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Discussions

Real-time monitoring of nitrate transport in deep vadose zone under a crop field—implications for groundwater protection

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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. In this study, water flow and solute transport through the deep vadose zone underlying an agricultural field were monitored using a vadose-zone monitoring system (VMS). Data obtained by the VMS over a period of 6 years allowed detailed tracking of water percolation and nitrate migration from the surface through the entire deep vadose zone to the water table at 18 m depth. The temporal variations in the vadose zone sediment water content were used to evaluate the link between rain patterns and water fluxes. 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 these isotopes were barely affected by natural soil or industrial nitrogen components. Total nitrate mass estimations and simulated pore-water velocity using the analytical solution of the convection–dispersion equation indicated dominance of nitrate vertical transport, and excluded the possibility of lateral nitrate input. Accordingly, prevention of groundwater pollution from surface sources such as agriculture has to include effective and continuous monitoring of the entire vadose zone.

Keywords: Nitrate transport, Deep percolation, Vadose zone, Groundwater pollution

1 Introduction

Groundwater contamination by nitrate originating from agricultural land use is a global problem. The World Health Organization guideline for maximum level of nitrate in the drinking water is 50 mg/L NO_3. The US Environmental Protection Agency (EPA) regards nitrate as requiring immediate action whenever its concentration exceeds drinking-water standards (US EPA, 1994). A detailed framework was established by the Nitrate Directive of the EC (European Community, 1991) to prevent water pollution by nitrate. Nevertheless, nitrate contamination has disqualified drinking-water wells in Israel (local standard: 70 mg/L NO_3) more than any other contaminant at the beginning of the 21st century (Elhanany, 2009). To prevent excessive leaching of nitrate and its arrival to the groundwater, it is essential to investigate and quantify the mechanism controlling nitrate migration in the unsaturated zone with respect to the specific agrotechnical regime implemented on land surface.

Nitrate fate in the subsurface has been investigated by various approaches, such as: (i) isotopic signature in groundwater systems (Kaplan and Magaritz, 1986; Wassenaar, 1995; Oren et al, 2004; Wassenaar et al., 2006; Showers et al., 2008; Dejwakh et al., 2012; Baram et al., 2013), (ii) crop-management strategies, which combine crop production and nitrate leaching to the subsurface (Leenhardt et al., 1998a, 1998b; Hanson et al., 2006; Doltra and Muñoz, 2010; Beggs et al., 2011), and (iii) studies based on data from the deep vadose zone (Onsoy et al., 2005; Green et al., 2008; Dann et al., 2010; Nolan et al., 2010; Botros et al., 2012; Kurtzman et al., 2013; Dahan et al., 2014; Turkeltaub et al., 2014, 2015a, 2015b).

Nitrogen is an essential nutrient for crop growth and is widely used as a fertilizer. Although the dominant forms of soluble nitrogen fertilizer are reduced (e.g. urea, ammonia) and evolve into other nitrogen species through biochemical processes in the soil, nitrate is the most common chemical contaminant in the deep vadose zone and groundwater (Green et al., 2008; Rupert, 2008).The transfer time of nitrate within the deep vadose zone has been estimated to take from weeks to decades, depending on the water regime, thickness of the unsaturated zone and lithological characteristics of the subsurface (Spalding et al., 2001; Scanlon et al., 2010). Moreover, estimates of cumulative nitrate fluxes in the unsaturated zone have shown significant differences in the timing and magnitude of fluxes derived from different land uses (Green et al., 2008; Dahan et al., 2014; Turkeltaub et al., 2014, 2015a, 2015b). Therefore, the cumulative impact of nitrate leaching from the root zone through the unsaturated zone on nitrate level in the groundwater is blurred by mixing and dilution in the aquifer water. The tendency toward elevated nitrate concentration in aquifer water is thus a relatively slow process (Green et al., 2008).

Although sampling groundwater from wells is easy, the concentration of nitrate might already be at levels that will lead to disqualification of the aquifer water. Knowing the time lag between initiation of a pollution process in the unsaturated zone and its final impact on aquifer quality could give decision-makers more time to plan possible backups for alternative water supply (Baram et al., 2014). Accordingly, estimations of water and solute fluxes which are based on data from the deep unsaturated zone could better indicate their potential long-term impact on the groundwater (Scanlon et al., 2002; Onsoy et al., 2005; Green et al., 2008). Nevertheless, most of these estimates are based on data obtained from excavated soil profiles or pore-water sampling over a short period of time, which represent a snapshot in time of the sediment's chemical state rather than dynamic temporal variations. Furthermore, knowledge of nitrate's fate and transport below the root zone is restricted due to issues such as soil spatial variability and long travel times in the deep vadose zone (Onsoy et al., 2005).

