slight decrease, Diclofenac seems unaffected and Carbamazepine displays a very slight increase.

The next parameter reflecting WWTP size is daily flow and the relation between daily flow and removal efficiency is shown graphically in Figure 11.

Based on the results in Figure 11 there seems to be no major relation between the daily flow and removal efficiency. The results are very similarity to the observations made for number of connected inhabitants above in Figure 10.

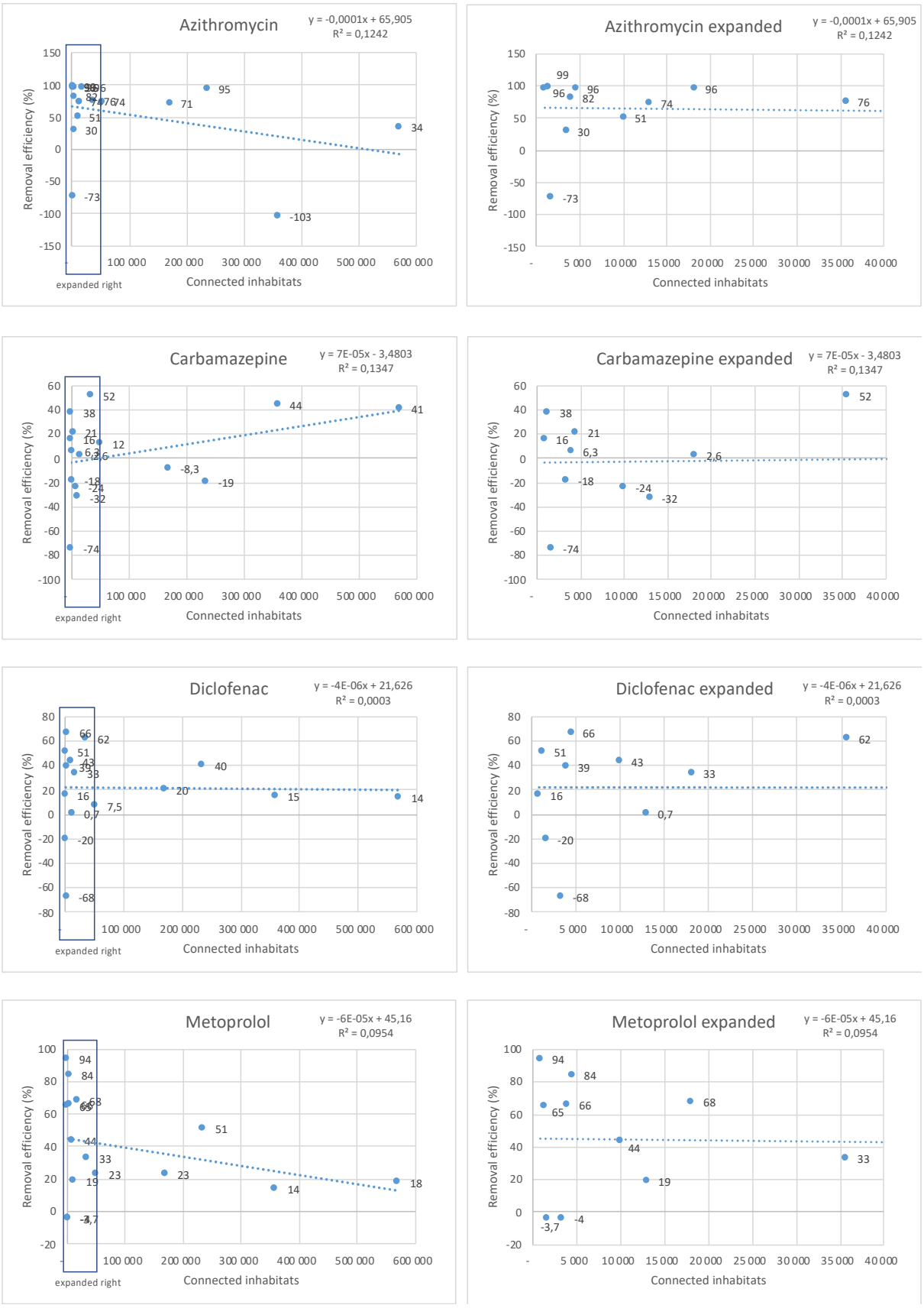

Figure 10. Relation between number of connected inhabitants and removal efficiency. Data from Table 4 and Table 5.

