Author's response

Dear editor,

Pleases find in the following the responses and revisions made in the manuscript following the referees and editor comments and the last editor report acknowledging the manuscript needs minor revisions.

This file includes:

1. Authors' response to Referee #1 Comments*
2. Authors' response to Referee #2 Comments* (responses to comments # 24 and 41 were changed from the original ones in the discussion forum following the last remarks of the editor).
3. A list of all relevant changes made in the revised manuscript*
4. Marked-up revised manuscript

   *Line numbers in the authors' responses and list refer to the revised manuscript (4).

Authors' response to Referee # 1 Comments

We thank the anonymous referee for his thoughtful review. We are sure the changes that will be made due to these comments will improve the manuscript significantly. The referee's comments and our responses are listed herein one by one.

1) **Title** – consider revising to emphasize the main scientific issue (spatial variability of nitrate?). One option is to replace “success, failure” (which can be misunderstood) with “spatial variability.”

Response 1 - We considered this comment for a long time (as well as before submission). The scientific issue in this work is not only spatial variability but also how can we model the fate of nitrogen from the agricultural fields to nitrate contamination in groundwater wells. Putting spatial variability in the title can raise an expectation that the paper focuses on heterogeneity and finds solutions in the stochastic-hydrology arena. Therefore we prefer not to use this term in the title. Nevertheless, we took the reviewers suggestion to put spatial variability up front at the beginning of the abstract (comment # 2).

The terms success and failure may be misunderstood, yet, on the other hand they raise positive curiosity in the reader. Reading the abstract is enough to understand the title. Furthermore, we think the success in modeling large scale while failing in point estimation and the reasons for that in this case, is a significant part of the scientific conclusions of this study.

2) **Abstract** - Consider stating the scientific problem early in the abstract (e.g. Can spatial variability of nitrate, be characterized on the basis of land use and standard agricultural practices?)

Response 2 – The reviewer's suggestion was accepted the following will be added to the text in line 14 after the first sentence of the abstract:

"Contaminated areas often show large spatial variability of nitrate concentration in wells. In this work we tried to assess whether this spatial variability, can be characterized on the basis of land use and standard agricultural practices." (L 14-16)

3) **61** - Consider also mentioning that nitrate is discharged to streams or other surface water receptors, which can be a major concern.

Response 3 – The paper is about nitrate leaching to aquifers under Mediterranean climate in which permanent surface flow are rare. Nevertheless, we will add a sentence that mentions the ecological problem of accesses nitrogen from agricultural sources in surface water bodies. (L 74-76)

4) **64** - Should this say “significant spatial variability?”

Response 4 - We thank the reviewer very much for this rightful correction. We will change "distribution" to "variability" in the revised manuscript. (L 71,72,73,84)

5) **75-80** - In the statement of objectives, consider making the scientific implications (e.g. explaining the spatial variability of nitrate) more prominent, and perhaps de-emphasize the model-specific and site-specific elements.

Response 5 - As mentioned above the spatial variability is one aspect of the scientific implications in this paper but not the only one. The term spatial distribution will be changed here to spatial variability, which will make it more prominent.

7) - Should “restore” be “estimate”?

Response 6 – We will change the text to “reconstruct the observed groundwater nitrate concentrations”. The term “reconstruct“ is used often to describe a model results that fit observations. (L 83)

8) 100 - Consider defining aerial coefficient of variation mathematically

Response 8: The sentence will be changed to: "The coefficient of variation (standard deviation / Average) of nitrate concentration in the wells in Fig. 2 is 38%." (L 106)

9) 145-150 - Are agricultural-chemical source of Cl important (e.g. KCl)? Are these accounted for in the mass balance?

Response 9 - Usually potassium fertilizers in this area are added according to soil or leaf analysis and most farmers use mixed type K fertilizers when needed (K_3PO_4 , KCl, K_2SO_4 etc.). Therefore, we couldn’t estimate the Cl concentration from fertilizer properly and it was neglected. In the worst case scenario of high need for K and using only KCl, the Cl mass contributed from the fertilizer will not exceed 15% of the total Cl mass input at the soil surface (Citrus orchard data, irrigation water contain 140 mg/l chloride and are the dominant source of Cl). This small contribution and rareness of such worst-case scenario justify neglecting this source of chloride in the chloride mass balance.

10) 227 - Consider spelling out “Israel Water Authority” here.

Response 10 - "IAW" will be changed to “Israel Water Authority”. (L 233)

11) 245 - Consider changing “strictly kept” to “kept constant” or something similar.

Response 11 - No, the recharge fluxes are not constant they are transient. They were calculated by the unsaturated zone flow models, and were not changed during calibration of the groundwater model.

12) 248 - Section 2.3.3. – This is quite brief and readers will have additional questions, e.g. about initial conditions and boundary conditions for NO3- concentrations.

Response 12- We accept the reviewers comment that this section is too brief and details on initial and boundary conditions are missing. The sentences will be changed to: "In the groundwater transport model the initial condition was the measured nitrate concentration at 2012. The transient nitrate-concentration boundary conditions were modified to account for similar reductions in nitrogen fertilization outside of the model domain. This was done by two steps: (1) run the model to the future with constant boundary condition and looking on the trends of the nitrate concentration of the wells inside the model domain; (2) adjusting these trends to the boundary condition and run the model to the future again with transient boundary conditions." (L 263-268)

13) 297 - Table 3 – Spell out “Crop Mass Balance” or define CMB in caption or table footnote.

Response 13 –"Chloride Mass Balance" will be spelled out in the Table caption (L 367)

14) 306 – spell out MAE (mean absolute error).

Response 14 – mean absolute error was defined as MAE in line 243.

15) 307 - It is not clear what is meant by "the improvement in the calibration ceased when…". Is the meaning that calibration efforts were stopped when MAE <0.5 and bias <0.1? And/or that it was difficult to improve results beyond those cutoff values?

Response 15 – The meaning is that the calibration efforts were stopped when the conditions of average MAE <0.5 m and bias <0.1 m, were achieved. It was assessed that in the framework of this research this target is appropriate. The hydraulic head gradient in the research area is relatively linear from east to west, with a magnitude of about 2.5 m/ 1 km, hence, the target of 0.5m is reasonable. We also do not want to elaborate further on the flow system in this paper because the main issue is the nitrogen transport and fate. Due to the reviewer's comment "met" will be changed to "achieved" (L 317)

16) 314 - Should this say "initial transport parameters"? (is this 500m value the one referred to in the previous sentence?)

Response 16 – No, this value of longitudinal dispersivity is the fitted value after the first stage of calibration. To emphasize this point we will change the text to: "The final transport parameters used in the calibrated model ..." (L 323)

17) 316 – Consider revising to "mean nitrate concentration for the entire modeled area"

Response 17 - We thank the reviewer for this good suggestion: "nitrate concentrations over the entire modeled area" will be changed to: "mean nitrate concentration for the entire modeled area". (L 325)

18) 318-319 - "The model reconstructed. . ." This seems repetitive and can be omitted.

Response 18 – We thank the reviewer for this comment. Nevertheless, although there is redundancy in this text we prefer to leave it because the first scentence describes the goodness of fit measures, while the second is the mechanistic interpretation of this result. The words "This means…" will be added to the beginning of the second sentence to emphasize the point. (L 331-332)

19) 326 and onward – It seems that the need for "multipliers" is a key result of the paper, because it indicates that nitrate variability is greater than can be explained by variation of crop-specific agricultural practices and physical processes, to the extent that they are simulated here. I suggest revising to emphasize this scientific significance, and to put less emphasis on the technical role of multipliers as an ad-hoc solution to a modeling problem. In other words, consider revising the language so that readers can see that the two models (with and without multipliers) address the scientific question of whether nitrate variability can be explained by general crop-type practices and the other factors considered in the numerical models.

Also, it would be helpful to further emphasize in the discussion how this result fits into the existing literature. For example, homogeneous NO3- input functions have been used with some success in local-scale (e.g.. single field) studies to explain spatially varying NO3- concentrations (e.g. Liao et al., 2012 http://onlinelibrary.wiley.com/doi/10.1029/2011WR011008/full ; Alikhani et al., 2016 http://www.sciencedirect.com/science/article/pii/S0022169416302098). In regional scale studies, it has been established that a homogeneous input function typically does not suffice, and multipliers similar to those of this study have been implemented (e.g. Green et al., 2016 http://www.sciencedirect.com/science/article/pii/S0022169416302852). This current study can be seen as a logical extension of the previous studies because it tests the extent to which the input function of NO3- can be improved, or even directly estimated from general agricultural practices and vadose zone characteristics. So in combining the current and previous studies, perhaps the authors could comment further on typical scales of variability (e.g. if intra-field variability of fertilizer applications were an issue, would the previous field-scale studies with homogeneous N-inputs have succeeded as well as they did?), the factors that may account for the variability (some already included in discussion), and/or related topics to inspire future research directions.

Response 19 – We thank the reviewer for this important comment. The significant conclusion concerning aquifer-nitrate spatial variability and land use spatial variability is written in lines 324-325 in the results section. We tried to separate results from discussion in this paper, nevertheless the reviewer’s suggestion to further discus the scientific (and practical) meaning here is accepted for improving reading flow. The following text will be added: "The meaning of this, is that nitrate spatial variability cannot be explained only by physical process of agricultural practice and land-use variability on surface. Other factors that are local and arbitrary, significantly affect nitrate concentration in some wells and therefore the measured spatial variability of nitrate in the aquifer. These factors were introduced into the numerical model as will be explained hereafter." (L 334-337)

As of the discussion, we accept the comment and will include more references in the revised version to better fit to existing literature, as suggested (e.g. Alikhani et al., 2016; Kourakos et al., 2012; Spalding and Exner., 1993; Liao et al., 2012; Green et al., 2016 etc.). (L 391-397, L46, L 64-65, and in the references)

20) 335 – Consider adding a sentence to note that the physical significance of the multipliers will be addressed in the discussion section.

Response 20 – A sentence before, the reader is directed to the discussion section (line 333). The term ”physical significance” will be used as suggested (L 344)

21) 349 – Change “on average for” to “as an average of”. Consider clarifying/acknowledging that even though the average is less than 70 mg/L, there would still be some wells exceeding that limit.

Response 21 – "on average for" will be changed to: "as an average of” as suggested. We thank the reviewer for the second remark. A sentence will be added saying: "Even in this case about half of the wells will still exceed the standard concentration.” (L 364, L 365)

22) 362 – Consider changing “coarse” to “approximate” or “first-order” or something similar

Response 22 – “Coarse” will be changed to “first-order” (L 378)

23) 368 – I suggest not using the word “failure”, as it can be misinterpreted as referring to the model itself, rather than to the relative smoothness of NO_3^- spatial gradients in the model as compared to measurements – a result which successfully addresses the scientific objectives of the study.

Response 23 - The strait-forward mechanistic modeling scheme that was used failed to reproduce the spatial variability in wells. Nevertheless, this result and modeling scheme led to the proof that unrealistic nitrate fluxes (much higher than application rates) are needed to support the most contaminated wells.

24) 370 – The specific explanation here (intra-field variability) seems to be given without consideration of additional possibilities that are discussed later in this section (E.g. rapid transport in bore hole annulus)

Response 24 - High fluxes of nitrate can be a consequence of high fluxes of water (hole annulus, leaks of irrigation system etc.) and/or high concentrations of nitrate (fertilizer tank leakage, compost pile forgotten to be distributed etc.). We will add a clarification on possible reasons for high nitrate inputs close to the well: "…and are a result of random failure of even fertilizer distribution in the field that can be due to one or more of the following reasons." (L 386)

25) 376-378 – I don't follow the logic of this text.

Response 25 – We understand the paragraph is somewhat unclear and will be clarified in the revised paper. The first sentence argues that heterogeneity of the porous medium may also cause local very high nitrate fluxes. The second sentence shows how data from this study supports such possibility. The text will be changed to the following: "Heterogeneity of the porous medium may cause extremely high nitrate fluxes likewise well failure discussed previously, and may be a source for local high contamination. The field survey reported here support this statement. Of the nine deep profiles reported here (Figure 6, Table 1), one showed extreme nitrate concentrations and calculated nitrate fluxes that were 4- to 5-fold higher than in the other profiles extracted from the same orchard (Persimmon A, Table 1). " (L 391-397)

26) 378 – I suggest changing "non-physical" to "heuristic" (L 397)

Response 26 - We accept the comment. "non-physical" will be changed to "heuristic"

Authors' response to referee # 2 comments

We thank referee #2 for his constructive review. We are sure that changes made in the manuscript due, will improve it. The referee's comments and our responses are listed herein one by one.

1) The manuscript is interesting and deals with a topic of great relevance around the world and some ideas are promising. However, the methodology seems to me a little deficient so the results and the conclusions are compromised. Some specific comments are listed below, indicating the letter 'L' the line in the original manuscript:

Response 1: Modeling of flow and transport from land surface to well- perforations (20 – 100 m deep, from which 5-50 are unsaturated) at an agricultural area with various crops of 13 KM^2, will always be deficient. Hence, model results and conclusions may be viewed as a compromise. We believe the data-based modeling in this work is a worthy and skill-full effort and the conclusions are significant, novel to some extent, and of high interest for the hydrology research and practice community.

2) L35-45: The sentences in this paragraph are true all of them but all of them are really strong statements and sometimes a little bite unconnected between them.