The recent development of a vadose-zone monitoring system (VMS) enables continuous monitoring of the hydrological and chemical properties of percolating water in the deep vadose zone. Data collected by the system comprise direct measurements of the water-percolation fluxes and the chemical evolution of the percolating water across the entire unsaturated domain. To date, the VMS has been successfully implemented in numerous studies on water flow and contaminant transport in the unsaturated zone in a variety of hydrological setups, including: (i) floodwater percolation in arid environments (Dahan et al., 2007, 2008, 2009), (ii) rainwater percolation through thick sand and clay formations (Rimon et al., 2007; Baram et al., 2012; Turkeltaub et al., 2015a), (iii) solute transport in the vadose zone (Rimon et al., 2011; Baram et al., 2013; Dahan et al., 2014; Turkeltaub et al., 2014, 2015b), and (iv) impact of agriculture on groundwater quality (Turkeltaub et al., 2014, 2015b).

A VMS was installed under a commercial crop field to study water flow and nitrate transport through the deep vadose zone with respect to rain pattern 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 (18 m deep) for over 6 years. The temporal variations in the vadose zone sediment water content were used to evaluate the link between rain pattern and water fluxes (Turkeltaub et al., 2014). 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. Stable nitrate isotope analysis identified the source of the nitrate in the subsurface. The vertical transport properties and total nitrate mass in the vadose zone were estimated according to the transient data. Results from the long-term monitoring were compared with earlier modeling efforts applied at the same site (Turkeltaub et al., 2014).

2 Methods

2.1 Study area

The study was conducted under a commercial crop field located in the southern part of the coastal plain of Israel and situated on the outcrops of a phreatic aquifer (34°41'13" E; 31°49'42" N). Mediterranean climate prevails in this area, with hot, dry summers (May–September) and rainy winters (October–April), 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 month, 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 (January) to 5.7 mm/day (July) (Israeli Meteorological Service, 2015).

From 2009 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 fallow. The crops were sown at the beginning of the wet season (November) and grew into the spring (April) with no

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additional irrigation. After harvest, plowing practice was implemented. Main fertilization application to the crop field was dairy-farm slurry manure. In September 2014, due to a change in the agricultural cultivation type, jojoba (*Simmondsia chinensis*) shrubs were planted, irrigation systems were installed, and the distribution of manure ceased.

2.2 Monitoring setup

The crop field site was selected as representative of the prevalent agricultural setting on the aquifer outcrops and was instrumented with a VMS (Fig. 1). Full technical descriptions of the VMS structure, performance and installation procedures can be found in other publications (Rimon et al., 2007, 2011; Dahan et al., 2008, 2009). For brevity, only a general description is given here.

The VMS is composed of a flexible sleeve installed in uncased slanted (35^o) boreholes hosting multiple monitoring units at various depths. Each monitoring unit has a flexible time-domain reflectometry (FTDR) sensor for continuous measurements of sediment water content, and vadose-zone sampling ports (VSPs) for frequent collection of pore-water samples from the unsaturated zone (Table 1). The slanted installation ensures that each monitoring unit faces an undisturbed sediment column that extends from land surface to the probe or sampling port depth. After insertion of the VMS into the borehole, the flexible sleeve is filled with a high-density solidifying material—liquid two-component urethane—that solidifies in the borehole shortly after its application, thereby ensuring proper sleeve expansion for attachment of the monitoring units to the borehole's irregular walls, sealing its entire void and preventing potential cross-contamination by preferential flow along the borehole.

The VMS under the crop field included eight monitoring units distributed vertically and laterally along the entire vadose zone cross section from a depth of 1 m to a depth of 18 m. Note that each monitoring unit is shifted vertically and horizontally from the others along the slanted orientation of the installation (Fig. 1). Since each monitoring unit is located under its own undisturbed sediment column, the integrated data from the VMS should be regarded as representative of a wider zone rather than a single vertical profile. Sediment water content was monitored daily. The pore-water sampling campaigns were conducted every 90 days on average.

2.3 Nitrate-transport simulations

The observed nitrate concentration dynamics at the 6.3 m depth (Table 1) was analyzed and compared with earlier modeling estimations conducted according to observations of water content under the crop field (Turkeltaub et al., 2014). Nitrate transport was modeled in terms of the convection–dispersion equation (CDE) equilibrium assuming resident concentration for a third-type inlet condition as follows (Toride et al., 1999):

$$R\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x} \qquad (1)$$

where c is the solution concentration, x is distance, t is time, D is the dispersion coefficient, v is the average pore water velocity (water flux q divided by the water content θ), and R is the retardation factor.

The nitrate concentrations obtained by the VSP located at the 4.2 m depth (Table 1) served as a series of successive applications of solute pulses (multi-pulse boundary condition). Both VSPs were located in a relatively homogeneous medium of sandy texture (Turkeltaub et al., 2014), following the intrinsic assumption of CDE analytical model homogeneity. The CXTFIT2 code (Toride et al., 1999) and the Levenberg–Marquardt-type optimization approach (Marquardt, 1963), both included in STANMOD (van Genuchten et al., 2012), were used for inversely estimating the pore-water velocity (*v*) and dispersion coefficient (*D*) according to observed concentrations. Both parameters were obtained by running CXTFIT2 multiple times for inverse optimization, each time with different initial values (Turkeltaub et al., 2015a,b).

