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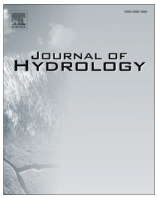

Uncertainty assessment of spatially distributed nitrate reduction potential in groundwater using multiple geological realizations

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SUMMARY

Spatially distributed nitrate reduction potential in groundwater was estimated for the clay till dominated Norsminde fjord catchment in Denmark using the distributed hydrological model MIKE SHE. The nitrate transport was simulated using particle tracking and nitrate was assumed to be instantaneously reduced at the redox interface. Spatially distributed depths of the redox interface were estimated based on the spatial patterns in groundwater recharge and sediment redox capacity. Uncertainty of the estimated nitrate reduction due to geological uncertainty was assessed using multiple geological realizations. The geological realizations were generated using the geostatistical software TProGS and either conditioned based on borehole data only or soft conditioned based on both borehole data and geophysical data. Finally an upscaling of the predicted nitrate reduction was done in order to evaluate the change in uncertainty with increasing scale. The study showed that the uncertainty (one standard deviation) of the estimated nitrate reduction potential (in percentage of nitrate input) on the original 100 m model scale was 25% if only using borehole data and 19% if combining the borehole data with geophysical data. The uncertainty on the model predictions decreased with increasing aggregation scale. The decrease in uncertainty was most apparent the first 500 m, where after the uncertainty started to level off. This scale corresponded well to the mean length of the sand units within the clay till. It is concluded that using geophysical data in combination with borehole data in generation of geological realizations can help decrease uncertainty on the estimated nitrate reduction and that the predictive capability of distributed models is constrained by the spatial resolution of key data such as geology.

1. Introduction

Leaching of excess nitrate from agricultural areas is an environmental problem in many parts of the world. This is also the case in Denmark, where the nitrate load from agriculture to surface waters constitutes one of the major problems in water resources management. In many Danish catchments more than 50% of the nitrate leaching below the root zone is naturally removed by nitrate reduction in the saturated zone before reaching surface waters (e.g. Ernstsen et al., 2006; Hansen et al., 2009; Styczen and Storm, 1993a,b). However, the present regulation on agriculture is applied uniformly for the whole of Denmark without considering this natural removal of nitrate in the subsurface and its spatial variability.

Nitrate (NO_3^-) can be transformed naturally under anaerobic conditions by reduction processes, where nitrate is reduced to nitrogen (N_2) (Appelo and Postma, 2005). In the subsurface nitrate is reduced by inherent reduced compounds in the sediments and nitrate reduction occurs at the redox interface (e.g. Ernstsen, 1996; Fujikawa and Hendry, 1991; Hansen et al., 2008), which defines the transition from oxic to anaerobic/reduced conditions in the subsurface. Around 50% of Denmark is covered by young clay till sediments (Weichselian age) and the redox interface here is normally found close to the surface. However, studies from both Denmark and North America have shown that the depth of the redox interface in tills can vary several meters over short horizontal distances (Ernstsen, 1996, 2013; Fujikawa and Hendry, 1991; Hansen et al., 2008; Keller et al., 1988).

The amount of nitrate reduction in an area depends on both the depth of the redox interface and the groundwater flow patterns as the nitrate needs to be transported below the interface. The shallow redox interface together with a groundwater dominated hydrology results in the high degree of nitrate reduction in the

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saturated zone in Denmark. But local variations in depths of the redox interface and in water flow patterns result in large spatial variations in the nitrate reduction within a catchment. Catchments comprise therefore both nitrate robust areas, where most of the nitrate leaching is transported below the redox interface and thereby reduced, and nitrate sensitive areas, where nitrate is transported directly to surface waters without significant reduction. A spatially differentiated regulation focused on these nitrate sensitive areas will be a more cost-effective approach than the current spatially uniform regulation.

Hydrological models have for many years been used for simulating the transport and fate of nitrate from agricultural areas. The models vary in complexity and process descriptions from lumped conceptual models (e.g. Arheimer and Brandt, 2000; Merz et al., 2009; Whitehead et al., 1998) to physically based models (e.g. Conan et al., 2003; Hansen et al., 2009; Styczen and Storm, 1993a,b; Wriedt and Rode, 2006). However, assessment of spatially distributed nitrate reduction in the saturated zone within a catchment has only been reported in the international literature by Hansen et al. (2009) and Merz et al. (2009). In order to use a hydrological model to delineate nitrate robust and sensitive areas the model must, first of all, be able to simulate nitrate reduction in groundwater and furthermore, have predictive capabilities at small spatial scale. To meet these requirements a distributed hydrological model which considers three-dimensional (3D) groundwater flow and reactive transport is required.