Response 2: The rational of this paragraph is to shortly describe the origin, a little history, and the magnitude of the global "nitrate problem" in groundwater with its relation to agriculture, hydrology and water quality. Following this comment we will slightly revise the last 3 sentences in the paragraph to increase the connection between the sentences, as follows: "Thus nitrate has become the most common groundwater contamination caused by agricultural activity in many countries (Jalali, 2005; Vitousek et al., 2009; Burow et al., 2010; Kourakos et al., 2012; Yue et al., 2014; Wheeler et al., 2015; Wang et al., 2016). In Israel for example, more than half of all the wells that have been disqualified as sources of drinking water were disqualified due to nitrate contamination (Israel Water Authority; IWA, 2015a). The process of groundwater contamination by nitrate occurs mainly below light soils and less under cultivated clays (Spalding and Exner, 1993; Kurtzman et al., 2016)" (L 41-46)

3) **L46-50:** Even when it is true, different crops and different regions present different efficiencies. I recommend you to try to explain this variability but also conclude with some results for Israel (or other semiarid regions) with some management more similar to your region (trees, vegetables (not in green houses),. . .)

Response 3: We agree that different crops at different regions present different efficiencies, exactly because of that we mentioned a large range of nitrate leaching percentages and we wrote: "...in different crops and countries" and cited many studies from around the world. From Israel we cited a large research that was made over many years (35) for many crop types (18) and a more recent work in modern greenhouses. However, as of the comment we will add "(18 crop varieties)" after "vegetables and fields crop" (L 51))

4) **L51:** urea is mainly considered a synthetic fertilizer.

Response 4: Synthetic or natural, urea ($CO(NH_2)_2$) is an organic compound and as of processes in the soil (and model) it undergoes mineralization, like the other organic nitrogen forms.

5) **L53:** the importance of mass transport process is usually referred to NO3, but NH4 uptake usually occurs by diffusion.

Response 5: Mass transport include both advection and dispersion, in which molecular diffusion is dominant in low velocities. We will add "(advective and diffusive)" after process. (L 55)

6) **L55:** consider changing light soils by aerated, dry,. . .

Response 6: The sentences will be corrected to: "…and in aerated light soil," (L 57)

7) **L55:** define 'relatively thin', because you are considering in your calibration at least 45 cm, and it could be even deeper.

Response 7: The nitrification occurs mainly at the upper part of the unsaturated zone, close to the land surface. Data of nitrification potential in orchards from this area shows that most of the potential is in the top 15 cm and almost negligible below 45 cm.

0-45 cm as the layer of nitrification and reference to Kurtzman et al., 2013 will be added to the revised manuscript. (L 58)

8) **L58:** denitrification to N oxides could be negligible in aerated soils, but complete denitrification to N2 is not so negligible and it is very difficult to measure, so there are a lack of real data.

Response 8: In this sentence we cited studies that neglected denitrification in their models. Denitrification is small but not negligible in the models of this work (Table 2, 3b). Denitrification is a sink for N-NO3 in our models and the further fate of the N species are not part of the scope of this work.

9) **L59:** this sentence is partially true, because nitrate leaching is not only the result of the nitrification, it is also the result of the poorly fixation of the nitrate molecule (negative charge) to the soil complex, mostly dominated by negative charges (clay and organic matter), whereas the ammonium (positive charge) presents a stronger retention to the soil and leaching is more difficult.

Response 9: We thank the reviewer for the good comment. The following sentence will be added to the revised manuscript: "Moreover, ammonium is a cation and tends to adsorb to the soil solids (clay fraction, organic matter)" (L 58-59).

10) **L83-86:** These two sentences fit better in the introduction.

Response 10: We wrote these sentences in the methods (research area section) to explain the choice of research site. This makes the text flow better.

11) **L86:** if it is unconfined, do you know how much water and nitrate leave the system? It is important in order to predict if the new entries are greater or smaller.

Response 11: We mentioned the aquifer is unconfined because a major part of this work deals with water and nitrate fluxes entering the aquifer from the unsaturated zone above. Water and nitrate enter/exit the modeled aquifer domain from the side boundaries and exits also through pumping wells.

The nitrate budget in the aquifer is always calculated, when concentration increase entries exceed exits and when concentrations decreases exits are higher than entries (e.g. scenario of reduced N fertilization).

12) **L100:** Do you know if all the wells are extracting at the same depth? In some aquifers has been reported nitrate stratification, suggesting contamination from different time periods.

Response 12: We show in Fig. 5(c) the depth of well-screens in the groundwater modeled area. Data does not show a trend of nitrate with screen depth. We discuss the possibility of denitrification in deep parts of the aquifer earlier in the text in lines (60-64) including a citation.

13) **L118-120:** how is the irrigation applied? Is not the same if is homogeneous (surface, furrow, sprinkle) or if it is drip irrigation.

Response 13: We will refer to the irrigation technique in the revised manuscript and text will be changed to: "The potato field was irrigated by sprinklers with an average…" "The strawberry field was irrigated by micro-sprinklers (at the early stage of growing) and drip irrigation after, to an average…" "The persimmon orchard was irrigated by micro-sprinklers to an average…" (L 123-127)

14) **L119:** and what happen with the citrus?

Response 14: As mentioned a few lines before, the data and unsaturated flow and transport model for citrus were taken from Kurtzman et al. (2013).

15) **L120:** please, define the size of the plastic tunnels, because there are many kinds. Moreover, the rainfall over the plastic should go somewhere, perhaps is draining with a reduced amount of nitrate, so this could lead to a reduction of the nitrate concentration in the aquifer. Please, discuss or consider it.

Response 15: We do not wish to overload the manuscript with more agro-technical data, which does not contribute to the main theme (nitrogen fate from ground surface to wells). After the tunnels are set irrigation is by drippers within the tunnel, and rain drains on top of the continuous plastic sheets out of the field, hence, it has no effect on the transport of nitrogen below the field.

16) **L125:** was it the same fertilizer rate when the petrol was cheaper some years ago? N fertilizers use to be highly related to energy price.

Response 16: No. As mentioned before, our knowledge about the agricultural practice is based on the farmers reports concerning the 10-20 years prior to soil sampling (where needed, we used the extension-service recommendations). There was no mentions of changes in the rate of N fertilization due to its price by the farmers.

17) **L128:** I do not understand why each crop should be correlated to each soil type. To me it makes more sense to have a soil map, combined with a weather map (you are presenting different precipitations) and with a nitrate concentration in the irrigation water map, and all them combined with the cop map, resulting in multiple combinations. Perhaps some of the variation that you cannot explain is due to your simplification.

Response 17: The soil-map which represent the soil type at surface will show the same soil for the entire modeled area as the first order classification – Red Mediterranean relatively sandy - "Hamra" soil (sandy loam - loamy sand up to sandy texture in some places). Therefore, in the root zone the sediment is quiet homogenous, yet heterogeneity in deep profiles is much larger. The 2 main limitations from having a calibrated model for the unsaturated zone for each plot are: 1) deep sampling cost and availability; 2) time and skills needed for model calibration. Therefore, for upscaling from only one field that was sampled (per land-use) to the large region, we decided to simplify by using one calibrated unsaturated zone model (which was extended to dozens of models for each unsaturated-zone thickness) for each crop type. This methodology is described in lines 157-158 and 186-202, and the simplifications are discussed in lines 361-378.

18) **L128:** Have you try to make first a comparison of the observed concentration at each well with the percentage of each crop in the well proximity? Because if it is not related, the rest of the assumptions could be not true.

Response 18: We didn't make this comparison because we aim at a much higher target than showing a 2D relationship between nitrate concentration in wells and land use which maybe misleading (unsaturated-zone, perforation depths and pumping rates are variable, there is a natural gradient, etc.,). We aim at describing the 3D flow and transport phenomenon from field surface to well perforations mechanistically, accounting for the above issues (see lines 75-80).

19) **L129-138:** Please define better the process. If I understood well you obtained for each crop three cores per depth; but, how do you divide them in order to get samples dried at 105◦C and at 40◦C at the same time. Moreover, do you think that if you dry the sample during 3 days at 40◦C the soil nitrate and ammonium is going to be the same than analysed in fresh samples conserved in the refrigerator few hours/days? And mostly in deeper layers, because as you say mineralization, nitrification, denitrification and all the N processes could be enhanced by this temperature increase, doesn't they? Please discuss or define better.

Response 19: As written in lines 129-130, for each crop 3 continuous cores from land surface to depth of 10 m depth were cored with the direct push technique. The cores were then cut into 30 cm segments and the segments were analyzed in the laboratory. From each segment a sample was taken for the gravimetric water content analysis (drying at 105 °C) and the rest of the soil in the segment was taken to drying in 40 °C, grinding ,sieving at 2 mm and extractions for the chemical analysis.

Drying is essential for grinding and sieving the soil and getting a representative and extractable sample before extraction, (we will insert “grind and” before “sieved”, L 142).

We used the same method here as performed in the work on citrus orchards which we use some of its results in the current analysis (Kurtzman et al. 2013). Prior to that work tests were made comparing extraction from fresh – dry – dry + grined and sieved samples and the average results of nitrate and ammonium were mostly the same (or lower in the fresh samples due to less effective extraction). The main problem with fresh samples was the large differences between replicates from the same core-segment, relative to the good homogenization of the sample after drying and sieving.

The choice of drying at 40 °C rather than warmer or cooler is to balance among the necessity of drying, the impact on the concentrations (mainly due to ammonium volatilization) and the drying duration. Drying soil in 40 °C for nitrate analysis is a common practice in soil science (e.g. Bottomley, P.S., Angle J.S., and Weaver. R.W.: Methods of Soil Analysis: Part 2—Microbiological and Biochemical Properties SSSA Book Ser. 5.2., SSSA, Madison, WI., 1994.; Unkovich, M., Herridge, D., Peoples, M., Cadisch, G., Boddey, R., Giller, K., Alves, B. and Chalk, P.: Measuring plant-associated nitrogen fixation in agricultural systems, Australian Centre for International Agricultural Research (ACIAR), Canberra, Australia, 2008).

20) **L184:** I understand modelling simplification, but if there are some farms close to the region (as you propose as cause of the well differences) I have some doubts about the application of only this kind of compost. Could you discuss a little bit.

Response 20: In the referenced study in line 184 (Ben Hagai et al., 2011) many types of composts from different compost producers were analyzed and averages of composts physical and chemical properties are published. We use these average numbers for modeling nitrogen inputs due to compost application in our regional analysis. Of course this may be another source of variability, nevertheless we believe compost that is distributed well in the field will not cause major differences in nitrate concentration in wells in regional scale.

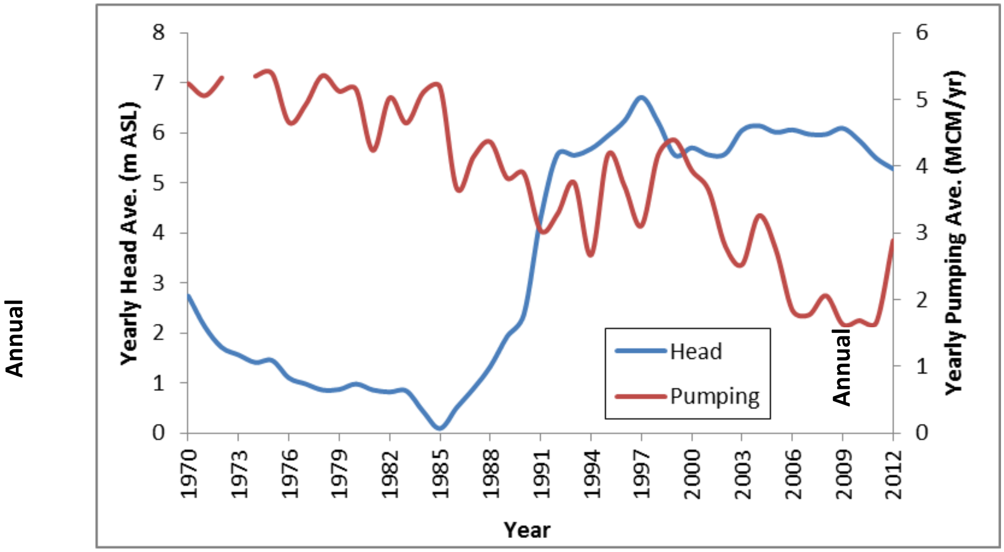
21) **L189:** I understand that you try to fix your simulated data after 50 years to the values observed in the 12 cores; however, what are your initial data? Moreover, you said that the same crops have been cultivated during 15 years but you simulate 50.

Is the same water table now than 50 years ago? (probably with smaller irrigated area). Could you discuss that?

Response 21: The idea of going way back to the past with the unsaturated-zone models is that the initial conditions will not play a significant role in the water and nitrate fluxes that feed the groundwater model starting in 1992 (30 years are sufficient for practice on the ground to reach the water table). Therefore, the actual initial conditions in the unsaturated zone assigned for 1962 are not important. Nevertheless we used reasonable conditions knowing the N concentrations - profile in 2012 and assuming the profile was poorer with N, 50 years before.

Orchards (citrus, persimmon) go back for a few decades in most plots, for the annuals (vegetable), of course more changes were made, in the surface inputs during the years. We took plots that farmers reported their agricultural practice for 15 years because our calibration data (10 m depth) is influenced from this period. This work did not aim at collecting exact history of cultivation for the last 50 years and a steady agro-practice was assumed. We will add: "… under the assumption of steady crop and agricultural practice during the 50 years" (L 195)

As for the water-table, its depth is relatively stable between 1992-2012 therefore these years were chosen as the modeling period of the groundwater model (see figure in the file attached to this response). Doing so the fluxes from the unsaturated zone models were taken as the fluxes at a fixed depth (no water-table influence). Before 1992 the water-table was at lower elevation due to more pumping in this region (see figure in the file attached to this response), data from the Israel water authority). The aforementioned details are in the manuscript in lines 231-235, and we believe further elaboration on this issue will not contribute to the manuscript.



22) L193: Can you define better what slightly means?

Response 22: Slightly means changes in the saturated hydraulic conductivity were in the same order of magnitude. This means the initial hydraulic properties suggested by the pedotransfer functions were not changed dramatically during calibration.

After "slightly changes" we will add: "...(i.e. only Ks within the same order of magnitude)..." (L 198)

23) L200-202: And what do you expect to happen with the N movement? Because in the no crop plots there also mineralization and nitrate leaching.

