Supporting information (appendices S1 and S2)

Spatial differences and temporal changes in illicit drug use in Europe quantified by wastewater analysis

Christoph Ort[1]*, Alexander L. N. van Nuijs[2]*, Jean-Daniel Berset[3], Lubertus Bijlsma[4], Sara Castiglioni[5], Adrian Covaci[2], Pim de Voogt[6,7], Erik Emke[6], Despo Fatta-Kassinos[8], Paul Griffiths[9], Félix Hernández[4], Iria González-Mariño[10], Roman Grabic[11], Barbara Kasprzyk-Hordern[12], Nicola Mastroianni[13], Axel Meierjohann[14], Thomas Nefau[15], Marcus Östman[16], Yolanda Pico[17], Ines Racamonde[10], Malcolm Reid[18], Jaroslav Slobodnik[19], Senka Terzic[20], Nikolaos Thomaidis[21] & Kevin V. Thomas[18]

Swiss Federal Institute of Aquatic Science andTechnology (Eawag), Dübendorf, Switzerland,[1]
Toxicological Center, University of Antwerp, Antwerp, Belgium,[2]
Water and Soil Protection Laboratory, Bern, Switzerland,[3]
Research Institute for Pesticides andWater, University Jaume I, Castellón de la Plana, Spain,[4]
Department of Environmental Health Sciences, IRCCS—Istituto di Ricerche Farmacologiche Mario Negri, Milan, Italy,[5]
KWR Watercycle Research Institute, Nieuwegein, the Netherlands,[6]
Institute for Biodiversity and Ecosystem Dynamics, University of Amsterdam, Amsterdam, the Netherlands,[7]
NIREAS-InternationalWater Research Center, University of Cyprus, Nicosia, Cyprus,[8]
European Monitoring Centre for Drug and Drug Addiction, Lisbon, Portugal,[9]
IIAA—Institute of Food Analysis and Research, University of Santiago de Compostela, Santiago de Compostela, Spain,[10]
University of South Bohemia in Ceske Budejovice, Faculty of Fisheries and Protection ofWaters, South Bohemian Research Center of Aquaculture and Biodiversity of Hydrocenoses, Vodnany, Czech Republic,[11]
Department of Chemistry, University of Bath, Bath, UK,[12]
Water and Soil Quality Research Group, Department of Environmental Chemistry, IDAEA-CSIC, Barcelona, Spain,[13]
Laboratory of Organic Chemistry, Åbo Akademi University, Åbo, Finland,[14]
Laboratoire Santé Publique Environnement, University of Paris Sud, Châtenay-Malabry, France,[15]
Department of Chemistry, Umeå University, Umeå, Sweden,[16]
Food and Environmental Safety Research Group, University ofValencia, Valencia, Spain,[17]
Norwegian Institute for Water Research (NIVA), Oslo, Norway,[18]
Environmental Institute, Kos, Slovak Republic,[19]
Division for Marine and Environmental Research, Rudjer Boskovic Institute Bijenicka, Zagreb, Croatia,[20]
Laboratory of Analytical Chemistry, Department of Chemistry, University of Athens, Athens, Greece[21]

*Co-first authors.

Correspondence to:

Christoph Ort, Urban Water Management, Swiss Federal Institute of Aquatic Science and Technology, Überlandstrasse 133, CH-8600 Dübendorf, Switzerland. Email: christoph.ort@eawag.ch

Alexander van Nuijs, Toxicological Centre, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium. Email: alexander.vannuijs@uantwerpen.be

Appendix S1

Questionnaire 2012

Eawag
Überlandstrasse 133
P. O. Box 611
8600 Dübendorf
Switzerland

Urban Water Management
Dr sc. techn. Christoph Ort
Researcher
Phone direct +41 (0)58 765 52 77
Fax direct +41 (0)58 765 53 89
christoph.ort@eawag.ch

eawag
aquatic research

Duebendorf March 2012

To whom it may concern

Emerging contaminants - i.e. household chemicals, pharmaceuticals, personal care products and illicit drugs - can be subject to high short-term variations in raw wastewater in the influent of a WWTP.

This questionnaire was designed to conduct an interview with professional wastewater treatment plant and sewer operators or a representative of local authorities with profound knowledge on the catchment and wastewater treatment plant under investigation. The interviewer must have read and understood the entire questionnaire before conducting the interview. Prefill information if possible to safe time at the interview and confirm your answers during the interview. It is recommended to conduct the interview in a personal meeting. In the dialogue a lot of additional valuable pieces of information can be obtained. The fields can be filled in directly in the protected word document or by hand in a printed version.

The questions are grouped according to specific aspects and categorized according to importance. It will help the interviewer to:

1) assess catchment characteristics,

2) understand the dynamics of wastewater in the catchment under investigation and

3) evaluate the suitability of the current sampling protocol.

We constantly aim at improving the questionnaire and, therefore, we highly appreciate any sort of feedback. Please do not hesitate to contact me if there are any questions.