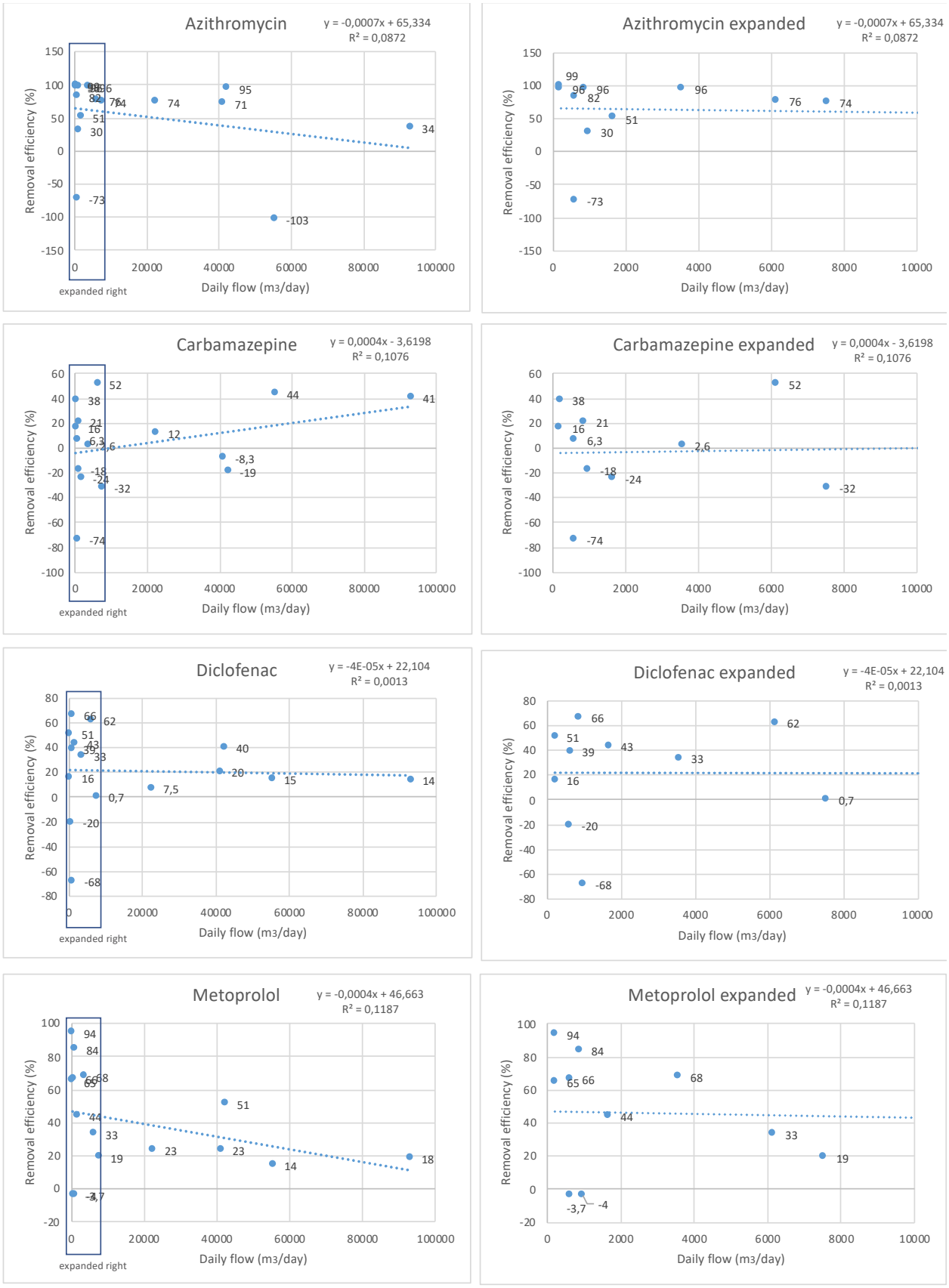

Figure 11. Relation between daily flow and removal efficiency. Data from Table 4 and Table5.

5.2 Sludge age, sludge digestion and removal efficiency

Two parameters that reflect the process of the WWTPs are sludge age and sludge digestion.

