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Veterinary pharmaceutical contamination and fecal contamination in mixed-land use watersheds: from agricultural headwater watersheds to water-monitoring watersheds

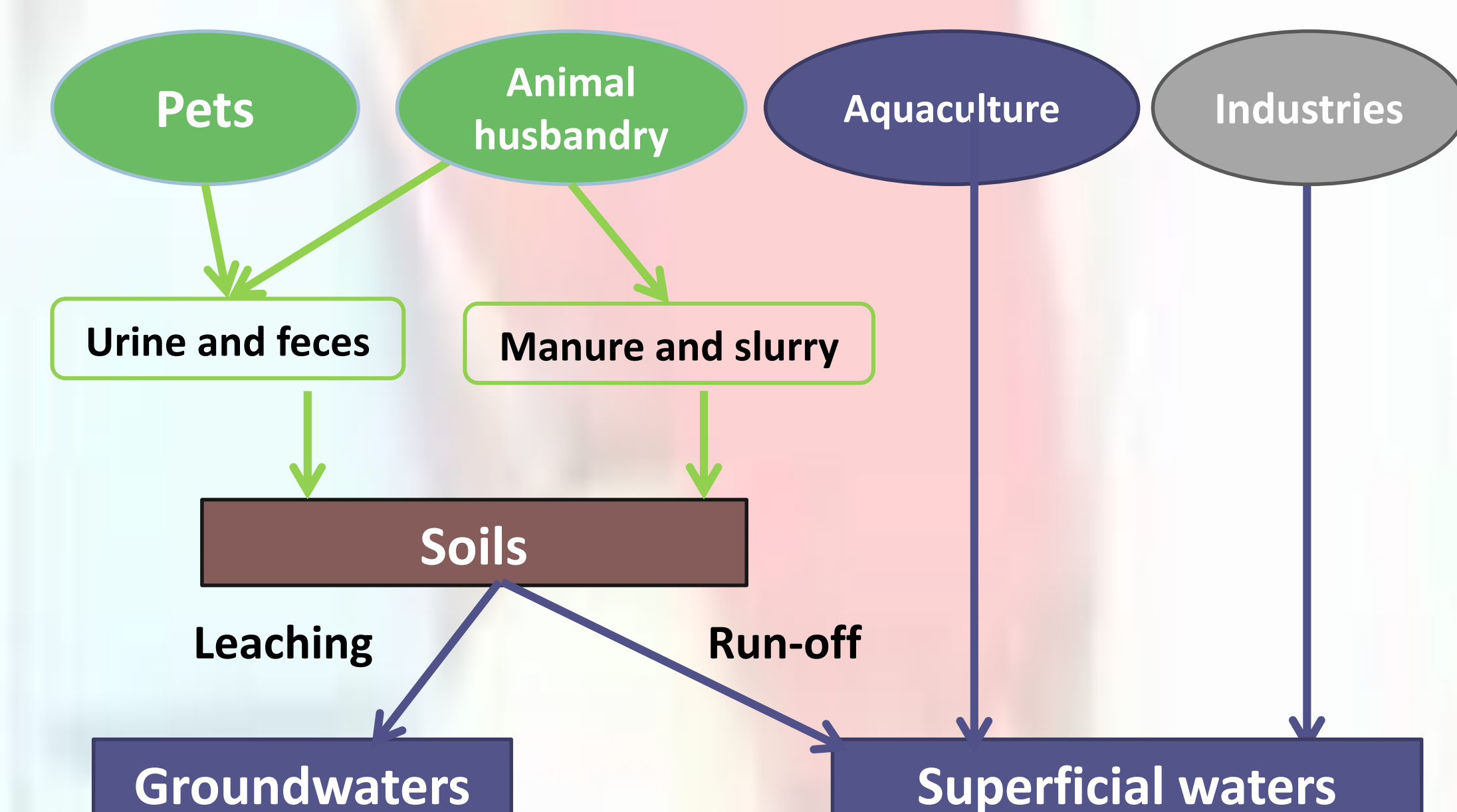


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Background & Objectives

Transfer of veterinary pharmaceuticals from sources to water



- ✓ Intensive livestock : curative and prophylactic use of pharmaceuticals
- ✓ 30 to 90% of (VP) excreted in feces and urine as parents molecules
- ✓ Degradation of the chemical quality of water is controlled by the presence of organic contaminants such as veterinary pharmaceuticals residues (VPR)
 - ⇒ Vulnerability of water resources

