Abstract

Nitrate is considered the most common non-point pollutant in groundwater. It is often attributed to agricultural management, when excess application of nitrogen fertilizer leaches below the root zone and is eventually transported as nitrate through the unsaturated zone to the water table. A lag time of years to decades between processes occurring in the root zone and their final imprint on groundwater quality prevents proper decision-making on land use and groundwater-resource management. This study implemented the vadose monitoring system (VMS) under a commercial crop-field. Data obtained by the VMS for of 6 years allowed, for the first time known to us, a unique detailed tracking of water percolation and nitrate migration from the surface through the entire vadose zone to the water table at 18.5 m depth. A nitrate concentration time series, which varied with time and depth, revealed—in real time—a major pulse of nitrate mass propagating down through the vadose zone from the root zone toward the water table. Analysis of stable nitrate isotopes indicated that manure is the prevalent source of nitrate in the deep vadose zone and nitrogen transformation processes have little effect on nitrate isotopic signature. The total nitrogen mass calculations emphasized the nitrate mass migration towards the water table. Furthermore, the simulated pore-water velocity through analytical solution of the convection–dispersion equation shows that nitrate migration time from land surface to groundwater is relatively rapid, approximately 5.9 years. Ultimately, agriculture land uses, which are constrained to high nitrogen application rates and coarse soil texture, are prone to induce substantial nitrate leaching.

Summary of Comments on hess-2016-63-manuscript-version3_ed.pdf

Page: 2

Number: 1 Author: Subject: Inserted Text Date: 6/28/16, 10:47:41 AM groundwater recharge behavior and tendency in the deep vadose zone of two agricultural settings, a grapefruit orchard and a crop field (Turkeltaub et al., 2014). Unsaturated flow models were calibrated to the water content observation and were used for groundwater recharge ~~fluxes simulations~~.

The objective of the present study was to demonstrates the water flow and nitrate transport through the deep vadose zone underlie the crop field, with respect to rain patterns as well as the agricultural and fertilization setup. Continuous data on variations in the sediment water content and nitrate concentrations were collected from the entire vadose zone for over 6 years. The nitrate concentration time series, which included variation of nitrate in time and at multiple depths, revealed, in real time, a major pulse of nitrate mass propagating down through the vadose zone toward the water table. These results indicate that nitrate fluxes in the unsaturated zone underlie agriculture land-uses were associated with high nitrogen application rates and coarse texture soils. Furthermore, pollution events originated from agriculture land-uses can be monitored in their early stages, long before pollution accumulates in the aquifer water.

2 Methods

2.1 Study area

A commercial crop field site was selected as a representative prevalent agriculture setting in the southern part of the coastal plain of Israel (34°41'13" E; 31°49'42" N) and is part of an array of VMSs that were installed under different representative land-uses situated above the southern part of the phreatic costal aquifer

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(Dahan et al., 2014, Baram et al., 2013, 2014, Turkeltaub et al., 2014,2015a, 2015b). The study was conducted between 09/2009 and 04/2015. Mediterranean climate prevails in this area, with hot, dry summers (May–September) and rainy winters (October–April), with an average annual rainfall of 512 mm and average temperatures of 31.2 °C (August) and 17.8 °C (January) in the hottest and coldest months, respectively (Israeli Meteorological Service, 2015). Reference evapotranspiration rates calculated according to the Penman–Monteith method (suggested by the Food and Agriculture Organization) range from 1.5 mm day^{-1} (January) to 5.7 mm day^{-1} (July) (Israeli Meteorological Service, 2015).

The crop field cultivation history includes alternation between rainfed agriculture, as wheat and irrigated agriculture as watermelon for seeds and cotton as summer crop (personal communication). From 2005 to 2013, the crop field site was cultivated with rainfed winter crops—spring wheat (*Triticum aestivum* L.) and pea (*Pisum sativum* L.) (Fig. 1). Then for 1 year (2013/2014), the field was uncultivated. The crops were sown at the beginning of the wet season (November) and grew into the spring (April). After harvest, disk plow and roller practices were implemented. Since 2005, the main fertilization application to the field was dairy-farm slurry manure, which was distributed over the 10 ha field for 60 days during May and June (Fig. 1). The total nitrogen concentration in the dairy slurry is 900 mg L^{-1} (Water Authority, 2012). In September 2014, jojoba (*Simmondsia chinensis*) shrubs were planted and irrigation systems were installed.

2.2 Monitoring

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year^{-1} (Herridge et al., 2008), which is about 43% of the nitrogen applied by the dairy slurry. Thus, application of dairy farm slurry combined with a legume crop (pea) seemed to have enriched the top soil with excess nitrogen, as compared to cultivation of cereal-type crops (Fig. 3a).

