

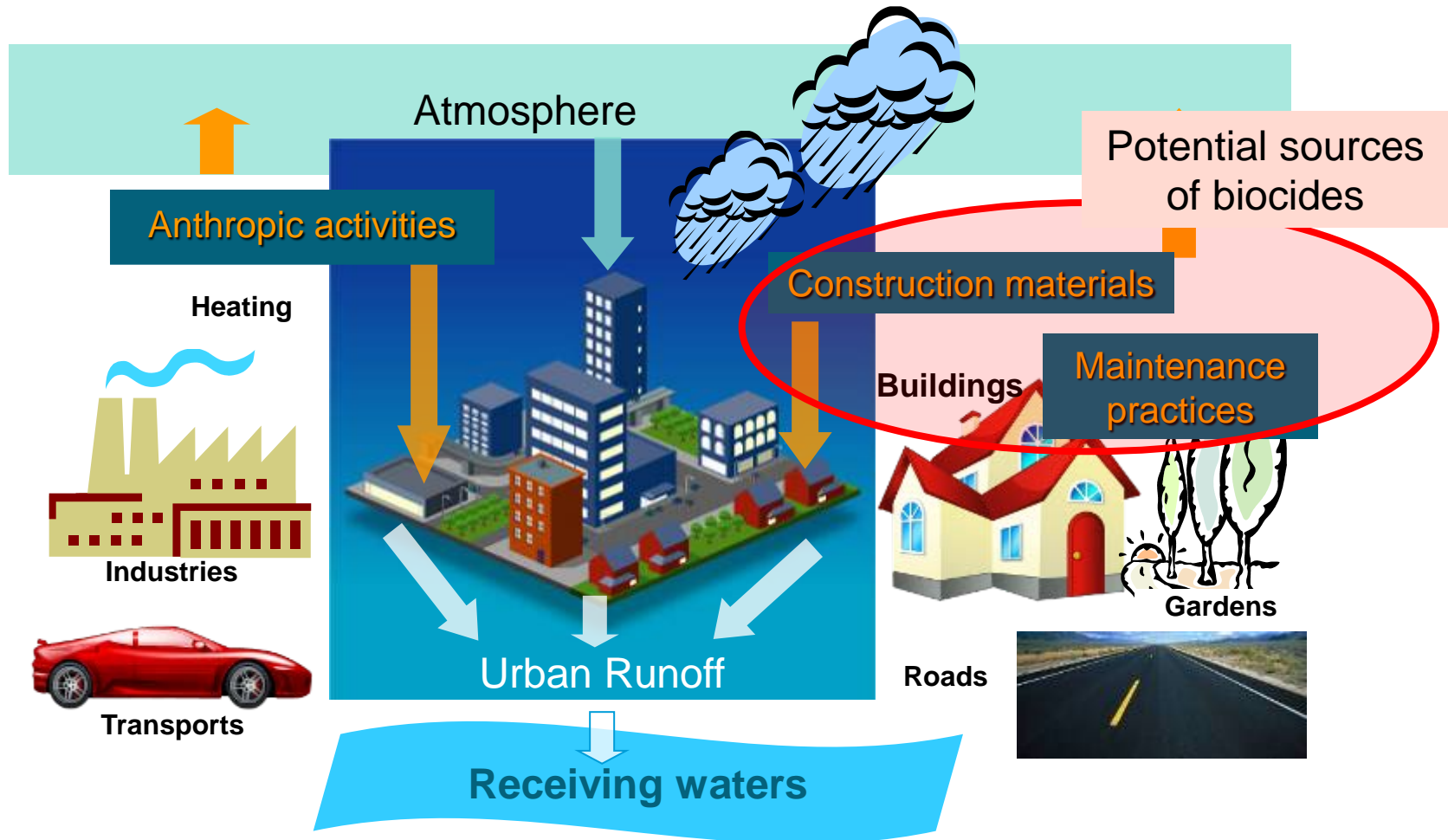
Benzalkonium runoff from roofs treated with biocides