GOALS of the study

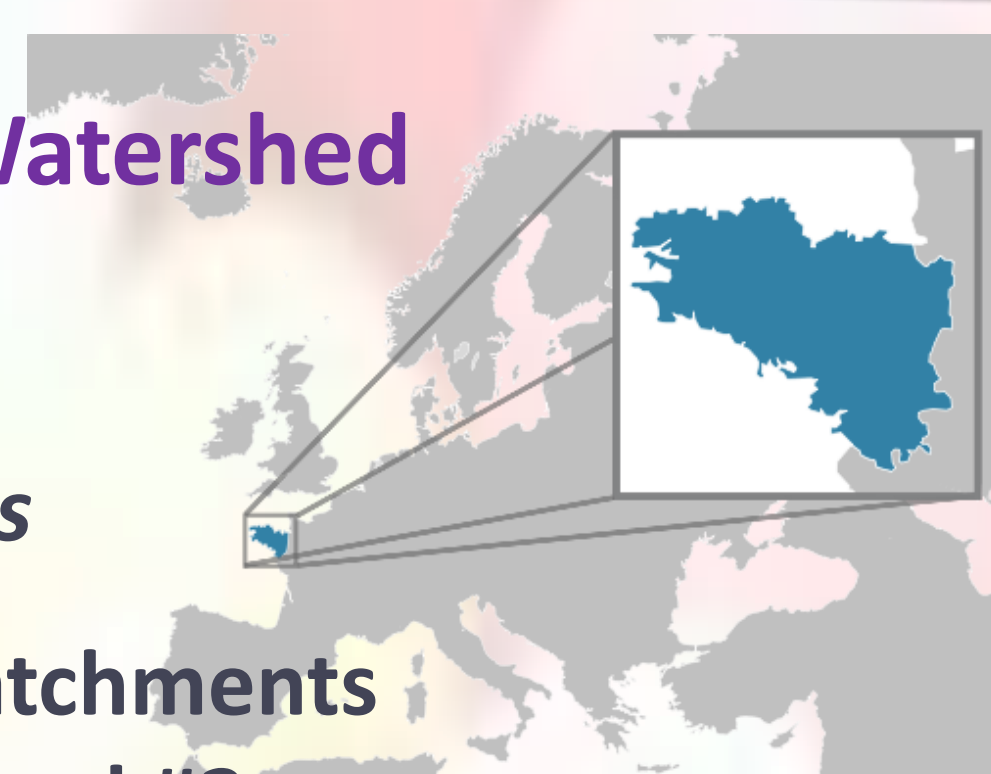
- 1/ Spatiotemporal occurrence of VPR in nested agricultural catchments,
- 2/ Spatiotemporal variability of VPR fluxes upstream and downstream from a WWTP.
- 3/ Fecal contamination and origin of the contamination (human, porcine, bovine).
- 4/ Contribution of livestock vs human fluxes in agricultural watersheds.

