Drug residues removal capacities by surface flow treatment wetlands: pharmaceutical compounds distribution in soil, water and plants compartments

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Contents

- Context and issues for SFTW around Strasbourg,
- Materials & method for sampling sessions in SFTW and drug quantification
- Results on the drug residues removal capacities and drug distributions to plants and mud in SFTWs

Context and issues for SFTW around Strasbourg

Context

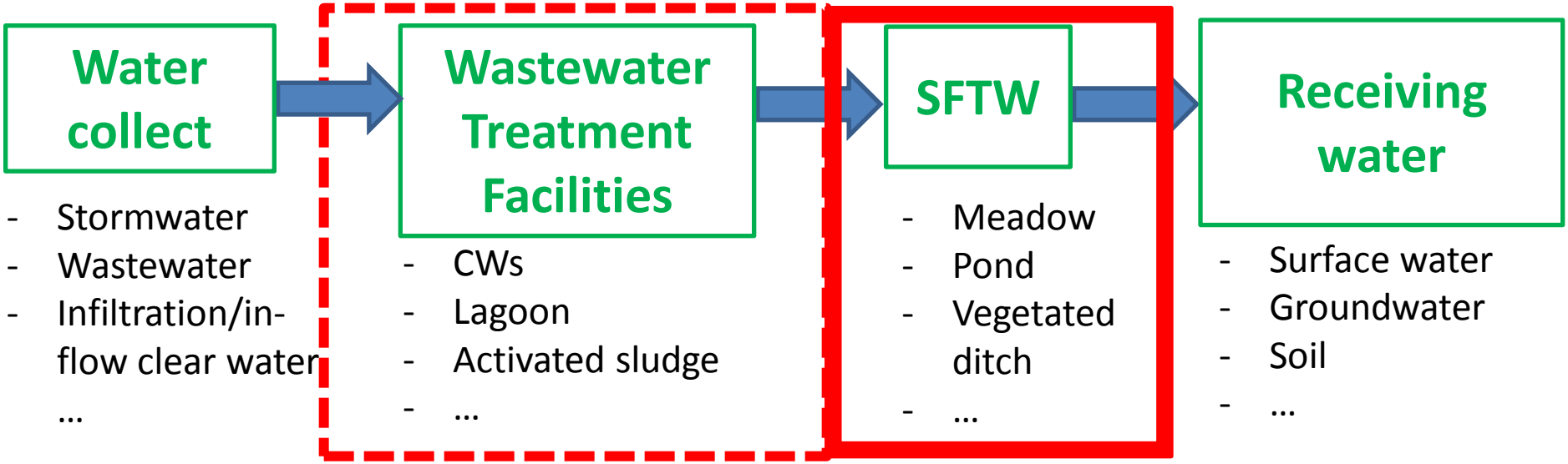
- **Nuel** PhD study : **« Dynamics of pharmaceutical compounds in Surface Flow Treatment Wetland (SFTW)»**
- PhD aims:
  - To quantify 81 phamaceutical compounds from different SFTW compartments (water, plants, soil, invertabrates);
  - To determine long term purifying capacities of 2 full-scale SFTWs (a pond and a vegetated ditch) ;
  - To optimise the sizing of these systems after a comprehensive understanding of the SFTW hydraulic behavior.

➢ **Are different typologies influence the removal abilities of SFTW ?**

➢ **Are there pharmaceutical compounds distribution in soil, water and plants compartments ?**

- Partners: **Water Agency Rhin-Meuse**, Région Alsace, **ICUBE** (UMR7357,) , IBMP (UPR2357), ENGEES, Strasbourg University

Introduction: What is SFTW ?



SFTW: 4 main functionalities:

1. **Dispersion of releases,**
2. **Sludge & SS retention,**
3. **Hydraulic peak attenuation,**
4. **Additional pollutant mitigation.**

Processes and key issues related to SFTW

Processes:

- Infiltration,
- Evapotranspiration,
- Biological degradation
- Nutrients storage by plant,
- Photo-degradation,
- Settling particulate matter,
- …

Key issues:

- No rule for design.
- What is the relative impact of the above processes on the SFTW good functioning ?
- What are their removal abilities for drug compounds ?

Aim of this study

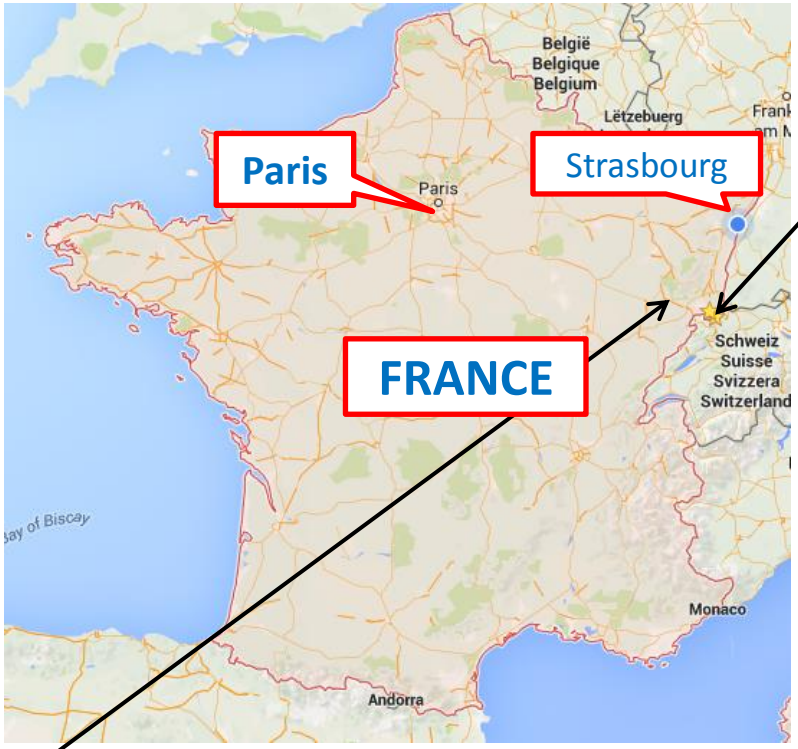
Highlight the drug residues removal abilities and their distributions to plants and mud, throughout the sampling sessions in 2 different SFTWs: a pond and a vegetated ditch.

Method

- **Regular sampling sessions** during two years on 2 different SFTWs: a pond and a vegetative ditch
- **81 pharmaceutical compounds and metabolites scaned and** quantified by Ultra Performance Liquid Chromatography coupled to mass spectrometry (UPLC-MSMS)

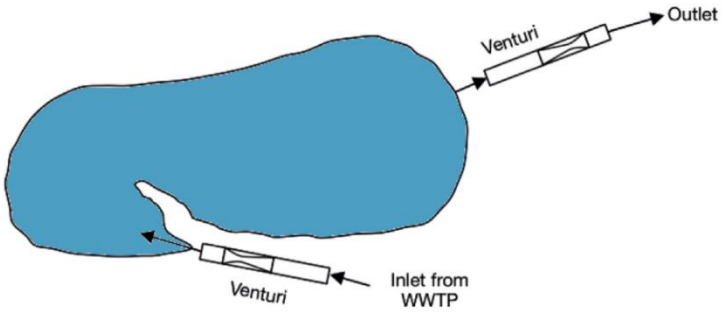
Materials & method for sampling sessions in SFTW and drug quantification

