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title: **Development of a mass flow-based spring capture zone delineation tool for drinking water pollution risk management**

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INTRODUCTION

In southern Luxembourg as in many regions where intensive farming takes places, groundwater from a large fracture sandstone aquifer has become increasingly at risk from pollution by pesticides. In order to avoid further deterioration of this essential drinking water resource, the immission situation at the surface has to be reduced or modulated by adapting land use and management practices. This requires the correct attribution of contributing surfaces to a specific spring or group of springs. As it is uneconomical to try calibrating fractured rock transport models for each spring, a mixing-cell like tool (Klaus et al., 2008) was developed that links predicted leached concentrations in the soil to the spring chemical fingerprinting via an inverse optimization procedure based on consistent mass flow equations.

EXPERIMENTAL SETUP AND METHOD

The three main axis of the project were:

- To define the input signal using the physically-based model PEARL. What pesticide leaching concentration C_{mean} should be considered typical for the sandy soils overlying the sandstone? What spatial and temporal variability should be considered?
- To characterize the aquifer's transfer function. Which transport dynamics are worth taking into account? As the mixing-cell approach requires pseudo steady-state, what are the probable residence time distributions in the soil and aquifer compartments?
- To use these results for catchment delineation.

Field work took place over two years focused around a sandstone plateau north of Luxembourg City. To characterize the pesticide sources from the agricultural areas, bromide infiltration tests were performed, TDR arrays installed, and a soil sampling campaign performed to collect soil physico-chemical parameters as well as pesticide residues. Thirty springs draining the plateau and the surrounding area were sampled monthly for water chemistry, pollutants, stable isotopes, and four times over two years for tritium, while a subset of seven springs were sampled weekly. Additionally, an observation borehole was drilled on the plateau downgradient of the agricultural area, and sampled weekly as well.

RESULTS AND DISCUSSION

Input signal

The soil's leaching dynamics and leaching potential was studied with the calibrated PEARL model. Although such process-based approach was limited by the amount of data available, it nonetheless yielded essential information concerning the soil's leaching behaviour.

Results showed that the inertia of the soil system meant leaching would go on at a significant rate for about five years after the last application. This is of importance, as the main pesticide of interest, atrazine, has been banned in Luxembourg in 2005. The leaching rate was dependent upon the crop rotation, with a factor ten between annual and triennial applications (Figure 1).

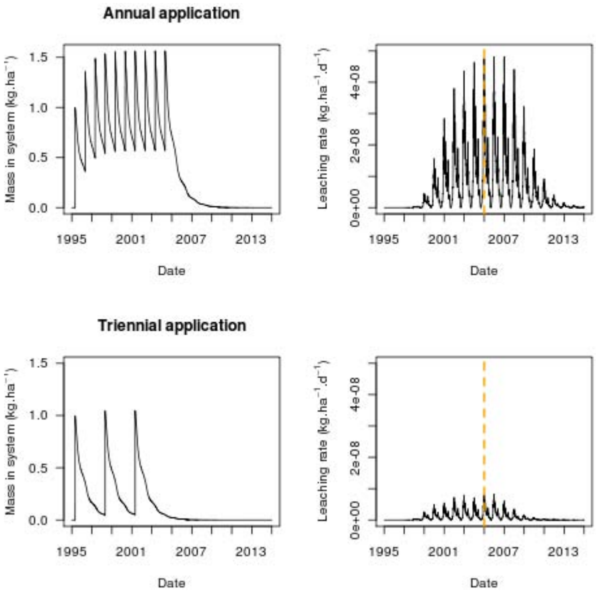

Figure 1. Comparison of the effect of crop rotation on pesticide leaching. The dashed orange lines mark the last field application.

Input parameters describing water transport are relatively straightforward to derive from water balance calculations, using a simple groundwater recharge metamodel for plausibility check. On the contrary, the number of governing processes controlling pesticide leaching (sorption, degradation), the relatively high uncertainty in the values of the process parameters as well as spatial and temporal variability regarding these (to name but a few: organic carbon content for sorption; soil water content, nutrient availability for microbial activity responsible for degradation) makes the prediction range of probable mean pesticide input to the groundwater system immensely larger, all the more so that even the apparently simple question of the initial pesticide application dynamic depends in a non-trivial way on agricultural practices, which cannot necessarily be regarded as unique on the plateau, let alone on individual fields.

Transfer function

Groundwater dating with both tritium and stable isotopes has helped shedding light on the aquifer's hydraulic and contaminant transport behaviour. The springs' stable physico-chemical parameters, as well as a discharge displaying little short term variations are first indicators of a well buffered groundwater system. Stable isotopes measurements have allowed rejecting the hypothesis of a karst-like system for the fractured sandstone in the study area, according to which large fractures rapidly would react to rain event by filling up and drain water quickly to the spring. Hydrograph separation using deuterium clearly showed that even strong precipitation events have little direct influence on spring discharge (quickflow amounting to no more than 10 % of total discharge at all time). Rejecting the karst hypothesis proved an essential step in the study, as this meant that the representative equivalent volume (REV) could be considered small enough to treat the entire formation as a porous-equivalent media, still possibly inhomogeneous and anisotropic, but continuous at the problem scale.