M.C. Gromaire

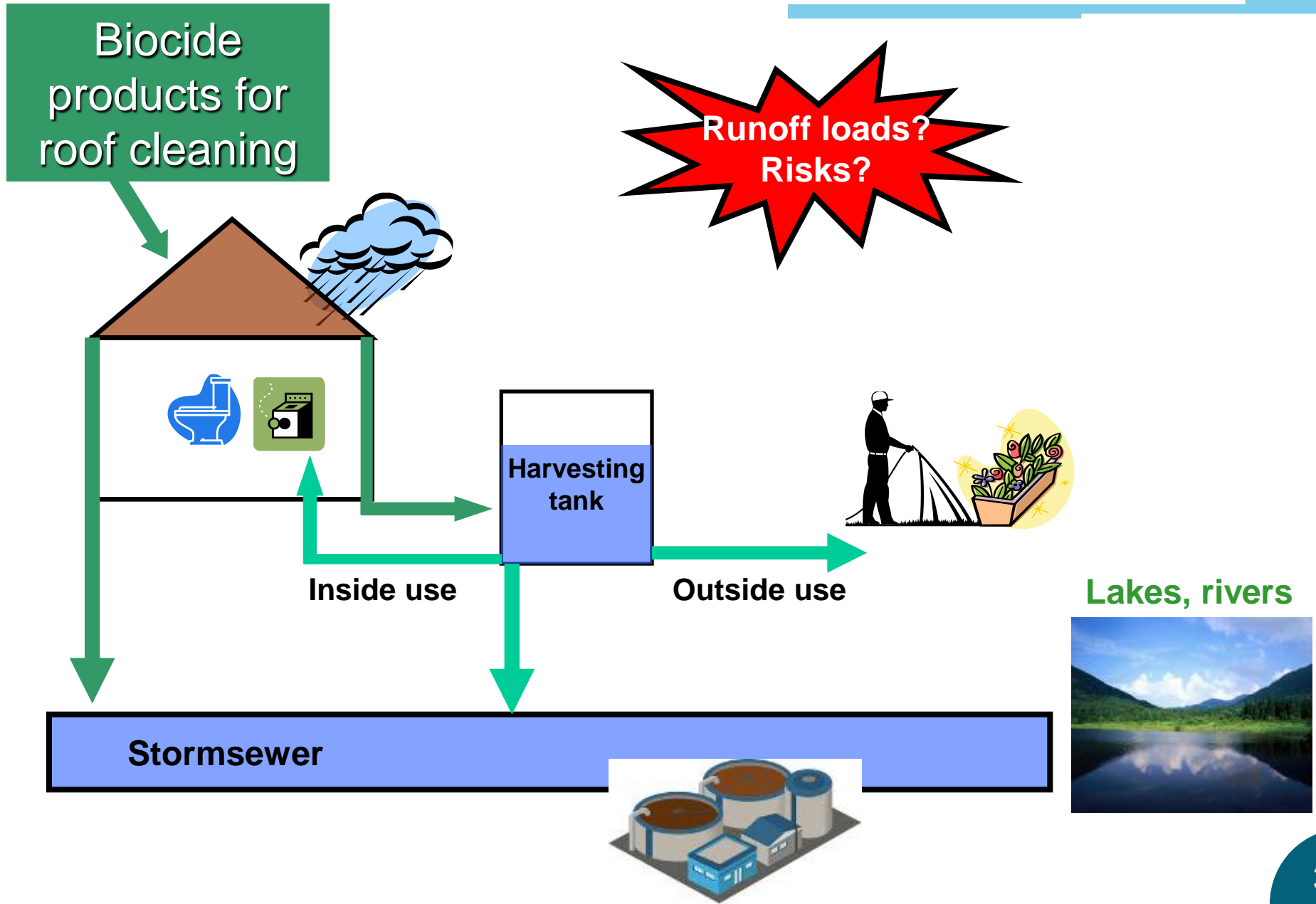
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Context / Objectives