The relation between sludge age and removal efficiency is shown graphically in Figure 12.

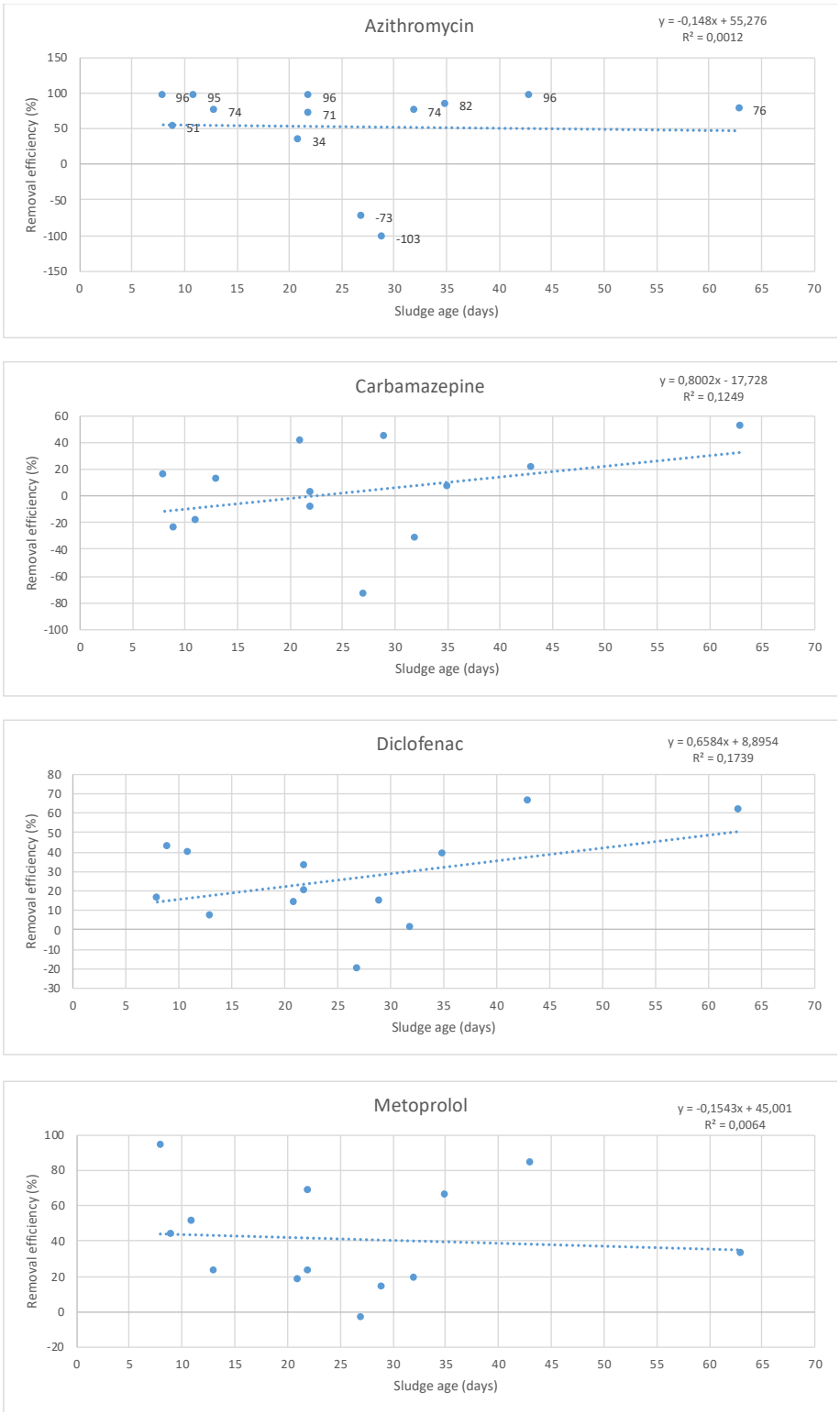

Figure 12. Relation between sludge age and removal efficiency. Data from Table 4 and Table5.

The results in Figure 12 reveal no major relation between sludge age and removal efficiencies. Azithromycin and Metoprolol seem completely unaffected by sludge age with a very slight negative slope. Carbamazepine and Diclofenac exhibit a very weak increase in removal efficiency with sludge age. A possible explanation to this would be that these persistent compounds might benefit from a longer sludge age. However, the indications are very weak.