Progression of the nitrate migration deeper into the vadose zone can be divided into two periods. In the first period, October 2010 to January 2013, at depths of 2.7, 4.2, 9.5 and 15.6 m (Fig. 3b,c,e,g), the increase in nitrate concentration was moderate and continuous; whereas, at depths of 6.3 and 18 m, there was no major change in nitrate concentrations (Fig. 3b-d). In the second period, starting from July 2013 following the rainy winter of 2012/13, substantial nitrate breakthroughs were noticeable throughout most of the vadose zone cross section (marked with arrows in Fig. 3). This rapid nitrate progression to the deeper parts of the vadose zone could be related to the soil's physical characteristics. In the top 3 m, the soil comprised of fine-textured layers (sandy-loam and loamy sand), and from 3 to 18.5 m (water table), the soil consisted of a coarser sand-textured layer (Turkeltaub et al., 2014). Thus, as a consequence of substantial water percolation, which induced intensive water flux across the coarse-textured soil, nitrate transport could be detected at deeper depths of the vadose zone.

Here, as well in previous studies in literature, nitrate fluxes in the unsaturated zone underlie agriculture land-uses were associated with nitrogen application rates and soil physical properties (Green et al., 2008; Botros et al., 2012; Turkeltaub et al., 2015b). Therefore, to attenuate nitrate leaching to aquifers, ~~search~~ should be dedicated to locate the ‘hot spots’ where ~~these conditions prevailed~~ (Liao et al., 2012).

3.2 Nitrate sources

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conditions favor higher rates of transport

The $\delta^{15}N$ values clearly showed that manure is the main source of nitrate in the vadose zone pore water (Fig. 4). Nitrate isotope composition in the vadose zone pore water depends on nitrogen sources and transformation processes (Böhlke, 2002). Examination of the isotopes values suggested that transformation processes such as denitrification and mineralization of soil nitrogen sources have little effect on nitrate isotopic signature. As discussed in the previous section, the relatively rapid nitrate transport downward to deeper parts of the vadose zone is controlled by soil properties and nitrogen application rates. These factors reduce the potential for transformation processes and plant uptake to occur (Liao et al., 2012). Moreover, ~~V~~various studies conducted under similar conditions (soil types and agriculture land use) as in the current study, presented insignificant nitrogen transformation processes and doubt the ability of attenuating nitrate within the deeper vadose zone (Green et al., 2008; Burow et al., 2010; Gautam and Iqbal 2010; Dann et al., 2013; Zhang et al., 2014; Turkeltaub et al., 2015b). Yet, other studies ~~suggested contrast~~ conclusions. Salazar et al. (2012) reported on low nitrate leaching rates in spite of high nitrogen application rates and Lockhart et al. (2013) claimed that depth to groundwater provided a significant control on nitrate concentration in groundwater regardless of soil type or crop type. Thus, a holistic approach ~~comprises~~ all potential factors that control nitrate fluxes to groundwater ~~should be held to identify the dominant ones~~.

3.3 Nitrate storage in the vadose zone

The yearly nitrate mass calculations (Eq. 2) displayed an increase from 2009 to 2010 (Fig. 5), at the same time as NO_3 concentration increased in the upper part of the vadose zone (Fig. 3a). Subsequently, the highest increase in nitrate mass was calculated for 2011 following the combination of cultivation of the pea crop and

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would combine

Number: 7 Author: Subject: Cross-Out Date: 6/28/16, 11:29:59 AM excessive application of dairy slurry (Fig. 5). It seems that the yearly fluctuations in calculated nitrate mass can be explained by the lag time in the transport process between the sampling points. Hence, the peak in nitrate mass observed in the upper parts during 2011 remained in the vadose cross section and eventually reached the deeper parts of the vadose zone ~~as a breakthrough type~~ (Fig. 5).

3.4 Nitrate transport model

Using nitrate time series obtained from deeper part of the vadose zone for model simulations allowed avoiding the highly dynamic nature of the root zone. Furthermore, transport calculations are less effected by mass balance uncertainties as according to previous section, nitrate attenuation processes are insignificant in deep vadose zone.

The results indicated relatively good agreement between observed and simulated nitrate concentration trends (Fig. 6). Nevertheless there were discrepancies in the absolute values and with the simulated nitrate concentrations increasing before the observed concentrations at the 6.3 and 18 m depths (Fig. 6a, d). These gaps could be explained by the assumptions that are intrinsic to the CDE model (Eq. 1) —homogeneous medium and average velocity—along with the assumption of even distribution of the nitrogen source on the surface. Nevertheless, the CDE provided an approximation that could be compared with earlier numerical modeling results (van Genuchten et al., 2012). The calculated hydrodynamic dispersion coefficient was 81 $cm^2\ day^{-1}$ and the pore-water velocity was 0.836 $cm\ day^{-1}$, which is about 305 $cm\ year^{-1}$. Multiplying the velocity by the weighted average water content, 0.060 $cm^3\ cm^{-3}$ (Fig. 2c-h), the Darcian flux equaled 18.3 $cm\ year^{-1}$, which is very similar to earlier average flux estimation of 19.9 $cm\ year^{-1}$ averaged for 24 years (Turkeltaub et al.,

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2014). If neglecting the diffusion term in the hydrodynamic dispersion coefficient, the estimated longitudinal dispersivity (D/*v*) is 97 cm. The calculated dispersivity value is relatively large compared with reported values from earlier solute transport investigations in sandy texture soils (e.g. Toride et al., 2003; Dann et al., 2010). However, it was showed that dispersivity increases with travel distance (Vanderborght and Vereecken, 2007).

