



Supplement of

Effect of topographic slope on the export of nitrate in humid catchments: a 3D model study

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Contents of this file

- 1. Text S1
- 2. Figures S1-S3

Additional Supporting Information (Files uploaded separately)

1. Captions for Datasets S1 to S6 $\,$

Introduction

The supporting information includes: Text S1, Figure S1-S3, Captions for Datasets S1-S7

Text S1. Numerical tracer experiments to calculate TTDs

To calculate the transit time distributions (TTDs) for Q and ET for the cross-sectional aquifers, numerical tracer experiments were performed with the numerical transport solution using the finite-element method in HydroGeoSphere. We assumed that inert tracers of uniform concentration existed in precipitation, therefore the fluxes of the tracers were applied to the land surface as a third-type (Cauchy) boundary condition for transport modeling. Tracer can exit the aquifer with outfluxes (Q and ET), neglecting the evapoconcentration effect associated with ET.

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October 5, 2022, 9:50pm

We considered a 200 years period for tracer experiments, which was sufficient to ensure convergence of computed residence/transit times. The 200 years were partitioned into 2400 periods of one-month intervals Δt . A different tracer was used for each of the periods resulting in a total of 2400 distinct tracers. The tracers were assumed to be injected at the beginning of their associated period t_0^i . The advective-dispersive multi-solutes transport was simulated using HydroGeoSphere. Longitudinal and transverse dispersivity values were 8 m and 0.8 m, respectively. The first 199 years of the simulation period were used as a spin-up phase. The last year was used for actual observation and computation of TTDs. Because the climate (boundary) conditions were identical for each year, the transport simulation was actually performed only for the first 12 tracers (covering the course of year), based on which the results for the other 2388 tracers can be reproduced. In this way, the breakthrough curves for the mass fluxes of the tracers in Q, ET, and for the stored mass of the tracers in the aquifer were determined. For a specific time t, the TTD for Q (or ET) was computed by calculating the mass fraction of each tracer in Q (or ET), using:

$$p_{Q/ET}(T,t) = \frac{F^i(t)}{\Delta t \sum F^i(t)} \tag{1}$$

where $F^{i}(t)$ is the mass flux of the tracer *i* in Q/ET at time t, $\sum F^{i}(t)$ is the sum of the mass flux of all tracers in Q/ET. *T* is the transit time that equals $t - t_{0}^{i}$ for tracer *i*. Similarly, the age distribution of storage was computed by calculating the mass fraction of each tracer stored in the aquifer, using:

$$p_S(T,t) = \frac{M^i(t)}{\Delta t \sum M^i(t)} \tag{2}$$

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total mass of all tracer stored in the aquifer. Based on the TTDs, the (mean) age and young water fraction of the Q/ET can be calculated using:

$$T_{Q/ET}(t) = \int_0^\infty p_{Q/ET}(T, t) T dT$$
(3)

$$YF_{Q/ET}(t) = \int_{0}^{90days} p_{Q/ET}(T, t)dT$$
 (4)

where $T_{Q/ET}(t)$ and $YF_{Q/ET}(t)$ are the age and young water fractions of Q (or ET), respectively. Note that both of them are time-variant and their temporal mean, e.g., annual mean values, can be further calculated.



Figure S1. The simulated and measured in-stream nitrate concentration using calibrated best-fit parameter values. The calibration period is from Jul 1999 to Jul 2003.

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October 5, 2022, 9:50pm



Figure S2. The spatial distributions for the loads of SIN and DIN (nitrate) in the catchment of base scenario (original topography), for (a) low peak of SIN, (b) high peak of SIN, (c) high peak of DIN and (d) low peak of DIN. The subplots are corresponding to the markers a, b, c and d in Figure 4c, respectively.

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October 5, 2022, 9:50pm

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Figure S3. The cross-sectional view for the distributions of saturation, flow paths, and exchange fluxes between surface and subsurface in the dry season (August) for the catchments with topographic slope (a) 1:20, and (b) 1:1000, (c) 1:100, and (d) 1:1000. The black lines represent the flow paths. The red curves describe exchange fluxes (along the cross-sectional profiles), positive values indicate seepage to the land surface and negative values indicate infiltration to the subsurface.

October 5, 2022, 9:50pm

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Data Set S1. Precipitation for the year 2005.

Data Set S2. Daily potential ET for the year 2005, estimated by Yang et al. [2018].

Data Set S3. Simulated Q, surface run-off, water ages, young flow fractions for the catchment of topographic slope 1:20 (Data for Figure 3). Data format is ready for Tecplot.

Data Set S4. Simulated C_Q , N loads and N fluxes for the catchment of topographic slope 1:20

(Data for Figure 4). Data format is ready for Tecplot.

Data Set S5. Simulated variables (Q, ET, groundwater table depth, young streamflow fraction,

N loads and fluxes, C_Q) in response to various topographic slopes (Data for Figure 6 and 8). Data format is ready for Tecplot.

Data Set S6. The TTDs for Q in the wet period and dry period, for the catchments with topographic slope of (a) 1:20 (b) 1:1000 (Data for Figure 7c). Data format is ready for Tecplot.