

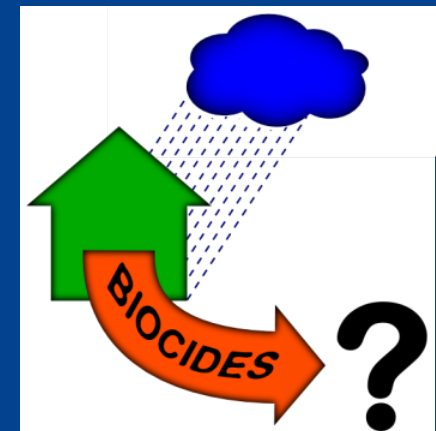


# DYNAMICS OF BIOCIDES EMISSIONS FROM BUILDINGS IN A SUBURBAN STORMWATER CATCHMENT

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U.E. BOLLMANN AND K. BESTER

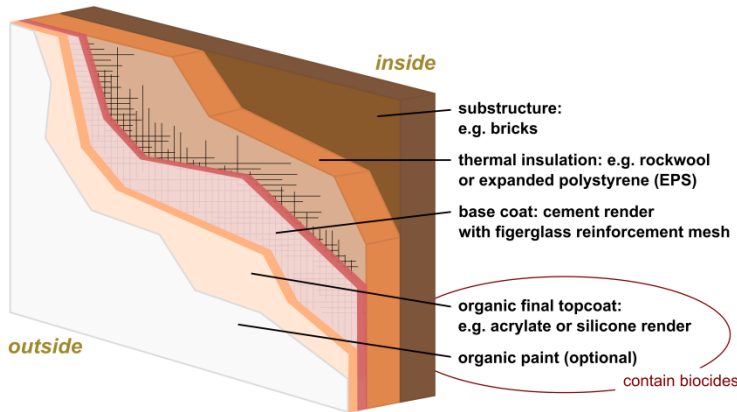
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# Biocides in building material

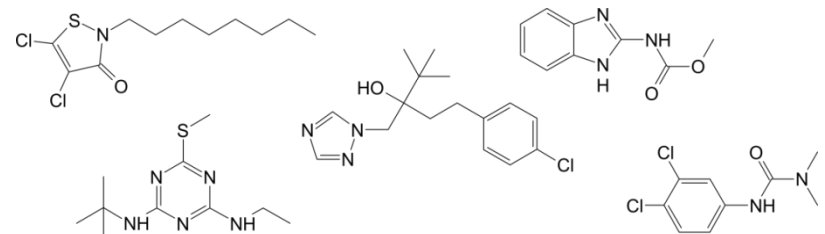


- State-of-the-art in material protection
  - In-can preservatives (Product type 6)
  - Film preservatives (Product type 7)
  - Wood protection (Product type 8)
  - Masonry preservatives (Product type 10)
  - Roof protection (flat roofs)
- Content in exterior renders and paints
  - 2-4 different biocides
  - 0.2% in render, 0.2-0.4% in exterior paints



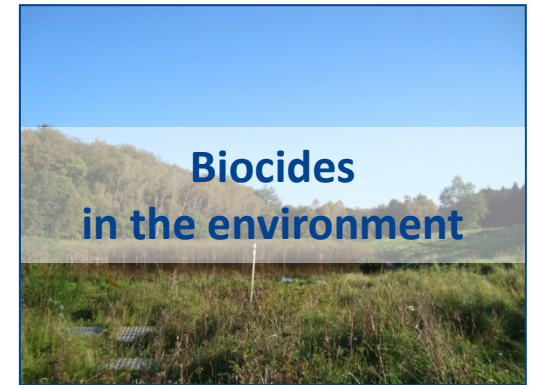
Principal construction of external thermal insulation composite system (ETICS)

- Different compounds with biocidal effects
  - Isothiazolinones, carbamates, phenylureas, triazines, triazoles



- » Hydrophilic molecules with toxic effects

# Biocides in building material



# Catchment

- Silkeborg (Denmark)

- 21.5 ha, 7 ha connected to sewer, separated sewer system
- Suburban, residential, 140 well-kept single family houses
- 5 % equipped with renders/paints, 20 % (painted) wood, 75 % brick-facades



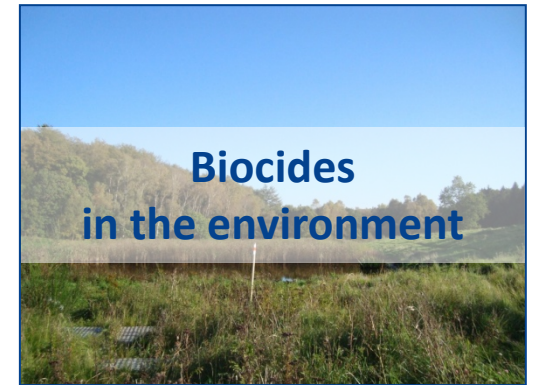
# Sampling and Analysis



- Flow proportional high resolution sampling with automatic water sampler
  - October 2011-June 2012
- Solid phase extraction
- High performance liquid chromatography tandem mass spectrometry (HPLC-MS/MS)