Sites presentation



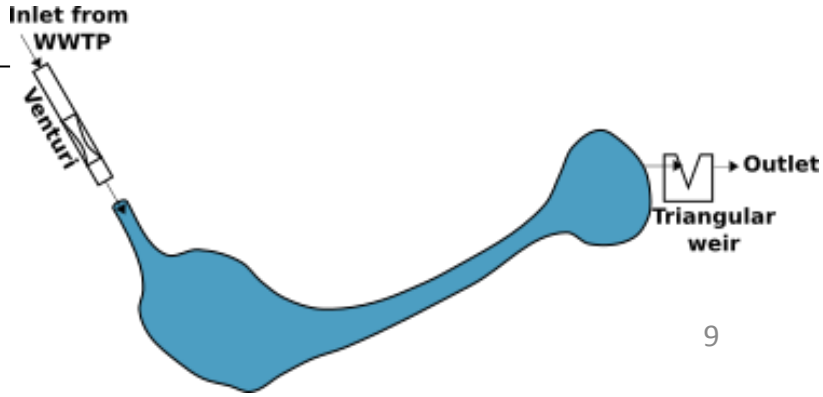
LUTTER

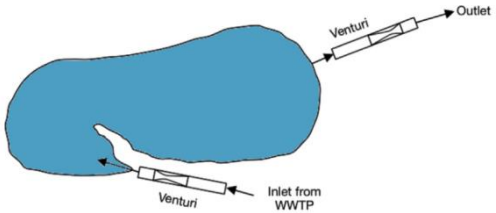
Two cities connected: Lutter & Raedersdorf
Treatment capacity: 808 PE
Sewage collection system: Wastewater and runoff
Treatment facility: VFCW +SFTW

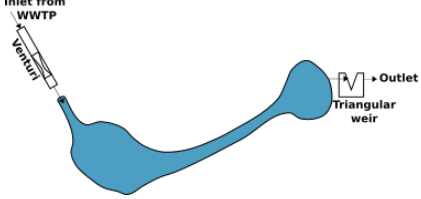


FALKWILLER

City connected: Falkwiller, Gildwiller & Hecken
Treatment capacity: 1 450 PE
Sewage collection system: wastewater and runoff
Treatment facility : VFCW + SFTW

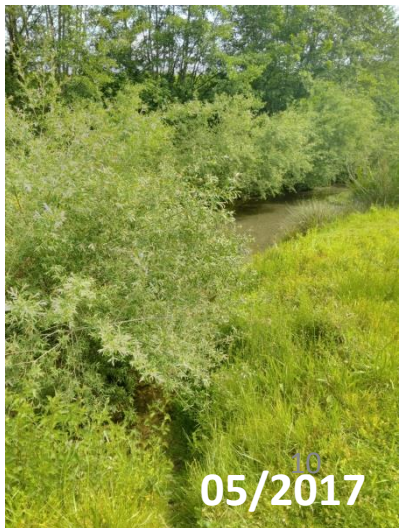






Sites presentation

Lutter SFTW		Falkwiller SFTW
2009	Construction	2010
450 m^3/day	Reference flow rate	1 080 m^3/day
Local plants	Plant	Local plants
Pond	Type	Vegetated ditches
1 to 4	Slope	1 to 1
0,3 – 0,9	deep (cm)	0,3
750	Surface (m^2)	140
425	Volume (m^3)	60
Peak attenuation Sedimentation Photodegradation Evaporation Infiltration	Expected mechanisms	Sedimentation Evapotranspiration Plant uptake Peak attenuation Infiltration



Materials

Weather stations

Ultrasound sensors (inlet & outlet)