2.4 Total nitrate mass

The total nitrate mass in the unsaturated zone estimations was calculated to emphasize the nitrate mass that will eventually contaminate the groundwater. The following equation was used for yearly nitrate mass (per area) in the vadose zone:

$$M = \int_{Z=water\_table}^{Z=ground\_surface} \overline{\theta_i} \times C_i \times dz_i \qquad (2)$$

where *M* is nitrate mass in the vadose zone under a unit area, *i* indexes the depth interval for which the corresponding VSP is at its centre, C_i is the nitrate concentration [M/L^3] sampled with the VSP at that depth interval, θi is the average water content measured by the nearest FTDR sensor, and dz_i is the interval length (Fig. 2).

3 Results and discussion

3.1 Nitrate migration in the unsaturated zone

From September 2009 to the time of the study, water contents were monitored at multiple depths by the VMS in the vadose zone of the crop field. The continuous monitoring of the vadose zone indicated temporal variations in measured water contents (Fig. 2). Throughout the monitoring period, most of the rainstorms caused a rise in the water content measured by the shallowest water sensor (0.5 m, Fig. 2). At the 2.1 m and 3.1 m depths, the rise in water contents corresponded mainly to significant rain events (Fig. 2b,c). The sensors at the deeper depths displayed temporal variability with respect to the cumulative annual rain pattern. In some years, a lag between the end of the rainy season and the rise in water content was recorded, whereas in other years, the rise in water content occurred throughout the entire vadose zone following a significant rain event (Fig. 2d–h). A more detailed description of the sequential rise in water content with depth following a wetting event on land surface, and a clear indication of propagation of a wetting wave down through the vadose zone are presented in our earlier study at the site (Turkeltaub et al., 2014), and in other studies at different sites as well (Rimon et al., 2007, 2011; Dahan et al., 2008, 2009; Baram et al., 2012, 2013).

During the monitoring period, slurry from a nearby dairy farm was often spread over the field, serving as a nitrogen source (Fig. 1). This practice was stopped in 2014 when the agricultural setting in the field was changed to a jojoba orchard. The dairy slurry was distributed throughout May and June, 60 days after harvesting, and was scattered across the crop field.

A time series of nitrate concentrations at various depths under the field was obtained by frequent sampling of the vadose zone pore water using the VSPs at multiple depths (Fig. 3). Throughout 6 years of continuous monitoring, different scales and magnitudes of the variations in nitrate concentration were observed. An overview of the nitrate concentration time series with depth (Fig. 3) reveals a major pulse of elevated concentrations, initiating close to the surface, and gradually progressing down the vadose zone toward the water table at a depth of ~18 m. The process was first monitored at the uppermost VSP at 1 m depth, where nitrate concentrations displayed a significant increase during the winter of 2010/2011. Then a gradual trend of reduction in nitrate concentration was observed at this depth until March 2014. A close examination of the nitrate concentrations at the 1 m depth indicated repeating fluctuations, high nitrate concentrations after harvest times due to application of the dairy slurry, and then a reduction in concentrations. Although hard to notice at the illustrated scale, the nitrate concentrations between September 2009 and September 2010 were still relatively high and fluctuated around ~600 mg/L (Fig. 3a). Then they escalated to ~3200 mg/L after cultivation of the pea crop. Following this tremendous increase in nitrate concentration in May 2011, a decline was observed until January 2012, to ~1500 mg/L (Fig. 3a). This phenomenon repeated itself in April 2012, when the nitrate concentration increased again to 2800 mg/L and then decreased to the lower value of 78 mg/L in April 2015 due to cessation of slurry application. Thus, application of dairy farm slurry combined with a legume crop (pea) seemed to have enriched the top soil with excess nitrogen, compared to cultivation of cereal-type crops.

Progression of the nitrate migration across deeper parts of the vadose zone could be divided into two periods (Fig. 3). In the first period, October 2010 to January 2013, at depths of 2.7 m, 4.2 m, 9.5 m and 15.6 m (Fig. 3b,c,e,g), the escalation in nitrate concentration was moderate and continuous, whereas at depths of 6.3 m and 18 m, there was no significant change in nitrate concentrations during this period (Fig. 3d,h). In the second period starting from July 2013, following the wet season of

2012/13, substantial nitrate breakthroughs were noticeable throughout most of the vadose zone cross section (marked with arrows on 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 domain is comprised of fine-textured layers (sandy-loam and loamy sand), and from 3 m down to 18 m (water table), the soil consists 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.

3.2 Nitrate sources

Nitrate isotope composition in the vadose zone pore water depends on nitrogen sources and transformation processes (Böhlk, 2002). Moreover, it can provide an indication of downward transport rates within the vadose zone (Turkeltaub et al., 2015b). Nitrogen isotopic signature analyses were conducted on water samples extracted by the VSPs (Fig. 4). The δ^{15}N values clearly showed that manure is the main source of nitrate in the vadose zone pore water (Fig. 4). Moreover, these values suggested that transformation processes such as nitrification and mineralization of soil nitrogen sources have little effect on nitrate isotopic signature. Nitrate seems to behave like a conservative ion in most parts of the vadose zone and there is relatively rapid nitrate transport downward to deeper parts of the vadose zone, in agreement with previous studies (Onsoy et al., 2005; Green et al., 2008). Ultimately, nitrate concentration time series and water content measurements from multiple depths in the deep vadose zone demonstrated a leaching process and migration of a nitrate plume from the soil surface toward the groundwater.