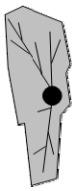
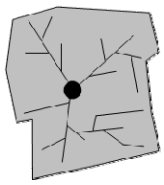
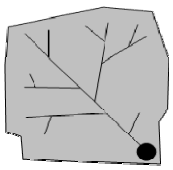
On behalf of the research team

Christoph Ort

Instructions
• Please fill in the grey shaded, underlined fields and tick boxes where appropriate. • For a sound assessment of the WWTP and sewer system under investigation and a meaningful interpretation of analytical results all pieces of information are valuable, we do not ask for unnecessary data. However, if time or willingness to fill in the questionnaire should be limited please answer the questions in the following order of priority: ★★★★ **All these questions must be answered to allow for a *minimal* evaluation**. ★★★ **Answers required for a *meaningful* evaluation.** ★★ Answers necessary to perform modeling and plausibility checks. ★ Additional information for a comprehensive analysis of additional aspects. • The grey numbers are the references for each field to facilitate the evaluation. **It would be highly appreciated if you could transfer the data in the accompanying excel sheet to share the work load** (one column is prefilled as an example)**. *All analytical results should also be entered into this excel file to facilitate data evaluation.*** • Please send all three files - i.e. 1. this file, 2. the large table with data of the monitoring campaign and 3. the excel sheet - plus any additional files (e.g. high resolution flow data) to christoph.ort@eawag.ch

1 General		
☐ Location and name		★★★★
1.1	Country _____	
1.2	City _____	
1.3	Name of WWTP _____	
☐ Email address of the person conducting the interview		★★★★
1.4	_____	
☐ Date of interview		★★★★
1.5	_____	
☐ Contact at WWTP		★
1.6	Name: _____	
1.7	phone: _____	
1.8	email: _____	

2 Catchment properties	
☐ What is the estimated population the WWTP serves?	★★★★
2.1 Number of people connected to the WWTP (based on census): _____ [-]	
2.2 Year of census: _____	
Other estimation methods to determine number of inhabitants if available:	★★
2.3 Design capacity: _____ [population equivalents]	
2.4 Planning horizon for design capacity (year): _____	
Number of people estimated based on number of house connections:	
2.5 _____ [-]	
2.6 Assumption for number of persons per house: _____ [-]	
2.7 Other method (please describe): _____	

☐ Are there significant net population differences between weekdays and weekends (i.e. commuters)?	★★★
2.8 ☐ Yes ☐ No	
If yes, can you quantify an increase (+) or decrease (-) compared to the weekend: 2.9 _____ 2.10 ☐ people per day [-] 2.11 ☐ daily flow [m^3/d]	
2.12 Explanation for the increase/decrease: _____	
☐ Which shape best represents your catchment (circle=location of WWTP)?	★
2.13 ☐ **1**  ☐ **2**  ☐ **3**  ☐ **4** 	
If possible, please provide a digital map of your catchment (sewer plan).	★★
2.14 ☐ A digital map of the catchment is provided in a separate file.	

Sewer system

☐ What type of sewers drains your catchment? Please specify fractions of total flow (2.15-2.18 should add up to 100). ★

	Gravity flow		Pressurized	
Separate sewers[1]	2.15	_____ [%]	2.16	_____ [%]
Combined sewers[2]	2.17	_____ [%]	2.18	_____ [%]

☐ How many lift stations are operated in the entire catchment area? ★★★★

2.19 _____ [-]

☐ Do you suspect significant exfiltration - i.e. wastewater losses - from sewer lines (including house connections)? ★★★★

2.20 ☐ Yes ☐ No

2.21 If yes, can you estimate how much (as % of total inflow)? _____ [%]

☐ What are travel distances (closest and most remote connected household) and the mean residence time of wastewater in your sewers? ★★

2.22 Shortest travel distance _____ [km]

2.23 Longest travel distance _____ [km]

2.24 Mean residence time _____ [hours]

☐ Is there any special infrastructure except pump/lift stations (e.g. actuators, retention basins) to regulate wastewater flow in the catchment before the influent of the WWTP (real time control)? ★★★

2.25 ☐ Yes ☐ No

Is it possible that water can be stored "offline" over a significant period of time, which could imply that this volume of wastewater occurring on e.g. Wednesday would not be sampled for in the Wednesday sample but maybe only on Thursday or any time later? Please comment:

2.26 _____

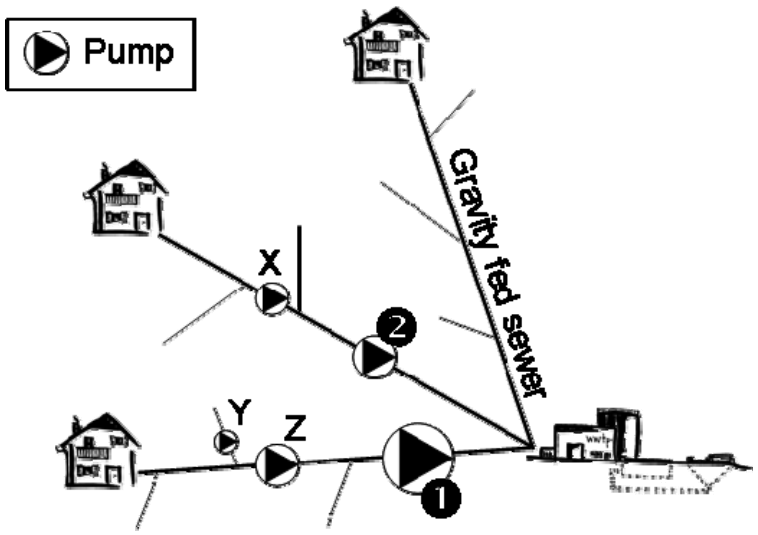
[1] Predominantly domestic/industrial wastewater, not diluted with surface runoff during rain events or infiltration after snow melt

[2] During rain events or snow melt significant amounts of surface runoff is collected together with the wastewater

Instruction for next questions: For (partly) pressurized drainage systems (pumps or lift stations in catchment)

What is the approximate average amount of the total flow to the WWTP that has been pumped at some stage in the catchment? **Please only consider the "final pumps/lift stations",** these are the ones directly delivering to the WWTP (i.e. ❶ and ❷ in the example below) and **not any sub-stations** farther upstream in the catchment (X, Y and Z in the example below). The illustration and table below serve as an example, please fill in the table on the next page as it applies to the drainage system under investigation.