Automatic and chilled samplers (inlet & outlet) controlled by the flow rate

Multiparameters probes for: PH, OD, T°, redox, Salinity,…

Sample bags, a cooler, laboratory gloves, pruning shears, …

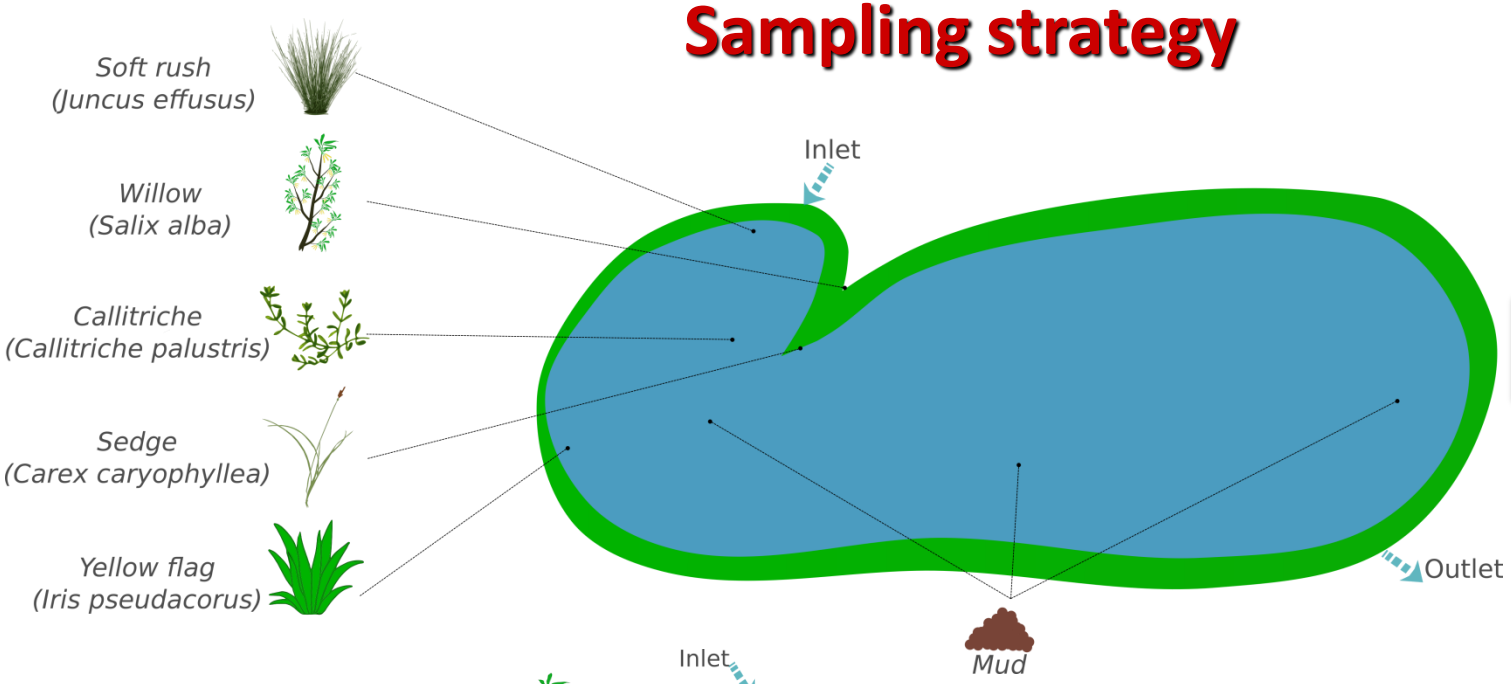
Sampling strategy

- Sessions every two months during two years

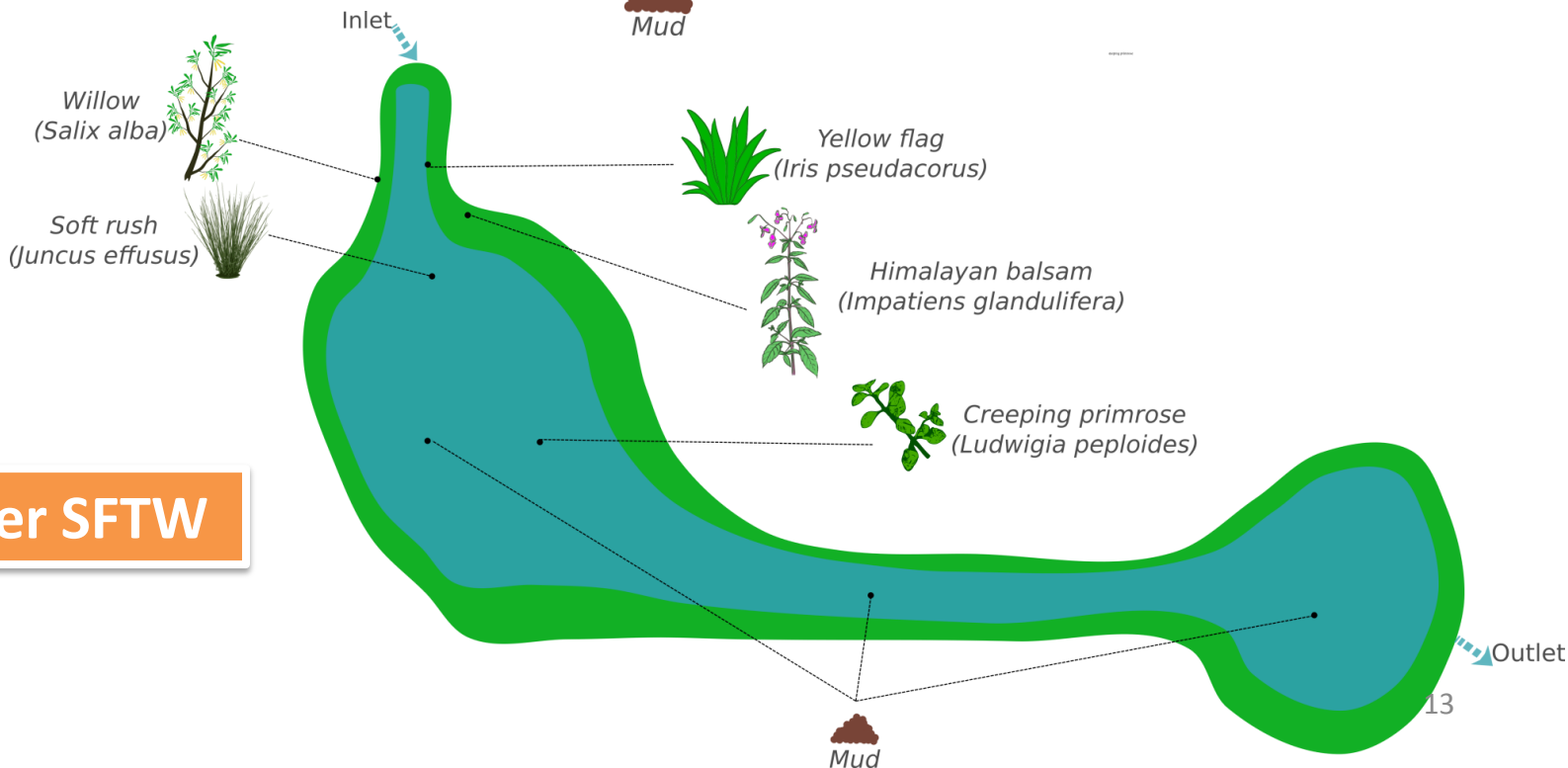
- Samples:
  - Inlet and oulet water ≈> 7 Liters
  - 5 different plant specis => 150 g per plant
  - 1 composite mud sample => 100 g

- Data acquired in situ:
  - Weather parameters
  - Inlet and outlet flowrates
  - Inlet and outlet physicochemical parameters (PH, OD, T°, redox, Salinity,…)

Sampling strategy



Lutter SFTW

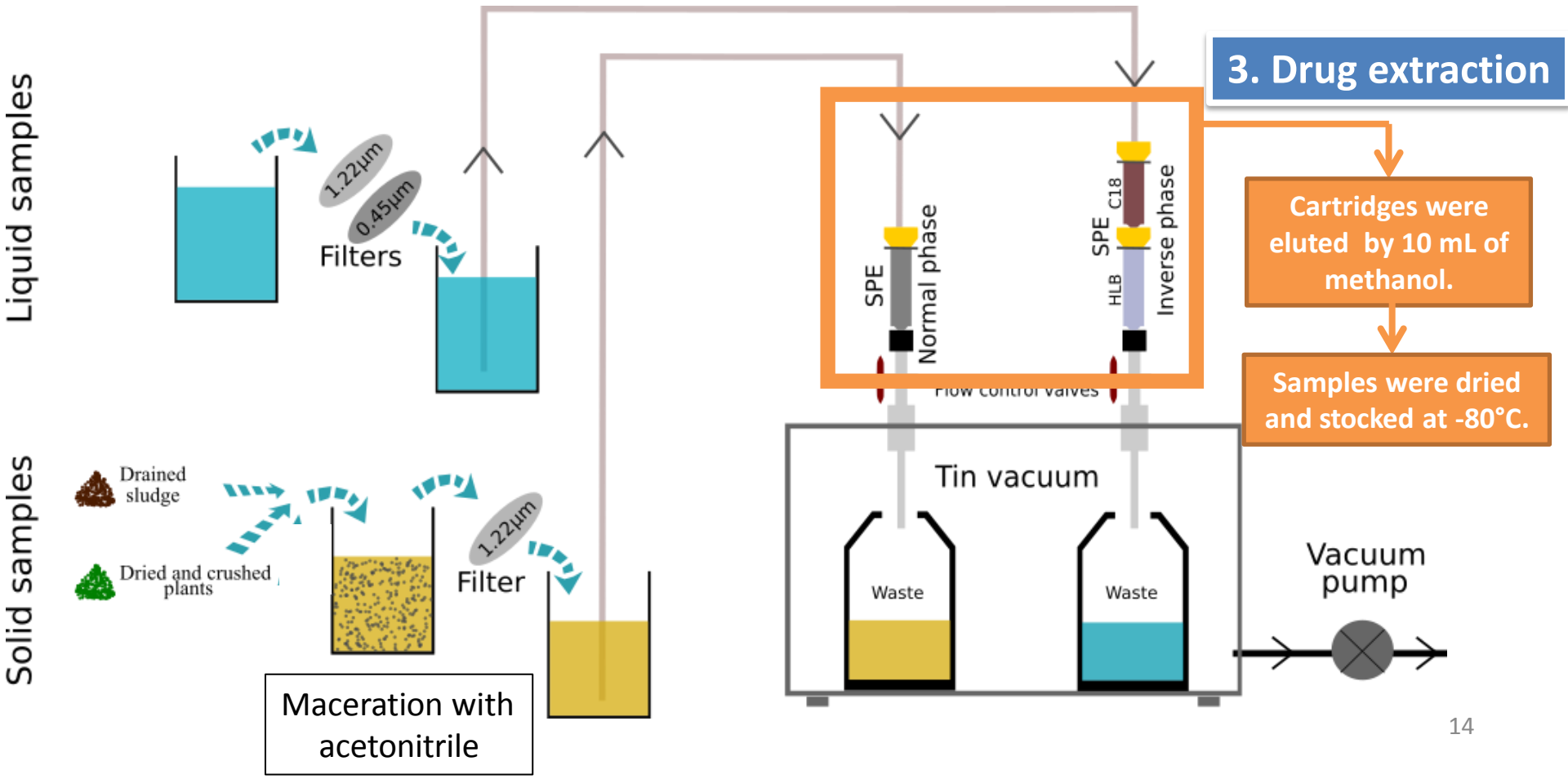


Falkwiller SFTW

Drug extraction from liquide and solide samples

1. Conditioning

2. SPE cartridges concentrate and stock drugs



3. Drug extraction

Cartridges were eluted by 10 mL of methanol.

