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Nitrogen fate during agricultural managed aquifer recharge: linking plant

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response, hydrologic, and geochemical processes

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12 Abstract

13 Agricultural managed aquifer recharge (Ag-MAR, on-farm recharge), where farmland is 14 flooded with excess surface water to intentionally recharge groundwater, has received increasing 15 attention by policy makers and researchers in recent years. However, there remain concerns 16 about the potential for Ag-MAR to exacerbate nitrate (NO_3) contamination of groundwater, and 17 additional risks, such as greenhouse gas emissions and crop tolerance to prolonged flooding. 18 Here, we conducted a large-scale, replicated winter groundwater recharge experiment to quantify 19 the effect of Ag-MAR on soil N biogeochemical transformations, potential NO₃⁻ leaching to 20 groundwater, soil physico-chemical conditions, and crop yield. The field experiment was 21 conducted in two grapevine vineyards in the Central Valley of California, which were each 22 flooded for 2 weeks and 4 weeks, respectively, with 1.31 and 1.32 m³ m⁻² of water. Hydrologic, 23 geochemical, and microbial results indicate that NO_3^- leaching from the first 1 m of the vadose 24 zone was the dominant N loss pathway during flooding. Based on pore water sample and N₂O 25 emission data denitrification played a lesser role in decreasing NO_3^- in the root zone but 26 prolonged anoxic conditions resulted in a significant 29% yield decrease in the 4-week flooded 27 vineyard. The results from this research, combined with data from previous studies, are 28 summarized in a new conceptual model for integrated water-N dynamics under Ag-MAR. The 29 proposed model can be used to determine best Ag-MAR management practices.

30 Keywords: MAR, groundwater recharge, nitrate, denitrification, soil, crop tolerance

31 1. Introduction

One-quarter of the world's population and 40% of agricultural production relies on overdrafted groundwater sources (Connor, 2015), with an expected increase in groundwater reliance due to climate change and growing water demand (Gorelick and Zheng, 2015; Haddeland et al., 2014; Siebert et al., 2010; Wada et al., 2010). Thus, reducing pressure on overdrafted groundwater systems is crucial to increase global resilience of food and drinking water in response to growing human population and climate change pressures.

38 Agricultural managed aquifer recharge (Ag-MAR, or on-farm recharge) is a form of managed 39 aquifer recharge (MAR) where farmland is flooded with excess surface water to recharge 40 groundwater intentionally (Grinshpan et al., 2021; Kocis and Dahlke, 2017; Waterhouse et al., 41 2020) and it has been increasingly used across the globe to address groundwater overdraft 42 (Dillon et al., 2019; Levintal et al., 2022). The purpose of Ag-MAR, in comparison to more 43 traditional MAR methods, is to transfer large amounts of surplus surface water from rivers or 44 reservoirs to agricultural land (e.g., idle land, agricultural fields and orchards) that serve as 45 spreading grounds for the recharge (Dahlke et al., 2018).

46 Ag-MAR adoption has increased in recent years, particularly in the USA and Europe 47 (Grinshpan et al., 2021; Negri et al., 2020; Niswonger et al., 2017). In California, for example, 48 Ag-MAR is implemented as one of the methods to overcome ongoing groundwater depletion 49 (Kocis and Dahlke, 2017). However, using farmland as spreading grounds bears the risk to leach 50 contaminants from the water or soil to groundwater which can impact drinking water quality 51 (Bachand et al., 2014), waterlogging of the root zone for long periods that can reduce crop health 52 (Ganot and Dahlke, 2021a), and ecosystem service tradeoffs, such as short and long-term effects 53 on in-stream flows (Levintal et al., 2022). Out of the above, leaching of legacy nitrogen (N), 54 mainly in the form of nitrate (NO_3) , is possibly the most widespread environmental risk of Ag-55 MAR. Consumption of drinking water above the maximum contaminant level (MCL; 10 mg L⁻¹ 56 NO₃-N in California) can increase the risk of cancers, birth defects, and other adverse health 57 effects (Weitzman et al., 2022). Globally, nitrate is the primary nonpoint source pollutant of 58 groundwater (Beganskas et al., 2018; Bishayee et al., 2022; Richa et al., 2022), whereby 59 agricultural lands serve as the main source for NO_3^- due to the buildup of legacy NO_3^- resulting 60 from years of fertilizer use inefficiencies (Van Meter et al., 2016; Waterhouse et al., 2020).

