DYNAMICS OF BIOCIDES EMISSIONS FROM BUILDINGS IN A SUBURBAN STORMWATER CATCHMENT

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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

• Different compounds with biocidal effects
  – Isothiazolinones, carbamates, phenylureas, triazines, triazoles

  » Hydrophilic molecules with toxic effects

Principal construction of external thermal insulation composite system (ETICS)
Biocides in building material

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

Catchment Sampling & Analysis

Biocides in the environment

Leaching
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)

Bollmann et al., Wat Res 56 (2014) 66
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\(^{-1}\)
- Diuron: 200 ng L\(^{-1}\)
- Terbutryn: 65 ng L\(^{-1}\)
- Cybutryn: 2.5 ng L\(^{-1}\)

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

Bollmann et al., Wat Res 56 (2014) 66
Mass loads in stormwater runoff

![Graph showing mass loads for various compounds in stormwater runoff.](image-url)
Mass loads in stormwater runoff

Assessment for terbutryn emissions

New facade: 2-4 mg m\(^{-2}\) event\(^{-1}\)
Burkhardt et al., ES&T 46 (2012) 5497

peak event corresponds to 19 and 39 m\(^2\) freshly treated wall about \(\frac{1}{4}\) of a normal house in the catchment

Aged facade: 0.1 mg m\(^{-2}\) event\(^{-1}\)
Burkhardt et al., ES&T 46 (2012) 5497

normal events represent about 80 m\(^2\) treated facades one wall out of four exposed to driving rain: 3-4 houses

Bollmann et al., Wat Res 56 (2014) 66
Biocides in building material

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

Biocides in the environment

leaching
Emission dynamics

Biocide leaching in the weather chamber

Runoff concentrations in first and 61st cycle of a forced rain experiment.

Burkhardt et al., UWSF 21 (2009) 36

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

Bollmann et al., Wat Res 56 (2014) 66
Emission dynamics

>> First flush: occurs irregularly

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

Bollmann et al., Wat Res 56 (2014) 66
Emission dynamics – combined sewer

>> Diurnal cycle

(a) Wet weather

(b) Dry weather

(1) Hydraulic flow

(2) Propiconazole

(3) Terbutryn

(4) Mecoprop

Sampling interval

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

Bollmann et al., Wat Res 60 (2014) 64
Weather as influencing parameter
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

Bollmann et al., Wat Res 56 (2014) 66
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: Methylisothiazolinone, PPZ: Propiconazole, TBU: Tebuconazole

Bollmann et al., Wat Res 56 (2014) 66
Conclusions

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

- 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

Stormwater treatment necessary!
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Thank you for your attention!
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