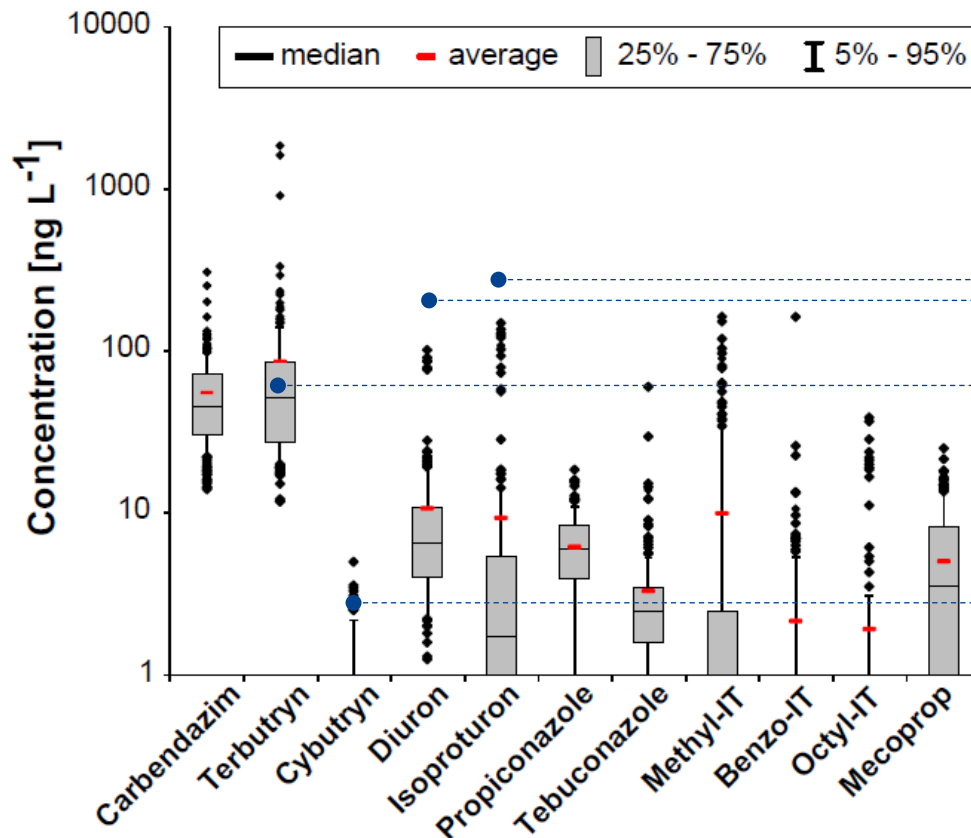
# Biocides in building material



**Which compounds? Concentrations?**  
**Emission scenarios correct?**  
Dynamics? First flush emissions?



# Concentrations in stormwater runoff



AA-EQS for fresh water  
DIRECTIVE 2013/39/EU