3.3 Nitrate storage in the vadose zone

Total nitrate mass in the unsaturated zone (as calculated by equation 2) indicates the nitrate's fate in the vadose zone under the crop field (Fig. 5). The mass calculation is based on the concentration time series from eight points across the unsaturated zone. Initially, the yearly nitrate mass calculations displayed a drastic increase in 2010, at the same time as its identification 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 a legume crop type and 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 (Fig. 5) remained in the vadose cross section and reached the deeper parts of the vadose zone as a breakthrough.

3.4 Validity of the transport model

To verify that the observed dynamics of the nitrate concentrations compare with earlier numerical model simulation results (Turkeltaub et al., 2014), the pore-water velocity and the hydrodynamic dispersion were inversely estimated by solving the analytical solution for the CDE (equation 1) (Fig. 6). The observed nitrate concentrations at the 4.2 m depth (Fig. 3c) were used as the nitrate source and input to the model in multi-pulse manner, and the CDE was calibrated according to nitrate concentrations obtained at the 6.3 m depth (Fig. 3d). Close examination of the results indicated relatively good agreement between observed and simulated nitrate concentration trends (Fig. 6). Nevertheless there were discrepancies in the absolute values, with the simulated nitrate concentrations increasing before the observed concentrations. These gaps could be explained by the assumptions that are intrinsic to the CDE model—homogeneous medium and average velocity—along with the assumption of even distribution of the nitrogen source on the surface. However, none of these assumptions could be found in the field. Nevertheless, the CDE provided an approximation that could be compared with earlier numerical modeling results. The calculated hydrodynamic dispersion was 50 cm^2/day and the pore-water velocity was 0.915 cm/day, which is ~333.97 cm/year. Multiplying the velocity by the average water content observed at 3.1 m, ~0.075 cm^3/cm^3 (Fig. 2c), the Darcian flux equaled ~25 ± 9 cm/year, which is an underestimation of the earlier average flux estimation of 19.9 cm/year averaged over 24 years (Turkeltaub et al., 2014).

3.5 Practical implications of vadose-zone monitoring

There is much evidence indicating that agricultural practices have led to alterations in unsaturated sediment pore-water chemicals. Moreover, agricultural land use is one of the major non-point sources for groundwater contamination, in particular contamination by nitrate. 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. Today's wide use of nitrate detection in groundwater by standard traditional monitoring wells might be misleading due to the mixing of waters from uncultivated and cultivated areas, which results in lower nitrate concentrations and masks the pollution process. Hence, protection of groundwater from potential pollution originating from agricultural land uses has to include effective and continuous monitoring of the vadose zone. In this way, pollution events can be monitored in their early stages, long before large-scale nitrate contamination of the groundwater becomes inevitable.

Lastly, the VMS presented here was installed under a crop field which was fertilized by the distribution of dairy slurry, a method that is commonly used worldwide. This method, and its potential impact on groundwater contamination, have been previously investigated (e.g. Basnet et al., 2001; Olson et al., 2009; Salazar et al., 2012). However, the results presented here from continuous monitoring of the vadose zone's hydraulic and chemical properties leave no doubt that this method causes groundwater pollution.

4 Conclusions

- Application of a VMS under an agricultural field enabled real-time tracking of water flow and nitrate transport from the surface through the entire deep vadose zone to the water table at 18 m depth.
- The leaching process and migration of a nitrate plume were demonstrated by nitrate concentration time series and water-content measurements from multiple depths in the deep vadose zone underlying a crop field fertilized by dairy slurry application.
- Isotopic composition of nitrate in the water samples indicated that manure is the main nitrogen source for nitrate in the vadose-zone pore water. Nitrogen

transformation processes such as nitrification and mineralization seem to have only little effect under an intensively fertilized crop field.

- Total nitrate mass estimations and simulated pore-water velocity using the analytical solution of the convection–dispersion equation indicated dominance of vertical nitrate transport.
- 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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Table 1

Depth distribution of the vadose-zone monitoring system (VMS) units.

Vertical depth from land surface (m)	
VSP[1,2]	FTDR[1,3]
1	0.5
2.7	2.1
4	3.1
6.3	5.7
9.5	8.9
12.6	12
15.7	15.1
18	17.4

[1] Depth measured relative to land surface at the site.

[2] Vadose zone pore-water sampling port.

[3] Flexible time-domain reflectometry probe.

Figures

Figure. 1. Crop field site with monitoring location during two periods: crop growth during the wet season (a), and after harvesting and during slurry application (b). (c) Schematic illustration of the vadose-zone monitoring system installed under the crop field. VSP, vadose-zone sampling port; FTDR, flexible time-domain reflectometry sensor.