61 In comparison to the deep vadose zone, NO_3^{-1} concentrations are highest either in the topsoil (0-62 10 cm) or just below the root zone (~1-2 m) where NO_3^{-1} is transported out of reach of roots with 63 irrigation (Waterhouse et al., 2021, 2020). NO₃⁻ in soils can originate from N-based fertilizer or 64 from N transformations, such as nitrification of ammonium (NH₄⁺)-based fertilizer or nitrification 65 of NH₄⁺ from mineralized soil organic-N (Stein and Klotz, 2016). In contrast, NO₃⁻ removal 66 pathways are denitrification and immobilization (controlled by microbial activity), leaching 67 (controlled by infiltration rate), and plant uptake (Kurtzman et al., 2021; Long et al., 2013; 68 Schmidt et al., 2011; Zhang et al., 2018). The governing processes during Ag-MAR are NO₃⁻ 69 leaching, mineralization, nitrification, and denitrification (Murphy et al., 2021; Schmidt et al., 70 2012; Waterhouse et al., 2021). The latter is favored under suboxic to anoxic conditions, as 71 expected during soil saturation of Ag-MAR events (Ganot and Dahlke, 2021b). N transformation 72 processes, excluding leaching, occur in the soil dependent on electron donor availability (e.g., 73 dissolved organic carbon (DOC)) and microbial community abundance and composition

74 (Peterson et al., 2013; Scarlett et al., 2021). Ambient conditions may also influence NO_3^{-1} 75 transformations, including soil moisture, carbon/nitrogen ratio, pH, soil texture, temperature, 76 vegetation, oxidation-reduction potential (ORP), and water residence time in the topsoil (Kraft et 77 al., 2014; Stein and Klotz, 2016). The underlying assumption adopted by previous researchers is 78 that NO_3^- -related processes are negligible below the root zone; therefore, NO_3^- leached below the 79 root zone will eventually reach the groundwater (Baram et al., 2016b; Gurevich et al., 2021). 80 Thus, in Ag-MAR, the aim is to reduce legacy NO_3^- via denitrification before NO_3^- leaching 81 occurs below the root zone.

NO₃⁻ leaching is an environmental risk that is not unique to Ag-MAR, but can occur in any MAR method (e.g., infiltration basins; Beganskas et al., 2018), since it mainly depends on the source of NO₃⁻ in the recharge environment. For example, Beganskas et al. (2018), Gorski et al. (2019), and Schmidt et al. (2011) recharged stormwater runoff from upslope agricultural fields in an infiltration basin in the Pajaro Valley, California, which contained 22-25 mg L⁻¹ NO₃⁻-N. If low-N source water is used in Ag-MAR (e.g., mountain runoff or snowmelt), elevated NO₃⁻ in soil pore water often originates from the soil matrix from recurring fertilizer applications.

Only a few studies exist to date that have estimated soil NO_3^- biogeochemical transformations and NO_3^- leaching under Ag-MAR. As one of the first, Bachand et al. (2014) investigated $NO_3^$ fate in a large farm-scale experiment where they flooded alfalfa, wine grapes, tomatoes, and fallow land for various periods ranging from 10 days to one month. They proposed a general framework for NO_3^- fate under Ag-MAR, concluding that NO_3^- will be leached to groundwater, but pore water and groundwater NO_3^- concentrations will be diluted following consecutive flood applications (assuming the use of low- NO_3^- water). However, they did not investigate any N 96 transformations. Waterhouse et al. (2020) estimated the potential risk of NO₃⁻ leaching under Ag97 MAR using only data from 10 m deep soil cores from 12 fields (no flooding was performed),
98 focusing on three crops (almonds, processing tomatoes, and wine grapes) and two soil groups
99 (low- and high-infiltrating soils). They concluded that vineyards were the most suitable crop for
100 Ag-MAR due to low legacy NO₃⁻ resulting from the crop's deep roots and overall low N inputs.

101 Murphy et al. (2021) investigated the role of flooding frequency (i.e., three flooding events of 102 0.15 m depth each) and soil texture on N dynamics and potential NO_3^{-1} leaching in laboratory 103 column experiments. Each flooding event was on the scale of hours followed by a drying period 104 of several days to two weeks. They observed that the majority of initially present soil NO_3^{-1} 105 leached during the first few hours of the first water application when conditions for 106 denitrification (i.e., removal of NO_3^{-1}) were unfavorable due to oxic conditions. NO_3^{-1} leaching was 107 quantified only for the laboratory column experiments ranging between 0.028 and 0.072 g NO_3 -108 N m⁻² for every 1 cm³ cm⁻² of applied water. Based on the combination of lab assay and field data 109 Murphy et al. (2021) showed that using only soil core data from pre- and post-flooding is 110 insufficient to quantify NO_3^{-1} leaching, emphasizing the need for continuous field measurements 111 during Ag-MAR.