Isoproturon: 300 ng L<sup>-1</sup>

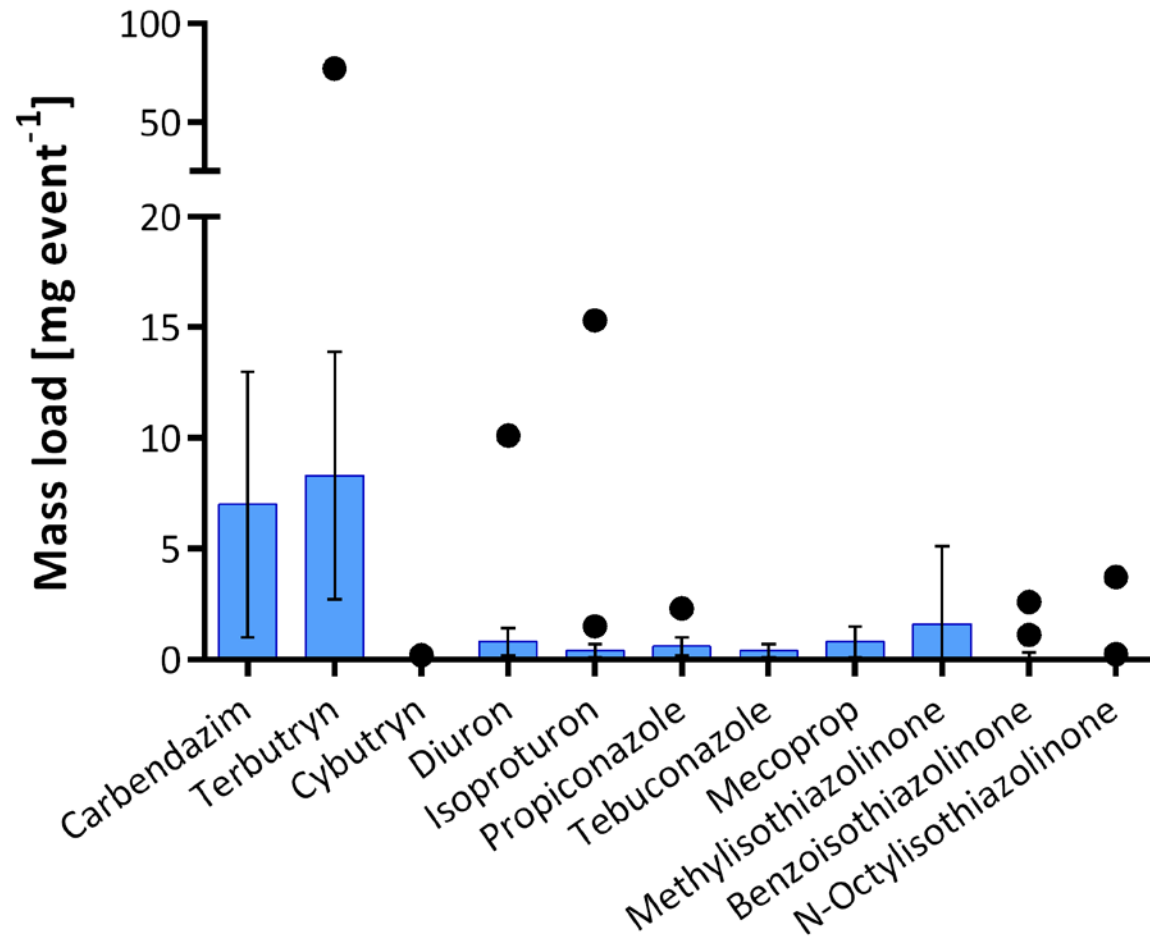
Diuron: 200 ng L<sup>-1</sup>

Terbutryn: 65 ng L<sup>-1</sup>

Cybutryn: 2.5 ng L<sup>-1</sup>

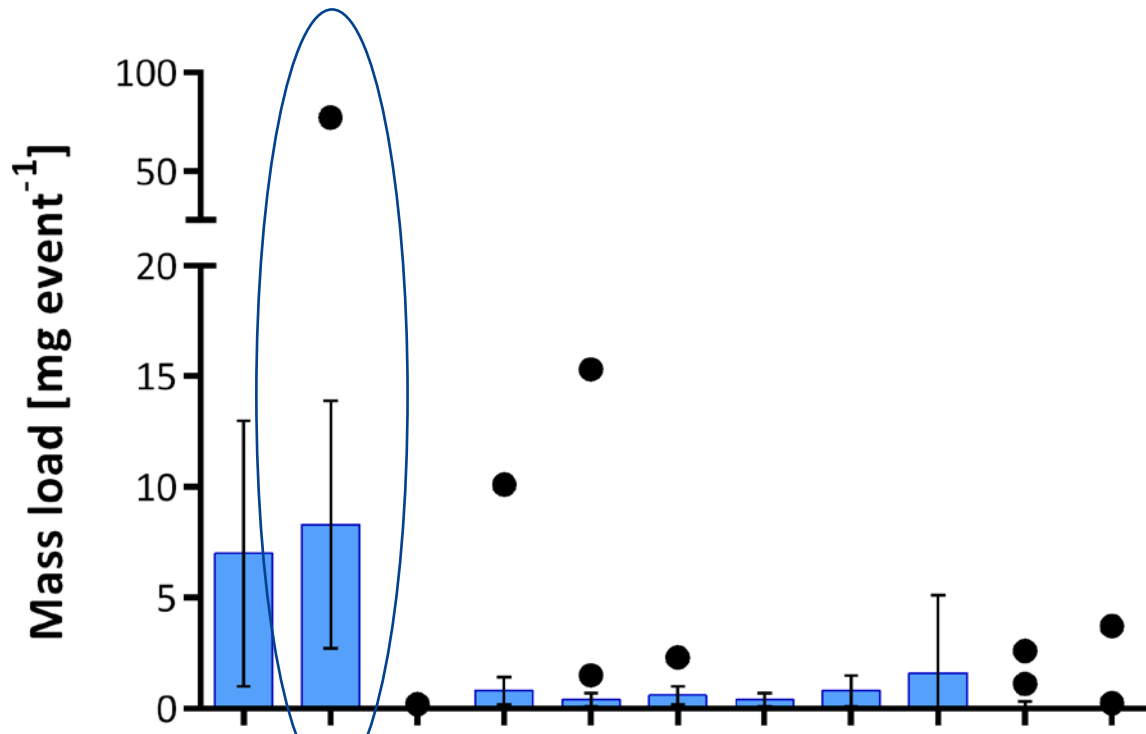
Oct. 2011-June 2012; 191 flow controlled samples, IT: Isothiazolinone