Illustration



Sample table for the system depicted above:

Pump/ lift station	Mode of operation	Pump cycle		Number of pump cycles per day**	Volume of pump sump	Pump capacity (flow rate***)	Distance of pump station to WWTP	Length of pressurized section after pump station	Daily flow through this station
ID	c=continuous i=intermittent	ON* [min]	OFF [min]	[-]	[m^3]	[L/s]	[km]	[km]	[m^3/d]
❶	☒ i ☐ c	15	30	30	180	200	1	0.1	5'500
❷	☐ i ☒ c	-	-	-	100	var.	0.5	0.5	1'000

*shortest duration **to be expected on average on a typical dry weather day ***value or "var." for variable

☐ Please indicate how the pumps/lift stations in the catchment under investigation are operated

Pump/ lift station	Mode of operation	Pump cycle		Number of pump cycles per day	Volume of pump sump	Pump capacity (flow rate)	Distance of pump station to WWTP	Length of pressurized section after pump station	Daily flow through this station
ID	c=continuous i=intermittent	ON [minutes]	OFF [minutes]	[-]	[m^3]	[L/s]	[km]	[km]	[m^3/d]
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____

2.27	Shortest time in column 3 (shortest ON time):	_____	[minutes]	★★★
2.28	Approx. daily volume for the pump station with shortest ON time (2.27)	_____	[m^3/d]	
2.29	Sum of all volumes in last column:	_____	[m^3/d]	

3 WWTP influent data			
☐ Please provide a brief description of where the flow meter is located to measure raw wastewater influent:			★★★
3.1 _____			
☐ Is the wastewater lifted at the influent of the WWTP?			★★★
3.2 ☐ Yes ☐ No			
If yes, how is the lift station operated?			★★
3.3 ☐ intermittently	3.4 ☐ continuously	volume of pump sump 3.5 _____ [m^3]	
☐ If possible, please attach a file (Excel or csv) with flow data at high temporal resolution (one month including some dry weather days at intervals of 1 to 5 minutes would be most informative).			★★★
3.6 ☐ Flow data at high temporal resolution are provided in a separate file.			
3.7 Temporal resolution of flow data _____ [minutes]			
If no file can be provided, please provide estimates for *a typical dry weather day* for:			★★★★
3.8 Total wastewater volume _____ [m^3/d]			
3.9 Minimum flow _____ [L/s]			
3.10 Maximum flow _____ [L/s]			
3.11 Maximum flow (wet weather) _____ [L/s]			★★

☐ How is flow measured (multiple answers possible)		★★★
3.12	☐ in an open channel	
3.13	☐ in a parshall flume (venturi)	
3.14	☐ with an echo sounding (from above water level)	
3.15	☐ measuring water level and velocity (submerged below water level)	
3.16	☐ with an MID or similar in a completely filled, pressurized pipe	
3.17	☐ other, please specify: _____	
☐ Calibration of flow meter		
3.18	Interval of flow meter calibration _____ [months]	★★★★
3.19	Calibration method (e.g. tracer experiment, volumetric (filling a tank)), please specify: _____	★★★★
3.20	Manufacturer's specifications for accuracy _____ [%]	★★
3.21	Estimated operational accuracy of flow meter (note that this is different from the manufacturer's specification of accuracy of the device, it includes installation and operation under real conditions): _____ [%]	★★★★
3.22	☐ The protocol of the last calibration is attached in a separate file.	★★★

4 Sampling (raw wastewater at the influent of the WWTP)	
☐ Exact location	
Please specify where exactly the raw wastewater samples are collected in the influent (e.g. after sand trap or fine screen, before/after primary clarifier): 4.1 ☐ After fine screen 4.2 ☐ After primary clarifier 4.3 ☐ Other, please specify: _____	★★★★
4.4 ☐ A schematic representation of the WWTP flow scheme is provided in a separate file.	★
☐ Sampling device	
4.5 Manufacturer _____	★
4.6 Type _____	★
4.7 Is the sampling bottle refrigerated ☐ Yes ☐ No	★★★★
4.8 Number of bottles used in the sampling device _____ [-]	★★
4.9 Total storage capacity in sampling device _____ [L]	★★
4.10 Composite sample extends over _____ [hours] (typically 24 hours)	★★★★
4.11 Material of sample container _____	★★★
☐ Sampling mode, only tick one (see next page for a description of sampling modes)	★★★★
4.12 ☐ Time-proportional composite (i.e. fixed volume/set frequency 4.13 ☐ Volume-proportional (i.e. variable frequency/set volume) 4.14 ☐ Flow-proportional (i.e. variable volume/set frequency) 4.15 Manual sampling (please provide how many samples per day): _____ 4.16 Other, please specify _____	
☐ Sampling interval	★★★★
4.17 In the case of time- or flow-proportional every _____ [minutes]	
4.18 In the case of volume-proportional every _____ [m^3]	

☐ How long did it take to conduct the interview?	★
4.19 _____ [minutes]	

Additional notes (e.g. suggestions for improving the questionnaire or aspects that were not covered but seem to be relevant at this specific wastewater treatment plant or the urban catchment).	
4.20 _____	

Descriptions of sampling modes* (for question on previous page)

Table 1. Visualization and brief description of different sampling modes (adapted from ISO Water quality - Sampling - Part 2: Guidance on sampling techniques ISO 5667-2 1991, 731 ISO,Genève, Switzerland).