Samples were dried and stocked at -80°C.

Drug quantifications: UPLC-MSMS

4. Solubilization

400 µL
10 % Methanol

30 µL
analysed

5. Detection and quantification

UPLC-MSMS

Samples	Drug concentration units
Liquid	µg/L
Solid (plants & Mud)	pg/g

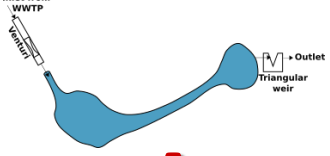
Thanks to specific drug standard curves

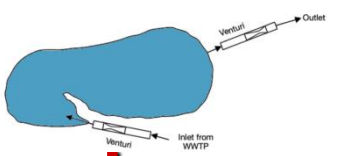
6. Removal capacities

$$= \frac{C_{Inlet} \cdot V_{Inlet} - C_{Outlet} \cdot V_{Outlet}}{C_{Inlet} \cdot V_{Inlet}}$$

C: Drug concentrations ; V: Daily volumes

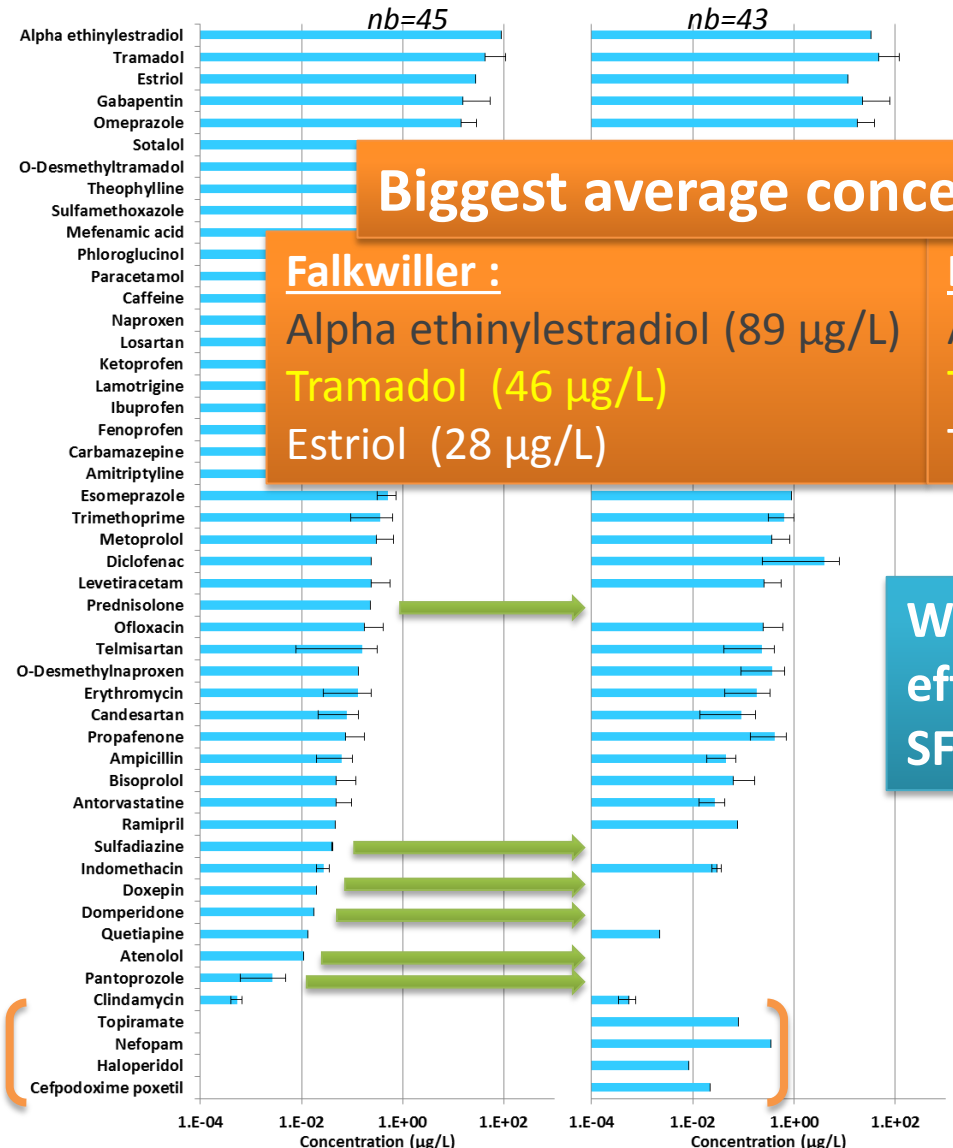
Results on the drug residues removal capacities and drug distributions to plants and mud in SFTWs