# Mass loads in stormwater runoff





# Mass loads in stormwater runoff



## Assessment for terbutryn emissions

New facade: 2-4 mg m<sup>-2</sup> event<sup>-1</sup>

Burkhardt et al., ES&T 46 (2012) 5497



peak event corresponds to 19 and 39 m<sup>2</sup> freshly treated wall about 1/4 of a normal house in the catchment

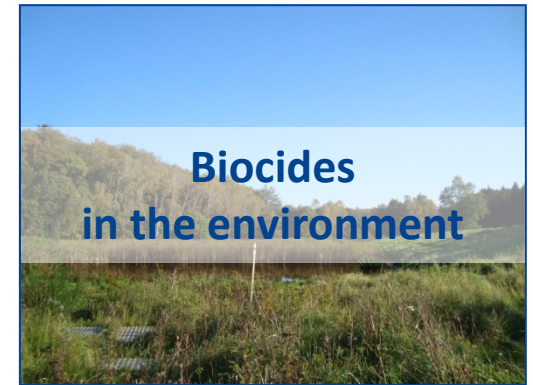
Aged facade: 0.1 mg m<sup>-2</sup> event<sup>-1</sup>

Burkhardt et al., ES&T 46 (2012) 5497



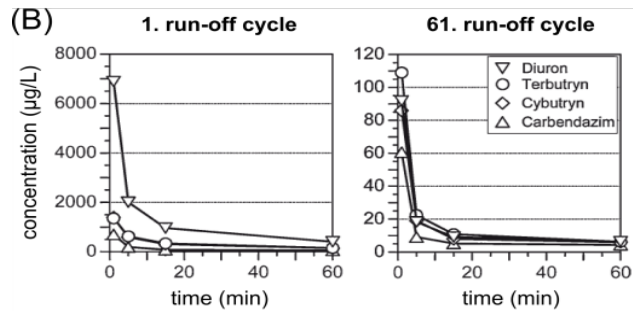
normal events represent about 80 m<sup>2</sup> treated facades one wall out of four exposed to driving rain: 3-4 houses

# Biocides in building material



Which compounds? Concentrations?  
Emission scenarios correct?  
**Dynamics? First flush emissions?**

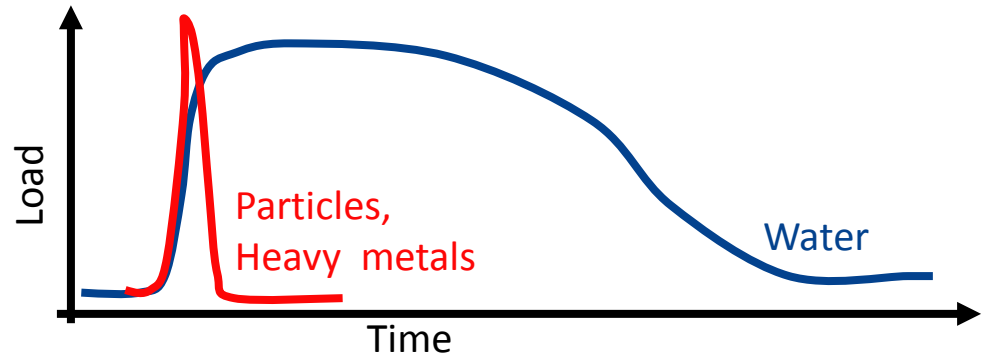
## Biocide leaching in the weather chamber



Runoff concentrations in first and 61<sup>st</sup> cycle of a forced rain experiment.