In a recent modeling study, Waterhouse et al. (2021) investigated Ag-MAR effects on denitrification rates and NO_3^- leaching in a heterogeneous, layered deep vadose zone (~15 m). They found that denitrification rates were highest in response to one continuous extensive water application due to the development of suboxic conditions compared to small incremental recharge events. However, this continuous water application scenario also leached NO_3^- deeper into the vadose zone. None of the above studies measured N-related biogeochemical processes 118 during continuous, long (e.g., several weeks) flooding applications for Ag-MAR in different 119 soils. This suggests that understanding of N fate and NO_3^- leaching at the field scale is still 120 elusive in Ag-MAR; thus, further research is needed.

121 This study aims to investigate the environmental impacts of Ag-MAR using a large-scale field 122 experiment conducted in the Central Valley, California (semiarid Mediterranean climate). Two 123 vineyards, each with a different soil texture, were simultaneously flooded during late winter, one 124 for four weeks (V1) and the other for two weeks (V2). Combining hydrologic, geochemical, and 125 microbial process analyses, we quantified the effects of Ag-MAR on the soil N biogeochemical 126 transformations, potential NO_3^{-1} leaching to groundwater, soil physico-chemical conditions, and 127 crop response. The results from this research, combined with data from previous studies, are 128 summarized in a new conceptual model for integrated water-N dynamics.

129 2. Materials and Methods

130 2.1. Description of site, water application, and measurements

131 A replicated field experiment was conducted at the Kearney Agricultural Research and 132 Extension Center (http://kare.ucanr.edu/) located 20 km southeast of the city of Fresno in the 133 Central Valley of California, USA. Two mature (>40-year-old) own-rooted 'Thompson Seedless' 134 grapevine (Vitis vinifera L.) vineyards were flooded; the first vineyard for four weeks (V1) and 135 the second vineyard for two weeks (V2) (February-March 2020). V1 is on a very deep fine 136 sandy loam (Hesperia, coarse-loamy, mixed, nonacid, thermic Xeric Torriorthents) with a 137 saturated hydraulic conductivity (K_{sat}) of 0.02 m hr⁻¹ for the first 1 m depth (USDA-NCSS soil 138 survey data), and V2 is on a fine sandy loam (*Hanford*) with a K_{sat} of 0.1 m hr⁻¹ for the first 1 m depth. Using a randomized complete block design, each vineyard was divided into six individual subplots, of which three were artificially flooded and three were kept under the natural precipitation regime (i.e., control) (**Fig. 1a**). Individual subplots were separated from one another using berms approximately 0.5 m high and 0.8 m wide (see **Fig. 1b**). The groundwater table was ~23 m below the surface measured in October 2019. The site has a semiarid, Mediterranean climate (Onsoy et al., 2005). Mean air temperature and total precipitation during the flooding period was 21.9 °C and 0.03 m, respectively.

146 Flooding started automatically at 06:00, 14:00, and 22:00 for 2-3 hrs at both vineyards each day, 147 except for the first four days during which manual operation was used to adjust flow rates to 148 prevent overflow to adjacent fields. Flooding of the vineyards started on 02/25/2020 and ended 149 on 03/10/2020 (V2) and 03/24/2020 (V1). Two flow meters were used at the water inlet point of 150 each vineyard to measure total applied water. Flooding was done using a single water outlet 151 located at the west side of each row within the flooded subplots. A total of 7298 and 4659 m³ 152 were discharged at V1 and V2, respectively. Commonly Ag-MAR would be conducted using 153 surface water. However, due to drought during the winter of 2019-2020 no surface water was 154 available at our experimental site, therefore, pumped groundwater was used as an alternative 155 water source for Ag-MAR. Groundwater was determined as a suitable alternative to surface 156 water because N species concentrations in groundwater were found to be comparable to those of 157 surface water in this area, as previously reported by Bachand et al. (2014).