- Sources of stormwater contamination



Context / Objectives



Methodology

Roof treatment typology

Roof treatment practices
(*survey among private / professionals*)

Treatment products, biocides, toxicity data
(*documentary research*)

Runoff monitoring

Biocide runoff under real meteorological conditions (**Semi-field tests**)

Major factors influencing the wash-off
(**Lab studies under simulated rainfall**)

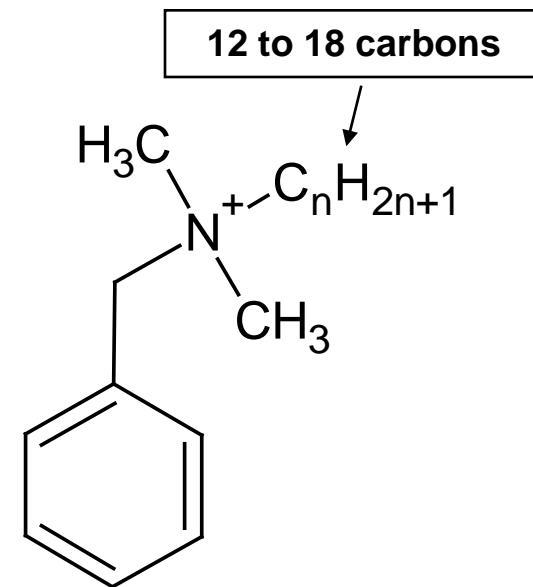
Potential effects

At local scale (**tests**)

Stormwater contamination at catchment scale (**modeling**)

Treatment Typology

- Most frequent treatment: **de-mossing**
 - 1 / 4 private house owners already did it
 - Every 5 years on average
 - Professionals or DIY
- Biocide used: **benzalkonium chloride**
 - Numerous trademarks
- Normal dosage: **4 to 7g/m²**



Alkyldimethylbenzylammonium
or benzalkonium
(CAS: 8001-54-5)



EC₅₀ Fishies = 280µg/L (Sütterlin, 2008)
EC₅₀ Invertebrates = 5.9µg/L (USEPA, 2006)