Burkhardt et al., UWSF 21 (2009) 36

## Particle mobilisation in road runoff with first flush



### First Flush

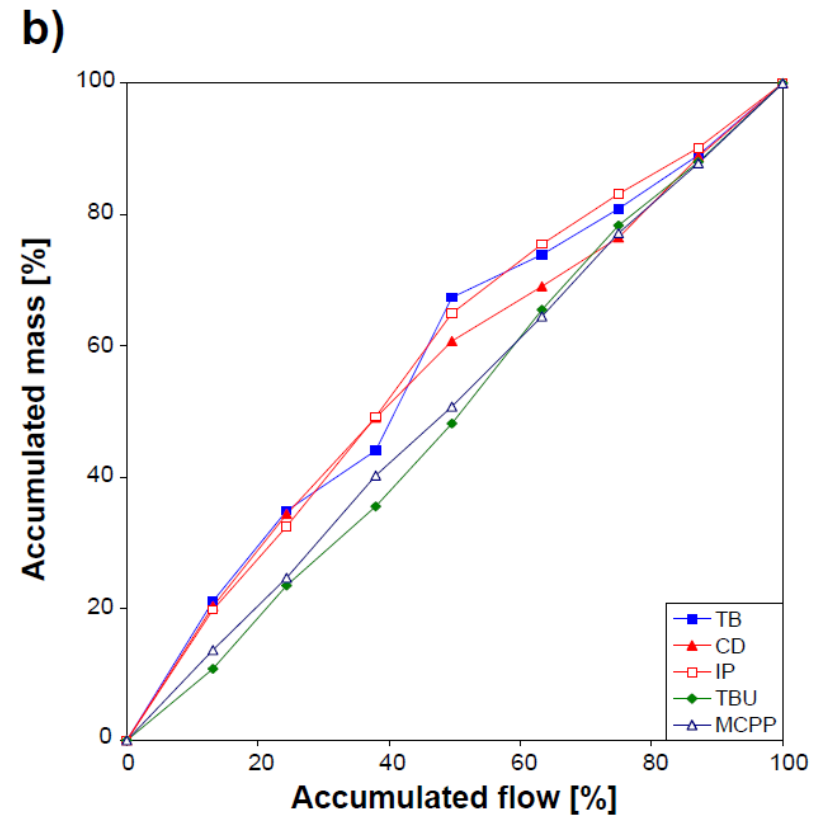
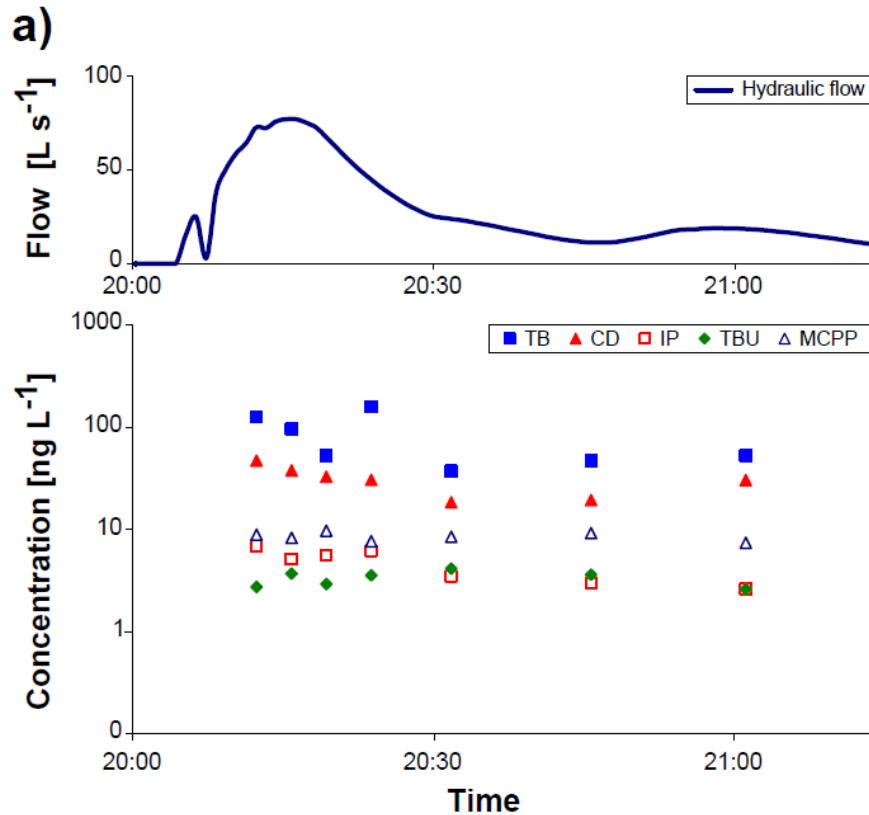
80% of the pollutant mass is transported in the first 30% of the volume of rainfall events