Conceptual example

Sampling mode		Short description (see "Sampling guide" to find out which sampling mode is suitable in which situation).	Illustration (F=Flow in sewer, S=sampling volume)	Specific equipment [1]	Flow meter [2]
Continuous	**flow-proportional**	Divert a side stream, proportional to the flow in the sewer		Pump with speed control (proportional to external flow signal)[3]	Yes
	constant	Divert a constant side stream from the sewer		Pump	No
Discrete	**T:** time-proportional	Take a constant sample volume at constant time intervals		Standard auto-sampler	No
	F: flow-proportional	Make sample volume proportional to the flow in the sewer taking them at constant time intervals		Auto-sampler with adjustable sampling volume [4] (proportional to external flow signal)	Yes
	V: volume-proportional	Take a constant sample volume at variable time intervals, after a certain volume of wastewater has passed the sampling point		Auto-sampler totaling an external flow signal up to a predefined volume and then triggering a sample [5]	Yes
	g: grab sample	Take one (or a number of) grab sample		Scoop, no power supply.	No

[1] Indicates what equipment is required besides sampling bottles, hose and power supply.
[2] Is a flow meter required for taking samples (external flow signal)? To calculate environmental loads from sampled (average) concentrations it always needs a flow meter.
[3] Ideally a robust peristaltic pump with fine increments to accurately control speed with external flow signal. Linearity of pump speeds and performance (suction and pressure height) need to be checked for conditions that apply in the field.
[4] Sampling volume of individual samples needs to be checked for linear behavior over the whole range of discharge in the sewer.
[5] Check variation of individual sample size.

*Ort C., Lawrence M.G., Rieckermann J. and Joss A. (2010) Monitoring Pharmaceuticals and Per-sonal Care Products (PPCPs) and Illicit Drugs in Wastewater Systems: Are Your Conclusions Valid? A Critical Review. Environmental Science & Technology 44 (16), 6024–6035.

5 Sampling period 2012 (to be filled in during/after sampling campaign)	
☐ Time of day when sample bottles are changed for the next 24-hour period.	★★★★
5.1 Time: _____	
☐ Which period do the total daily wastewater volumes represent (5.33 - 5.47, see second row in separate file)?	★★★★
5.2 ☐ The 24 hours prior to the time provided in point 5.1 (preferred) 5.3 ☐ Midnight to midnight of the day before sample collection (not suitable) 5.4 ☐ Midnight to midnight of the day when sample collection took place (not suitable)	
☐ Did the catchment area change for your WWTP during the sampling period? For instance, we know that in some municipalities wastewater is directed to different WWTPs during different times of the year.	★★★★
5.5 ☐ Yes ☐ No 5.6 e If yes, Please briefly comment on any date specific issues: _____	
☐ Was there anything special during the monitoring period (e.g. public/school holidays or special events such as festivals etc.)?	★★★★
5.7 ☐ Yes ☐ No 5.8 If yes, Please briefly comment on any date specific issues: _____	
☐ Please provide influent flow data at high temporal resolution (i.e. minute intervals) for the entire monitoring period.	★★★★
5.9 ☐ A file in a similar format as in 3.6 is provided.	

☐ Sample handling (to be filled in by the researcher, not for WWTP staff)			★★★★
5.10	Storage time until analysis	_____ [days]	
5.11	Storage temperature (in freezer, not in sampler)	_____ [°C]	
5.12	Filtration onsite before transport (within 1 hour)	☐ Yes ☐ No	
5.13	Filtration in laboratory	☐ Yes ☐ No	
5.14	Filtration (pore size)	_____ [µm]	
5.15	Filtration (filter material)	_____	
5.16	Acidification onsite before transport (within 1 hour)	☐ Yes ☐ No	
5.17	Acidification during storage	☐ Yes ☐ No	

☞ Please fill in the table with relevant data for the monitoring campaign in the table (entries 5.18 - 5.233) provided in an extra file (suitable for printing in A3 for example, or directly electronically in the protected word document).

☞ It would be highly appreciated if you could transfer all collected information into the accompanying excel sheet to share the work load (one column is prefilled as an example). We can then compile all WWTPs' data in one file to allow an efficient, consistent data evaluation.

☞ All analytical results should also be entered into the accompanying excel sheet to facilitate data evaluation (6.1 - 17.16).

☞ Please return the questionnaire as soon as sections 1-4 are completed and then send an update once section 5 is completed and another update with all analytical results once the analyses are completed.

Appendix S1

Questionnaire 2013

Eawag
Überlandstrasse 133
P. O. Box 611
8600 Dübendorf
Switzerland

Urban Water Management
Dr sc. techn. Christoph Ort
Researcher
Phone direct +41 (0)58 765 52 77
Fax direct +41 (0)58 765 53 89
christoph.ort@eawag.ch

Dübendorf, March 2013

Dear colleagues

If you already participated in one of our previous monitoring campaigns, you know how this works. As there are not too many changes since 2012, we provide you with a prefilled version of the evaluation file from 2012. It was corrected and updated with your feedback. Please carefully check if all entries for your wastewater treatment plant (WWTP) are still valid for 2013.

Please note that there are some (new) fields that need to be filled in anyway:

1. **date of interview or re-checking the data (field 1.5)**
2. **newly structured section on population in WWTP catchment (field 2.1 and 2.5-2.12)**
3. **information on presence of hospitals in the WWTP catchment (field 2.19 and 2.20)**
4. **if possible, please provide information that could not be obtained in previous years**

If you have never participated before please read the following details and instructions.