Semi field testing

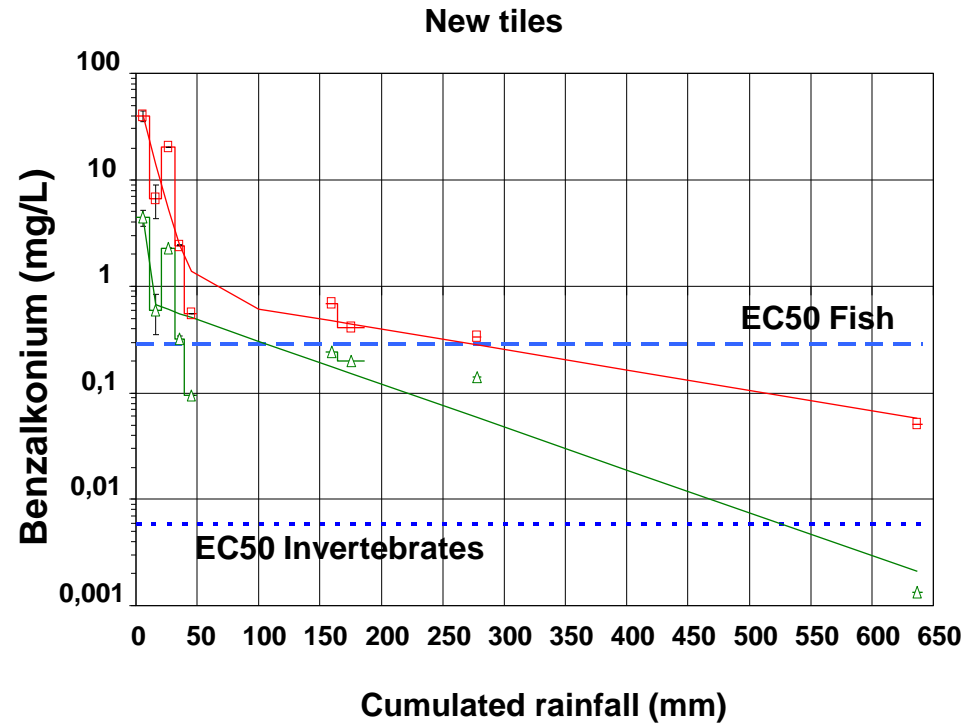
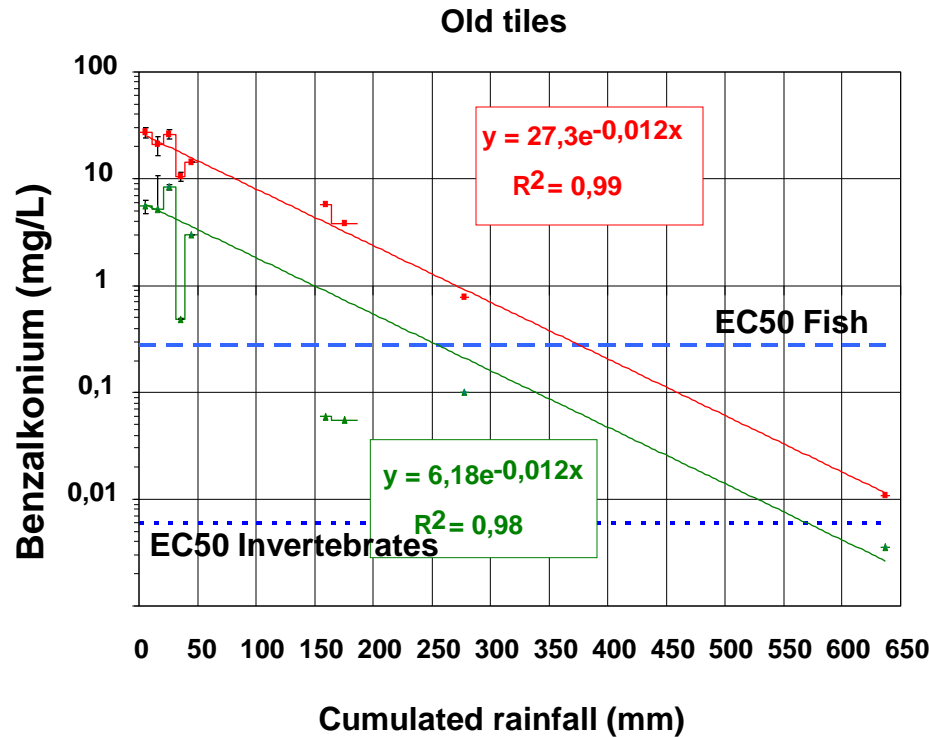
- 12 test roof frames

Materials	De-mossing treatment	Aspect	Exposition	Surface	Slope
Ceramic Tiles (CI) Concrete Tiles (Co)	No (controls)	Old	S	~ 1m ²	40°
	Yes	Old	N, S, E, W		
	Yes	New	N		

- 13 month follow up after biocide treatment
 - First 5 successive rain events (50 mm, 1 month)
 - 4 rain events later on