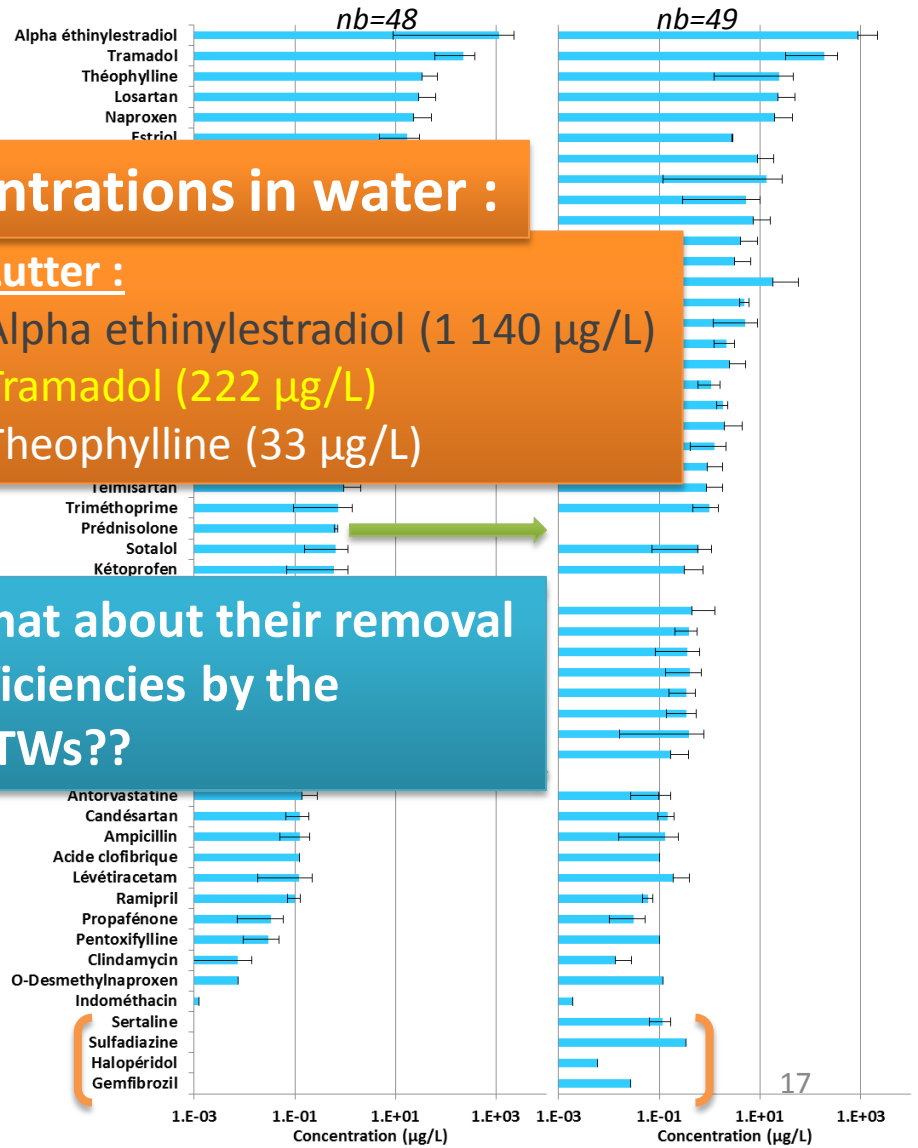




Average drug concentrations in liquid samples

Falkwiller SFTW : Inlet Outlet **Lutter SFTW :** Inlet Outlet



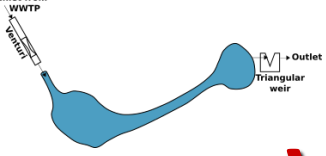


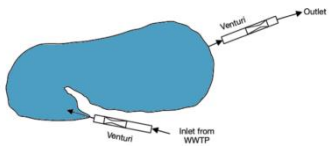
Biggest average concentrations in water :

Falkwiller :
Alpha ethinylestradiol (89 µg/L)
Tramadol (46 µg/L)
Estriol (28 µg/L)

Lutter :
Alpha ethinylestradiol (1 140 µg/L)
Tramadol (222 µg/L)
Theophylline (33 µg/L)

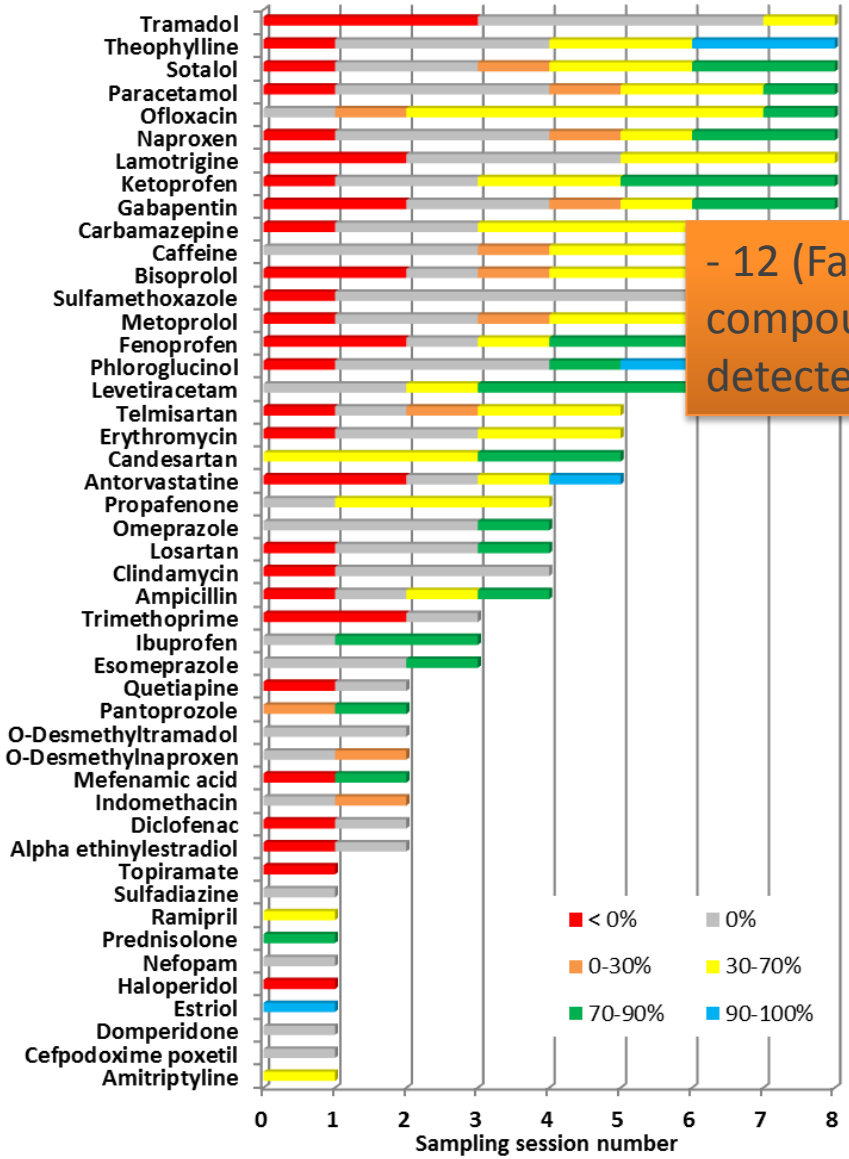
What about their removal efficiencies by the SFTWs??



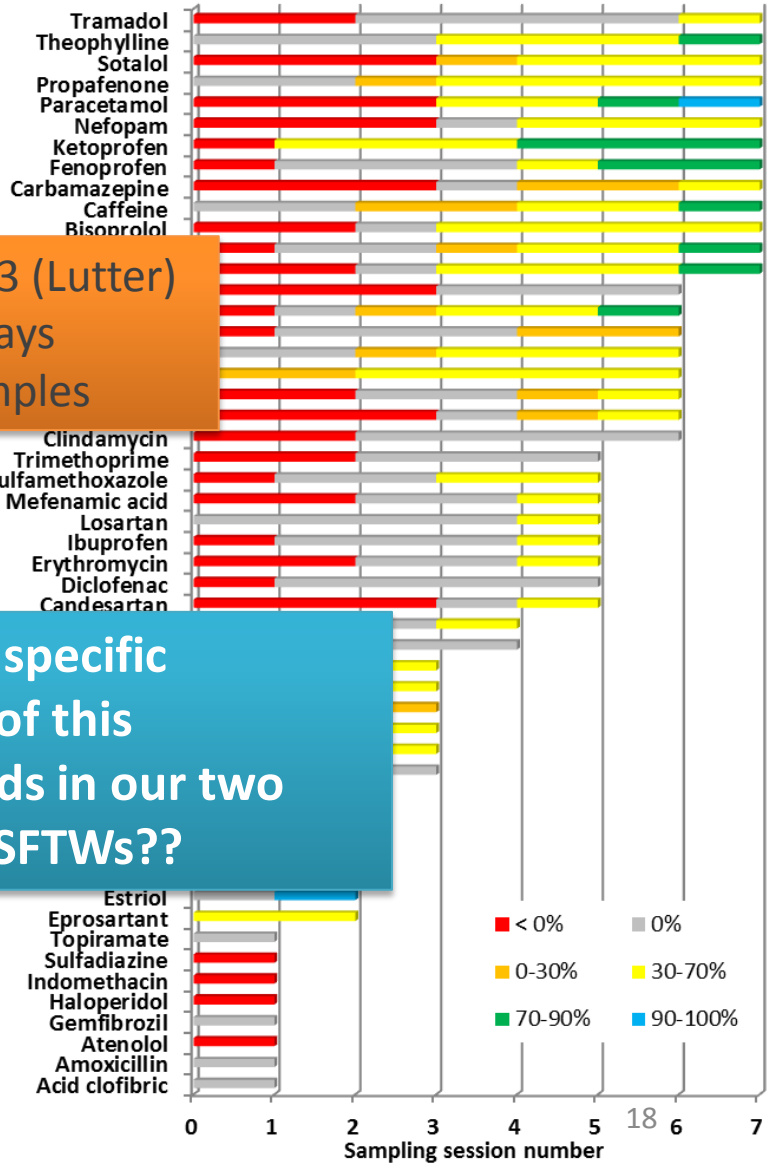


Variable drug removal efficiencies

Falkwiller SFTW :