Response 23: We thank the referee for this good comment. Small surface fluxes of nitrogen exist in the non -cultivated areas. In the no-crop model we applied 10 kg ha^{-1} yr^{-1} nitrogen with some spreading during the year.

We will add after “water flow”: “and nitrogen transport (10 kg ha^{-1} yr^{-1} nitrogen applied on ground surface)..." (L 206)

24) **L208:** calibration is a main part in a modelling process, but independent validation also is. Because you calibration can sometimes be tricky, because you have many ways to get to a good result if you are combining many different parameters, but not only one of this ways is the more accurate. Because of that I suggest you to divide some observed results (in time, space or whatever) and use them for a new calibration and confirm that (validate) simulating for the other points and getting also an accurate adjust to the observed results.

Response 24: We changed our response to this comment after the rightful remark of the editor concerning our original response in the discussion stage (response to comment # 41 was clarified as well). In his remark the Editor stated the need for validation; and encouraged us to show the difficultness of validation here and to discuss the significance of the conclusions drawn with calibrated models that were not validated (with data that was left-out and not used for calibration). Therefore, the following paragraph was added to the discussion in the revised manuscript:

"The workflow in this study did not include model validation after each calibration stage (i.e. for each land-use unsaturated: flow, conservative-transport, and nitrogen reactive-transport; groundwater flow and groundwater transport), which is of course a disadvantage. Validation tests for each calibration would have given a statistical measure of the goodness-of-fit of the calibrated model with independent (left-out of calibration) observations, rather than only the calibration fit. Even though, the conclusions of this work are highly significant because they are based on entirely independent data. The total mass of nitrate that crossed the water-table (unsaturated zone models) is verified by the groundwater well data. Whereas the failure to reproduce the spatial variability would have not been changed with validation fit estimates." (L 400-406)

25) **L234:** Again, why are you using now 20 years instead of the 15 that are sure with the same management?

Response 25: The 15 years which the farmers in the sampled plots reported the agricultural practice were needed for the calibration of the 10 m deep unsaturated zone models. Moving to the regional analysis where the unsaturated-zone models were extended/shortened in depth for each cell of the saturated zone model – the 15 years reported by the specific farmers of the sampled plots, has no meaning any more. The reason for modeling the aquifer for the years 1992-2012 is explained in lines 231-235 and further here in response 21.

26) **L234-235:** Consider including a reference for this statement.

Response 26: After "stable" we will add "(Israel Water Authority data),…" (L240-241)

27) **L237:** the figure labels for each zone (particularly Bnei-Zion) are oversized and do not allow to see at least one sampling point and the scale.

Response 27: We thank the reviewer for this comment the graphics of Figure 5 will be changed in a way that no spatial data will be hidden.

28) **L238-239:** the figure caption for b) and c) are changed.

Response 28: Oops, the caption numbering (b,c) in Figure 5 will be corrected, thanks.

29) **L242:** please, define the period observed for wells.

Response 29: After "in the wells" we will add "(1992-2012)". (L 249)

30) **L250-252:** How do you expect that this change affect the crops? Do you have a crop module? And some of these data fit better in the results than here.

Response 30: This estimation is based on the calculations of Kurtzman et al. (2013) which is cited. Both agricultural and hydrological consequences were discussed in that study, whereas, in the current study we concentrate on the hydrological. Kurtzman et al. 2013 emphasize the point, that the reduction in N-root-uptake is smaller than the reduction in nitrate-leaching when N input is reduced from its high rates, because of the root-up-take data-supported model that is used there for citrus and in the same form (different parameters) here for vegetables and deciduous. From Kurtzman et al., 2013: a decrease of 25 % in the nitrogen fertilization mass results in a decrease of only 4 % - 9 % of root nitrogen uptake yet it resulted in a decrease of 50 % nitrate-nitrogen flux at the water table. Decrease of 50 % in the nitrogen fertilization mass results in a decrease of 22 % of root nitrogen uptake and a decrease of 72 % nitrate-nitrogen flux at the water table.

This short paragraph is included in the Materials and methods section because it explains the method used for the scenario simulations in this work. The results reported here are from Kurtzman et al. (2013) and not a results of this work.

31) **L256:** are they similar or are you using the same data? Define.

Response 31: Thanks for this comment. The same data was used. We will change the wording in the revised manuscript so it will be clear that the same data was used for future atmospheric boundary conditions. (L 263)

32) **L274:** Why do you think that nitrate flux is 540 kg ha in the persimmon A if you only apply 200 kg?

Response 32: The N-NO3 deep fluxes reported in the bottom line of Table 1 are those calculated for each sampled profile using the Cl and N-NO3 concentrations of the deep sediment (Eq. 1 & 2). In this profile (persimmon A) the deep samples contained very high N-NO3 concentrations resulting in high calculate N-NO3 flux. As the referee commented rightfully, this flux is almost 3 folds higher than the surface N input the farmer apply on average on the orchard surface. This is a good example of the heterogeneity of N deep fluxes that is needed to support the concentrations in the contaminated wells, and it is discussed later (lines 375 – 377). If the N-NO3 fluxes to the water table were everywhere 20-50% of the applied-N rate (crop dependent) as they are on average, the concentrations of NO3 in wells was much more uniform than observed – this is a major result of this study.

33) **L315:** consider including units.

Response 33: the porosity and the dispersivities-ratio are dimensionless.

34) **L330-332:** how do you define which region should be multiplied by which factor? I do not understand this arbitrary correction.

Response 34: After the first round of calibration of the NO3 transport model we knew we get good the total mass of nitrate that enters the aquifer from the unsaturated zone. We also knew these fluxes are too uniform to produce the variability in nitrate concentrations in wells. We still assumed the variability in wells can only be caused by variable top fluxes. Therefore, we first asked in how much should we multiply the nitrate fluxes to get the high concentrations that we got in the most contaminated wells ([NO3]>100 mg/l). We found we need multipliers of 5 and even 10 for the fluxes near these wells. To keep the total flux in the region the same, the small areas that got high multipliers had to be compensated with relatively large areas with low multipliers – these were naturally distributed around wells were the nitrate concentrations are relatively low. Most of the calculated fluxes from the field profiles (Table 1) were not very far from the average flux of our representative transient model for each crop (Table 3), hence there is good reasoning for keeping most of the area multiplied by 1. The range of 2 orders of magnitude for the multipliers was not chosen arbitrary it was shown in the work of Kourakus et al 2012 that nitrate fluxes cover 2 order of magnitude, this will be clarified in the revised text.

35) **Table 5:** define which coefficient for each one, please.

Response 35: The comment is not clear, what coefficient? what one?

If the referee meant what multiplier was assigned to each well in the 2^{nd} calibration, the answer is that there is no one multiplier for each well. There are different multipliers for different model cells around each well. Generally, the high multipliers were assigned close to the most contaminated wells.

36) **L343-345:** this has been already defined in the material and methods

Response 36: That is correct. Nevertheless, we prefer to repeat shortly here (2 lines) for the reading -flow of the results section, and for readers who are only interested in the scenario simulations results.

37) **L369-370:** It could be many other things, the simplification level of the system, the soil variability (you only sampled 12 cores for a 13.3 km2 surface), different soil/rainfall/management/nitrate in the well irrigation water/. . . for the same crop.

Response 37: We generally agree and thank the referee for this comment and will change the wording here to be less strict. Yet, it should be emphasized that fluxes of nitrate that are a few folds larger than any reasonable fertilization rate (like the referee noticed in comment # 32), are needed to reconstruct the contamination in wells. This conclusion should be said somehow loud. The following discussion explains (and will explain better in the revised text) possible reasons for uneven distribution of N by close-to-well high loads. Further, heterogeneity of porous-medium is mentioned as possible cause for variability in concentrations in groundwater. See also response to comment # 24 of Referee # 1 and the changes we suggested there.

38) **L370-373:** if you do not completely trust your data; how do you expect that we could do?

Response 38: We completely trust the nitrate-concentration of water sampled from wells and analyzed by the Israel Water Authority or authorized organizations and are reported in their database. These are the concentration in wells' water; the lines mentioned by the referee discus, what may lead to high concentrations in some wells and has nothing to do with the goodness of this data.

We also completely trust farmers report on level of fertilization in the following manner. For example, if a farmer uses liquid NH4NO3 as N-fertigation in his drip-irrigation system. We are positive he calculates well the mass of N he brings to the field in the fertilizer tank considering the irrigated area, and his irrigation/fertigation schedule to report X kg N ha-1 y-1 is applied (our models, of daily resolution, breakdown this with additional data from the farmer, to N applied at each irrigation). What we say in these lines is that in the vicinity of some wells, the fertilizer for some time, did not find its way to evenly be distributed in the area and higher fluxes occurred near the contaminated wells.

39) **L379:** I could agree with you, but you should validate these coefficients in order to see that they are not just a mathematical trick.

Response 39: If the referee means by "mathematical trick" that this specific set of coefficients and their distribution are not a unique solution for fitting the observed and modeled nitrate concentration, we partly agree. The # of multipliers, their values, and their spatial distribution may be further optimized, yet the optimized scheme will lead to the same important conclusions: 1) The most contaminated wells require local fluxes of nitrate that cannot be explained by regular agricultural practice and representative physically-based modeling ; 2) In most of the agricultural area the fluxes produsced by the representative data-based models are a good estimate of the local nitrate fluxes at the water table;

As of the validation see response to comment # 24 here.

40) **Figure 10:** check the image quality.

Response 40: We want to preserve the original figure from Mercado 1976 (we believe the "old looking" gives the reader a good feeling of the post audit analysis 40+ year old predictions).

41) **L417:** I do not think that this is a "significant success" without some kind of validation.

Response 41: Here there is no question of validation with unsaturated-zone data that was left-out of the calibration and used for validation of the unsaturated zone models. The unsaturated zone analysis and models gave a very good estimate of the total mass of nitrate that entered the aquifer from above. This successful estimate is verified by comparing the average nitrate concentrations data in groundwater wells with these concentrations in the groundwater model that is fed by the unsaturated zone models.

List of all relevant changes made in the revised manuscript

L 14-16: "Contaminated areas often show large spatial variability of nitrate concentration in wells. In this study we tried to assess whether this spatial variability, can be characterized on the basis of land use and standard agricultural practices."

L 41-46: "As a consequence, nitrate has become the most common groundwater contamination caused by agricultural activity in many countries (Jalali, 2005; Vitousek et al., 2009; Burow et al., 2010; Kourakos et al., 2012; Yue et al., 2014; Wheeler et al., 2015; Wang et al., 2016). In Israel for example, more than half of all the wells that have been disqualified as sources of drinking water were disqualified due to nitrate contamination (Israel Water Authority; IWA, 2015a). The process of groundwater contamination by nitrate occurs mainly below light soils and less under cultivated clays (Spalding and Exner, 1993; Kurtzman et al., 2016)."

L 51: "(18 crop varieties)"

L 55: "(advective and diffusive)"

L 57: "aerated"

L 58: "(0-45 cm, Kurtzman et al., 2013)"

L 58-59: "Moreover, ammonium is a cation and tends to adsorb to the soil solids (clay fraction, organic matter)"

L 64-65: "(e.g., Liao et al. 2012; Thayalakumaran et al., 2015, Green et al., 2016)"

L 74-76: "In environments that are more humid accesses nitrogen from agricultural sources in surface water bodies is an ecological concern, however, under Mediterranean climate, the problem of groundwater contamination is the major problem concerning N leaching from agricultural land."

L 71,72,73: "variability"

L 83: "reconstructing the observed groundwater nitrate concentrations"

L 84: "variability"

L 106: "(standard-deviation average^{-1})"

L 123-124: "by sprinklers"

L 124: "field"

L 125: "by micro-sprinklers (at the early stage of growing) and drip irrigation after"

L 127: "irrigated by micro-sprinklers"

L 142: "grind and"

L 195: "under the assumption of steady crop and the same agricultural practice during the 50 years"

L 198: "(i.e. only Ks within the same order of magnitude)"

L 206: "and nitrogen transport (10 kg ha^{-1} yr^{-1} nitrogen applied on ground surface)"

L 233: "Israel Water Authority"

L 240-241: "Israel Water Authority data"

Fig. 5: The "Bnei Zion" label moved that no spatial data is hidden. The figure numbering (b,c) was corrected.

L 249: "(1992-2012)"

L 263: "the same as that of"

L 263-268: "In the groundwater transport model the initial condition was the measured nitrate concentration at 2012. The transient nitrate-concentration boundary conditions were modified to account for similar reductions in nitrogen fertilization outside of the model domain. This was done by two steps: (1) run the model to the future with constant nitrate-concentration boundary condition and looking on the trends of the nitrate concentration of the wells inside the model domain; (2) adjusting these trends to the boundary condition and run the model to the future again with transient nitrate-concentration boundary conditions."

L 367: "CMB - Chloride mass balance."

L 317: "achieved"

L 323: "final"

L 325: "mean nitrate concentration for the entire modeled area"

L 331-332: "This means that"

L 334-337: "The meaning of this, is that nitrate spatial variability cannot be explained only by physical process of agricultural practice and land-use variability on surface. Other factors that are local and arbitrary, significantly affect nitrate concentration in some wells and therefore the measured spatial variability of nitrate in the aquifer. These factors were introduced into the numerical model as will be explained hereafter."

L 344: "and some physical explanations"

L 364: "as an"

L 365: "Even in this case about half of the wells will still exceed the standard concentration."

L 378: "first-order"

L 386: "that can be due to one or more of the following reasons"

L 391-397: "Heterogeneity of the porous medium may cause extremely high nitrate fluxes likewise well failure discussed previously, and may be a source for local high contamination. The field survey reported here support this statement. Of the nine deep profiles reported here (Figure 6, Table 1), one showed extreme nitrate concentrations and calculated nitrate fluxes that were 4- to 5-fold higher than in the other profiles extracted from the same orchard (Persimmon A, Table 1). Multipliers that adjust nitrate fluxes to groundwater were used in models previously (e.g. Alikhani et al., 2016). The two orders of magnitude difference in nitrate multipliers (0.1 – 10) used in this work is not incomparable, Kourakos et al. (2012) used distributions with means of 1 to 100 mg L^{-1} nitrate, loading to groundwater for the same land-use."