Semi-field testing



■ Concrete ▲ Ceramic tiles

Emission level depends on the tile material
But not on the exposition

Very high concentrations in the first runoff events
Rapid first order decrease

Persistent emissions even after 13 month

Semi-field testing - mass balance after 13 month

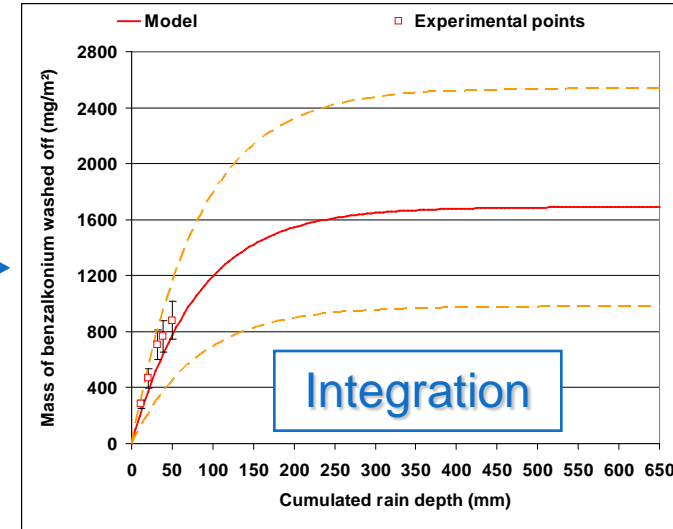
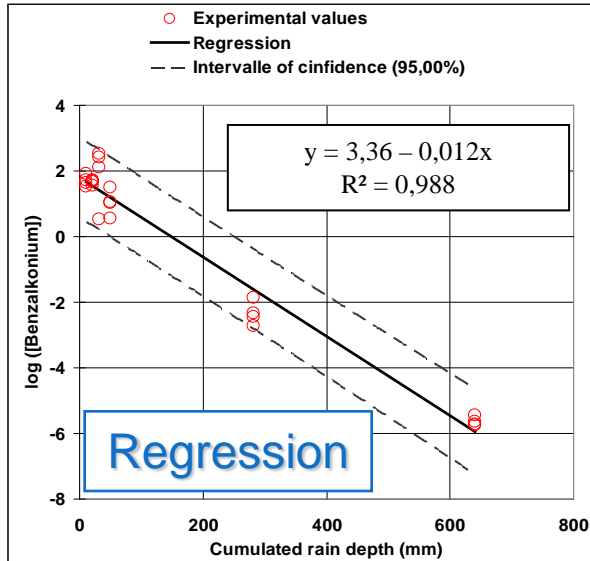
Initial mass spread

Total mass leached

Residual mass in the roof material

— Mass
— Balance

Product dilution and analysis



Extraction and analysis

Semi-field testing - mass balance after 13 month

- Benzalkonium trapped in material negligible compared to mass spread (0.5 - 2%)
- Mass washed off dominant for both concrete and clay
- Low recovery (especially for clay) → could indicate biodegradation processes

	Concrete	Ceramic
Mass spread (mg/m ²)	5900 ± 300	
Residual concentration (mg/m ²)	130 ± 17	30 ± 16
Benzalkonium washed off by the rain Min. - Moy. - Max. (mg/m ²)	980 - 1690 - 2540	65 - 150 - 300
Total benzalkonium recovery	25% < P < 38%	2% < P < 4%

Lab studies under simulated rainfall

- Identification of main factors controlling benzalkonium runoff



Emission patterns depend on the surface treatment of tiles

→ Impervious surface : fast leaching proportional of the mass spread

→ Pervious surface : product penetration and long term emission

Simulated rainfall

Influence of dosage (mass and concentration of biocide product), material, rain intensity on benzalkonium runoff

Material	Mass of biocide spread	Conc. of biocide product	Rain intensity
Concrete	Y	Y	N
Natural ceramic	N	Y/N	-
Siliconated ceramic	Y	Y/N	-