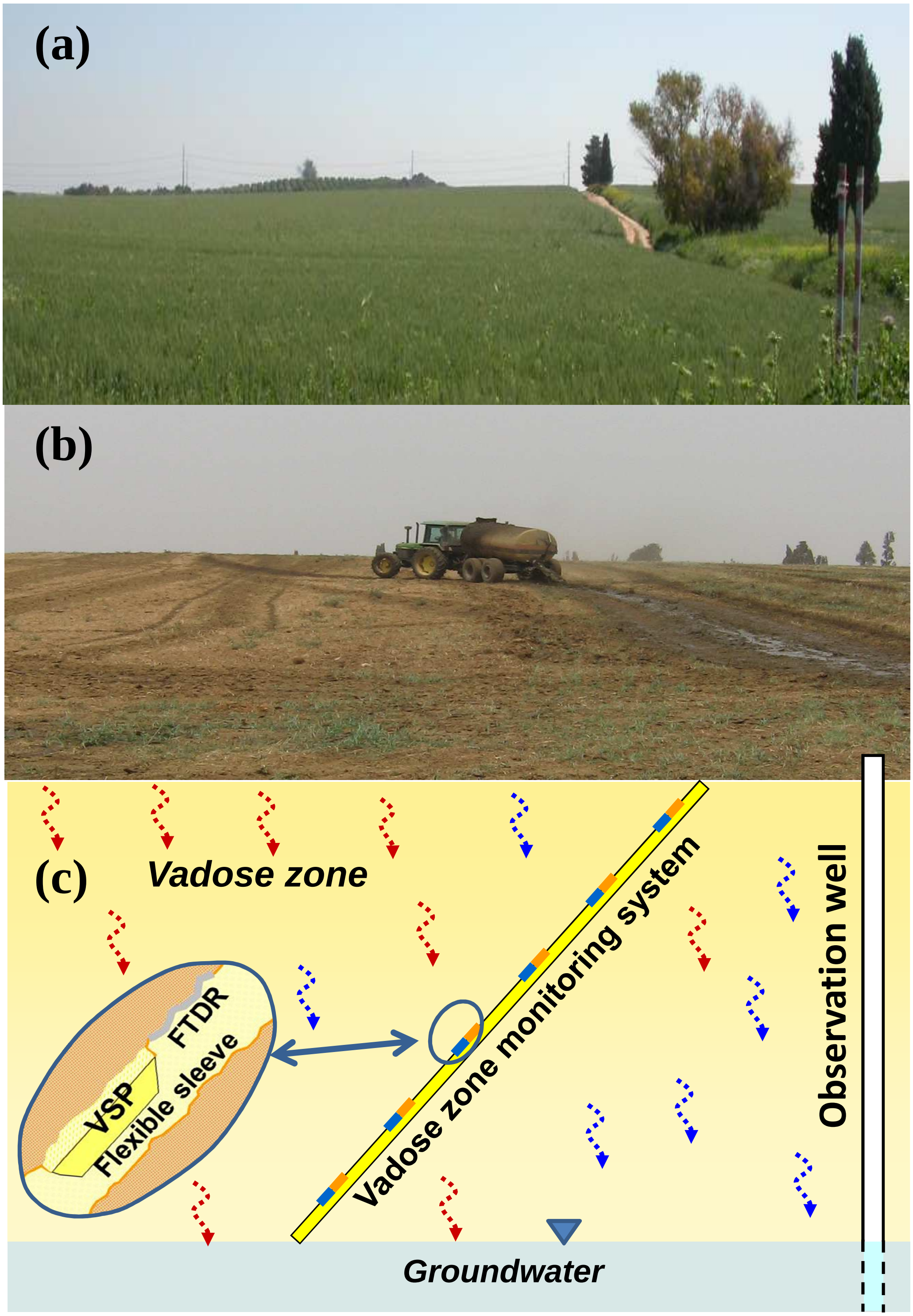
Figure. 2. Daily rainfall and water-content (θ) variations at different depths across the vadose zone as monitored by the flexible time-domain reflectometry sensors.