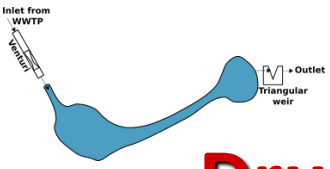


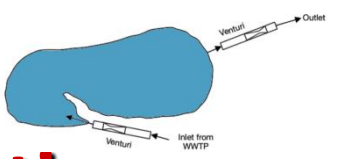
Lutter SFTW :



- 12 (Falkwiller) and 13 (Lutter) compounds were always detected in water samples

Is there a specific behavior of this compounds in our two different SFTWs??





Drug compounds behavior characterization

Falkwiller SFTW :

-Removal efficiencies ≈ 13 %

-Relatively low inlet and outlet concentrations

- High variability and high inlet and outlet concentrations

Lutter SFTW :

- Good removal efficiencies ≈ 40 %

There is different behaviors of the drug compounds due to the SFTW typologies

- High inlet and outlet concentrations (29 and 20 µg/L)

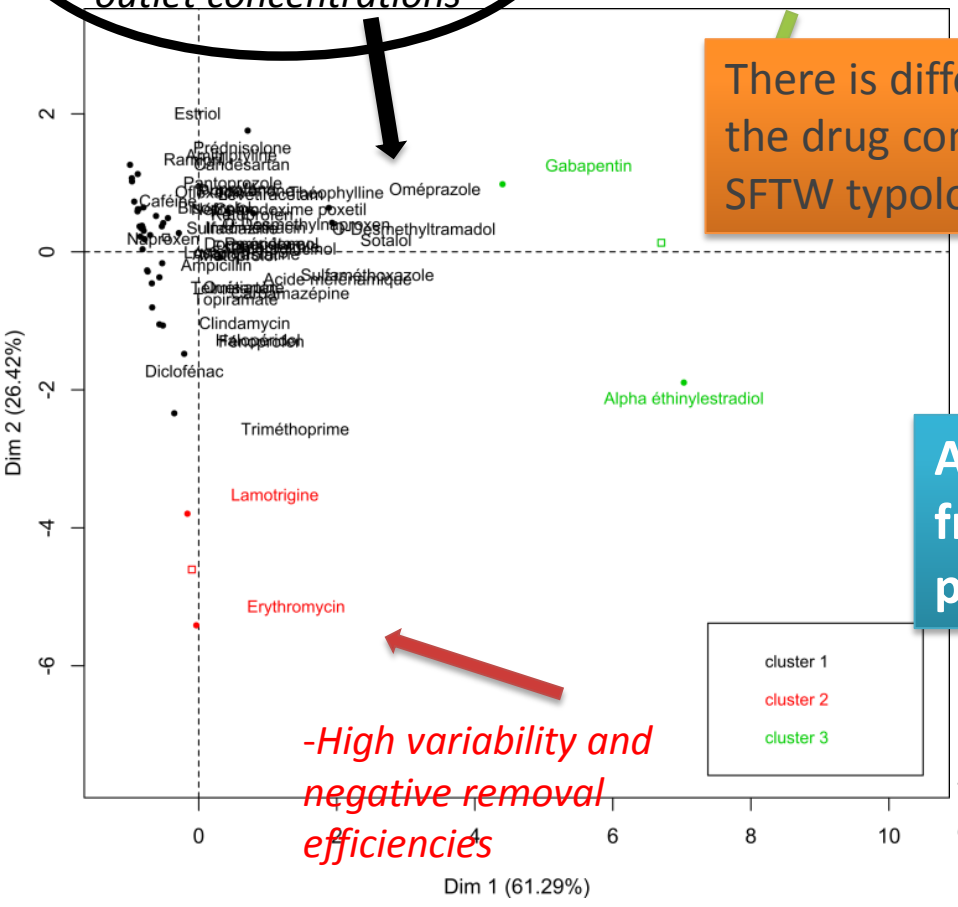
Are there drugs transfers from the water to the plants??

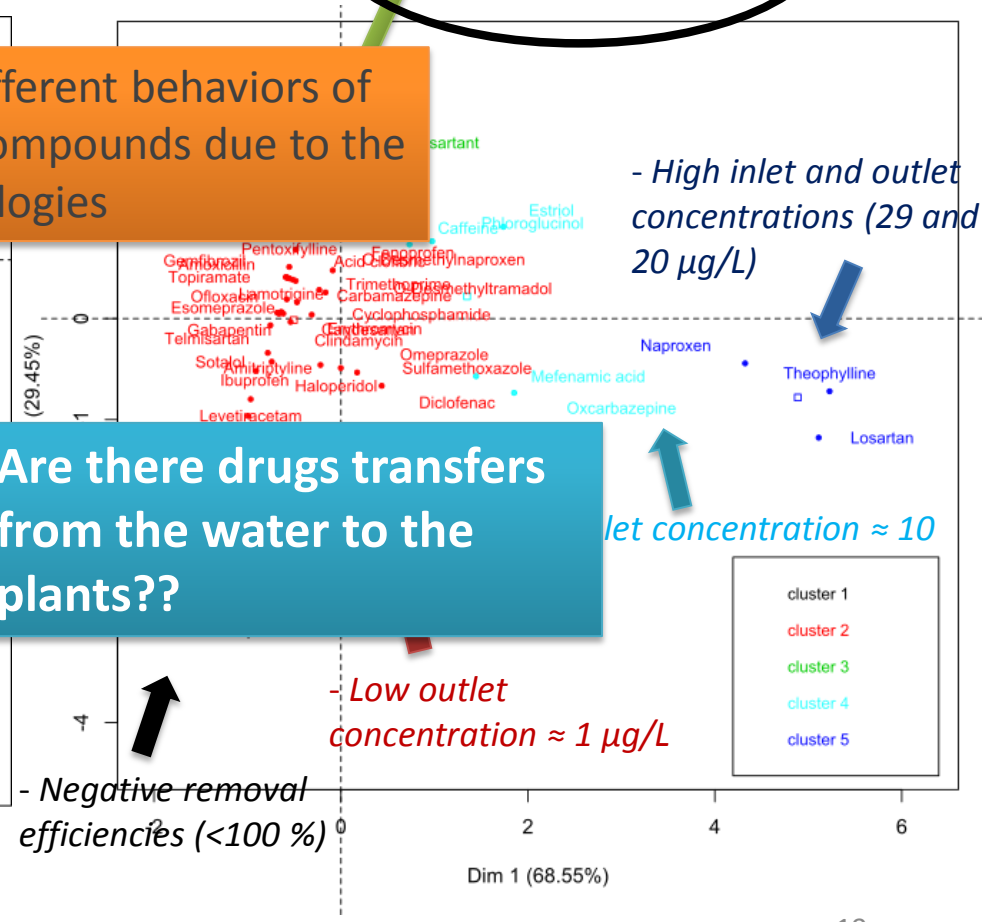
-High variability and negative removal efficiencies