The calculated nitrate transport time from land surface to groundwater is approximately 5.9 years. Yet, the increase in nitrate concentration at the 18 m depth occurred in July 2013, which is 8 years after the first slurry application. Olson et al. (2009) reported that there was a threshold amount of slurry application before nitrate accumulated in the soil. Hence, the gap of 2 years between the first application and nitrate arrival to 18 m depth might be related to the period before critical amount of manure was applied to the field.

3.5 Practical implications of vadose-zone monitoring

To prevent a long-term gradual degradation in groundwater quality, the link between sources of pollution on the surface and their migration pattern in the unsaturated zone should be understood long before their final cumulative imprint in the aquifer water. Herein, the application of a VMS under an agricultural field enabled, for the first time known to us, real-time tracking of water flow and nitrate transport from the surface through the ~~entire~~ [1] deep vadose zone. [2] Accordingly similar monitoring concepts for the vadose zone can be used as an alert apparatus for pollution events in their early stages while pollution is still migrating in the unsaturated zone, and long before accumulation in the aquifers water.

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This study demonstrates how nitrate concentrations in the vadose zone exceed the local standard for disqualified drinking-water wells and threaten the groundwater quality. Hence, agro-hydrologically sustainable manure application rates, i.e. sufficient crop production and minimizing nitrate leaching, could be satisfied by suitable regulation or adjustments to meet crop requirements (Olson et al. 2010). To optimize the efficiency of the manure distribution methodology, estimations should include the controlling factors as soil properties, crop type, season, nitrogen attenuation processes and the critical amount of manure application before nitrate accumulation in the soil occurs. Considering only part of the factors could lead to the opposite result. For example, the manure application in this study occurred during the beginning of the dry period, May and June (there are no rain events till October) to prevent nitrogen leaching due to rain events. However, the distributed nitrogen was retained in the soil till winter time and did not undergo significant attenuation processes. The incorrect assumption of manure distribution during the dry period resulted in intensive nitrate leaching. Furthermore, according to the observations presented in this study, the manure application should be reduced following legume crop type. Yet, in many cases, there is a surplus amount of manure to be disposed. Therefore, alternative methods for waste management have to be utilized, ~~coincided~~ with ~~regulating~~ manure application (Westerman and Bicudo, 2005; van Grinsven et al., 2012).

Nitrate transport from land surface to water table through a relatively thick vadose zone occurred within less than a decade. This is a considerably rapid pollutant migration when considering remediation strategies. Moreover, the nitrate observations obtained by the VMS and the isotopic signature analysis indicated that nitrate attenuation processes are insignificant. Hence, agriculture sites constrained to similar

Page: 17

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conditions as in this study, most of the nitrate mass that leaches under the root zone will eventually reach groundwater.

4 Summary and Conclusions

An intensive nitrate leaching beyond root zone was attributed to soil properties and nitrogen application rates. The implementation of a vadose zone monitoring system (VMS) under an agricultural field enabled real-time tracking of water flow and migration of a nitrate plume from the surface through the deep vadose zone to the water table at 18.5 m depth. Isotopic composition of nitrate-nitrogen in the water samples indicated that manure is the main nitrogen source for nitrate in the vadose-zone pore water. Nitrogen transformation processes seem to have only little effect under an intensively fertilized crop field. Total nitrate mass estimations displayed the nitrate mass advancement toward the deep vadose zone. Moreover, according to the simulated pore-water velocity, nitrate arrival to water table occurred within less than a decade.

As in this study, an array of VMSs was installed under other representative agriculture land-uses situated above the southern part of the Israeli costal aquifer. The findings from each site are combined to generate a comprehensive perspective on dominant factors controlling groundwater quality and quantities. Subsequently, these conclusions will be examined with a regional scale aquifer transport model.

Protection of groundwater from potential pollution originating from agricultural land uses has to include effective and continuous monitoring of the vadose zone. Pollution events can be monitored in their early stages, long before pollution accumulates in the aquifer water.

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This seems misleading because it implies that in this study, data from an array of VMSs was used; but my understanding is that the data in this paper are only from ONE VMS. I think you need to replace "are combined" to "will be combined in the future".