L 397: "heuristic"

L 400-406: "The workflow in this study did not include model validation after each calibration stage (i.e. for each land-use unsaturated: flow, conservative-transport, and nitrogen reactive-transport; groundwater flow and groundwater transport), which is of course a disadvantage. Validation tests for each calibration would have given a statistical measure of the goodness-of-fit of the calibrated model with independent (left-out of calibration) observations, rather than only the calibration fit. Even though, the conclusions of this work are highly significant because they are based on entirely independent data. The total mass of nitrate that crossed the water-table (unsaturated zone models) is verified by the groundwater well data. Whereas the failure to reproduce the spatial variability would have not been changed with validation fit estimates."

L 454-456: "Alikhani, J., Deinhart, A. l., Visser, A., Bibby, R.K., Purtschert, R., Moran, j. E., Massoudieh, A., Esser, B. K.: Nitrate vulnerability projections from Bayesian inference of multiple groundwater age tracers, J. Hydrol., 543, 167-181, 2016."

L 489-491: "Green, C. T., Jurgens, B. C., Zhang, Y., Starn, J. J., Singleton, M. J., Esser B. K.: Regional oxygen reduction and denitrification rates in groundwater from multi-model residence time distributions, San Joaquin Valley, USA, J. Hydrol., 543, 155-166, 2016."

L 528-529: "Liao, L., Green, C. T., Bekins, B. A., Böhlke, J. K.: Factors controlling nitrate fluxes in groundwater in agricultural areas, Water Resources Res., 48, W00L09, doi:10.1029/2011WR011008, 2012."

Modeling nitrate from land-surface to wells' perforations under agricultural land: success, failure, and future scenarios in a Mediterranean case study.

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Abstract. Contamination of groundwater resources by nitrate leaching under agricultural land is probably the most troublesome agriculture-related water contamination worldwide. Contaminated areas often show large spatial variability of nitrate concentration in wells. In this study we tried to assess whether this spatial variability, can be characterized on the basis of land use and standard agricultural practices. Deep soil sampling (10 m) was used to calibrate vertical flow and nitrogen-transport numerical models of the unsaturated zone under different agricultural land uses. Vegetable fields (potato and strawberry) and deciduous orchards (persimmon) in the Sharon area overlying the coastal aquifer of Israel were examined. Average nitrate-nitrogen fluxes below vegetable fields were 210–290 kg ha^{-1} yr^{-1}, and under deciduous orchards, 110–140 kg ha^{-1} yr^{-1}. The output water and nitrate-nitrogen fluxes of the unsaturated zone models were used as input data for a three-dimensional flow and nitrate-transport model in the aquifer under an area of 13.3 km^2 of agricultural land. The area was subdivided into four agricultural land uses: vegetables, deciduous orchards, citrus orchards and non-cultivated. Fluxes of water and nitrate-nitrogen below citrus orchards were taken from a previous study in the area. The groundwater flow model was calibrated to well heads by changing the hydraulic conductivity. The nitrate-transport model, which was fed by the abovementioned models of the unsaturated zone, succeeded in reconstructing the average nitrate concentration in the wells. However, this transport model failed in calculating the high concentrations in the most contaminated wells and the large spatial variability of nitrate concentrations in the aquifer. To reconstruct the spatial variability and enable predictions, nitrate fluxes from the unsaturated zone were multiplied by local multipliers. This action was rationalized by the fact that the high concentrations in some wells cannot be explained by regular agricultural activity, and are probably due to malfunctions in the well area. Prediction of the nitrate concentration 40 years in the future with three nitrogen-fertilization scenarios showed that: (i) under the "business as usual" fertilization scenario, the nitrate concentration (as NO_3^-) will increase on average by 19 mg L^{-1}; (ii)under a scenario of 25 % reduction of nitrogen fertilization, the nitrate concentration in the aquifer will stabilize; (iii) with a 50 % reduction of nitrogen fertilization, the nitrate concentration will decrease on average by 18 mg L^{-1}.

1 Introduction

1.1 Groundwater contamination by nitrate under agricultural land

Since the development of the Haber–Bosch process in 1910, in which ammonia (NH_3) is cheaply produced from atmospheric nitrogen (N_2), mineral nitrogen has become the most important and common fertilizer in modern intensive agriculture. This process earned Fritz Haber the Nobel Prize for chemistry in 1918 and its significance was emphasized for many decades thereafter (e.g. "the most important invention of the 20th century" – Smil, 1999; Erisman et al., 2008). However, nitrogen fertilization is commonly applied in surplus and leaches below the roots, mainly as the conservative anion nitrate (NO_3^-), which has strict limits under drinking-water standards worldwide. As a consequence, nitrate has become the most common groundwater contamination caused by agricultural activity in many countries (Jalali, 2005; Vitousek et al., 2009; Burow et al., 2010; Kourakos et al., 2012; Yue et al., 2014; Wheeler et al., 2015; Wang et al., 2016). In Israel for example, more than half of all the wells that have been disqualified as sources of drinking water were disqualified due to nitrate contamination (Israel Water Authority; IWA, 2015a). The process of groundwater contamination by nitrate occurs mainly below light soils and less under cultivated clays (Spalding and Exner, 1993; Kurtzman et al., 2016).

1.2 The path from nitrogen fertilizer to nitrate in groundwater

Many studies have reported leaching ranges of 25–90 % of the nitrogen applied to agricultural fields in different crops and countries (Guimerá et al., 1995; McMahon and Woodside, 1997; Neilsen and Neilsen, 2002; Kraft and Stites, 2003; de Paz and Ramos, 2004; Ju et al., 2006; Zhao et al., 2011; Venterea et al., 2011). In Israel, Bar-Yosef et al. (1999) reported nitrate leaching of 55–65 % for different vegetables and field crops (18 crop varieties) in a 35-year survey. More recently, Turkeltaub et al. (2015) calculated leaching ratios in the range of 15–35 % under a modern greenhouse for intensive growing of vegetables.

Applications of nitrogen fertilizers of different species: nitrate, ammonium (NH_4^+) or organic nitrogen (e.g. urea, manure, compost) or a combination of these, are practiced. Most crops up-take only the mineral species (nitrate, ammonium). The nitrate and ammonium are up-taken by plant roots mostly in a mass transport process (advective and diffusive), which is limited by a crop-specific threshold concentration (Sorgona et al., 2006; Kurtzman et al., 2013). Some of the organic nitrogen in the soil is mineralized to ammonium and in aerated light soils, most of the ammonium is oxidized to nitrate (nitrification) in a relatively thin layer in the upper part of the soil column (0-45 cm, Kurtzman et al., 2013). Moreover, ammonium is a cation and tends to adsorb to the soil solids (clay fraction, organic matter). Under anaerobic conditions, the nitrate is reduced to nitrogen gas via denitrification, which takes the nitrogen out of the system (Galloway et al., 2004). Nevertheless, denitrification is not a significant process in relatively aerated sandy soils and is frequently assumed to be negligible (Hanson et al., 2006; Doltra and Muñoz, 2010; Turkeltaub et al., 2015). Due to these processes, the nitrogen species that leaches down to the aquifer is mainly nitrate. In the groundwater, nitrate is diluted and transported mostly as a conservative anion that is often extracted out of the system by pumping wells. Denitrification in aquifers is an important process in some cases (e.g., Liao et al. 2012; Thayalakumaran et al., 2015, Green et al., 2016). Nevertheless, in the thick aquifer discussed here, dominated by sandy sediments and under Mediterranean climate, denitrification is negligible in the upper 95 % of the aquifer's depth (Kurtzman et al., 2012). In environments that are more humid accesses nitrogen from agricultural sources in surface water bodies is an ecological concern, however, under Mediterranean climate, the problem of groundwater contamination is the major problem concerning N leaching from agricultural land.

Nitrate contamination of the groundwater below agricultural land is often characterized by significant spatial variability of the nitrate concentrations in wells (Hu et al., 2005; Liu et al., 2005; Wheeler et al., 2015). This variability may evolve from the spatial variability of the soil properties. Nevertheless, in an area with relatively uniform soil, it is most likely related to variable land use (crops) and inconsistent agricultural practices (Almasri and Kaluarachchi, 2007; Bian et al., 2016).

Research of nitrate leaching from agricultural land can be divided into three scales and zones of interest. Agricultural aspects of root uptake of nitrate and its seepage below the root zone have been studied quite extensively in the agricultural research domain, where transient mechanistic models are often used for the analysis (e.g., Hanson et al., 2006; Doltra and Muñoz, 2010). The developing vadose-zone hydrology discipline looks at nitrate data and processes deeper in the unsaturated zone as well (Kurtzman et al., 2013; Dahan et al., 2014). Regional assessments of groundwater contamination with nitrate make use of varying degrees of simplification of vadose-zone processes (e.g., Mercado, 1976; de Paz and Ramos, 2004; Kourakos et al., 2012).

The objective of this research was to quantitatively assess the nitrate throughout its course from fertilization on the field surface through the flow processes in the root zone, down through the thick unsaturated zone, and in the aquifer toward the pumping wells. We further aimed at reconstructing the observed groundwater nitrate concentrations by calculated fluxes from the unsaturated zone, and to explain the spatial variability of the nitrate concentration in the groundwater by the spatial variability of the surface land use. Finally, we used the field- and regional-scale calibrated models for future assessment of aquifer contamination under different fertilization scenarios.

2 Materials and methods

2.1 Research area: nitrate contamination in the Sharon area, Israel

The nitrate problem in groundwater in Israel is concentrated under intensively cultivated areas of Mediterranean red sandy-loam (Hamra) soil overlying the coastal aquifer (IWA, 2015b; Kurtzman et al., 2016). Two main regions in which nitrate contamination has been a concern for several decades are Rehovot–Rishon (Mercado, 1976) and the Sharon region (Kurtzman et al., 2013). This research focuses on the Sharon area (Fig. 1). The Israeli coastal aquifer is an unconfined aquifer, one of the most important freshwater sources in Israel for both agriculture and domestic consumption.

The climate is semiarid with annual precipitation of 550 mm mainly during the winter season from November to April. The main land uses over the aquifer are agricultural and residential (cities, towns and villages). The aquifer is in the Kurkar group (Pleistocene) composed of sands, calcareous sandstone, and marine and continental silty and clay lenses. The aquifer lies over the thick clays of the Saqiye group, which are conceptualized as an aquiclude (Gvirtzman, 2002). The unsaturated zone thickness ranges from 3 to 80 m below ground surface.

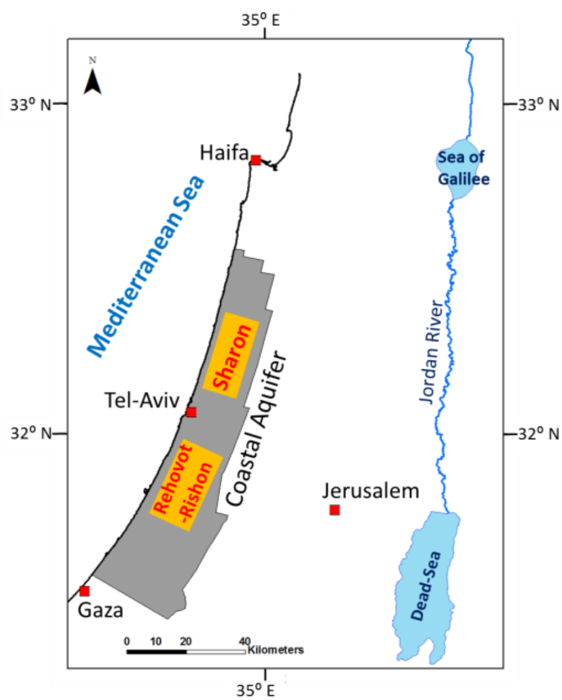

Figure 1: Location map of the Israeli coastal aquifer and two areas (in red) with major nitrate contamination of the groundwater. This work presents a case study focusing on the Sharon area.

This research concentrates on a 13.3 km^2 agricultural area in the Sharon region. Nitrate concentration in wells in this research area have been increasing by an average 1 mg L^{-1} yr^{-1} for more than 40 years (Kurtzman et al., 2013). Although generally considered contaminated, significant spatial variability exists in the nitrate concentration in wells over short distances. Heavily contaminated wells can be at as little as 500 m from a non-contaminated well (Fig. 2). The coefficient of variation (standard-deviation $average^{-1}$) of nitrate concentration in the wells in Fig. 2 is 38% (Levy, 2015). This spatial variability indicates local contamination sources rather than regional contamination. It might evolve from crop type, fertilization masses or the agricultural practice in the fields at ground surface. Therefore, the research area was divided into four characteristic land uses: vegetables (40 % of area, large masses of nitrogen fertilization), citrus (33 % of area, also transpiring in the winter season), deciduous (14 % of area, large volumes of irrigation) and no crop (13 % of area) (land-use data from Survey of Israel maps, 2000).