Tritium values in the sampled springs ranged from 6 to 9.5 TU, with most value around 8 TU. A parameter estimation exercise on these data with both a dispersion and an exponential-piston flow transfer function (Maloszewski, Zuber, 1982), describing the transformation of an input signal as it travels through the aquifer, yielded mean groundwater residence times of eleven to seventeen years, with similar values for both transfer functions.

Once a transfer function has been parameterized with environmental isotopes, predictions concerning pollutant transport become possible assuming the pollutant of interest behaves similarly to the isotope used for parameter estimation. As no data concerning degradation and sorption within the aquifer were available, these two processes have been ignored. The aquifer displays a large inertia to pollutant transport, with a flushing period of at least 15 years (without the possible additional retardation effect due to sorption processes). Adding this to the five years' response lag of the soil system means that the atrazine concentration in the springs should not be expected to start decreasing before 2020. On the other hand, this was favourable to the implementation of the optimization algorithm, as steady-state in spring pesticide discharge could be assumed.

Catchment delineation

The core of the catchment delineation tool is a linear optimization algorithm (Loucks et al., 1981), whereby the boundaries of the spring's capture zone are calculated by minimizing an objective function based on a cost-distance matrix and subject to a number of constraints (Fig. 2):

- a water balance constraint.
- a mass balance constraint for pesticides measured at spring level.

Different cost-distance matrices have been tried out, from a simplest non-directional linear increase to more complex anisotropic cases accounting for the main fracture directions. The major hurdle to a satisfactory implementation of the optimization algorithm is the large uncertainty concerning the spatial variability of pesticide leaching, and hence in the mean leaching value used in the optimization algorithm, as the total surface of contributing agricultural land is linearly related to that value. Because the high spring density draining the sandstone plateau allowed subsurface catchments to be relatively well defined by a water balance constraint alone, it was decided to turn the problem around and use these computed catchments to calculate the mean soil leaching on the plateau C_{mean}. This simple approach made use of the natural averaging effect of the aquifer. Spring concentration of a given pollutant is however due to two overlapping control factors:

- Varying proportion of agricultural surfaces in the zone of contribution,
- Varying intensities in pesticide application between fields in the zone of contribution.

While the first factor is precisely the study's focus, mapping the various leaching intensities required additional spatial information. The residues from the soil sampling were combined with crop rotation census data and then split into intensity classes. These classes were then interpolated using kriging, yielding a leaching map. The calibration done, the delineation tool could then be applied in forward mode to the other springs sampled during the project.

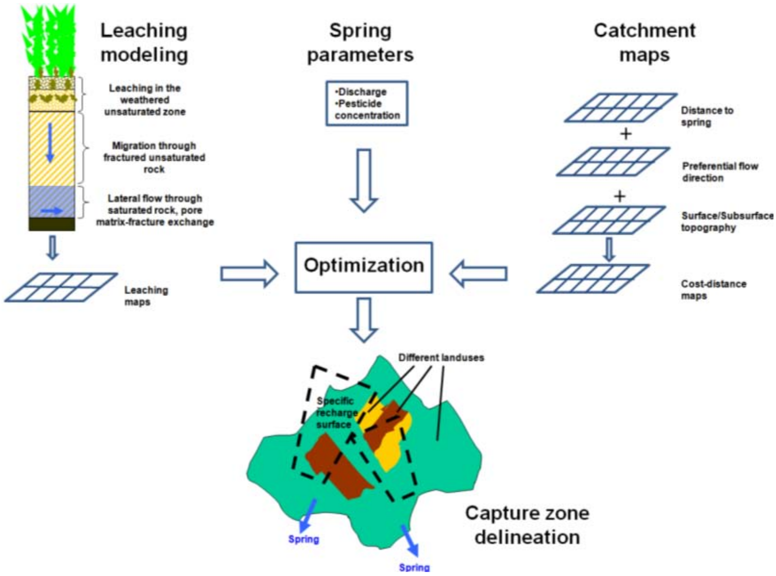

Figure 2. Optimization workflow.

Lastly, in order to study the sharpness of the obtained catchment boundaries, a pseudo spatially distributed approach was also simulated by running multiple optimizations with different leaching maps generated from randomly sampling the soil leaching's frequency distribution, making sure the distribution mean was equal to the mean soil leaching C_{mean} (Petach et al., 1991). The final capture zone resulted from the union of all optimal solutions.

OUTLOOK

As often the case in environmental problems, the stepping stone was to quantify the pollutant sources and their spatial distribution. This could be partially circumvented in the present study thanks to the particular morphology of the catchments, but precludes a wider application of the delineation algorithm for other soils and regions until the uncertainty concerning pesticide leaching prediction has been significantly reduced.

Despite this major drawback, the proposed methodology has a number of interesting features.

- Use of available physico-chemical parameters measured at spring level in a quantitative way.
- Integration on a scientific basis of water balance (catchment's surface area) and mass balance (contributing areas) in a physically consistent distance-based model.
- Substantial leeway to add additional geomorphological and geological information (anisotropy, faulting, folding) of different quality and origin (from surveys, tracer tests, risk mapping) to the cost-distance matrix.
- Computer-based tool to assist the delineation process.

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