158 Ten monitoring profiles were instrumented for continuous measurements, five within each 159 vineyard (Fig. 1a). Out of the five profiles in each vineyard, only one profile was installed in the 160 control (i.e., the non-flooded subplots) to allow higher spatial measurement resolution in the Ag161 MAR treatment subplots, which are the focus of this study. Each profile was installed 0.7 m from 162 the vine row toward the furrow and included three sensor clusters at 0.2, 0.6 and 1 m depth. Each 163 cluster included measurements of soil moisture and temperature (TEROS12, Meter, WA, USA), 164 O₂ in gaseous phase (KE-25, Figaro, Japan), and ORP using constructed platinum electrodes 165 (following Owens et al., 2005) and commercial Ag/AgCl reference electrodes (Accumet, Thermo 166 Fisher Scientific, MA, USA). Additional measurements included three pressure transducers 167 within stilling wells to record ponding levels (CS451, Campbell Scientific, UT, USA). Sensors 168 were placed in hand augered holes that were backfilled with soil while compacting it at 169 approximately 0.2 m intervals. Data were logged (CR1000 and CR800, Campbell Scientific, UT, 170 USA) at a 10-min interval. The complete site and sensor descriptions are detailed in **Figs. 1a** and 171 1c, respectively, and a photo taken during the flooding is presented in Fig. 1b. Atmospheric 172 measurements (temperature, precipitation) at 60-min intervals were taken from the California 173 Irrigation Management Information System station (CIMIS; station 39 Parlier, CA) situated 400 174 m from the experimental site.

175 Crop management practices followed standard recommendations with grape harvest in 176 September and cane pruning on 20 January 2020. Both vineyards were fertilized once per year, 177 two months after the flooding (end of April 2020), when 336 kg ha⁻¹ of N fertilizer (ammonium 178 sulfate 21-0-0 with 24% sulfur) were applied. Vines were manually harvested on 23 September 179 2020. The entire amount of fruit harvested from each plot was recorded and used for statistical 180 analyses.

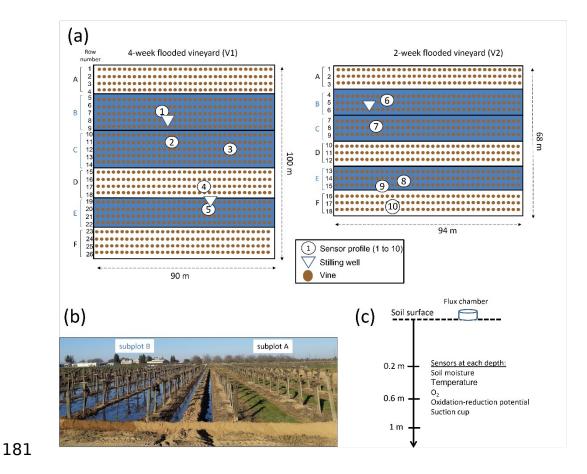


Fig. 1. Experimental setup. (a) Sensor locations in each vineyard. Flooded subplots are marked
in blue. (b) A photo of the 4-week flooded vineyard (V1) showing subplots A and B during the
flooding. (c) Sensor locations within a single profile.

185 2.2. Pore water, gas emission, and soil sampling

Pore water, greenhouse gas (GHG) emission, and soil sampling campaigns were carried out to better understand and quantify the N biogeochemical processes within the soil. Pore water samples were taken using suction cups (LT-DBL, Irrometer, CA, USA), installed at each of the ten profiles and at the same depths as the sensor clusters (0.2, 0.6, and 1 m). After installation, suction cup boreholes were backfilled with sieved soil slurry, followed by 0.15 m of soil, and sealed with 0.05 m bentonite; shallow 0.2 m suction cups were installed without bentonite. Pore water was sampled on average twice a week during the flooding period and once a week at other times, except for one continuous 5-day measurement campaign after fertilization to capture the fertilization effect on N processes. Samples were stored on ice until storage in a 4 °C cold room. Pore water was analyzed for NO_{3}^{-} , NH_{4}^{+} , and DOC. Analytical protocols are detailed in the supporting information.