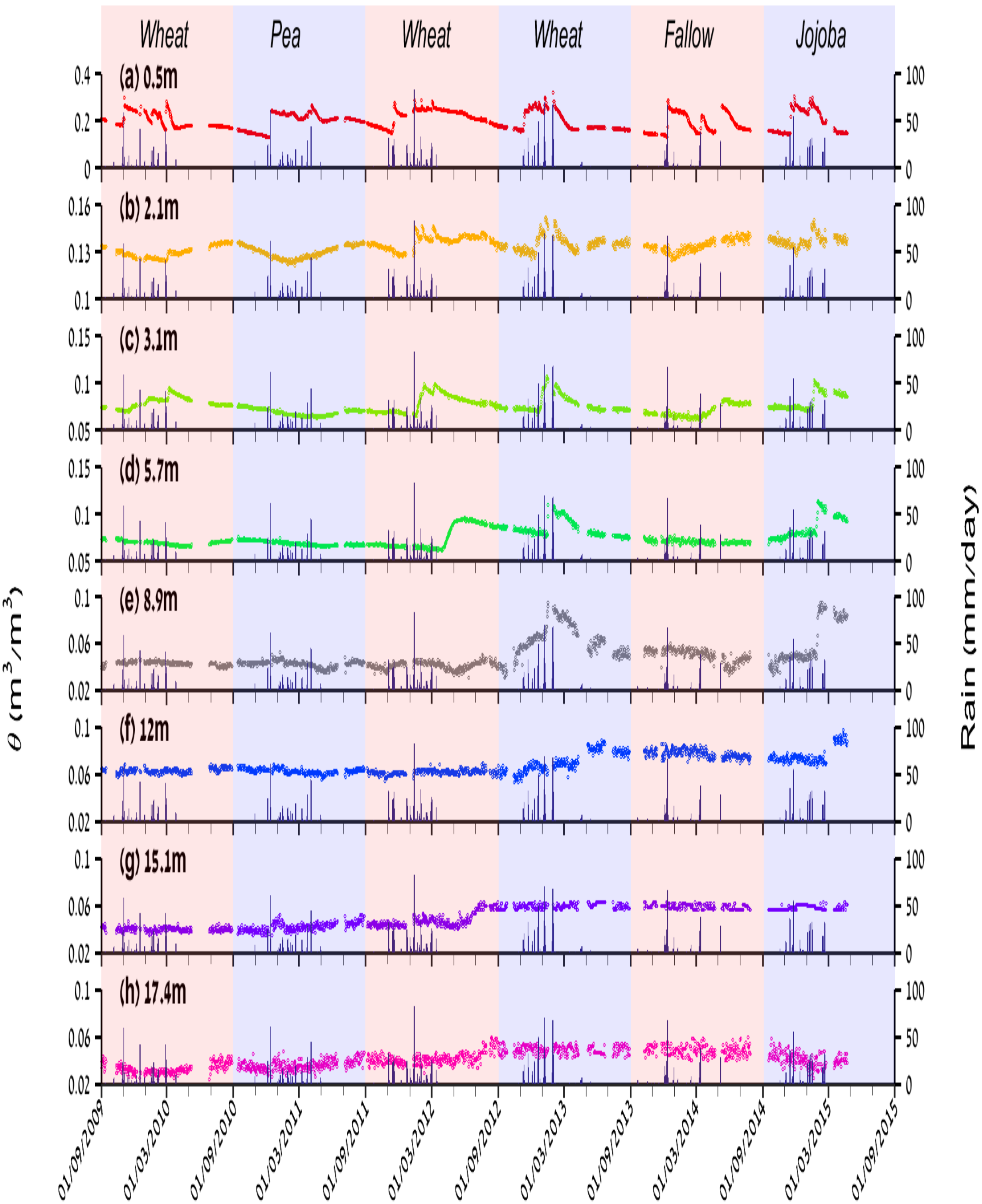
Figure. 3. Daily rainfall and time series of observed nitrate (NO_3) concentrations which were obtained by the vadose-zone sampling ports (VSPs) at multiple depths for 6 consecutive years.

Figure. 4. $\delta^{15}N$ profile of nitrate in the water samples obtained from the vadose zone under the crop field.

Figure. 5. Yearly total nitrate mass of the entire vadose zone over the years of sampling.

Figure. 6. Observed (red circles) and simulated (dashed blue line) nitrate concentrations for the vadose-zone sampling port (VSP) at the 6.3 m depth. The nitrate concentrations obtained by the VSP at the 4.2 m depth served as a multi-pulse input boundary condition.


(a)
(b)
(c)
Vadose zone
FTDR
VSP
Flexible sleeve
Vadose zone monitoring system
Observation well
Groundwater