A distributed hydrological model makes predictions at the grid scale defined for the model. However, due to the lack of data it is in practice not possible to describe spatial variations of model parameters and input data at the grid scale. Studies have shown that distributed models in fact do not have predictive capability at grid scale, but only at the scale for which they were calibrated (Hansen et al., 2013, 2008; Hansen et al., 2009). In these studies the main reason for the lack of predictive capability at grid scale is hypothesized by the authors to be inadequate data to describe the geological heterogeneity at a sufficiently detailed scale.

Geological models are most often constructed based on borehole data. However, borehole data are in most areas too sparse to represent the geological heterogeneity at small horizontal scales. The description of the local scale geological heterogeneity can be improved by means of geophysical methods, where especially airborne methods such as transient airborne electromagnetic (AEM) systems are useful for large areas. Because geophysical data are indirect measurements, the conversion of geophysical data to lithology is associated with considerable uncertainty. The geological uncertainty can be evaluated using a number of geological models that are equally plausible, which can be stochastically generated using a geostatistical approach (He et al., 2014; Koch et al., 2013).

When applying a distributed hydrological model it is interesting to evaluate at what spatial scale the model in fact has predictive capability and thereby at what scale the model results should be used. Wood et al. (1988) and Beven (1995) introduced the concept Representative Elementary Area (REA) as the minimum area at which local patterns of parameters and input variables are sufficiently well integrated to produce a similarity in response. At the REA only the statistical properties (mean and variance) are important, while a correct description of the actual spatial distribution does not matter. Refsgaard et al. (2014) suggested a generalized form of the REA concept, the Representative Elementary Scale (RES), where the uncertainty at grid scale is assessed by stochastic modelling of the variable/parameter(s) that are considered the dominant source of uncertainty. The RES is the minimum scale at which a model, at best, has predictive capability corresponding to a given accuracy.

The objectives of this study were to (i) estimate spatially distributed nitrate reduction potential in the saturated zone in a Danish till area to enable delineation of nitrate robust and sensitive areas, (ii) assess the uncertainty on the estimated nitrate reduction potential due to uncertainty on the geology and on the location of the redox interface, (iii) analyze how the uncertainty changes with increasing scale in order to evaluate the relationship between predictive model capability and spatial scale, (iv) evaluate whether the uncertainty can be reduced by using an extensive geophysical dataset in combination with borehole data, and finally (v) evaluate whether the uncertainty on the geology or on the location of the redox interface contributes most to the total uncertainty.

2. Study area

The study area was the 101 km^2 Norsminde fjord catchment located on the east coast of Jutland in Denmark (Fig. 1). The catchment is intensively farmed with more than 70% of the catchment area being agricultural land. Norsminde fjord is classified as having a poor ecological status due to a high nutrient load consisting mainly of nitrogen from agriculture (Danish Nature Agency, 2013). The nitrate leaching (estimated) from the root zone was on average 25 kg/ha/year with a spatial variation within the catchment between 2 and 180 kg/ha/year for the period 2000–2003 (Fig. 1c). The total nitrogen load to the fjord was on average 134 ton N/year in the period 2000–2003.

The catchment is dominated by a moraine landscape from Weichsel with mainly clayey soils (Fig. 1b) that are heavily tile drained. The topography varies from around 100 m to sea level (Fig. 1a). An extramarginal stream valley from Weichsel divides the catchment into a western more elevated and rather hilly part and an eastern part consisting of a flat low-lying plain. The climate is temperate with a mean precipitation and evapotranspiration for the period 1995–2003 of 773 mm/yr and 555 mm/yr respectively. Odder-Rævs stream system contributes to the main part of the discharge from the catchment to the fjord with a mean discharge rate of 232 mm/yr for the period 1995–2003. A large fraction of the stream discharge originates from tile drainage.

The stratigraphy in the area consists of Paleogene and Neogene sediments covered by a sequence of Pleistocene glacial deposits. The Paleogene layers consist of fine-grained low-permeable marl and clay. The Neogene layers above are only found in the western part of the catchment and comprise a clay-dominated Miocene sequence with large sand units interbedded. In the southern part of the area, the Paleogene and Miocene deposits are cut by the buried Boulstrup tunnel valley. The upper glacial sequence is dominated by clayey till sediments with glaciofluvial sandy sediments occurring as small and distributed units within the clay. The glacial sequence is in some areas heavily tectonically deformed (He et al., 2014).

3. Materials and methods

The workflow of the study consisted of several steps. Step 1 to step 4 involved the generation of nitrate reduction potential maps for a set of geological models and is illustrated in Fig. 2. Step 5 involved the uncertainty and scale analysis of the nitrate reduction potential maps. The individual steps are explained in the following.

3.1. Step1: Geological models

Multiple geological models were constructed stochastically to evaluate the effect of geological uncertainty on the nitrate reduction potential in the saturated zone. Two different ensembles of each 10 geological models were constructed to evaluate the advantages of using a comprehensive geophysical dataset in combination with borehole data compared to using only borehole data. The geological models were constructed in several steps in the studies