Finally, the effect of sludge digestion on removal efficiencies was investigated. For each of the four pharmaceuticals the removal efficiency was listed from highest to lowest and compared to whether sludge digestion was applied (indicated by YES) or if it wasn't applied (indicated by NO). The results are shown in Figure 13.

		Azithromycin				Carbamazepine				Diclofenac				Metoprolol
Satow	NO	99		Swarzewo	YES	52		Laage	NO	66		Degeberga	NO	94
Kretinga	NO	96		Gdynia-Debogorze	YES	44		Swarzewo	YES	62		Laage	NO	84
Laage	NO	96		Gdansk-Wschod	YES	41		Satow	NO	51		Kretinga	NO	68
Degeberga	NO	96		Satow	NO	38		Jastrzebia-Gora	NO	43		Krakow	NO	66
Rostock	YES	95		Laage	NO	21		Rostock	YES	40		Satow	NO	65
Krakow	NO	82		Degeberga	NO	16		Krakow	NO	39		Rostock	YES	51
Swarzewo	YES	76		Kristianstad	YES	12		Kretinga	NO	33		Jastrzebia-Gora	NO	44
Palanga	NO	74		Krakow	NO	6,3		Klaipėda	YES	20		Swarzewo	YES	33
Kristianstad	YES	74		Kretinga	NO	2,6		Degeberga	NO	16		Klaipėda	YES	23
Klaipėda	YES	71		Klaipėda	YES	-8,3		Gdynia-Debogorze	YES	15		Kristianstad	YES	23
Jastrzebia-Gora	NO	51		Tollarp	NO	-18		Gdansk-Wschod	YES	14		Palanga	NO	19
Gdansk-Wschod	YES	34		Rostock	YES	-19		Kristianstad	YES	7,5		Gdansk-Wschod	YES	18
Tollarp	NO	30		Jastrzebia-Gora	NO	-24		Palanga	NO	0,7		Gdynia-Debogorze	YES	14
Nida	NO	-73		Palanga	NO	-32		Nida	NO	-20		Nida	NO	-3,7
Gdynia-Debogorze	YES	-103		Nida	NO	-74		Tollarp	NO	-68		Tollarp	NO	-4

Figure 13. Relation between sludge age and removal efficiency. Data from Table 4 and Table5.

Based on the results in Figure 13 there is no clear trend that sludge digestion has any major impact on the removal efficiency of the four pharmaceuticals.

Overall there are no clear indications that any of the WWTP parameters listed above have any large effect on removal of pharmaceuticals from the wastewater.

6 Conclusion

The Predicted Incoming Load (PIL) values, using regional pharmaceutical consumption data, were in general lower than Measured Incoming Load (MIL) values determined by chemical analysis of incoming wastewater of the 15 WWTPs.

Of the four investigated pharmaceuticals Carbamazepine and Metoprolol showed the best overall correspondence between consumption and occurrence data in most WWTPs, while for Azithromycin and Diclofenac, the differences between MIL and PIL values were much larger.

Carbamazepine is a good candidate to be used as predictor of expected chemical load to a WWTP using consumption data in a certain region. For Carbamazepine relation between MIL and PIL is described by the function PIL=0.9107*MIL with an R^2 of 0.9272.

The highest average removal efficiency was observed for Azithromycin with 53%, followed by Metoprolol with 39%. Diclofenac had a removal efficiency of 21% while Carbamazepine had the lowest value of only 4% on average. It should be noted that there are large differences between WWTPs and the average value only serves as an indication of the degree of removal. However, it is not unexpected that Carbamazepine has the lowest removal efficiency since it is known to be persistent in WWTPs and in the environment.

No major effects were observed between removal efficiency and the number of connected inhabitants. Likewise, the daily flow did not seem to impact the removal efficiency.

No major relation between sludge age and removal efficiencies was observed. Azithromycin and Metoprolol seemed completely unaffected by sludge age. Carbamazepine and Diclofenac showed a very weak increase in removal efficiency with sludge age, which possibly could be explained by the fact that these two compounds are persistent and might benefit from a longer sludge age. Yet, these indications are very weak.

Finally, there was no clear trend that sludge digestion had impact on the removal efficiency of the four pharmaceuticals.

7 References

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Project partners

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