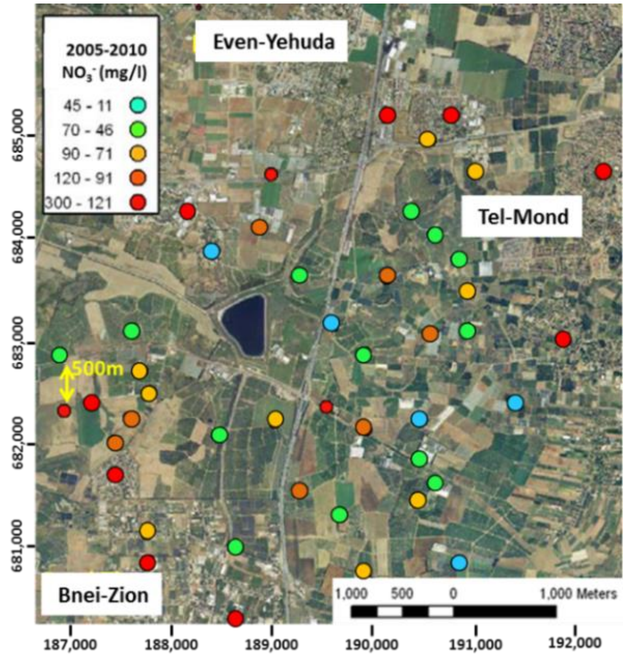

Figure 2: The agricultural area selected for modeling and 5-year average nitrate concentration in wells. Note the high spatial variability. Nitrate concentration data are from the Israel Water Authority. Coordinates system: Israeli Transverse Mercator (ITM).

2.2 Nitrate fluxes from the fields to the deep unsaturated zone

2.2.1 Fields, irrigation, fertilization and meteorological data

For the three aforementioned crop types, representative fields were selected for deep sampling in the Hamra soils of the Sharon region: potato and strawberry fields representing the vegetable land use; a persimmon plantation representing the deciduous crop, and data from an orange orchard reported in Kurtzman et al. (2013) representing citrus. In each field, data of irrigation and fertilization regimes (quantities and timing in daily resolutions) were collected from the farmers. Data on irrigation water quality (nitrate and chloride concentrations) were collected from the Israel Water Authority. The potato field was irrigated by sprinklers with an average irrigation depth of 480 mm yr^{-1}, and fertilized with 450 kg N ha^{-1} yr^{-1}. The strawberry field was irrigated by micro-sprinklers (at the early stage of growing) and drip irrigation after to an average depth of 1000 yr^{-1}, and fertilized with 450 kg N ha^{-1} yr^{-1}. Strawberries were grown under plastic tunnels and the field was completely covered with a plastic sheet during the winter, hence precipitation was not counted in the water balance for this field. The persimmon orchard was irrigated by micro-sprinklers to an average depth of 850 mm yr^{-1}, and fertilized with 200 kg N ha^{-1} yr^{-1}. The nitrogen forms of the applied fertilization were ammonium-nitrate solution in the irrigation water (persimmon and strawberry) and dry scattering of urea (potato). Nitrogen in the compost was accounted for in the strawberry and potato fields where this organic amendment was applied. The farmers in all representative fields reported that the same crop was cultivated for at least 15 years before sampling (with minor exceptions for the potato field). Time series of daily precipitation and reference evapotranspiration (Penman–Monteith equation, Allen et al., 1998) for each field were collected from nearby automated meteorological stations operated by the Israel Ministry of Agriculture.

2.2.2 Field sampling and soil analysis

In each of the three fields (persimmon, strawberry and potato), three sampling coreholes were drilled using the direct push technique, and a continuous core was obtained from 0–10 m depth (Fig. 3). The coreholes were drilled at a distance of 50–200 m from each other. Soil (and sediment) cores were cut into 30-cm segments. Drilling was done in June 2012. Core segments were sealed with caps and tape and kept in a cooler until reaching the laboratory, where the core segments were analyzed for the following variables: gravimetric water content (105 °C), bulk density (core dry mass per volume), gravimetric particle-size distribution (hydrometer method), chloride concentration of a 1:2 soil:water extract (with Sherwood 926 chloridometer), nitrate and ammonium concentrations in a 1 M KCl 1:5 soil:water solution extract (Kachurina et al., 2000, with Quickchem 8000 autoanalyzer, Lachat Instruments, Loveland, CO). The soil samples that were used for extraction were grind and sieved to 2 mm after drying (40 °C for 3 days).

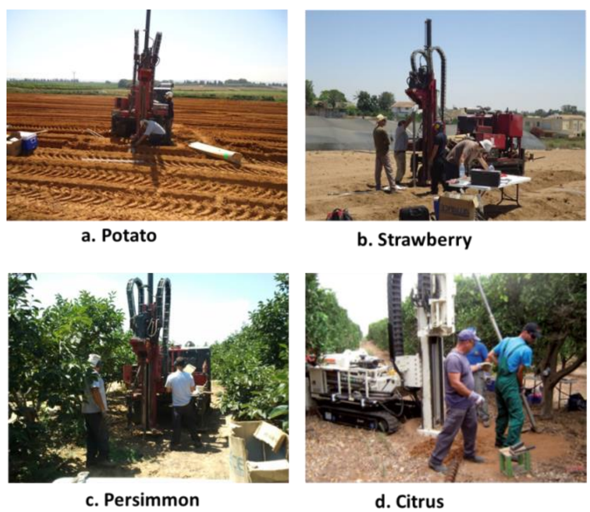

Fig. 3. Direct push sampling of the unsaturated zone (0–10 m below ground surface) under the different agricultural land uses. (a–c) Sampled in the current study. (d) Sampled for Kurtzman et al. (2013): the unsaturated model developed there was used here.

2.2.3 Modeling water flow and nitrogen transport in the unsaturated zone

Steady-state approximations:

Average fluxes of water and nitrate-nitrogen toward the groundwater under the fields were calculated in a steady-state approximation with the chloride mass balance (Allison and Hughes, 1983; Scanlon et al., 2007):

$$R = \frac{\left(P\cdot Cl_p + I\cdot Cl_I\right)\int_{z=10m}^{z=2m}\theta(z)dz}{\int_{z=10m}^{z=2m}\theta(z)\cdot Cl_{PW}(z)dz} \ , \quad (1)$$

where R [L T^{-1}] is the mean annual groundwater recharge flux, P [L T^{-1}] is the mean annual precipitation flux, I [L T^{-1}] is the mean annual irrigation application, Cl [M L^{-3}] is the steady-state approximation of the chloride concentration with subscripts P, I and PW referring to precipitation, irrigation water and unsaturated-zone pore water, respectively, and θ [L^3 L^{-3}] is the volumetric water content. The interval of integration for calculating deep unsaturated-zone averages was from z = 2 m (below the root zone) to z = 10 m depth (deepest available data). The steady-state approximation of nitrate flux to the groundwater was obtained by multiplying the water flux (R, Eq. 1) by the depth- and θ-weighted average of nitrate-nitrogen concentrations below the root zone:

$$F_{NO_3} = \frac{R\int_{z=10m}^{z=2m}\theta(z)\cdot \mathrm{NO_3}-N_{PW}(z)dz}{\int_{z=10m}^{z=2m}\theta(z)dz} \ , \quad (2)$$

where F_{NO3} [M L^{-2} T^{-1}] is the mean annual flux of nitrate-nitrogen to the groundwater and NO_3-N_{PW} [M L^{-3}] is the nitrate-nitrogen concentration in the deep vadose zone pore water.

Transient models:

Transient vertical 1D numerical models of water flow and nitrogen transport were calibrated to data of one drill hole in each field: potato, strawberry and persimmon. The numerical code HYDRUS-1D was used for the calibration and simulations (Šimůnek et al., 2009). The 1D vertical Richards' equation with a root water-uptake sink was used for modeling flow in the unsaturated zone:

$$\frac{\partial\theta(h)}{\partial t} = \frac{\partial}{\partial z}\left[K(h)\cdot\left(\frac{\partial h}{\partial z}+1\right)\right] - S(h) \ , \quad (3)$$

where t [T] is the time, z [L] is the vertical coordinate, h = h(z,t) [L] is the pressure head, θ(h) is the volumetric water content, K(h) [L T^{-1}] is the unsaturated hydraulic conductivity, and S(h) [T^{-1}] is a root water-uptake sink term which is non-zero in a transpiring root zone. The van Genuchten–Mualem model (Mualem, 1976; van Genuchten, 1980) was used for the θ(h) and K(h) relationships of the different sediment layers, and Feddes et al.'s (1978) functions, fitted to each crop, were used for S(h) (Šimůnek et al., 2009).

One dimensional advection–dispersion equations representing chain reactions of the nitrogen system are presented in Eqs. (4–6). Only ammonium is accounted for in the solid phase. Sink/source terms for: root uptake of ammonium and nitrate, urea/compost mineralization, ammonium volatilization, ammonium nitrification and nitrate denitrification complete the right-hand side of this equation system.

$$\frac{\partial\theta C_{Ur}}{\partial t} = \frac{\partial}{\partial z}\left(\theta D\frac{\partial C_{Ur}}{\partial z}\right) - \frac{\partial q C_{Ur}}{\partial z} - \mu_{min}\theta C_{Ur} \ , \quad (4)$$

$$\frac{\partial\theta C_{NH4}}{\partial t} + \frac{\partial\rho K_d C_{NH4}}{\partial t} = \frac{\partial}{\partial z}\left(\theta D\frac{\partial C_{NH4}}{\partial z}\right) - \frac{\partial q C_{NH4}}{\partial z} - f_{NH4}SC_{NH4} + \mu_{min}\theta C_{Ur} - \mu_{nit}\theta C_{NH4} - \mu_{vol}\theta C_{NH4} \ , \quad (5)$$

$$\frac{\partial \theta C_{NO3}}{\partial t} = \frac{\partial}{\partial z}\left(\theta D \frac{\partial C_{NO3}}{\partial z}\right) - \frac{\partial q C_{NO3}}{\partial z} - f_{NO3} S \cdot C_{NO3} + \mu_{nit} \theta C_{NH4} - \mu_{dnit} \theta C_{NO3} \ , \qquad (6)$$

where C_{Ur} C_{NH4}, and C_{NO3} [M L^{-3}] are concentrations of the nitrogen species (urea, ammonium and nitrate, respectively) in the porewater solution, ρ [M L^{-3}] is the soil's bulk density, θ [L^3 L^{-3}] is volumetric water content, D [L^2 T^{-1}] is the hydrodynamic dispersion coefficient, q [L T^{-1}] is the water flux, $f_{NH4}SC_{NH4}$ and $f_{NO3}SC_{NO3}$ [M T^{-1} L^{-3}] are the root ammonium-nitrogen- and nitrate-nitrogen-uptake sinks, respectively, where f_{NH4} and f_{NO3} are user-defined functions relating solute uptake to the water uptake S and solute concentrations; μ_{min} [T^{-1}] is a first-order urea/compost mineralization rate (sink term in Eq. 4 and source term in Eq. 5), μ_{nit} [T^{-1}] is a first-order nitrification rate (sink term in Eq. 5 and source term in Eq. 6), μ_{vol} [T^{-1}] is a first-order ammonium-nitrogen volatilization rate, μ_{dnit} [T^{-1}] is a first-order denitrification rate and k_d [L^3 M^{-1}] is the ammonium-nitrogen partition coefficient. Application of compost (strawberry) was treated with equations (4–6) as follows: farmers' reports of annual application of compost (m^3 ha^{-1}) were converted to mineralized nitrogen (Eq. 4) according to 15 % and 5 % nitrogen by mass mineralized in the first and second year after application, respectively (Eghball et al., 2002). A dry compost density of 600 kg m^{-3} with 2 % of the dry mass consisting of nitrogen were used (Ben Hagai et al., 2011).

Fifty years (1962–2012) of daily precipitation, reference evapotranspiration (approximated from pan evaporation before 2002), irrigation water (with appropriate chloride and nitrate concentrations) and nitrogen fertilization were set as the upper boundary condition. A "Free Drainage" boundary (pressure gradient = 0) was used as the bottom boundary condition throughout. The calibration was aimed at fitting the measured profiles on the day of sampling, which was the last day of the 50 years of model runs, under the assumption of steady crop and the same agricultural practice during the 50 years.

Rosetta pedotransfer functions (Schaap et al., 2001) were used with particle-size distribution and bulk-density data to obtain initial values of the parameters of the hydraulic function θ(h) and K(h) for the model layers in the top 10 m (which were sampled and analyzed). These initial values were slightly changed (i.e. only Ks within the same order of magnitude) during the calibration of the flow model in which the error between measured and modeled water contents was minimized. Dispersion coefficients of the soil/sediment layers were calibrated in the transport models with the unsaturated zone chloride observations. Nitrate-nitrogen data were used for calibrating the nitrate, mostly by changing the function of nitrate uptake, f_{NO3} (Eq. 6). All calibrations were performed manually by trial-and-error runs.

To account for the actual unsaturated zone thickness in each cell of the groundwater model, the unsaturated models were extended/shortened to fit steady-state approximations of the actual unsaturated thickness (4–50 m below the ground, at 1m resolution, Fig. 4). This extension was also applied to the citrus orchard model from Kurtzman et al. (2013). Another model was constructed for water flow and nitrogen transport (10 kg ha^{-1} yr^{-1} nitrogen applied on ground surface) in the unsaturated zone below uncultivated areas using the hydraulic properties of the citrus orchard drill holes (this sampling point is at the center of the modeled area).

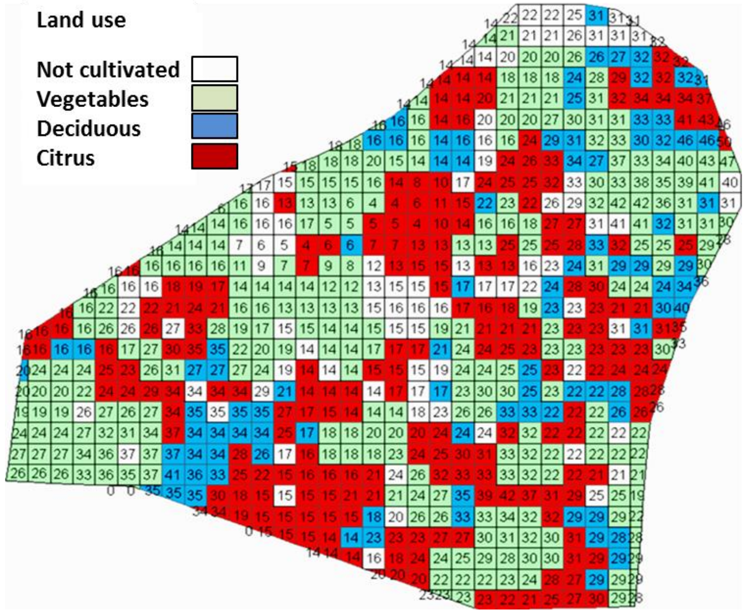

Figure 4: Land use (color) and depth to water table in meters (number) for each grid cell of the modeled area.