Y : important effect

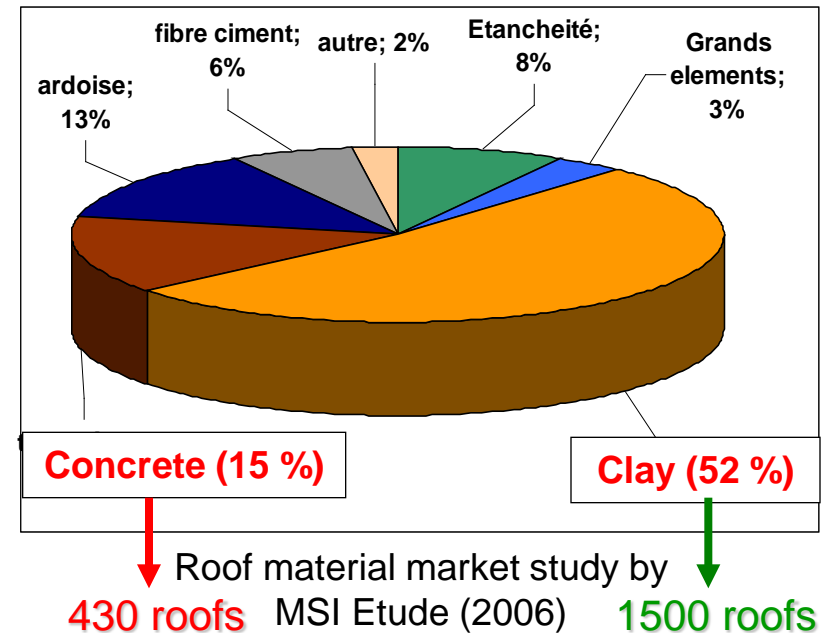
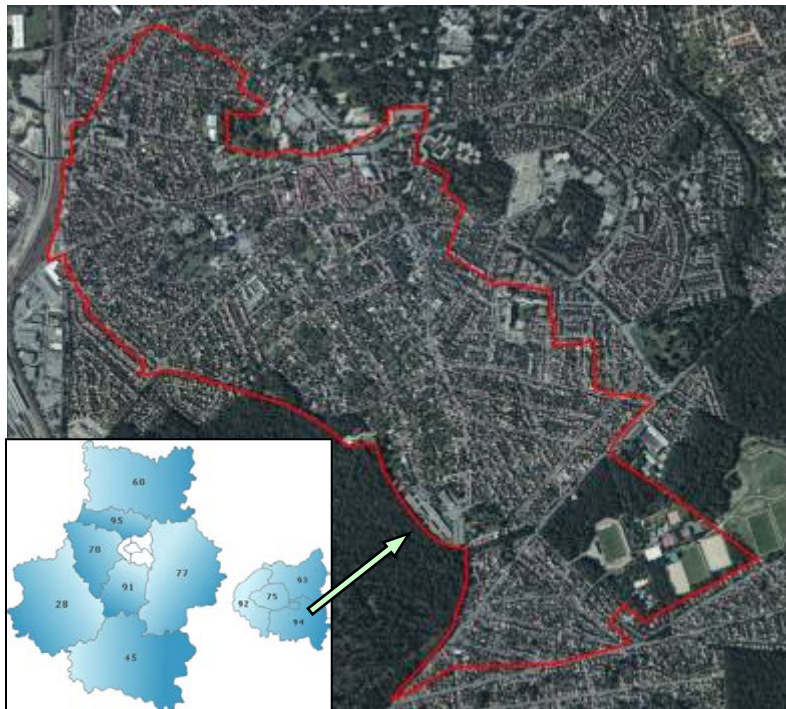
O/N : few effect