Samples & Methods

Study site and sampling

Brittany (Northwest of France) : Haut-Rance Watershed

- ✓ 1st french region of livestock activities
- ✓ Recycling of farming waste on soils
- ✓ 75% of tap water is produced with surface waters



Headwater catchments
Points #1 and #2

River grab sampling and
filtration (0.7 µm)

Water monitoring station
Point #7 (84 km²)



⇒ 6 campaigns from February to reflect spatial and temporal variation for the analysis of pharmaceutical concentration and sources in nested watersheds

Analytical methods

Twenty pharmaceuticals

SPE → RRLC reversed phase → Tandem MS

16 antibiotics :

amoxicillin, ampicillin, benzylpenicillin, ceftiofur, danofloxacin, doxycycline, enrofloxacin, flumequine, lincomycin, marbofloxacin, oxytetracycline, spiramycin, sulfadiazine, sulfamethazine, tetracycline, and trimethoprim,

3 non-steroidal anti-inflammatory drugs:

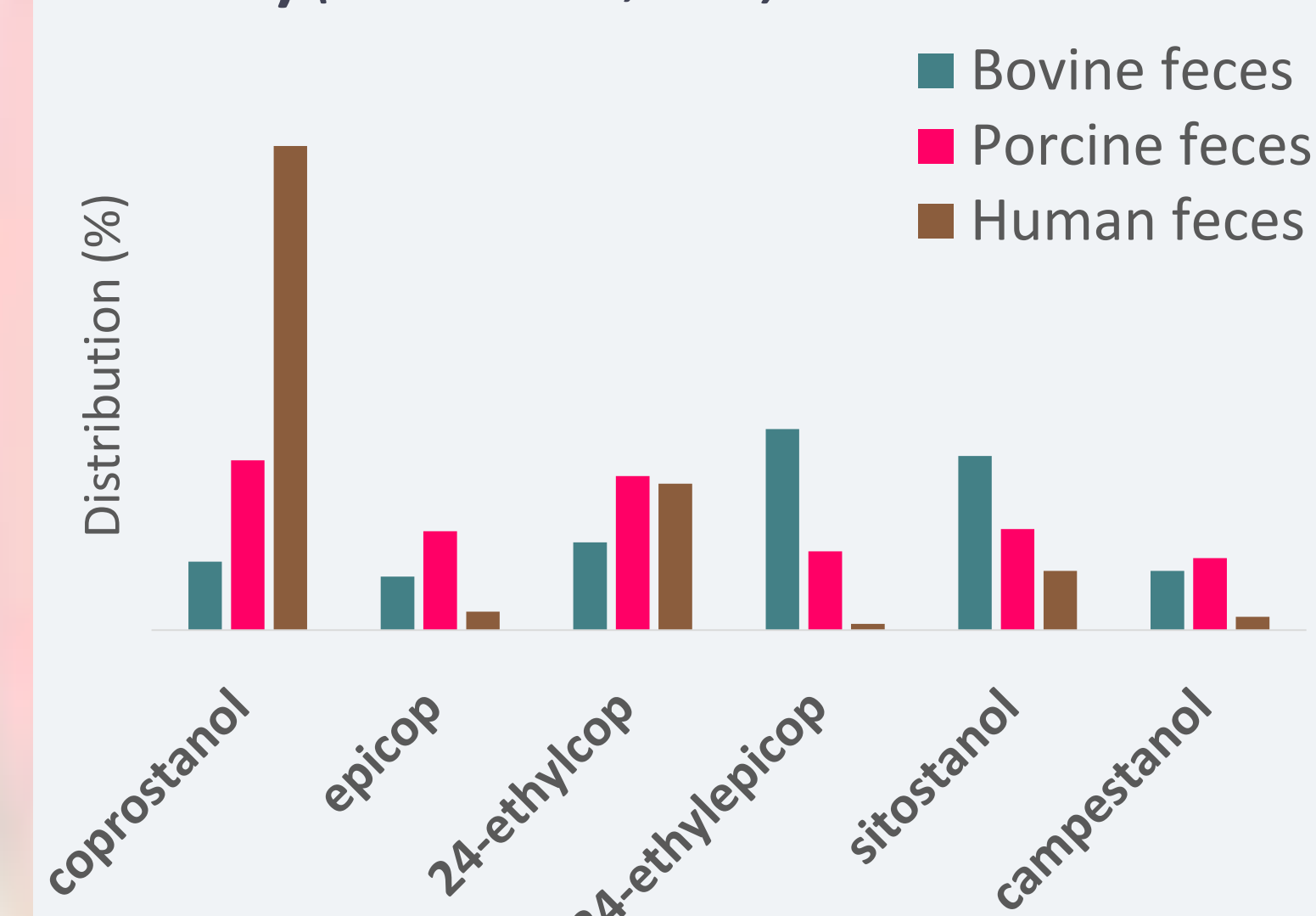
diclofenac, flunixin, ketoprofen,

1 anticonvulsive drug :

carbamazepine.

Faecal indicator and markers:

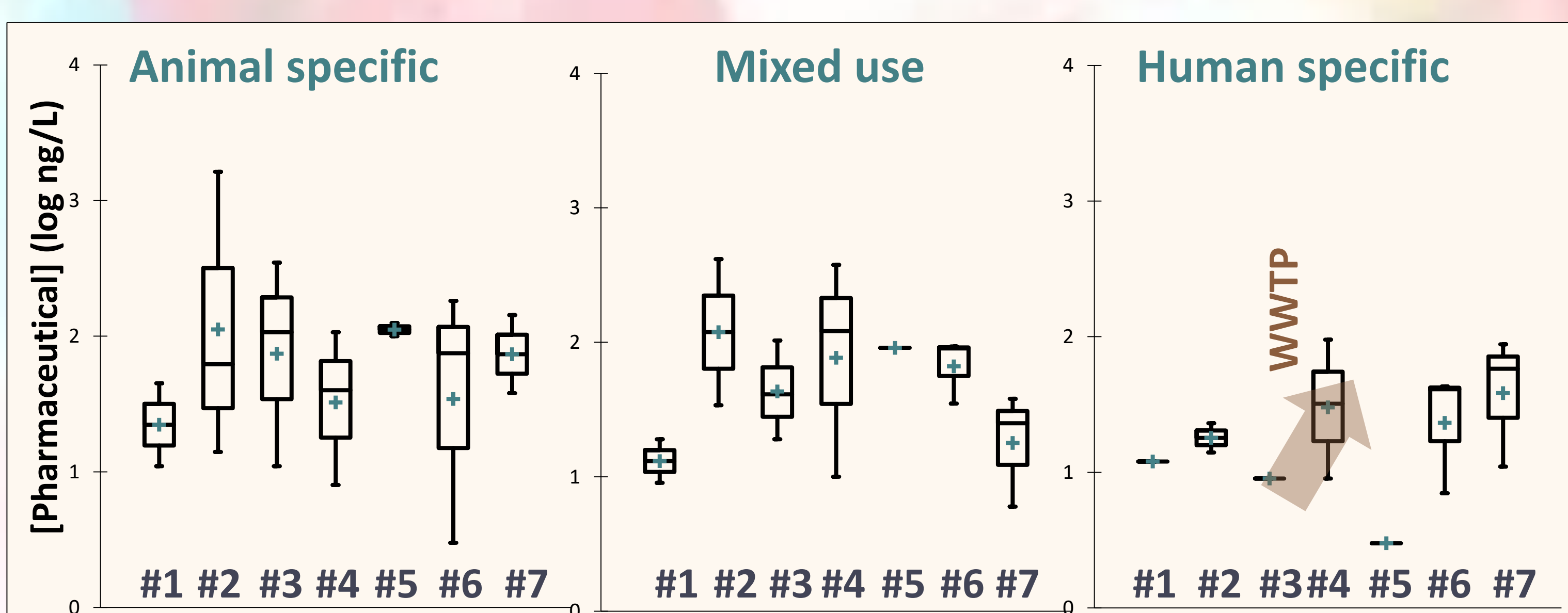
E. Coli and fecal markers to discriminate the origin of the contamination (human, porcine or bovine) (Derrien et al., 2011)



1 animal species = 1 stanol fingerprint
⇒ Stanol fingerprint specific of animal feces

Results & Discussion

Occurrence of pharmaceuticals



⇒ Spatial occurrence of pharmaceutical according to their medical use:

✓ Animal-specific and mixed-use pharmaceuticals were detected at all 7 points, upstream and downstream WWTP.