Thus, we created a "data library" of transient water and nitrate fluxes at the water table beneath the four land uses (posteriorly the potato model was used for the vegetable land use because the strawberry deep fluxes were similar and the potato field covered a greater area).

2.3 Modeling of water flow and nitrate transport in the aquifer

2.3.1 Boundaries, data, spatial discretization, and simulation period

A water flow and nitrate transport numerical model in the groundwater below the agricultural area in the Sharon region was developed. The model was constructed with GMS 8.2 software (AQUAVEO, 2012), the MODFLOW model for water flow (McDonald and Harbaugh, 1988) and the MT3D model for transport (Zheng, 1990). The model solves the water flow and advection–dispersion equations in the groundwater numerically (Eqs. 7 and 8):

$$S_S \cdot \frac{\partial h}{\partial t} = \frac{\partial}{\partial x}\left(K_{xx}\frac{\partial h}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_{yy}\frac{\partial h}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_{zz}\frac{\partial h}{\partial z}\right) + R - P \,, \qquad (7)$$

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D_x\frac{\partial C}{\partial x}\right) - \frac{\partial (v_x C)}{\partial x} + \frac{\partial}{\partial y}\left(D_y\frac{\partial C}{\partial y}\right) - \frac{\partial (v_y C)}{\partial y} + \frac{\partial}{\partial z}\left(D_z\frac{\partial C}{\partial z}\right) - \frac{\partial (v_z C)}{\partial z} + \frac{R \cdot C_{duz}}{n} - \frac{P \cdot C}{n} \,, \qquad (8)$$

where S_s [L^{-1}] is the specific storage, h [L] is the hydraulic head, t [T] is the time, x,y,z [L] are the three-dimensional coordinates, K_{xx}, K_{yy} and K_{zz} [L T^{-1}] are the hydraulic conductivities along the x, y, z axes, P and R [T^{-1}] are volumetric fluxes per unit volume that represent sinks of water pumping in wells (P) and sources of water from recharge (R). C [M L^{-3}] is nitrate concentration in the aquifer, D_x, D_y and D_z [L^2 T^{-1}] are hydrodynamic dispersion coefficients, v_x, v_y and v_z [L T^{-1}]

are the velocities, n is porosity, and C_{duz} [M L^{-3}] is the nitrate concentration in the deep unsaturated zone (in the recharge flux). The last term on the right in Eq. (8) is the nitrate sink due to pumping.

The modeled area was a polygon of 13.3 km^2 of agricultural land in the Sharon region of Israel. There has been no significant residential land use in this area in the last 60 years and all nitrate fluxes from the ground surface were assumed to be from agricultural sources. The boundary conditions were transient hydraulic heads and nitrate concentrations based on data from wells near the model boundaries. Model calibration was based on transient measured data in wells inside the polygon (Fig. 5a). Time series of well heads and nitrate concentrations for the boundary conditions and calibration were obtained from the Israel Water Authority.

The area was discretized to cells of 150:150 m. Vertically, the model is of 13 layers with thicknesses set according to the wells' perforations (Fig. 5b and 5c). Each cell in the top layer is fed with specific transient fluxes of water and nitrate from the unsaturated zone, according to the unsaturated zone land-use model and its thickness (Fig. 4).

The groundwater model was run for 20 years (1992–2012). The input source/sink fluxes and boundary conditions were inserted into the model as monthly values (stress period = 1 month). By choosing this period, we ensured that the fluxes from the unsaturated zone (model runs start in 1962) represent the land use and not an artifact of initial conditions of the unsaturated zone models. Moreover, during the years 1992–2012, the average water level in the model regions was relatively stable (Israel Water Authority data), supporting the steady-state approximation of the unsaturated zone thickness.

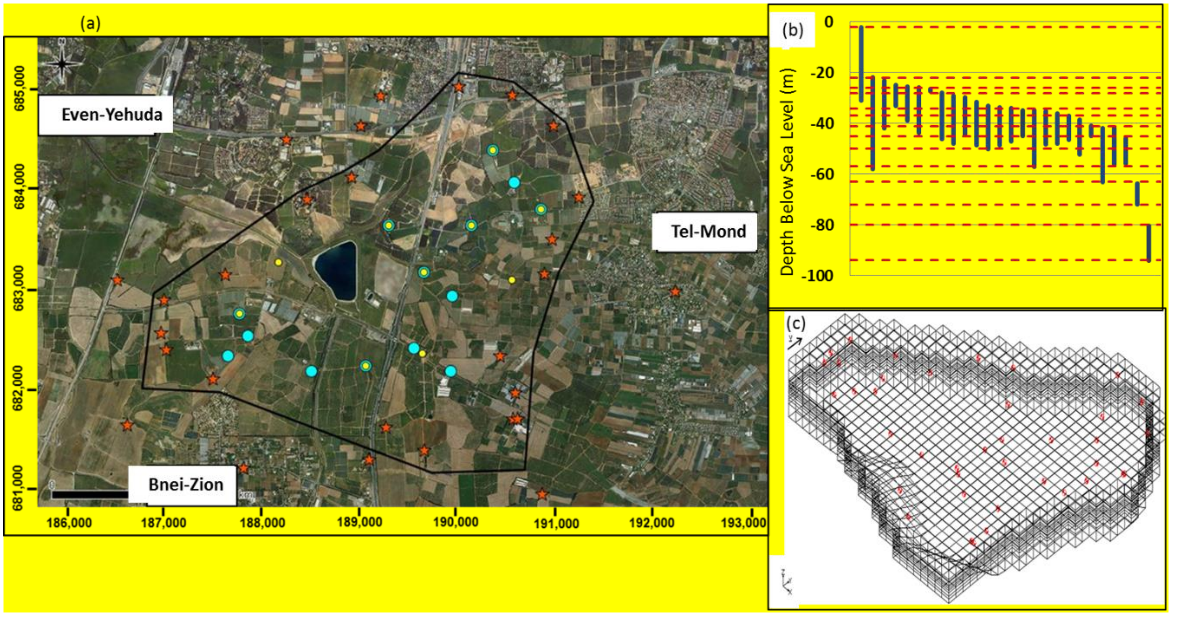

Figure 5: (a) Groundwater model boundaries and wells: red stars – well data used for transient boundary conditions; yellow spots – well data used for calibration of the flow model; blue spots – well data used for calibration of nitrate transport model. (b) Depth of well screens (blue vertical bars) and model layers (red horizontal dashed lines). (c) 3D view of the model domain (finite difference discretization) and wells (red).

2.3.2 Groundwater model calibration

The water flow model was calibrated against measured water levels in the wells (1992-2012). The model was run with some zonation of horizontal hydraulic conductivity and a constant value of the storage coefficient until the mean absolute error (MAE) between measured and calculated water levels over the years was less than 0.5 m, and the mean error (bias) was close to zero. Recharge fluxes from the unsaturated zone models were strictly kept. In the first calibration stage of the nitrate transport model, dispersivity was fitted. Further steps in the calibration of this model were strongly related to the results and are elaborated upon in section 3.

2.3.3 Simulations of future nitrate contamination under various fertilization scenarios

An approximation based on the unsaturated modeling results reported by Kurtzman et al. (2013) was used to estimate the nitrate fluxes at the water table under different fertilization scenarios: a decrease of 25 % in the nitrogen fertilization mass results in a decrease of 50 % nitrate-nitrogen flux at the water table, whereas a reduction of 50 % in nitrogen fertilization results in a 72 % reduction in nitrate-nitrogen at the water table. Time series of nitrate-nitrogen fluxes at the water table were constructed using these ratios and the previously mentioned unsaturated flow and transport models (fitted to land uses and depth of the unsaturated zone). Three scenarios were tested: "business as usual", and 25 % and 50 % reduction in nitrogen application for the years 2012–2052. In these scenarios, it was assumed that land use would not change and that the precipitation and potential evapotranspiration would be the same as that of 1972 to 2012. In the groundwater transport model the initial condition was the measured nitrate concentration at 2012. The transient nitrate-concentration boundary conditions were modified to account for similar reductions in nitrogen fertilization outside of the model domain. This was done by two steps: (1) run the model to the future with constant nitrate-concentration boundary condition and looking on the trends of the nitrate concentration of the wells inside the model domain; (2) adjusting these trends to the boundary condition and run the model to the future again with transient nitrate-concentration boundary conditions.

3 Results

3.1 Unsaturated zone

3.1.1 Sediment data, and steady-state approximations of fluxes

Some spatial variability within the plot of each land use was observed, with one extremely different nitrate profile under the persimmon orchard (Fig. 6). Steady-state recharge and nitrate-nitrogen fluxes (Eqs. 1 and 2) were calculated for the data from each corehole. The spatial variability seen in the profiles (Fig. 6) was reflected by the variable deep fluxes within the plots (Table 1). Transient models were constructed for one corehole in each field. Nitrate-nitrogen fluxes under the strawberry and potato fields were relatively similar (~ 210 kg N ha^{-1} yr^{-1}), hence the transient potato model that was calibrated to Profile C was used for all areas of vegetable land use. The transient model representing the deciduous land use was Persimmon C (Table 1, Fig. 4). Hydraulically-significant lithologic data of the sediment profiles as gravimetric percentage of the clay texture (<0.002 mm) is displayed in table 2.

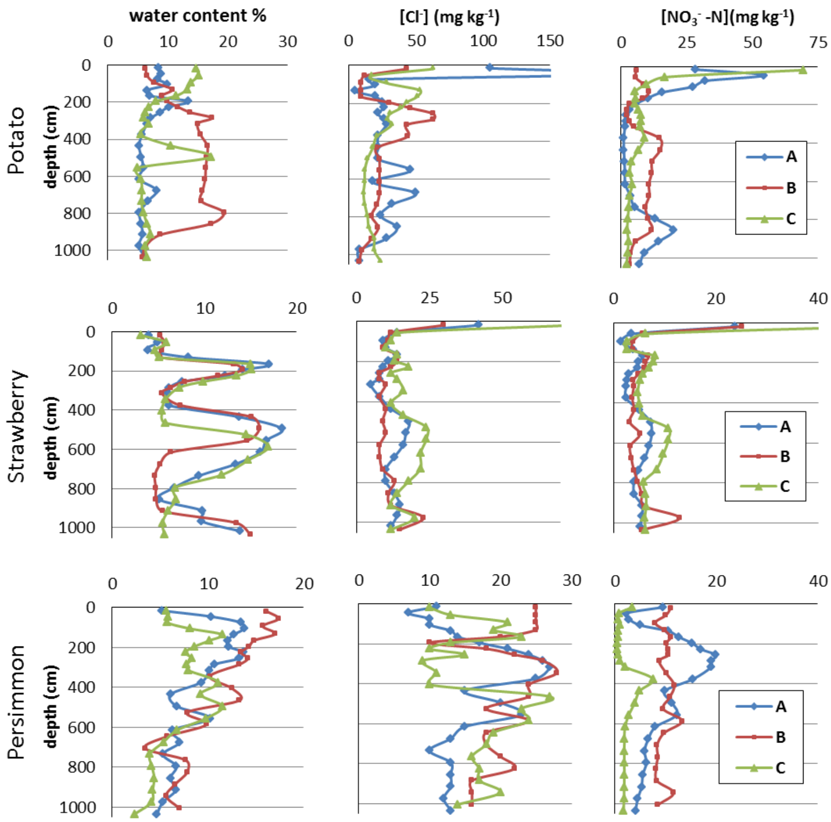

Figure 6: Gravimetric water content and concentrations of chloride and nitrate-nitrogen in the sediment profiles. Three sampling coreholes (A – blue, B – red, C – green) in each field (potato, strawberry and persimmon).

Table 1: Average deep (2–10 m) porewater concentrations and steady-state approximations of water and nitrate-nitrogen fluxes calculated for each profile.

	Potato			Strawberry			Persimmon		
	A	**B**	**C**	**A**	**B**	**C**	**A**	**B**	**C**
Pore water mean chloride concentration (mg L^{-1})	421	192	266	198	179	188	234	232	263
Pore water mean Nitrate-Nitrogen concentration (mg L^{-1})	96	63	63	47	53	76	130	25	38
Water recharge flux (mm yr^{-1})	208	457	330	359	397	378	421	424	370
Nitrate-Nitrogen flux (kg ha^{-1} yr^{-1})	200	290	210	170	210	290	540	110	140

3.1.2 Transient unsaturated zone flow and nitrogen transport models

Table 2a–2c present the hydraulic, transport and reaction model parameters that were calibrated to the observed unsaturated zone data. The partition coefficient for ammonium, k_{d-NH4} = 3.5 L kg^{-1} was used in all layers, and the first-order mineralization rate was set to μ_{min} = 0.56 day^{-1} (Hanson et al., 2006). The relation of nitrate-nitrogen uptake to root-zone concentration and water uptake ($f_{NO3}SC_{NO3}$) was of the form used by Kurtzman et al. (2013) with limiting nitrate-nitrogen concentrations of 45 mg L^{-1}, 35 mg L^{-1} and 20 mg L^{-1} for potato, strawberry and persimmon, respectively. Limitation of the nitrogen reactions to the top layers of the soil was based on previous work in which nitrification potential was analyzed in orchard soils from this region (Kurtzman et al., 2013).

Table 2. Measured clay content and parameters of the calibrated unsaturated zone flow and transport models under (a) potato field, (b) strawberry field, and (c) persimmon orchard. Note that in some layers hydraulic parameters were modified during calibrations (nd – no data).