197 In-situ GHG emissions were measured using the closed-flux vented chamber method (e.g., 198 Garland et al., 2014). Twenty individual chambers were installed, each made from 0.26 m 199 diameter polyvinyl chloride (PVC) pipe, consisting of a bottom ring and a cap. Two chambers 200 were installed near each of the ten sensor profiles shown in **Fig. 1a** – one on the mount between 201 the vines and one in the furrow. Collars were inserted into the soil to a depth of 0.07 m and left 202 in the same location for the entire duration of the experiment. Chamber headspace was measured 203 after each sampling to account for any reduction in volume due to water within the chamber. At 204 sampling time, the chambers were sealed onto the collars with a rubber sleeve made from a tire 205 innertube. Gas samples were taken through a rubber septum at four times (0, 30, 60, and 80 min) 206 using a 20 mL air-tight syringe and injected into pre-evacuated 12 mL vials. Before each 207 sampling effort, a 12V fan (installed within each chamber) was activated for 20-30 seconds to 208 ensure a mixed representative headspace sample. Temperature was measured during each time 209 step in 10 out of the 20 chambers. Samples were analyzed using a gas chromatograph (Shimadzu 210 Trace Gas GC, Shimadzu Corp., Kyoto, Japan) for carbon dioxide (CO_2), methane (CH_4), and 211 nitrous oxide (N_2O). Fluxes were calculated according to Garland et al. (2011). Gas samples 212 were taken on similar days as the water samples and mostly around noon.

213 Soil samples were taken from four depths: 0-0.1, 0.1-0.2, 0.5-0.6, 0.8-1 m using a hand auger. 214 All samples were taken from the same rows in which the sensor profiles were installed and at 215 similar distances from the vines toward the furrow (~ 0.7 m). Samples were stored identical to 216 pore water samples and taken during four campaigns according to the different experimental 217 stages: 02/06/2020 (baseline data), 03/10/2020 (end of flooding at V2), 03/25/2020 (end of 218 flooding at V1, and two weeks post-flooding at V2), and 04/15/2020 (three weeks post-flooding 219 at V1). Soil samples were analyzed for NO₃, NH₄⁺, DOC, and texture. In addition, soil 220 subsamples were stored at -80 °C and later used for incubation experiments (see details in 221 section 2.4).

222 2.3. Nitrogen leaching estimates

The amount of NO_3^- leaching below 1 m (the root zone in the case of flooded-irrigated vineyards) during flooding was quantified using a vadose-zone-based water and N mass balance model (Baram et al., 2016b), in which M_A is the cumulative mass of NO_3^- per flooded area (as $NO_3^--N \text{ [g m}^{-2}\text{]}$) lost through leaching (**Eq. 1**) and *M* is the total mass of NO_3^- (as $NO_3^--N \text{ [g]}$) lost during the flooding (**Eq. 2**):

228
$$M_A = \sum_{i=1}^n L_i C_i \Delta t_i$$
(1)

229
$$M = \sum_{i=1}^{n} M_{A_i} A$$
 (2)

where *L* is the amount of water leaching below 1 m estimated using the infiltration rate (section 2.5) [m d⁻¹], *C* is the average NO₃⁻-N concentration in the leaching water at 1 m depth [g m⁻³], Δt is a given time period [d] between each *i*th measurement, and *A* is the flooded surface area [m²]. Because all pore water NH_4^+ samples measurements at both vineyards were < 1 g m⁻³, these values were considered negligible in the quantification of total inorganic N leached for modeling purposes.

236 2.4. Potential denitrification

237 Incubation experiments were conducted in the lab to assess denitrification rates during the 238 flooding period. Net denitrification rates were estimated using a modified method of Petersen et 239 al. (2012). Briefly, field-moist soil (15 g) collected one day before flooding and one day after 240 flooding was added to 100 ml serum bottle and sealed with rubber stopper and metal caps, and 241 then flushed with N₂ gas for 10 min to create anaerobic conditions and afterwards equilibrated 242 with atmospheric pressure using a syringe. Bottles were placed in an incubator for 7 days at 243 25°C. Incubated samples were analyzed for changes in NO₃⁻ concentrations on the initial and 7th 244 day to determine denitrification rates.

245 2.5. Hydrological analysis

Daily infiltration rates were calculated using two independent methods. In the first method, the total applied water was divided by the flooded surface area and total number of flooding days for each vineyard. The second method, also referred to as the falling head method, estimates the infiltration rate from the slope of the decreasing ponding level in each flood plot after water shutoff, which is then normalized for daily infiltration rates. Both methods provide bulk infiltration rates.