Bertrand-Krajewski et al., Wat Res 32 (1998) 2341

# Emission dynamics



## >> Common events



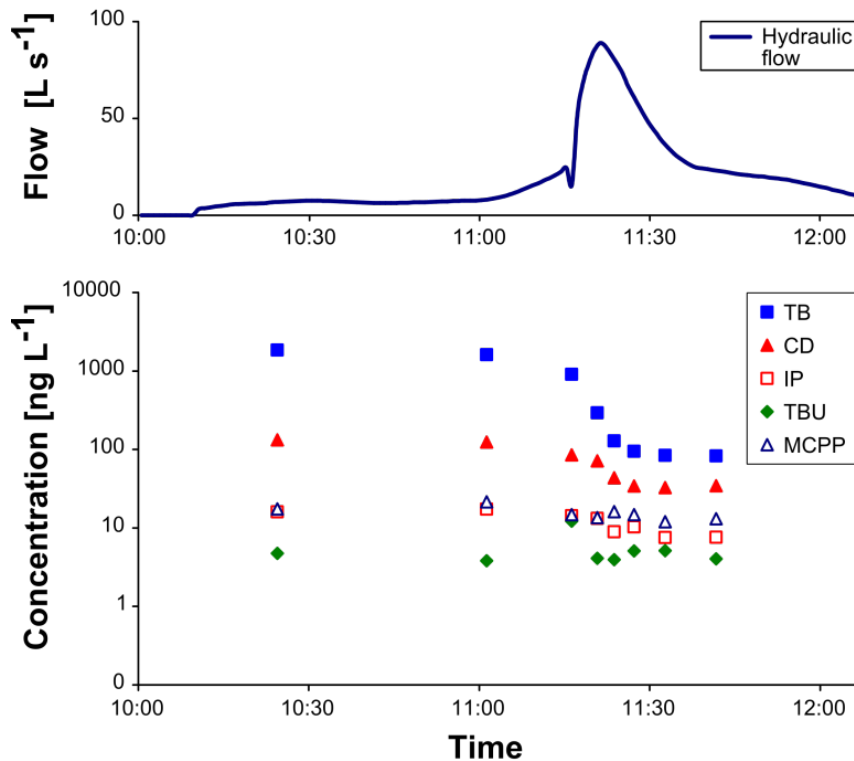
Flow controlled samples; TB: Terbutryn, CD: Carbendazim, IP: Isoproturon, TBU: Tebuconazole, MCPP: Mecoprop

# Emission dynamics

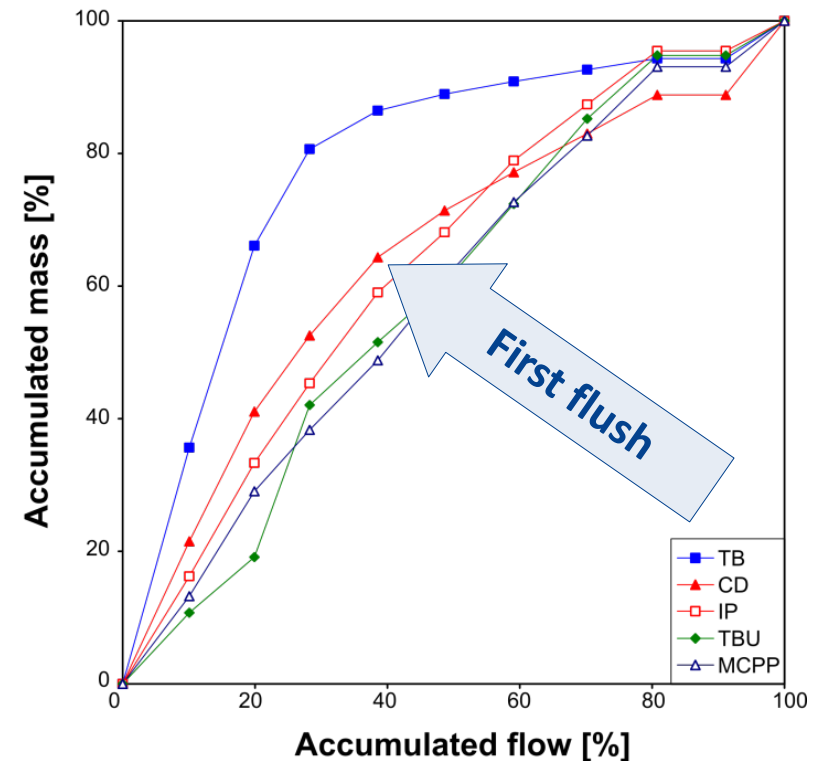


>> First flush: occurs irregularly

a)



b)

