



Development of a method for in situ measurement of denitrification in aquifers using ^{15}N tracer tests and membrane inlet mass spectrometry

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In NO_3^- -contaminated aquifers containing reduced compounds like organic carbon or sulfides, denitrification is an intense process. Its characterization is of interest because NO_3^- -consumption improves water quality and N_2O production can cause emission of this greenhouse gas to the atmosphere. Spatial distribution of NO_3^- - and N_2 produced by denitrification in groundwater (excess N_2) reflects the NO_3^- -input as well as cumulative denitrification during aquifer passage. Reaction progress (RP) at a given location, i.e. the relative consumption by denitrification of the NO_3^- that had been leached to the aquifers, characterizes the stage of the denitrification process. RP can be derived from the ratio between accumulated gaseous denitrification products and initial NO_3^- -concentrations. The amount and spatial distribution of reduced compounds within denitrifying aquifers is not well known. Recent findings from parallel investigations on in situ denitrification and reactive compounds suggests that single-well ^{15}N tracer tests might be suitable to characterize the stock of reduced compounds in aquifers (Konrad 2007).

The overall objective of our studies is measure the spatial dynamics of denitrification within two sandy aquifers in northern Germany. This includes measurement of the actually occurring denitrification process. Moreover we want to determine the long-term denitrification potential which is governed by the stock of reactive material.

Here we present a new approach for in situ-measurement of denitrification at monitoring wells using a combination of ^{15}N -tracer push-pull experiments with in situ analysis of ^{15}N -labeled N_2 and N_2O using membrane inlet mass spectrometry (MIMS). We will present first results from a laboratory test with aquifer mesocosms using the MIMS method. In this test we supplemented aquifer material of two depths (2 and 7 m below surface) of a drinking water catchment in Northwest Germany with K^{15}NO_3 solution. After tracer application we took water samples at regular intervals with an automated sampling device over 5 days. A small part of the sample was directly conducted in the membrane inlet of our mass spectrometer and the other part was collected in serum bottles which were immediately sealed with rubber septa and stored for later measurement by isotope ratio mass spectrometer (IRMS).

Results available up to now showed for both types of measurement a linear increase of denitrification products ($^{15}(\text{N}_2\text{O}+\text{N}_2)$) over time. At the end of our laboratory test we measured up to 270 and 2400 $\mu\text{g/L}$ $^{15}(\text{N}_2\text{O}+\text{N}_2)$ in the water samples from the supplemented aquifer material of 3 and 7 m depth respectively. Because of the online measurement with MIMS we were able to see during the experiment if and when the production of the labeled denitrification products started. Later-on this approach will be used in the field. Here, the MIMS-technique will be especially advantageous, because the success of tracer test can be immediately seen during in situ sampling.

Results of excess- N_2 measurements at the monitoring wells within the two aquifers showed a range of 0 to 30 mg L^{-1} excess- N_2 and a RP between 0 and 100%.

References:

Konrad, C. (2007): Methoden zur Bestimmung des Umsatzes von Stickstoff, dargestellt für drei Pleistozäne Grundwasserleiter Norddeutschlands, PhD thesis, Dresden Univ. of Techn., Germany, 157 pp.