(a)	Flow and transport parameters									Reaction parameters		
			Water content							Volatilization (NH_4), Nitrification, Denitrification		
Layer	Depth (m)	Clay (%)	Residual θr	Saturation θs	α (cm^{-1})	n	Saturate Hydraulic condictivity K (cm day^{-1})	Bulk density ρ (gr cm^{-3})	Disper-sivity (cm)	μ_{vol} (day^{-1})	μ_{nit} (day^{-1})	μ_{dnit} (day^{-1})
1	0-0.15	19	0.068	0.415	0.025	1.6	68	1.45	1.5	0.05	0.2	0.005
2	0.15-0.3	19	0.068	0.415	0.025	1.6	68	1.45	1.5	0	0.2	0
3	0.3-0.45	19	0.068	0.415	0.025	1.6	68	1.45	1.5	0	0.05	0
4	0.45-1.5	nd	0.058	0.420	0.031	3.1	675	1.45	10	0	0	0
5	1.5-4	11	0.065	0.445	0.028	1.8	165	1.46	25	0	0	0
6	4-5.2	4	0.057	0.409	0.031	3.3	775	1.43	12	0	0	0
7	5.2-9.4	5	0.057	0.406	0.031	3.3	766	1.6	38	0	0	0
8	9.4-10.15	2	0.068	0.415	0.025	1.6	68	1.57	9	0	0	0
9	10.15-10.3	nd	0.065	0.445	0.028	1.8	165	1.46	25	0	0	0

(b)			Flow and transport parameters							Reaction parameters		
Layer	Depth (m)	Clay (%)	Water content		α (cm^{-1})	n	Saturate hydraulic conductivity K ($cm\ day^{-1}$)	Bulk density ρ ($gr\ cm^{-3}$)	Disper-sivity (cm)	Volatilization (NH_4), Nitrification, Denitrification		
			Residual θr	Saturation θs						μ_{vol} (day^{-1})	μ_{nit} (day^{-1})	μ_{dnit} (day^{-1})
1	0 - 0.15	3	0.053	0.401	0.033	3.2	709	1.46	1.5	0.05	0.18	0.001
2	0.15 - 0.3	3	0.053	0.401	0.033	3.2	709	1.46	1.5	0	0.18	0.005
3	0.3 - 0.45	3	0.053	0.401	0.033	3.2	709	1.46	1.5	0	0.005	0
4	0.45 - 1.5	3	0.053	0.401	0.033	3.2	709	1.46	10.5	0	0	0
5	1.5 – 2.9	23	0.068	0.388	0.024	1.4	34	1.56	14	0	0	0
6	2.9 - 4.95	3	0.053	0.405	0.032	3.4	788	1.44	2	0	0	0
7	4.95 - 6.15	18	0.065	0.408	0.026	1.6	61	1.49	12	0	0	0
8	6.15 - 7	18	0.075	0.489	0.026	1.4	98	1.23	8.5	0	0	0
9	7 - 7.65	18	0.059	0.358	0.028	1.4	31	1.65	6.5	0	0	0
10	7.65 - 10.3	4	0.054	0.392	0.031	3.4	767	1.5	25	0	0	0

(c)			Flow and transport parameters							Reaction parameters		
Layer	Depth (m)	Clay (%)	Water content		α (cm^{-1})	n	Saturate hydraulic conductivity K ($cm\ day^{-1}$)	Bulk density ρ ($gr\ cm^{-3}$)	Disper-sivity (cm)	Volatilization (NH_4), Nitrification, Denitrification)		
			Residual θr	Saturation θs						μ_{vol} (day^{-1})	μ_{nit} (day^{-1})	μ_{dnit} (day^{-1})
1	0-0.15	12	0.06	0.404	0.028	2	159	1.48	1.5	0.08	0.1	0.0025
2	0.15-0.3	12	0.06	0.404	0.028	2	159	1.48	1.5	0	0.01	0.001
3	0.3-0.45	12	0.06	0.404	0.028	2	159	1.48	7	0	0	0
4	0.45-1.2	12	0.06	0.404	0.028	2	159	1.48	20	0	0	0
5	1.2-2.1	15	0.059	0.367	0.028	1.6	60	1.61	9	0	0	0
6	2.1-3.45	12	0.056	0.364	0.030	1.9	114	1.61	13.5	0	0	0
7	3.45-5.9	11	0.055	0.353	0.030	1.7	62	1.65	24	0	0	0
8	5.9-7.05	4	0.057	0.392	0.030	3.1	599	1.49	12	0	0	0
9	7.05-10.3	2	0.053	0.353	0.030	4.5	1357	1.5	32	0	0	0

Water and nitrogen balances resulting from the calibrated models showed significant recharge and deep nitrate-nitrogen leaching (40–55 % of total nitrogen input) under the investigated agricultural land (Table 3a and 3b). The yearly average (for 2002–2012) water and nitrate-nitrogen fluxes toward the water table calculated by the numerical models and those calculated by the steady-state approximation (chloride mass balance) matched well (Table 3a and 3b). The maximal difference between the two methods was 24 mm yr^{-1} (6.5 %) and 20 kg ha^{-1} yr^{-1} (7 %) for the water and nitrate-nitrogen fluxes, respectively. The average flux of nitrate-nitrogen toward the water table in citrus orchards in this area was found to be 30 % of the total nitrogen input (Kurtzman et al. 2013), lower than the leaching fraction under the vegetable and deciduous areas investigated here.

Table 3. Annual average (a) water and (b) nitrogen balance calculated by the unsaturated transient flow and transport models for 2002–2012, and comparison of deep fluxes to steady-state approximations. CMB - Chloride mass balance.

(a)		**Potato**	**Strawberry**	**Persimmon**
Average water input ($mm\ yr^{-1}$)	Irrigation	463	1050	822
	Rain	607	0	538
Average water output ($mm\ yr^{-1}$)	Root Uptake	467	367	639
	Evaporation	276	335	352
	Recharge	323	354	366
Recharge by CMB ($mm\ yr^{-1}$; wells C in Table 1)		330	378	370

(b)		**Potato**	**Strawberry**	**Persimmon**
Average nitrogen input ($Kg\ Ha^{-1}\ yr^{-1}$)	Fertilization	450	Mineral-350 Organic-100	200
	Nitrate-nitrogen in irrigation water	50	100	90
Average nitrogen output ($Kg\ Ha^{-1}\ yr^{-1}$)	Ammonia-volatilization	65	35	25
	Denitrification	65	75	35
	Root ammonium-nitrogen uptake	20	35	20
	Root nitrate-nitrogen uptake	165	125	110
	Nitrate-nitrogen flux toward groundwater	**200**	**310**	**130**
Nitrate-nitrogen flux toward groundwater by chloride mass balance ($Kg\ Ha^{-1}\ yr^{-1}$; wells C in Table 1)		210	290	140
Nitrate-Nitrogen leaching percentage		**40%**	**55%**	**45%**

3.2 Groundwater model

3.2.1 Model calibration

The flow model was calibrated by assigning different horizontal hydraulic conductivities, in the range of $K_{xx} = K_{yy} = 4.5 – 30$ m d^{-1}, to five subregions, where the higher values are in the western part of the modeled area. These hydraulic conductivity values are similar to previous studies in the Sharon region of the Israel coastal aquifer (Bachmat et al., 2003; Lutsky and Shalev, 2010). The calibrated anisotropy was $K_{xx}\ K_{zz}^{-1} = 5$ and the specific yield was Sy = 0.12.

The goodness-of-fit parameters between calculated and observed heads were the MAE and the mean error (the bias), calculated for each observation well (Table 4) and for all observations. The improvement in the calibration ceased when the target weighted-average MAE < 0.5 m and bias < 0.1 for all observations were achieved (Table 4, Fig. 7).

Table 4: Goodness of fit of the calibrated flow model (calculated–observed). MAE – mean absolute error; bias – mean error.

Well Name	# of observations	MAE (m)	Bias (m)
Tel Mond Ziv A	8	0.31	0.15
Tel Mond 8	6	0.40	0.21
Herut 41/3	20	0.48	0.29
Tel Mond 13	9	0.25	<0.01
Bnei Dror D	1	0.31	-0.31
Tel Izhak C	18	0.34	-0.31
Tel Izhak 41/2	20	0.61	0.49
Gan Efraim 3	27	0.72	-0.56
Gan Efraim 2	8	0.19	0.10
Gan Shlomo Berman-Cohen	6	0.45	0.45
Total observations and weighted-average errors	123	0.48	<0.01

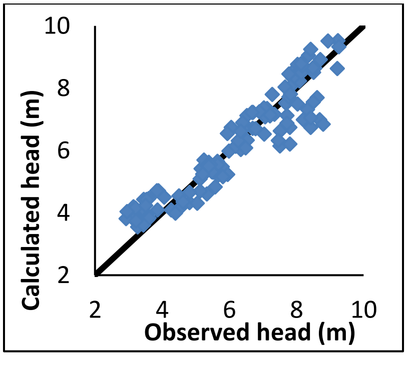

Figure 7: Calibrated flow model's calculated vs. observed heads in meters above mean sea level. Black line: calculated = observed.

The nitrate transport model was calibrated by changing the dispersivity value, starting with a value in line with Neuman's (1990) formula. The final transport parameters used in the calibrated model were: dispersivity = 500 m, ratio between longitudinal and transverse dispersivities = 10 and effective porosity = 0.12. This first stage of the calibration resulted in a good fit between observed and modeled mean nitrate concentration for the entire modeled area (i.e., spatially weighted average with weights for each well calculated by the Thiessen polygon method; Thiessen, 1911). However, the model showed poor fits between observed and calculated nitrate concentrations at each well separately (Table 5, Fig. 8a). This means that the model reconstructed well the entire mass of nitrate in the aquifer but it failed to describe the nitrate's spatial variability (bottom two lines in Table 5a vs. observed, averages and standard deviations). To test whether the nitrate inputs from the unsaturated-zone model are significant in comparison to nitrate flowing from the boundaries (variable-concentration boundary condition), the model was run with 0 nitrate flux from the unsaturated zone. The overall average nitrate concentration was 0.66 of the observed concentration (bottom two lines in Table 5b vs. observed, averages and standard deviations). These results led to the understanding that although the unsaturated model produces good values for overall nitrate flux, the contaminated wells cannot be modeled with fluxes resulting from “normal” agricultural practice. The meaning of this, is that nitrate spatial variability cannot be explained only by physical process of agricultural practice and land-use variability on surface. Other factors that are local and arbitrary, significantly affect nitrate concentration in some wells and therefore the measured spatial variability of nitrate in the aquifer. These factors were introduced into the numerical model as will be explained hereafter.

Simulations showed that observed nitrate concentrations above 100 mg L^{-1} cannot be simulated with the nitrate fluxes produced by the calibrated unsaturated zone model (Table 3b). Multiplication of fluxes by up to a factor of 10 was needed to produce high concentrations in the wells. On the other hand, we had to maintain the overall flux of nitrate over the entire model domain. Therefore, in the second stage of the calibration, nitrate fluxes that were calculated by the unsaturated zone model were multiplied by factors as follows: 1 % of the area – factor of 10 (near the most contaminated wells); 3 % of the area – factor of 5; 4 % – factor of 2.8; 55 % – factor of 1; 19 % – factor of 0.6 and in 18 % of the area, the fluxes were multiplied by a factor of 0.1. The reasoning and some physical explanations for these extreme fluxes in small areas surrounding some wells will be discussed later (in section 4). These local multipliers resulted in a reasonable fit between observed and modeled nitrate concentrations for both each well separately and the overall nitrate average and standard deviation (bottom two lines in Table 5c vs. observed).

Table 5: Observed vs. calculated nitrate concentrations during the calibration process. (a) After the first calibration stage (parameter fit). (b) A test model with 0 nitrate flux from the unsaturated zone. (c) After the second calibration stage (local multipliers). Avg. – average; MAE – mean absolute error; bias – mean error.

		Observed	(a) Model after 1st calibration			(b) Model without nitrate influx from unsaturated zone			(c) Model after 2nd calibration		
Well Name	# of observations	Avg. Value (mg L^{-1})	MAE (mg L^{-1})	Bias (mg L^{-1})	Calculated Avg. Value (mg L^{-1})	MAE (mg L^{-1})	Bias (mg L^{-1})	Calculated Avg. Value (mg L^{-1})	MAE (mg L^{-1})	Bias (mg L^{-1})	Calculated Avg. Value (mg L^{-1})
Bnei Dror D	14	20	48	-48	68	20.8	-20.1	40.1	8	2	18
Tel Mond 5	10	51	13	-12	64	6.8	6.6	44.5	4	-3	54
Tel Mond 8	31	53	15	-2	55	14.1	10.2	42.7	14	1	52
Herut 6	24	54	15	-15	69	12.2	12.2	41.9	10	-10	64
Tel Mond Ziv A	9	59	14	-14	73	4.1	3.6	55.7	5	-4	63
Tel Izhak C	13	61	15	-13	73	17.5	17.5	43.0	8	7	53
Gan Efraim 4	13	65	13	-10	75	13.0	12.0	53.1	11	-3	68
Tel Mond 13	17	66	9	4	63	24.9	24.3	42.2	11	-1	67
Gan Shlomo Man	10	70	11	-6	76	19.1	16.0	54.0	13	-9	79
Gan Shlomo Berman	15	75	10	-0.3	75	22.4	22.4	52.6	10	-2	77
Gan Efraim 2	13	87	12	10	77	40.4	40.4	46.6	11	-4	91
Gan Efraim Lapter	14	101	30	30	70	54.4	54.4	46.0	12	3	97
Gan Shlomo A	11	115	26	26	90	38.5	38.5	76.7	15	8	107
Tel mond 3A	14	130	59	59	72	86.7	86.7	43.7	19	12	119
All Wells	208	**73**	20	0.6	**72**	27	24.5	**48**	10	0.3	**72**
Standard Deviation		**27**			**8**			**9**			**25**

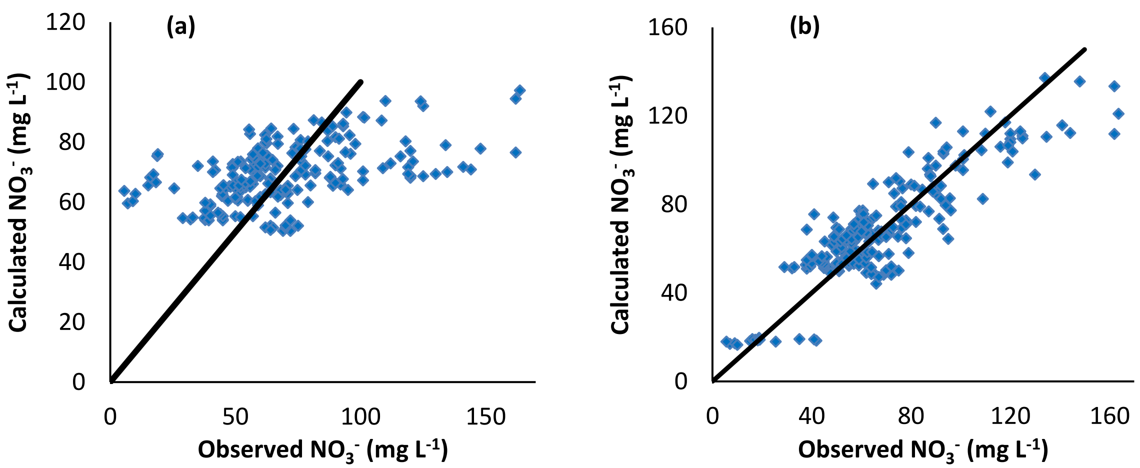

Figure 8: Calculated vs. observed nitrate concentrations. (a) After the first calibration stage (parameter fit). (b) After the second calibration stage (local multipliers). Black line is calculated = observed.