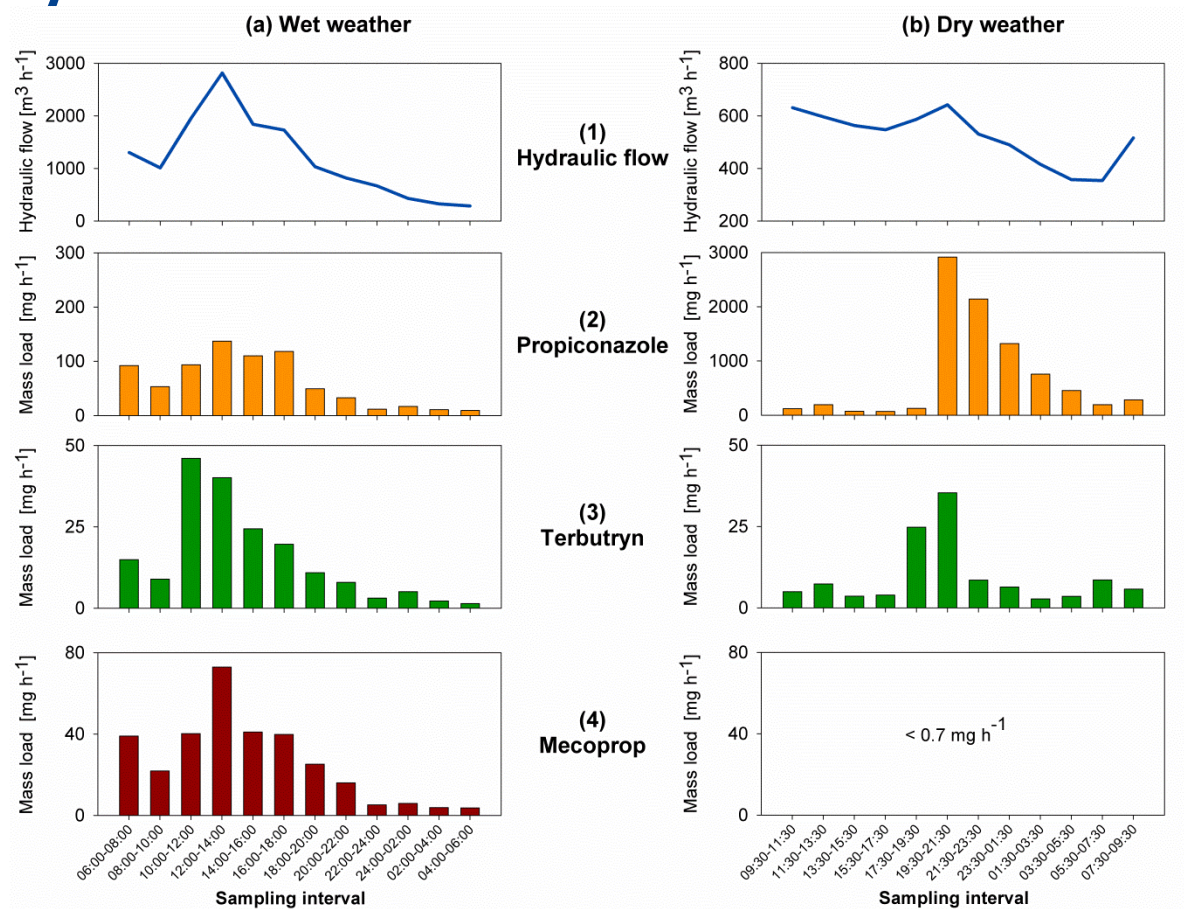


Flow controlled samples; TB: Terbutryn, CD: Carbendazim, IP: Isoproturon, TBU: Tebuconazole, MCPP: Mecoprop

# Emission dynamics – combined sewer



## >> Diurnal cycle



WWTP Bjermarken (Roskilde, Denmark),  
12x 2h combined samples of the influent

**Poster**  
Biocides in combined sewer systems



# Weather as influencing parameter

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# Weather as influencing parameter



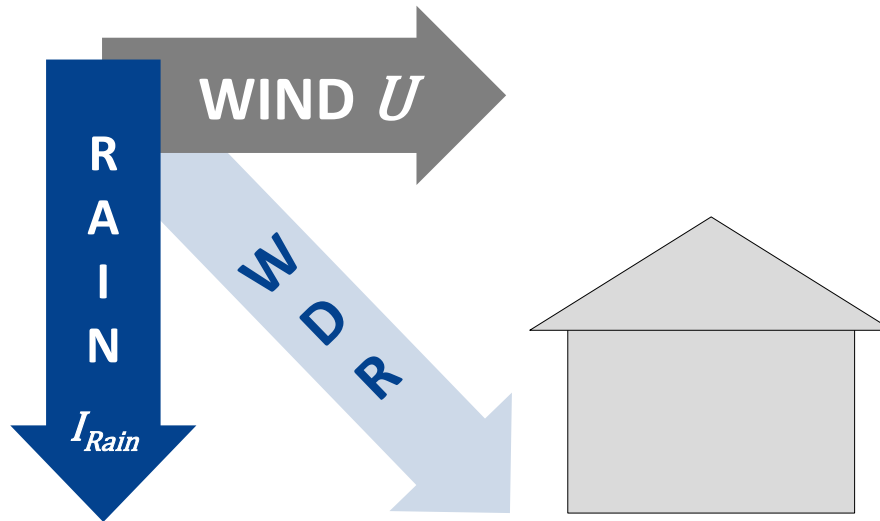
- What is influencing the leaching?
- Correlation of the concentration or acc. mass load per event with:
  - Rain amount / accumulated flow
  - Length of the event
  - Rain intensity
  - Length of the dry period prior to the event
  - Wind driven rain intensity



# Weather as influencing parameter



>> Run-off volume  $\propto$  Wind-driven rain (WDR)