Wheat
Pea
Wheat
Wheat
Fallow
Jojoba
(a) 0.5m
(b) 2.1m
(c) 3.1m
(d) 5.7m
(e) 8.9m
(f) 12m
(g) 15.1m
(h) 17.4m
θ (m³/m³)
Rain (mm/day)
0.4
0.2
0
0.16
0.13
0.1
0.15
0.1
0.05
0.1
0.06
0.02
100
50
0
01/09/2009
01/03/2010
01/09/2010
01/03/2011
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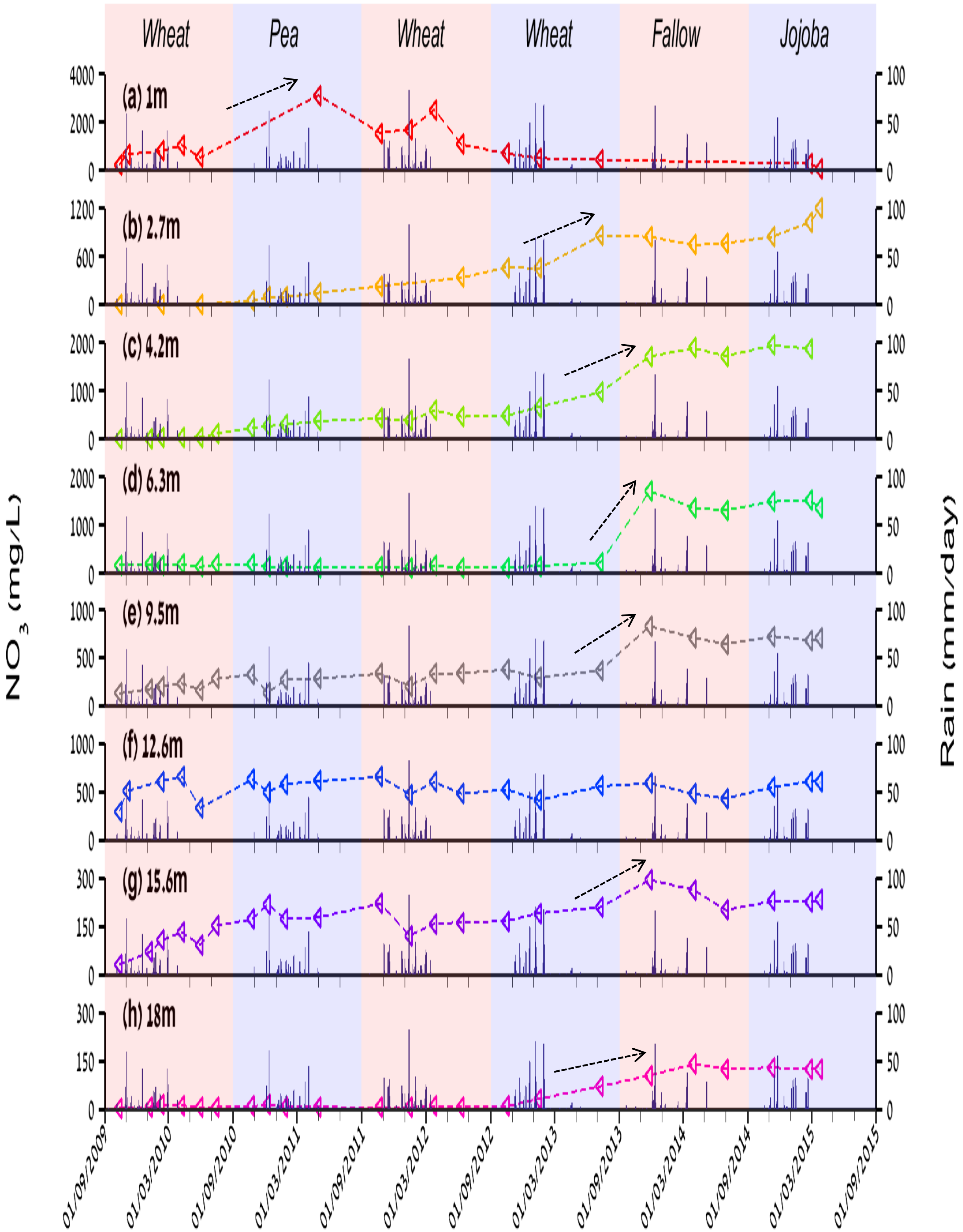

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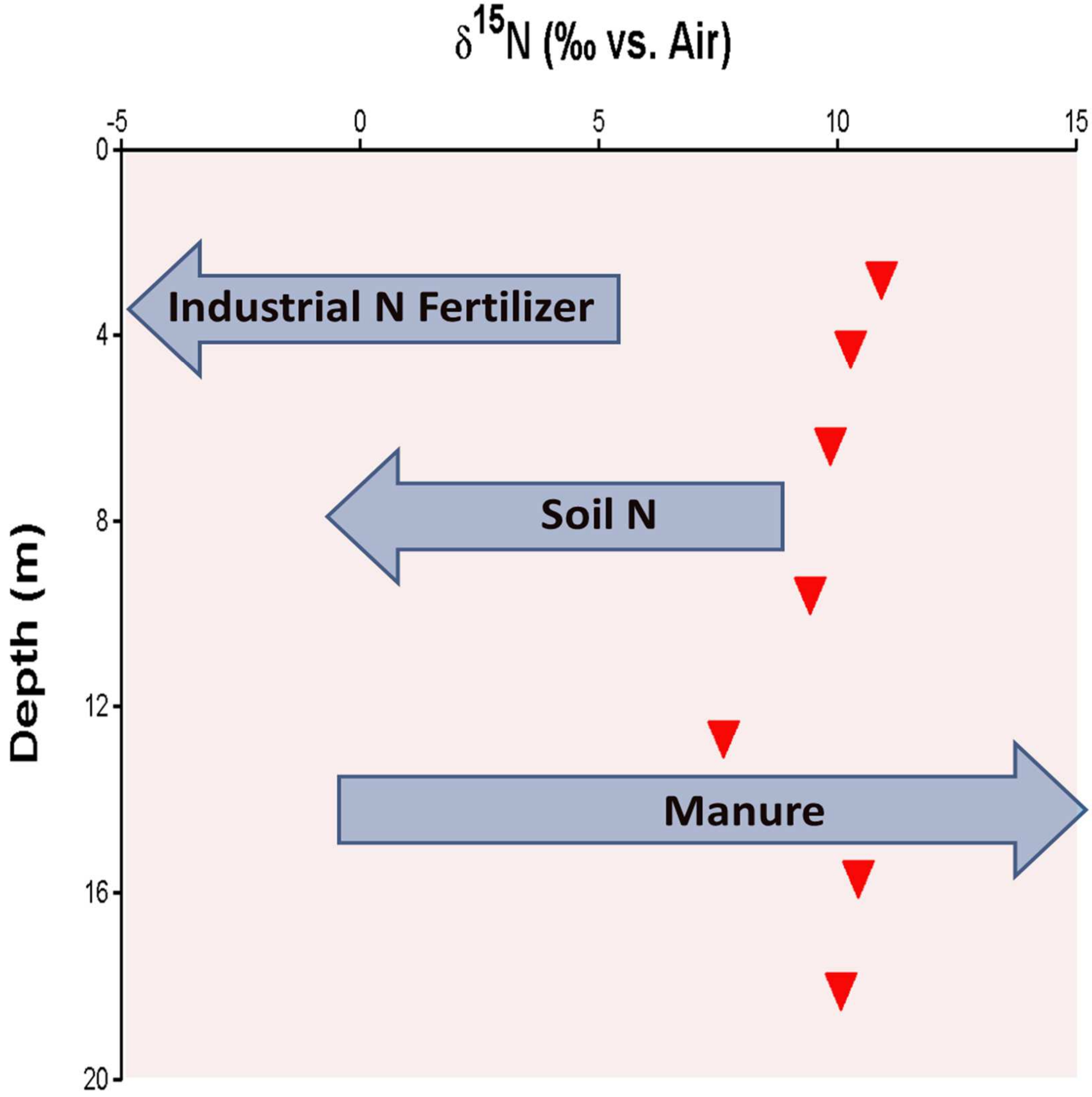