This questionnaire was designed to conduct an interview with professional wastewater treatment plant and sewer operators or a representative of local authorities with profound knowledge on the catchment and wastewater treatment plant under investigation. The interviewer must have read and understood the entire questionnaire before conducting the interview. Prefill information if possible to safe time at the interview and confirm your answers during the interview. It is recommended to conduct the interview in a personal meeting. In the dialogue a lot of additional valuable pieces of information can be obtained. The fields can be filled in directly in the protected word document or by hand in a printed version.

The questions are grouped according to specific aspects and categorized according to importance. It will help the interviewer to:

1) assess catchment characteristics,

2) understand the dynamics of wastewater in the catchment under investigation and

3) evaluate the suitability of the current sampling protocol.

We constantly aim at improving the questionnaire and, therefore, we highly appreciate any sort of feedback. Please do not hesitate to contact me if there are any questions.

On behalf of the research team

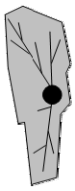
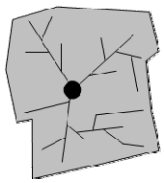
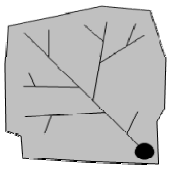
Christoph Ort

eawag aquatic research

Instructions
• Please fill in the grey shaded, underlined fields and tick boxes where appropriate. • For a sound assessment of the WWTP and sewer system under investigation and a meaningful interpretation of analytical results all pieces of information are valuable, we do not ask for unnecessary data. However, if time or willingness to fill in the questionnaire should be limited please answer the questions in the following order of priority: ★★★★ **All these questions must be answered to allow for a *minimal* evaluation**. ★★★ **Answers required for a *meaningful* evaluation.** ★★ Answers necessary to perform modeling and plausibility checks. ★ Additional information for a comprehensive analysis of additional aspects. • The grey numbers are the references for each field to facilitate the evaluation. It would be highly appreciated if you could transfer the data into the accompanying excel sheet to share the work load (data prefilled from 2012). *All analytical results should also be entered into this excel file to facilitate data evaluation.* • Please use the format of numbers/text provided in column C of the attached Excel evaluation file. • If you sample outside the common monitoring period 6.-12.3.2013 (day 1 – day 7), simply add your data for day 8 – day 15. For the evaluation it does not matter whether these days are prior or after the common monitoring period.
Please send all three files - i.e. 1. this file, 2. the table with data of the monitoring campaign and 3. the excel sheet - plus any additional files (e.g. high resolution flow data) to christoph.ort@eawag.ch

6 General		
☐ Location and name		★★★★
6.1	Country _____	
6.2	City _____	
6.3	Name of WWTP _____	
☐ Email address of the person conducting the interview or checking the data		★★★★
6.4	_____	
☐ Date of interview or re-checking the prefilled entries from previous years		★★★★
6.5	_____	
☐ Contact at WWTP		★
6.6	Name: _____	
6.7	phone: _____	
6.8	email: _____	

7 Catchment properties	
☐ What is the estimated **residential population** the WWTP serves (**commuters see 2.13**)?	★★★★
7.1 Residential population connected to the WWTP (based on official census **excluding commuters**): _____ [-] 7.2 Year of census: _____	
Other estimation methods to determine residential population <u>if available</u>:	★★★
7.3 Design capacity: _____ [population equivalents] 7.4 Planning horizon for design capacity (year): _____ 7.5 Current <u>total</u> loading: _____ [population equivalents] 7.6 Number of population equivalents expected to originate from <u>industrial sources</u>: _____ [population equivalents]	
Number of people estimated based on number of house connections: 7.7 _____ [-] 7.8 Assumption for number of persons per house: _____ [-]	
Other method (please describe type and period: e.g. "people estimated based on average BOD load during 2013 (with 60g BOD/d) subtracting 20% assumed to originate from industrial input"): 7.9 Description: _____ 7.10 Number of people based on this method: _____ [-]	
Which number should be used as best estimate for the <u>residential population</u> being connected to the WWTP: 7.11 _____ [-] 7.12 Please provide an explicit reason for this choice: _____	★★★★

☐ Are there net population differences between weekends and weekdays?	★★★
7.13 Do commuters substantially increase or decrease the residential population being connected to the WWTP? ☐ Yes ☐ No	
If yes, can you quantify a net increase (+) or decrease (-) compared to the weekend: 7.14 _____ 7.15 ☐ people per day [-] 7.16 ☐ daily flow [m^3/d]	
☐ Which shape best represents your catchment (circle=location of WWTP)?	★
7.17 ☐ **1**  ☐ **2**  ☐ **3**  ☐ **4** 	
If possible, please provide a digital map of your catchment (sewer plan). 7.18 ☐ A digital map of the catchment is provided in a separate file.	★★
☐ Are there any hospitals in the catchment of the WWTP?	★★
Please provide the number of hospitals discharging wastewater without pretreatment into public sewers connected to this WWTP (please write 0 if you know there are none, and leave it blank if you don't know it): 7.19 _____ [-] What is the total number of hospital beds (sum of all hospitals above)? 7.20 _____ [-]	

Sewer system

☐ What type of sewers drains your catchment? Please specify fractions of total flow (2.21-2.24 should add up to 100). ★

	Gravity flow		Pressurized	
Separate sewers[3]	7.21	_____ [%]	7.22	_____ [%]
Combined sewers[4]	7.23	_____ [%]	7.24	_____ [%]

☐ How many lift stations are operated in the entire catchment area? ★★★★

7.25 _____ [-]