3.2.2 Simulations of three fertilization scenarios 40 years into the future

The calibrated model was run to 40 years in the future (2012–2052) under three scenarios: (i) "business as usual"; (ii) application of 75 % of the currently applied nitrogen fertilization; (iii) application of 50 % of the currently applied nitrogen fertilization. The simulation results showed that (i) the average concentration in all wells in the simulated area will continue to increase in the "business as usual" scenario, reaching 106 mg L^{-1} in 2052 (vs. 87 mg L^{-1} in 2012); (ii) reducing the fertilization to 75 % will approximately maintain the present concentrations; (iii) reducing the fertilization to 50 % will lead to a trend of declining nitrate concentration to less than 70 mg L^{-1} (Israel's drinking water standard for nitrate) as an average for all wells in the modeled area (Fig. 9 and Table 6). Even in this case about half of the wells will still exceed the standard concentration.

Table 6: Current (2012) observed nitrate concentrations and those simulated for the year 2052 for three nitrogen-fertilization scenarios: 100 %, 75 % and 50 % of the current application used by farmers. In red are concentrations below the Israeli drinking water standard for nitrate.

Well Name	**Observed (2012, mg L^{-1})**	**Simulated concentrations at 2052 (mg L^{-1}) for fertilization scenario**		
		100 %	**75 %**	**50 %**
Bnei Dror D	16	27	21	19
Tel Mond 5	60	77	64	57
Tel Mond 8	60	82	68	61
Herut 6	69	79	67	60
Tel Mond Ziv A	70	83	68	59
Tel Izhak C	73	96	73	62
Gan Efraim 4	79	101	77	66
Tel Mond 13	78	99	80	71
Gan Shlomo Man	88	107	84	73
Gan Shlomo Berman-Cohen	90	109	86	75
Gan Efraim 2	106	138	101	84
Gan Efraim Lapter	122	139	98	78
Gan Shlomo A	128	130	103	89
Gan Efraim 3	129	157	108	86
Tel mond 3A	134	164	112	88
Average	**87**	**106**	**81**	**69**
Standard Deviation	26.7	29.1	16.5	11.5

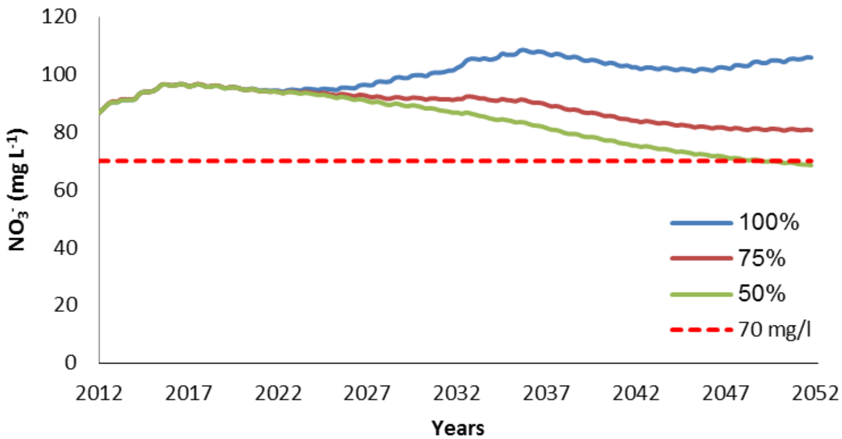

Figure 9: Simulations of future average nitrate concentrations in wells under three nitrogen-fertilization scenarios: 100 %, 75 % and 50 % of the current application used by farmers. 70 mg L^{-1} – Israel's drinking water standard for nitrate.

4 Discussion

Our results showed successful evaluation of the total mass of nitrate in the aquifer using data of agricultural practice and deep unsaturated-zone samples to calibrate flow and transport models of the unsaturated zone, which feed the aquifer. Nevertheless, this straightforward model failed to produce the observed spatial variability of nitrate concentrations in wells, which required a random non-mechanistic modeling approach.

Successful delivery of the total volumes of water and nitrate mass to the 13.3 km^2 aquifer under agricultural land was achieved despite the following first-order assumptions: only four types of land use (three crops); steady crops for 50 years; homogeneity of agricultural practices and similar profiles of porous medium within each crop. These assumptions neglect small-scale variability, yet work for the regional scale totals for the following reasons: the farmers generally follow irrigation and fertilization recommendations made by extension services; about half of the land is covered by orchards for which applications of water and fertilizer have been steady for decades; on a regional scale, if the soil properties are generally similar, the details of the different profiles of the deep unsaturated zone have only a minor effect.

Failure to reproduce the spatial variability of nitrate concentrations lay mainly in predicting the extreme concentrations in some wells. These nitrate concentrations cannot be explained by any rational agricultural practice, and are a result of random failure of even fertilizer distribution in the field that can be due to one or more of the following reasons. It should be acknowledged that water wells are often at the "logistic center" of the agricultural field, and organic and mineral fertilizers are stocked nearby; temporal leakage can cause high concentrations in the well for years afterwards. Furthermore, the immediate area of the well is susceptible to preferential flow paths due to incidental ponding (Gurdak et al., 2008) and/or shortcuts through the annulus of the boreholes. This is especially common in old private boreholes that are used mainly for irrigation, which are common in the investigated area. Heterogeneity of the porous medium may cause extremely high nitrate fluxes likewise well failure discussed previously, and may be a source for local high contamination. The field survey reported here support this statement. Of the nine deep profiles reported here (Figure 6, Table 1), one showed extreme nitrate concentrations and calculated nitrate fluxes that were 4- to 5-fold higher than in the other profiles extracted from the same orchard (Persimmon A, Table 1). Multipliers that adjust nitrate fluxes to groundwater were used in models previously (e.g. Alikhani et al., 2016). The two orders of magnitude difference in nitrate multipliers (0.1 – 10) used in this work is not incomparable, Kourakos et al. (2012) used distributions with means of 1 to 100 mg L^{-1} nitrate, loading to groundwater for the same land-use. Therefore, the heuristic multiplications used to calibrate the nitrate transport model were ultimately justified. Moreover, these multiplications were essential for simulating future scenarios (Fig. 9, Table 6).

The workflow in this study did not include model validation after each calibration stage (i.e. for each land-use unsaturated: flow, conservative-transport, and nitrogen reactive-transport; groundwater flow and groundwater transport), which is of course a disadvantage. Validation tests for each calibration would have given a statistical measure of the goodness-of-fit of the calibrated model with independent (left-out of calibration) observations, rather than only the calibration fit. Even though, the conclusions of this work are highly significant because they are based on entirely independent data. The total mass of nitrate that crossed the water-table (unsaturated zone models) is verified by the groundwater well data. Whereas the failure to reproduce the spatial variability would have not been changed with validation fit estimates.

In the case of the Israeli coastal aquifer, we are fortunate enough to be able to perform a post-audit analysis of nitrate-level predictions made 40 years ago in another part of the aquifer (Rehovot-Rishon region, Fig. 1). This region of the aquifer was overlain mainly by agricultural land (in 1950–1970), with similar sandy-loam (Hamra) soils (Mercado, 1976). The latter work predicted a continual increase in nitrate concentration in the groundwater below this area, from 50 mg L^{-1} in 1970 to a range of 120–180 mg L^{-1} in 2015 (Fig. 10). The observed average concentration in this area in 2014 was 90 mg L^{-1} (Israel Water Authority data). This is indeed an increase, but not the expected one. On the other hand, this increase of 40 mg L^{-1} over 45 years is similar to the nitrate concentration increase in the Sharon area (Kurtzman et al., 2013).

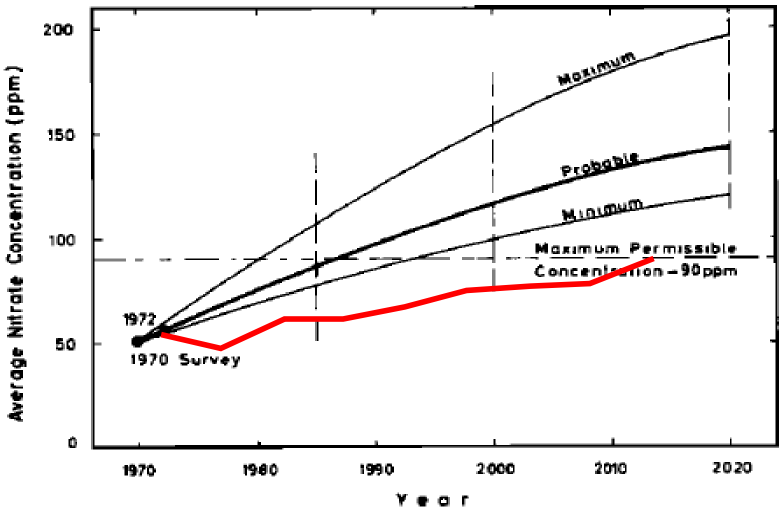

Figure 10: Post-audit of average nitrate concentration predicted in 1976 for another part of the Israeli coastal aquifer. All black lines and writing are original predictions from Mercado (1976). Red line is the historical average nitrate concentrations from the wells in that area that produced since 1970 (no new wells, data were obtained from the Israel Water Authority. Maximum permissible concentration of nitrate was reduced from 90 mg L^{-1} to 70 mg L^{-1} in 2001).

The main reason for the overshoot of Mercado's (1976) prediction is probably the very significant reduction in agricultural land due to urbanization in this area in the last 5 decades. Most of this urbanization are agricultural towns which became modern cities with tight sewage systems, where practically all the wastewater is piped to treatment plants. In the current work, the predictions were also made assuming steady agricultural land use with no urbanization processes that might lead to a similar overshoot in nitrate concentration predictions.

5 Summary and Conclusions

Groundwater under irrigated agricultural land over light soils commonly suffers from nitrate contamination. Nevertheless, significant spatial variability in nitrate concentrations in these parts of the aquifer exist, suggesting that it is caused by variability in nitrate fluxes from the unsaturated zone. An agricultural area (13.3 km^2) in the Sharon region overlying the Israeli coastal aquifer in which the abovementioned phenomena are observed was selected to investigate the process through calibrated flow and nitrate transport models from the agricultural land surface to the well screens (15 to 130 m below the surface). Unsaturated flow and nitrogen species transport models were calibrated to data from below the root zone that were obtained with direct push sampling under four typical crops in the area: citrus, persimmon, potato and strawberry. The flow and nitrate transport model in the aquifer was fed from water and nitrate fluxes from the unsaturated models, and calibrated to water levels and nitrate concentrations in the wells. The agricultural data and the flow and transport models of the unsaturated zone successfully predicted the total mass of nitrate in the aquifer. However, they failed to predict the spatial variability of nitrate in the wells, which was observed to be significantly larger than predicted. Therefore, the solution for calibrating the nitrate transport model was to multiply the modeled nitrate fluxes at the water table in small areas around the most contaminated wells with high multipliers (2.8–10), whereas nitrate fluxes in larger areas around the non-contaminated wells were multiplied by low factors (0.1–0.6) and in most of the area (55 %), the modeled fluxes from the unsaturated zone were conserved. The calibrated flow and transport model was then used to predict the development of nitrate concentrations in the aquifer 40 years in the future, with three nitrate-fertilization scenarios: business as usual (continuing present practice), or reducing nitrogen inputs by 25 % or 50 %. None of the scenarios showed any improvement in aquifer conditions in the next 10 years. Reducing nitrate application by 50 % will bring the average nitrate concentration in the aquifer to below drinking water standards in 40 years, whereas a cut of 25 % will only bring it back to the current level in 40 years. We conclude that the total mass of nitrate in an aquifer under agricultural land can be calculated with significant success from relatively limited land-use and deep unsaturated-zone data. Nevertheless, highly contaminated wells, are most probably effected by malfunction in the close vicinity of the well that cannot be predicted by a straight-forward agro-hydrological modeling scheme. Locally, it was shown that remediation of the aquifer in a half-century time scale requires reduction of the nitrogen fertilization input in the range of 25 % -50 %.

Competing interests. The authors declare that they have no conflict of interest.

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