- Negative removal efficiencies (<100 %)

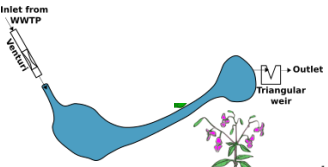
- Low outlet concentration ≈ 1 µg/L

let concentration ≈ 10

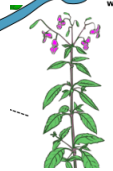




Average drug concentrations in plants samples



Falkwiller SFTW

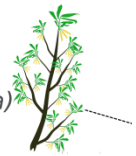


Himalayan balsam (Impatiens glandulifera) *nb=20*

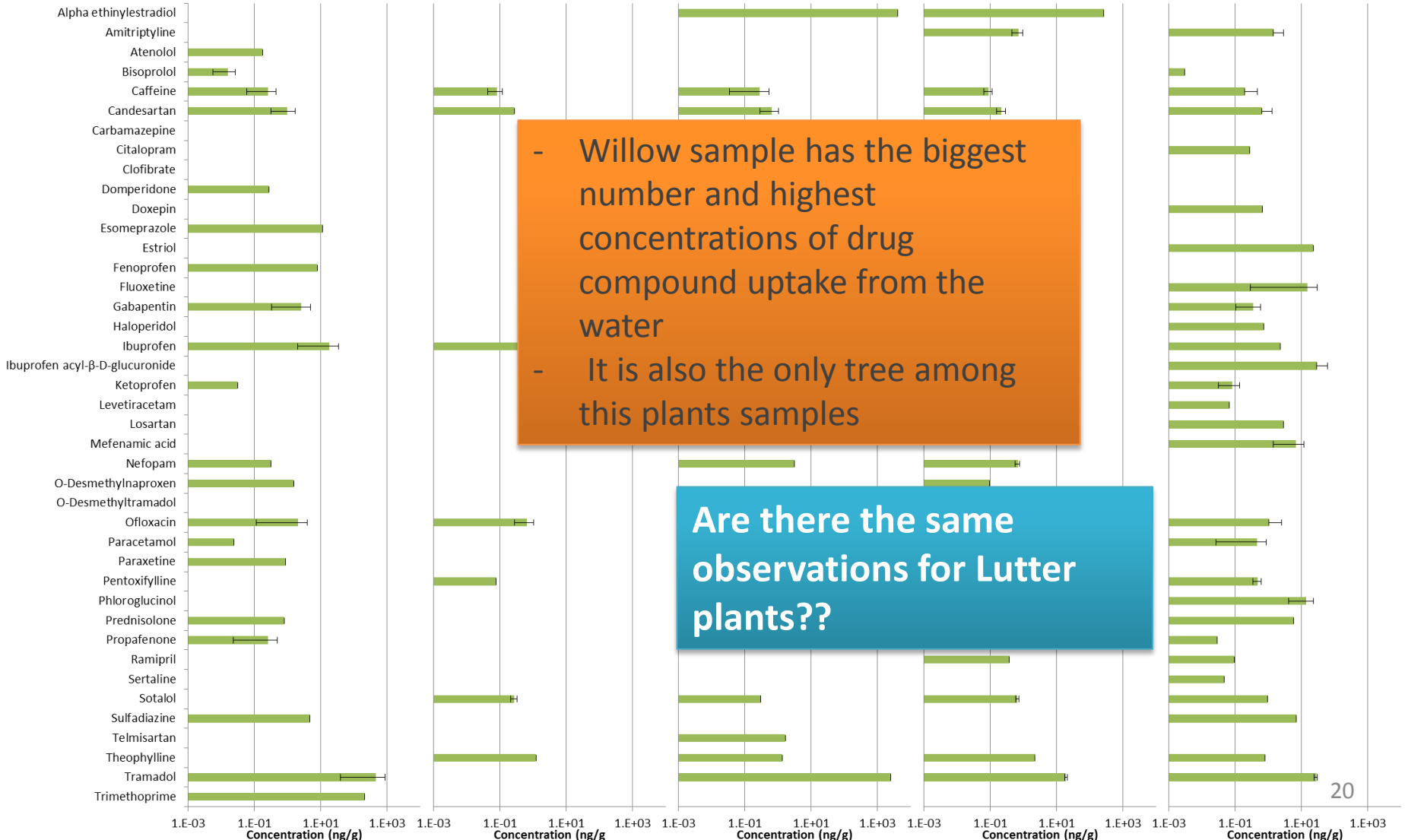
Yellow flag (Iris pseudacorus) *nb=7*

Soft rush (Juncus effusus) *nb=21*

Creeping primrose (Ludwigia peploides) *nb=21*



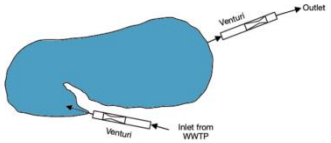
Willow (Salix alba) *nb=28*



- Willow sample has the biggest number and highest concentrations of drug compound uptake from the water
- It is also the only tree among this plants samples

Are there the same observations for Lutter plants??

Average drug concentrations in plants samples



Lutter SFTW

Callitriche (Callitriche palustris) — *nb=26*

Sedge (Carex caryophyllea) — *nb=20*

Yellow flag (Iris pseudacorus) — *nb=31*

Soft rush (Juncus effusus) — *nb=34*

Willow (Salix alba) — *nb=34*

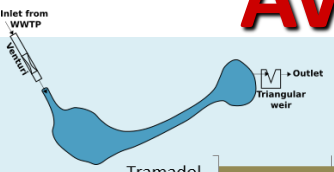
Acid clofibric
Alpha ethynylestradiol
Amitriptyline
Antorvastatine
Atenolol
Bisoprolol
Caffeine
Candesartan
Carbamazepine
Cefpodoxime poxetil
Citalopram
Clindamycin
Cyclophosphamide
Desloratadine
Domperidone
Doxepin
Eprosartant
Erythromycin
Estriol
Fenoprofen
Fluoxetine
Gabapentin
Haloperidol
Ibuprofen
Ibuprofen acyl-β-D-glucuronide
Indomethacin
Ketoprofen
L Thyroxine
Levetiracetam
Losartan
Mefenamic acid
Metoprolol
Naproxen
Nefopam
Ofloxacin
Oxcarbazepine
Paracetamol
Paraxetine
Pentoxifylline
Phloroglucinol
Prednisolone
Propafenone
Quetiapine
Ramipril
Sertaline
Sildenafil
Sotalol
Telmisartan
Theophylline
Tramadol
Trimethoprime

0.001 0.1 10 1000
Concentration (ng/g)