☐ Do you suspect significant exfiltration - i.e. wastewater losses - from sewer lines (including house connections)? ★★★★

7.26 ☐ Yes ☐ No

7.27 If yes, can you estimate how much (as % of total inflow)? _____ [%]

☐ What are travel distances (closest and most remote connected household) and the mean residence time of wastewater in your sewers? ★★

7.28 Shortest travel distance _____ [km]

7.29 Longest travel distance _____ [km]

7.30 Mean residence time _____ [hours]

☐ Is there any special infrastructure except pump/lift stations (e.g. actuators, retention basins) to regulate wastewater flow in the catchment before the influent of the WWTP (real time control)? ★★★

7.31 ☐ Yes ☐ No

Is it possible that water can be stored "offline" over a significant period of time, which could imply that this volume of wastewater occurring on e.g. Wednesday would not be sampled for in the Wednesday sample but maybe only on Thursday or any time later? Please comment:

7.32 _____

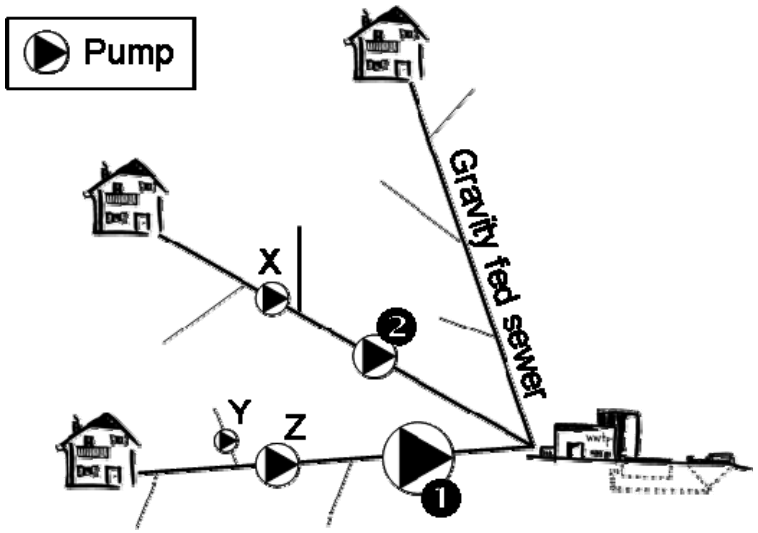
[3] Predominantly domestic/industrial wastewater, not diluted with surface runoff during rain events or infiltration after snow melt

[4] During rain events or snow melt significant amounts of surface runoff is collected together with the wastewater

Instruction for next questions: For (partly) pressurized drainage systems (pumps or lift stations in catchment)

What is the approximate average amount of the total flow to the WWTP that has been pumped at some stage in the catchment? **Please only consider the "final pumps/lift stations",** these are the ones directly delivering to the WWTP (i.e. ❶ and ❷ in the example below) and **not any sub-stations** farther upstream in the catchment (X, Y and Z in the example below). The illustration and table below serve as an example, please fill in the table on the next page as it applies to the drainage system under investigation.

Illustration



Sample table for the system depicted above:

Pump/ lift station	Mode of operation	Pump cycle		Number of pump cycles per day**	Volume of pump sump	Pump capacity (flow rate***)	Distance of pump station to WWTP	Length of pressurized section after pump station	Daily flow through this station
ID	c=continuous i=intermittent	ON* [min]	OFF [min]	[-]	[m^3]	[L/s]	[km]	[km]	[m^3/d]
❶	☒ i ☐ c	15	30	30	180	200	1	0.1	5'500
❷	☐ i ☒ c	-	-	-	100	var.	0.5	0.5	1'000

*shortest duration **to be expected on average on a typical dry weather day ***value or "var." for variable

☐ **Please indicate how the pumps/lift stations in the catchment under investigation are operated**

Pump/ lift station	Mode of operation	Pump cycle		Number of pump cycles per day	Volume of pump sump	Pump capacity (flow rate)	Distance of pump station to WWTP	Length of pressurized section after pump station	Daily flow through this station
ID	c=continuous i=intermittent	ON [minutes]	OFF [minutes]	[-]	[m^3]	[L/s]	[km]	[km]	[m^3/d]
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____
_____	☐ i ☐ c	_____	_____	_____	_____	_____	_____	_____	_____

7.33	Shortest time in column 3 (shortest ON time):	_____	[minutes]	★★★
7.34	Approx. daily volume for the pump station with shortest ON time (2.31)	_____	[m^3/d]	
7.35	Sum of all volumes in last column:	_____	[m^3/d]	

☐ Any important comments related to catchment and sewers not covered with 2.1-2.33: ★★★★

7.36

8 WWTP influent data			
☐ Please provide a brief description of where the flow meter is located to measure raw wastewater influent:			★★★
8.1 _____			
☐ Is the wastewater lifted at the influent of the WWTP?			★★★
8.2 ☐ Yes ☐ No			
If yes, how is the lift station operated?			★★
8.3 ☐ intermittently	8.4 ☐ continuously	volume of pump sump 8.5 _____ [m^3]	
☐ If possible, please attach a file (Excel or csv) with flow data at high temporal resolution (one month including some dry weather days at intervals of 1 to 5 minutes would be most informative).			★★★
8.6 ☐ Flow data at high temporal resolution are provided in a separate file.			
8.7 Temporal resolution of flow data _____ [minutes]			
If no file can be provided, please provide estimates for *a typical dry weather day* for:			★★★★
8.8 Total wastewater volume _____ [m^3/d]			
8.9 Minimum flow _____ [L/s]			
8.10 Maximum flow _____ [L/s]			
8.11 Maximum flow (wet weather) _____ [L/s]			★★