N : no effect

Stormwater contamination at catchment scale

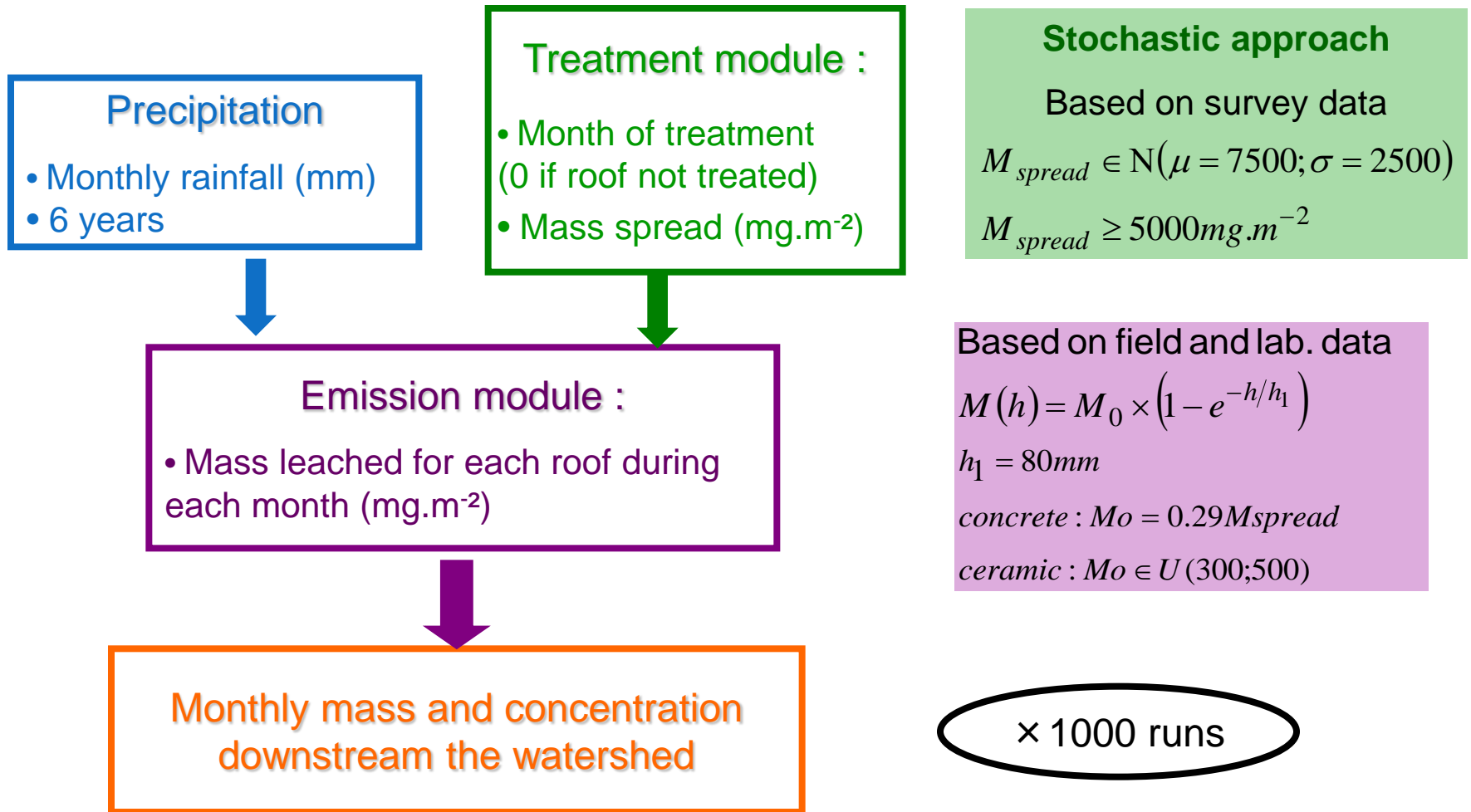
Sucy en Brie catchment

Total area (km ²)	Imperviousness (%)	Road area (km ²)	Roof area (km ²)	Number of treatable roofs
2.1	30	0.325	0.315	2850