**Mass flow  $M$  during respective event**

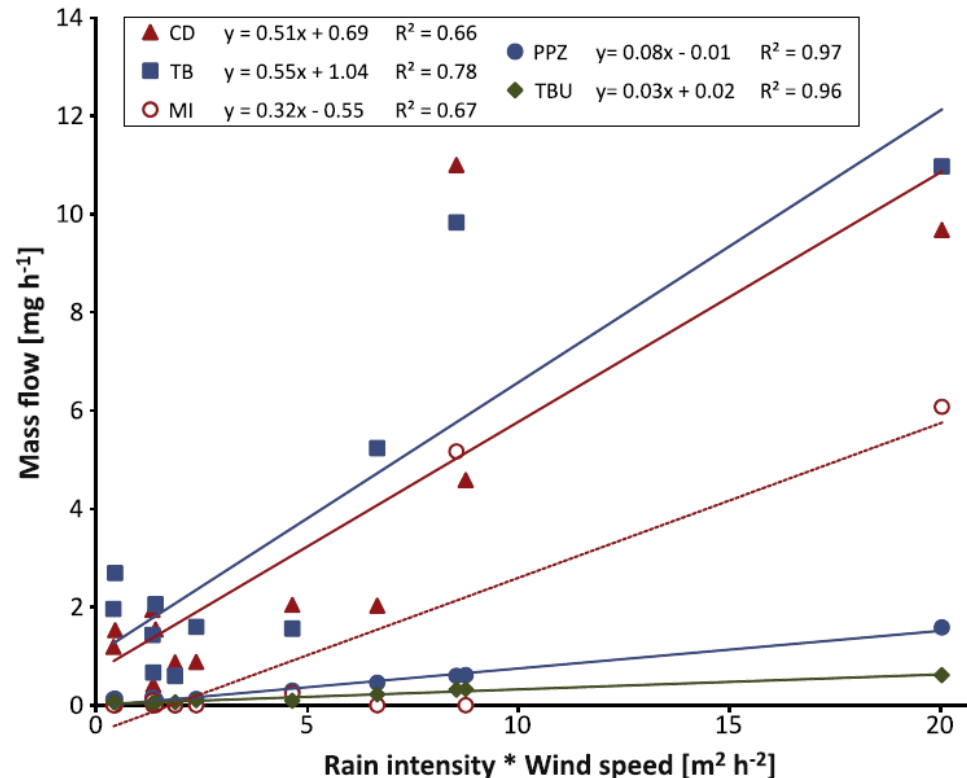
$$M_{WDR} = C_C C_S I_{Rain} U$$

with  $C_S$  = substance specific constant  
 $C_C$  = catchment specific constant

# Weather as influencing parameter



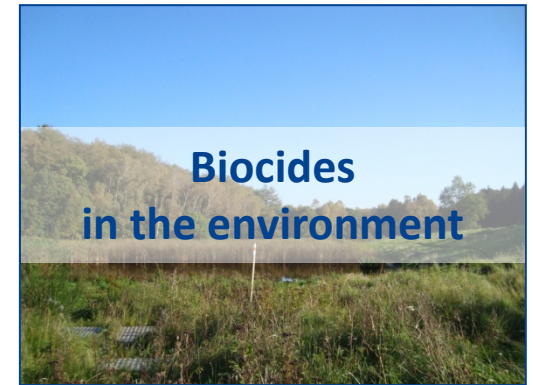
>> Wind-driven rain causes leaching of biocides from stormwater catchment



Stormwater catchment Silkeborg North, average mass flow per event

CD: Carbendazim, TB: Terbutryn, MI: Methyisothiazolinone, PPZ: Propiconazole, TBU: Tebuconazole

# Biocides in building material



**Conclusions**

# Conclusions

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- Monitoring

- Direct emissions via stormwater in suburban areas
- Biocides in urban areas can exceed quality standards
- Different footprint than in other European countries



Stormwater  
treatment  
necessary!

- Emission dynamics

- Usually rather constant throughout an event
- Irregular first flush phenomena
- Indication for correlation between occurrence in stormwater and wind driven rain



- No first-flush stormwater treatment possible
- Unnecessary loss of substance
- Fine tune application?





# Acknowledgement

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- Scientific cooperation:
  - Jes Vollertsen (Aalborg University)
  - Jan Carmeliet and Timothy Wangler (ETH Zurich, Switzerland)
  - Michael Burkhardt (HSR, Rapperswil, Switzerland)
- Diverse companies in the sector for the willingness to share materials and knowledge
- Financial support:
  - Danish EPA through the project *Methods for the improvement of scenarios concerning the emission of biocides from buildings* (667-00065 & 667-00066)
  - AUFF grant: *Advanced water purification using bio-inorganic nano catalysts and soil filters* ([www.waterpurification.au.dk](http://www.waterpurification.au.dk))

**Thank you for your attention!**



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