☐ How is flow measured (multiple answers possible)	★★★
8.12 ☐ in an open channel 8.13 ☐ in a parshall flume (venturi) 8.14 ☐ with an echo sounding (from above water level) 8.15 ☐ measuring water level and velocity (submerged below water level) 8.16 ☐ with an MID or similar in a completely filled, pressurized pipe 8.17 ☐ other, please specify: _____	
☐ Calibration of flow meter	
8.18 Interval of flow meter calibration _____ [months]	★★★★
8.19 Calibration method (e.g. tracer experiment, volumetric (filling a tank)), please specify: _____	★★★★
8.20 Manufacturer's specifications for accuracy _____ [%]	★★
8.21 Estimated operational accuracy of flow meter (note that this is different from the manufacturer's specification of accuracy of the device, it includes installation and operation under real conditions): _____ [%]	★★★★
8.22 ☐ The protocol of the last calibration is attached in a separate file.	★★★
☐ Any comments relevant for the inlet structure of the WWTP that have not been covered with 3.1-3.22:	★★★★
8.23 _____	

9 Sampling (raw wastewater at the influent of the WWTP)	
☐ Exact location	
Please specify where exactly the raw wastewater samples are collected in the influent (e.g. after sand trap or fine screen, before/after primary clarifier): 9.1 ☐ After fine screen 9.2 ☐ After primary clarifier 9.3 ☐ Other, please specify: _____	★★★★
9.4 ☐ A schematic representation of the WWTP flow scheme is provided in a separate file.	★
☐ Sampling device	
9.5 Manufacturer _____	★
9.6 Type _____	★
9.7 Is the sampling bottle refrigerated ☐ Yes ☐ No	★★★★
9.8 Number of bottles used in the sampling device _____ [-]	★★
9.9 Total storage capacity in sampling device _____ [L]	★★
9.10 Composite sample extends over _____ [hours] (typically 24 hours)	★★★★
9.11 Material of sample container _____	★★★
☐ Sampling mode, only tick one (see next page for a description of sampling modes)	★★★★
9.12 ☐ Time-proportional composite (i.e. fixed volume/set frequency 9.13 ☐ Volume-proportional (i.e. variable frequency/set volume) 9.14 ☐ Flow-proportional (i.e. variable volume/set frequency) 9.15 Manual sampling (please provide how many samples per day): _____ 9.16 Other, please specify _____	
☐ Sampling interval	★★★★
9.17 In the case of time- or flow-proportional every _____ [minutes]	
9.18 In the case of volume-proportional every _____ [m^3]	

☐ How long did it take to conduct the interview?	★
9.19 _____ [minutes]	

Additional notes	
9.20 _____	

Descriptions of sampling modes* (for question on previous page)

Table 1. Visualization and brief description of different sampling modes (adapted from ISO Water quality - Sampling - Part 2: Guidance on sampling techniques ISO 5667-2 1991, 731 ISO,Genève, Switzerland).

Conceptual example

Sampling mode		Short description (see "Sampling guide" to find out which sampling mode is suitable in which situation).	Illustration (F=Flow in sewer, S=sampling volume)	Specific equipment [1]	Flow meter [2]
Continuous	**flow-proportional**	Divert a side stream, proportional to the flow in the sewer		Pump with speed control (proportional to external flow signal)[3]	Yes
	constant	Divert a constant side stream from the sewer		Pump	No
Discrete	**T:** time-proportional	Take a constant sample volume at constant time intervals		Standard auto-sampler	No
	F: flow-proportional	Make sample volume proportional to the flow in the sewer taking them at constant time intervals		Auto-sampler with adjustable sampling volume [4] (proportional to external flow signal)	Yes
	V: volume-proportional	Take a constant sample volume at variable time intervals, after a certain volume of wastewater has passed the sampling point		Auto-sampler totaling an external flow signal up to a predefined volume and then triggering a sample [5]	Yes
	g: grab sample	Take one (or a number of) grab sample		Scoop, no power supply.	No

[1] Indicates what equipment is required besides sampling bottles, hose and power supply.
[2] Is a flow meter required for taking samples (external flow signal)? To calculate environmental loads from sampled (average) concentrations it always needs a flow meter.
[3] Ideally a robust peristaltic pump with fine increments to accurately control speed with external flow signal. Linearity of pump speeds and performance (suction and pressure height) need to be checked for conditions that apply in the field.
[4] Sampling volume of individual samples needs to be checked for linear behavior over the whole range of discharge in the sewer.
[5] Check variation of individual sample size.

*Ort C., Lawrence M.G., Rieckermann J. and Joss A. (2010) Monitoring Pharmaceuticals and Per-sonal Care Products (PPCPs) and Illicit Drugs in Wastewater Systems: Are Your Conclusions Valid? A Critical Review. Environmental Science & Technology 44 (16), 6024–6035.