✓ Human-specific pharmaceuticals concentration significantly increased from point #3 (upstream WWTP) to point #4 (downstream WWTP).

✓ Need to perform risk assessment studies of pharmaceuticals mixtures

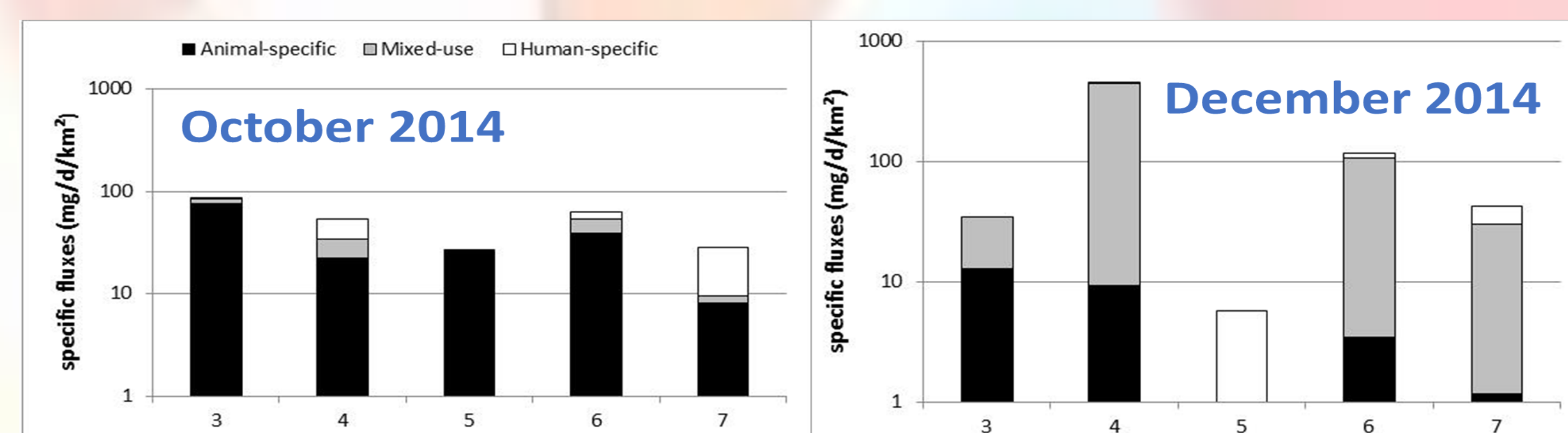
Fecal contamination and origin

⇒ Spatial origin of pharmaceutical according to co-markers of sources:

✓ Fecal stanols as co-tracers of pharmaceuticals (mixed-use) origin.

✓ Predominance of bovine faecal contamination, except after the WWTP at point #4

Livestock and human fluxes in watershed



⇒ Spatio temporal variations of fluxes of pharmaceuticals:

✓ October: Predominance of animal specific pharmaceuticals (in black) from headwater to water monitoring station after rainfall events and when runoff events connected agricultural soils to the river.

✓ December: Increase of mixed-use and human pharmaceuticals contributions with decrease of runoff, suggesting a dilution of agricultural sources.

Conclusion

Veterinary pharmaceutical quantification (more than 100 ng/L) from headwater catchment to water monitoring station

Headwater agricultural catchment are important sources of veterinary pharmaceuticals

Veterinary pharmaceuticals, *E coli* are transferred during rain events

Mitigation of diffuse pollutions : priority on headwater catchments.