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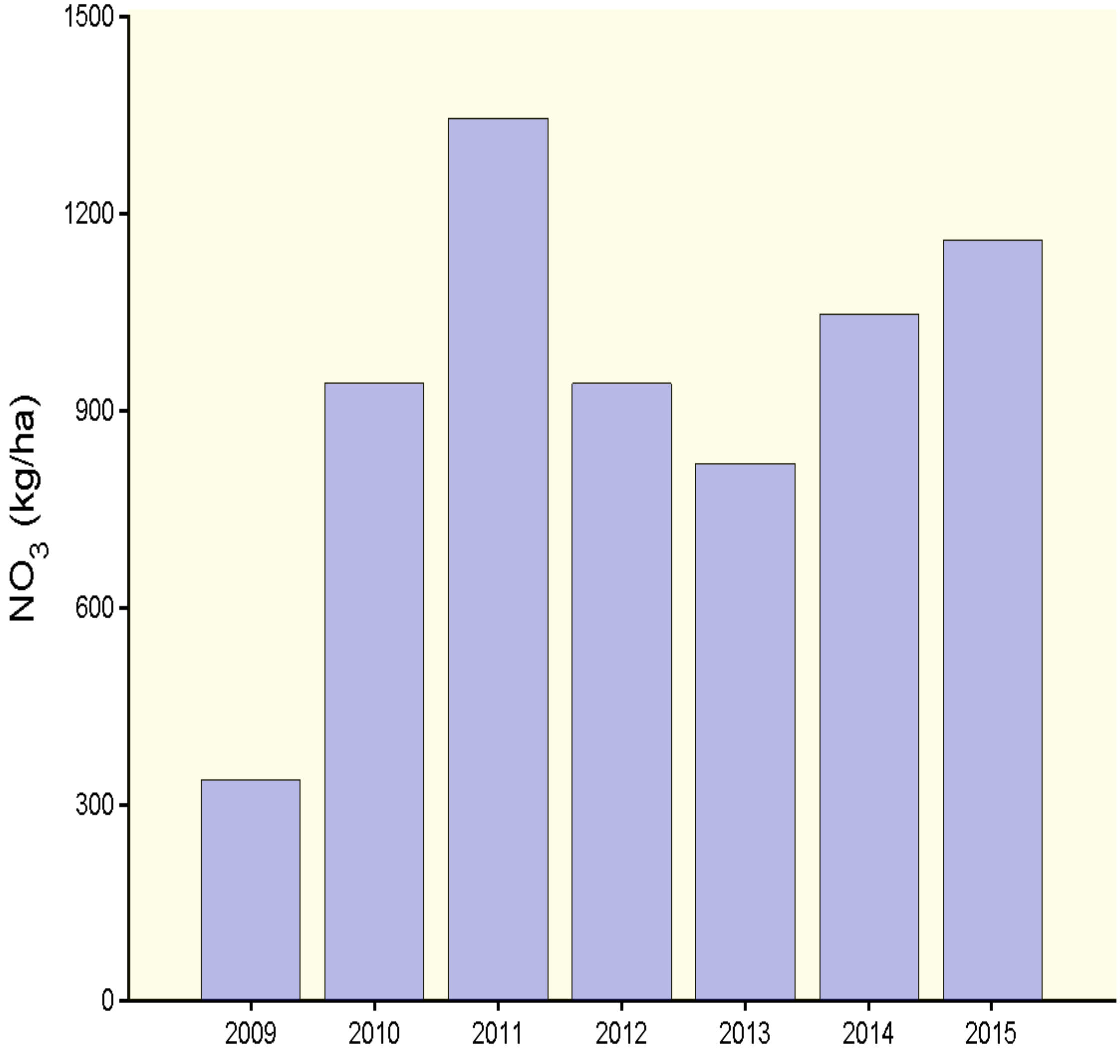

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