10 Sampling period 2012 (to be filled in during/after sampling campaign)	
☐ Time of day when sample bottles are changed for the next 24-hour period.	★★★★
10.1 Time: _____	
☐ Which period do the total daily wastewater volumes represent (5.33 - 5.47, see second row in separate file)?	★★★★
10.2 ☐ The 24 hours prior to the time provided in point 5.1 (preferred) 10.3 ☐ Midnight to midnight of the day before sample collection (not suitable) 10.4 ☐ Midnight to midnight of the day when sample collection took place (not suitable)	
☐ Did the catchment area change for your WWTP during the sampling period? For instance, we know that in some municipalities wastewater is directed to different WWTPs during different times of the year.	★★★★
10.5 ☐ Yes ☐ No 10.6 If yes, Please briefly comment on any date specific issues: _____	
☐ Was there anything special during the monitoring period (e.g. interruption of the operation of the WWTP, sampling failure, public/school holidays or special events such as festivals etc.)?	★★★★
10.7 ☐ Yes ☐ No 10.8 If yes, Please briefly comment on any date specific issues: _____	
☐ Please provide influent flow data at high temporal resolution (i.e. minute intervals) for the entire monitoring period.	★★★★
10.9 ☐ A file in a similar format as in 3.6 is provided.	

☐ Sample handling (to be filled in by the researcher, not for WWTP staff)			★★★★
10.10	Storage time until analysis	_____ [days]	
10.11	Storage temperature (in freezer, not in sampler)	_____ [°C]	
10.12	Filtration onsite before transport (within 1 hour)	☐ Yes ☐ No	
10.13	Filtration in laboratory	☐ Yes ☐ No	
10.14	Filtration (pore size)	_____ [µm]	
10.15	Filtration (filter material)	_____	
10.16	Acidification onsite before transport (within 1 hour)	☐ Yes ☐ No	
10.17	Acidification during storage	☐ Yes ☐ No	

☞ Please fill in the table with relevant data for the monitoring campaign in the table (entries 5.18 - 5.233) provided in an extra file (suitable for printing in A3 for example, or directly electronically in the protected word document).

☞ It would be highly appreciated if you could transfer all collected information into the accompanying excel sheet to share the work load (one column is prefilled as an example). We can then compile all WWTPs' data in one file to allow an efficient, consistent data evaluation.

☞ All analytical results should also be entered into the accompanying excel sheet to facilitate data evaluation (6.1 - 17.16).

☞ Please return the questionnaire as soon as sections 1-4 are completed and then send an update once section 5 is completed and another update with all analytical results once the analyses are completed.

Appendix S2

Uncertainty considerations

Approximative formula (linear uncertainty propagation):

$$U_{T,approx.form} = \sqrt{\left(\frac{U_S}{\sqrt{n}}\right)^2 + U_C{}^2 + U_F{}^2 + U_P{}^2}$$

equation 1

where

$U_{T,approx.form}$	overall uncertainty based on approximative formula
U_S	uncertainty from sampling for a one-day (24-h) composite sample
U_C	uncertainty from chemical analysis
U_F	uncertainty from flow measurements
U_P	uncertainty from population estimation

With the numbers used in the manuscript $U_{T,approx.form} = 0.42$

Monte Carlo simulation for uncertainty estimation:

$$L = U_{S,MC} \cdot \frac{C \cdot F}{P}$$

equation 2

where

L	load of illicit drug target residue in sewer
$U_{S,MC}$	uncertainty from sampling over n-day period
C	concentration of target residue
F	total flow for one composite sample (typically over 24h)
P	population in wastewater treatment plant catchment

with

$$U_{S,MC} \sim N(1, \frac{U_S}{\sqrt{n}})$$
$$C \sim N(100, U_C \cdot 100)$$
$$F \sim N(10000, U_F \cdot 10000)$$
$$P \sim N(1000000, U_P \cdot 1000000)$$

where $N(m,s)$ is a normal distribution with mean m and standard deviation s (uncertainty propagation is independent of absolute values of m)	

Calculate RSD from Monte Carlo simulation results:

$U_{T,MC} = \frac{sd(L)}{mean(L)}$ where $U_{T,MC}$ overall uncertainty based on Monte Carlo simulation	equation 3

R script for equations 1-3:

```
# Uncertainty/error propagation

# RSD of uncertainties U
# ======================

# S=sampling
# C=chemical analysis
# F=flow measurements
# P=population

US=0.2
UC=0.3
UF=0.2
UP=0.2

# number of days (for estimation of uncertainty of mean over m days)
# ==================================================================

n=7

#
================================================================================
# uncertainty/error propagation using linear approximation as e.g. used in
```

```
# Lai et al. (WR 2011)
#
================================================================================
UT_approx_formula=sqrt((US/sqrt(n))^2+UC^2+UF^2+UP^2) # equation 1

# ========================================================
# uncertainty estimation using Monte Carlo simulation (MC)
# ========================================================

# number of Monte Carlo simulations
# =================================
n_MC=1e6

# average values to be used in MC
# and absolute SD (instead of RSD)
# ================================
concentration=100
sd_concentration=UC*concentration
sd_sampling=US/sqrt(n)
flow=10000
sd_flow=UF*flow
population=1000000
sd_population=UP*population



repeat_MC <- function()  # equation 2
{
  # MC of individual components
  sampling_unc_MC=rnorm(n_MC,1,sd=sd_sampling)
  chemical_analysis_MC=rnorm(n_MC,concentration,sd=sd_concentration)
  flow_MC=rnorm(n_MC,flow,sd=sd_flow)
  population_MC=rnorm(n_MC,population,sd=sd_population)

  loads_MC=sampling_unc_MC*(chemical_analysis_MC*flow_MC)/population_MC



  UT_MC=sd(loads_MC)/mean(loads_MC)    # equation 3
  return(UT_MC)
}
```

```
UT_MC_vec <- rep(NA,1)

for (i in 1:length(UT_MC_vec))
{
UT_MC_vec[i] <- repeat_MC()
}



#Results for approximative formula and Monte Carlo simulation

UT_approx_formula
mean(UT_MC_vec)
```