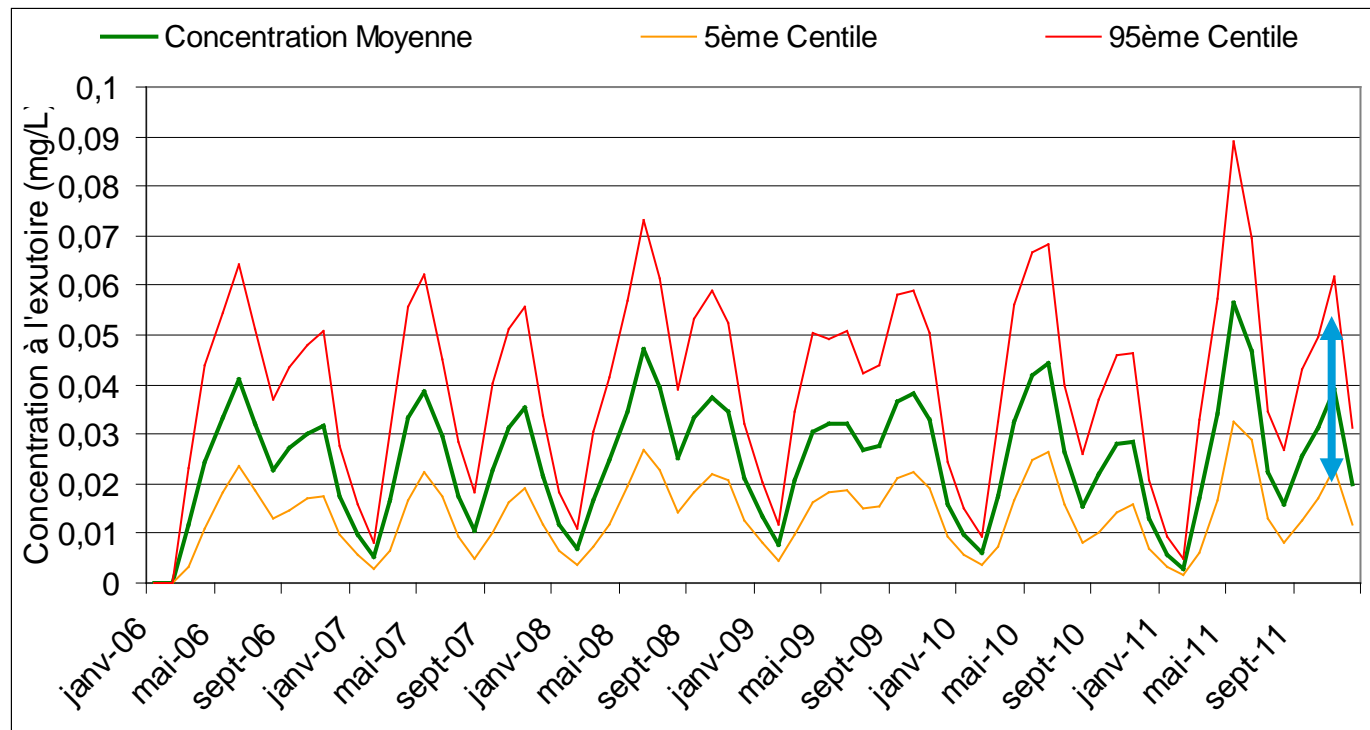
Stormwater contamination at catchment scale

Structure of the model



Stormwater contamination at catchment scale

- Simulated stormwater concentration ranges at the outlet of Sucy catchment



- *Strong annual pattern linked to treatment periods*
- *Rapid decrease in periods without treatment*
- *Strong uncertainty due to variability in treatment practices*

Stormwater contamination at catchment scale

- Comparison to measured stormwater concentrations at the outlet of Sucey catchment

	Model	Measurements		
	Monthly conc. (µg/L)	Total conc. (µg/L)	Dissolved conc. (µg/L)	Particulate conc. (mg/kg)
16/07/2011	13 to 35	28	0.13	50
19/07/2011		7	0.29	73
18/10/2011	17 to 50	28	0.28	84

Modeled values are consistent with measurements

but

Measured benzalkonium almost at 100% in particulate phase

Conclusion

- Roof treatment using **benzalkonium** biocides: a widespread practice
- **Huge runoff contamination** after a treatment (several mg/L)
 - Concentration 1,000 x EC_{50}
 - Fast decrease of contamination but concentration > EC_{50} even after 100s of mm
- Potential impacts at local scale (small ponds, harvesting tanks,...):
 - For harvesting : disconnect during the first months after a treatment
- At catchment scale
 - For separative sewers → Dilution factor 2 to 10 to avoid short term toxicity

A wooden gazebo with a tiled roof is situated on a grassy field. The gazebo has a gabled roof with brown tiles and a wooden frame. It is supported by four concrete blocks. In the background, there is a metal fence and trees. A green banner with white text is overlaid on the image.

Thank you for your attention