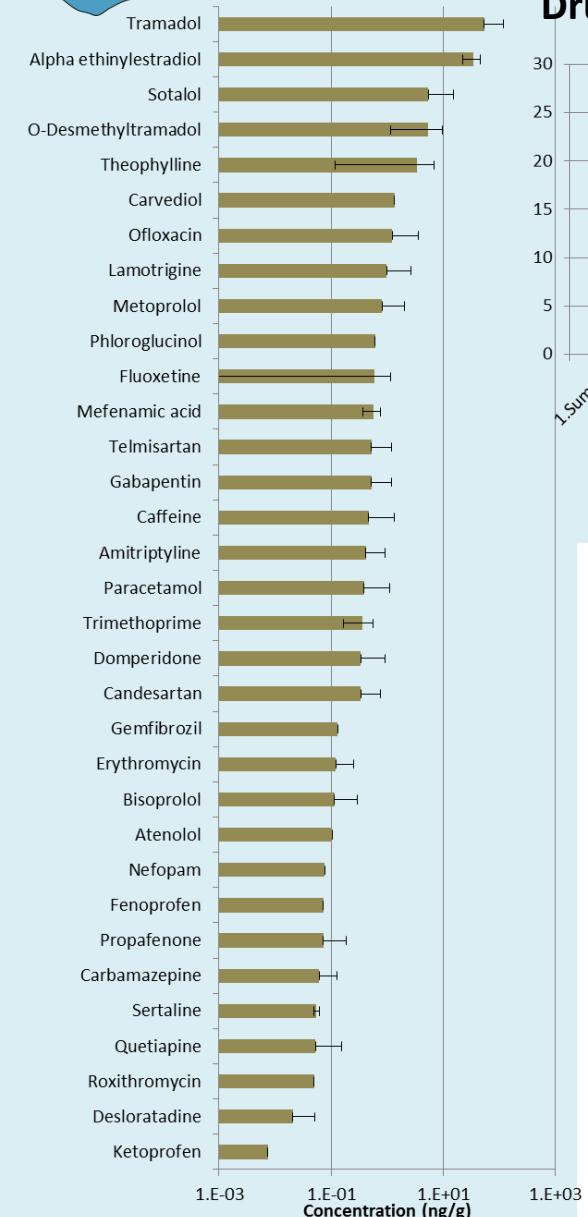
- Willow shows again good abilities to uptake drug compound from the water
- It is still the only tree among this plants samples

What about the mud stocked in the SFTWs ??

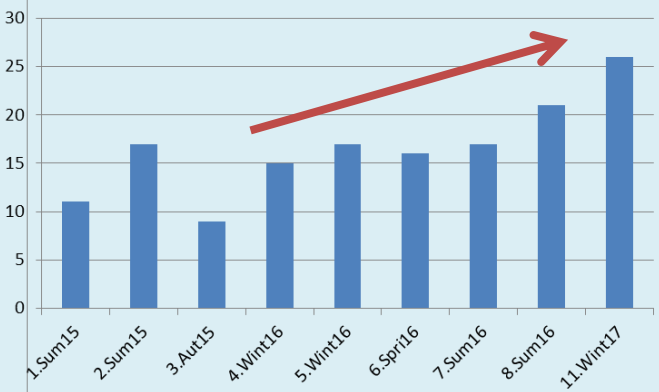
Average drug concentrations in mud



Falkwiller SFTW

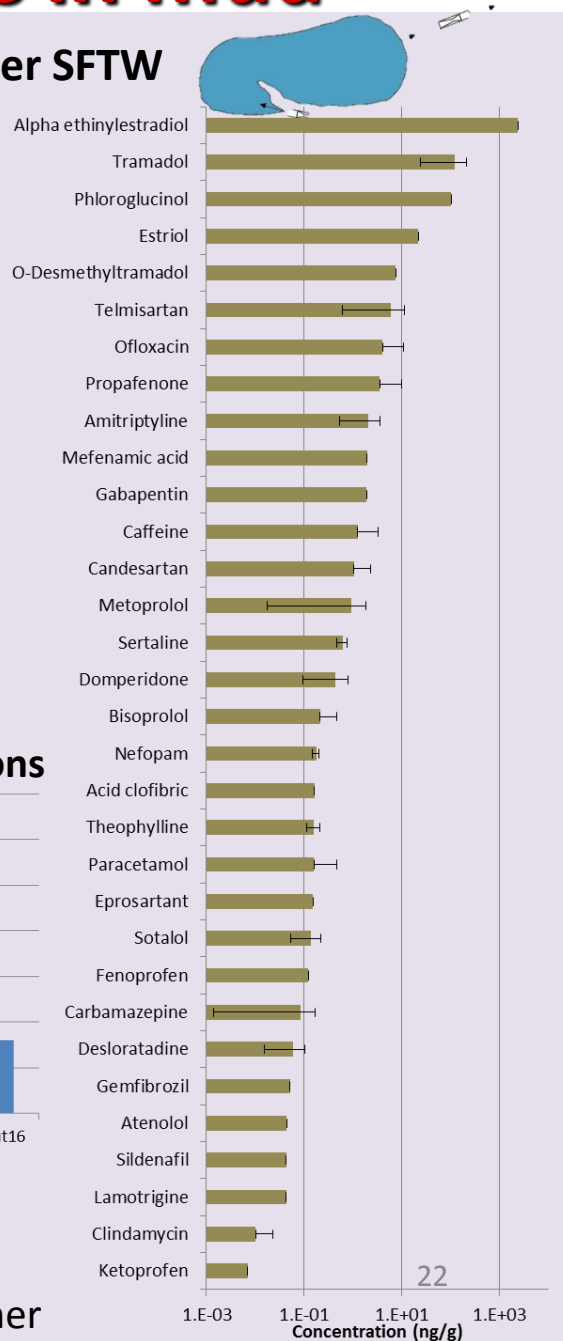


Drug number throughout the seasons

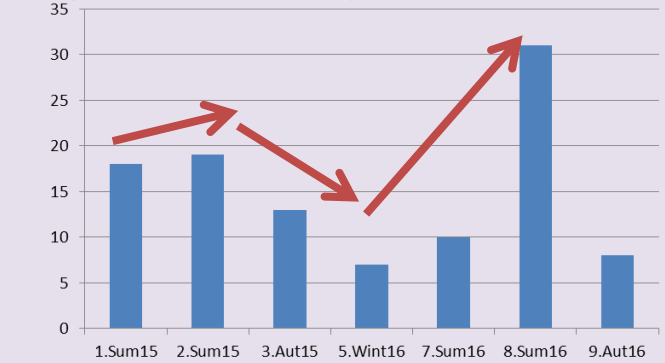


- Continuous drug adsorption

Lutter SFTW

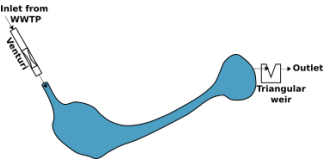


Drug number throughout the seasons



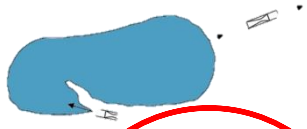
- Dynamic accumulation :
  - Release on Winter
  - Accumulation on summer

Drugs distribution to plant and mud



Falkwiller SFTW

Lutter SFTW



- There is a global transfer and distribution of drug compounds to all of the solid samples
- Willow plants also allow high uptakes of few compounds

- Low concentrations in all samples

- Low concentrations in all samples

Too high average concentrations
Alpha ethinylestradiol => Rush
Ibuprofen=> Sedge
Mefenamic acid => Willow
Tramadol => callitriche, iris and willow

CONCLUSION & OUTLOOK

- After wastewater treatment facilities, 86 drug compounds and metabolites were quantified
- SFTW removal efficiencies were highly variable for each drug compounds
- There are specific drug compounds transfers from the water to the plants
- Plants uptakes quantities of drugs are negligible in comparison to outlet drug flow
- In the pond, the drug adsorption in mud was dynamic and affected by seasonal effects whereas in the vegetated ditch, it was continuous.

Perspectives

Estimate the total mass of drug uptaken by plants and stocked in the mud before potential sludge extractions

Thanks for your attention

Contact

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