252 Groundwater recharge was calculated using a one-dimensional vertical water balance model253 (Eq. 1) (Dahlke et al., 2018). A single solution was solved for each of the eight flooded

monitoring profiles using input data averaged over an hourly interval. The fraction of applied
water going to deep percolation towards the groundwater table was calculated by accounting for
evapotranspiration and storage in pore space.

$$257 R_t = I_t + P_t - ET_t - \Delta S_t - Q_t (3)$$

where R_t [m] is recharge, I_t [m] is the amount of applied surface water, P_t [m] is precipitation, ET_t [m] is evapotranspiration, ΔS_t [m] is the change in soil storage, and Q_t [m] is surface runoff at time step t, which was assume to be negligible due to the use of berms. Eq. 3 was solved using the Thornthwaite-Mather procedure (Steenhuis and Molen, 1986) performed via Excel solver (Office 365 ProPlus, 2020). This procedure was used in previous Ag-MAR studies (Dahlke et al. 2018). The procedure was solved for the first meter assuming this is also the maximum depth of the major root zone (Araujo et al., 1995) in which evapotranspiration demand takes place.

265 **3.** Results

266 3.1. Recharge and soil physical characteristics

267 The flood treatments in the two vineyards showed different hydrologic and biogeochemical 268 responses during and after the flooding for recharge (V1 - Fig. 2 and V2 – Fig. 3). As a result of 269 field topography, flooding was not uniform in the flooded subplots. Dry areas occurred in the 270 eastern part of V1 (Fig. 1a; subplot C) and V2 (Fig. 1a; subplots C and E), effectively reducing 271 the flooded area to an average of 4613 m² (90%) and 3035 m² (75%) in V1 and V2, respectively. 272 Soil ORP and O_2 concentration decreased rapidly in V1, reaching anoxic conditions (ORP < 100 273 Eh) at 0.2, 0.6 and 1 m depth 1-3 days after flooding started (Figs. 2c and 2d). Reducing, anoxic 274 conditions were sustained for 12-18 days after flooding ended, leading to ~40 days of continuous

reducing conditions within the soil profile in V1. In contrast to V1, V2 maintained good aeration during the flooding and only experienced anoxic conditions at the shallowest depth of 0.2 m and for a short duration (2-5 days) (**Figs. 3c** and **3d**). O₂ and ORP averaged ~20% O₂ and ~400 Eh ORP at all other depths within the flooded soil profiles in V2. Although flooded plots at both vineyards reached near or fully saturated conditions as indicated by the soil moisture (**Figs. 2b** and **3b**), the observed differences in soil O₂ and ORP clearly indicate different environmental conditions that impacted biogeochemical processes in each vineyard.

As expected, control plots of both vineyards showed predominantly oxic ORP conditions (i.e., O₂ ~20% and ORP > 500 Eh) for the duration of the experiment and only abrupt soil moisture increases after rain events. The only exception was profile 4 in V1 (see **Fig. 1a** for location), which showed a soil moisture increase at 1 m depth followed by a decrease in O₂. Since no soil moisture increase was observed at 0.2 and 0.6 m, we attributed this change to the possibility of lateral flow from the flooded subplots, impacting the oxygen status in the deeper soil profile.

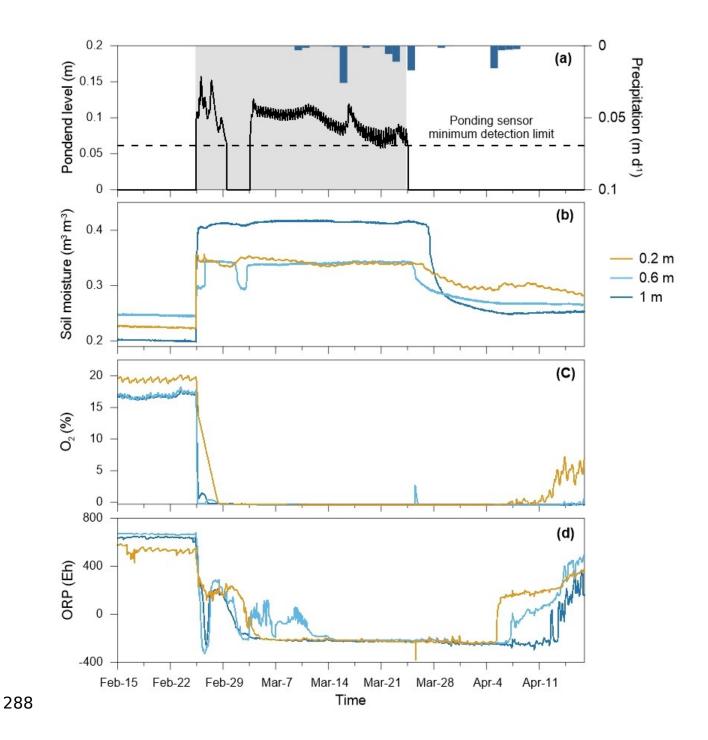
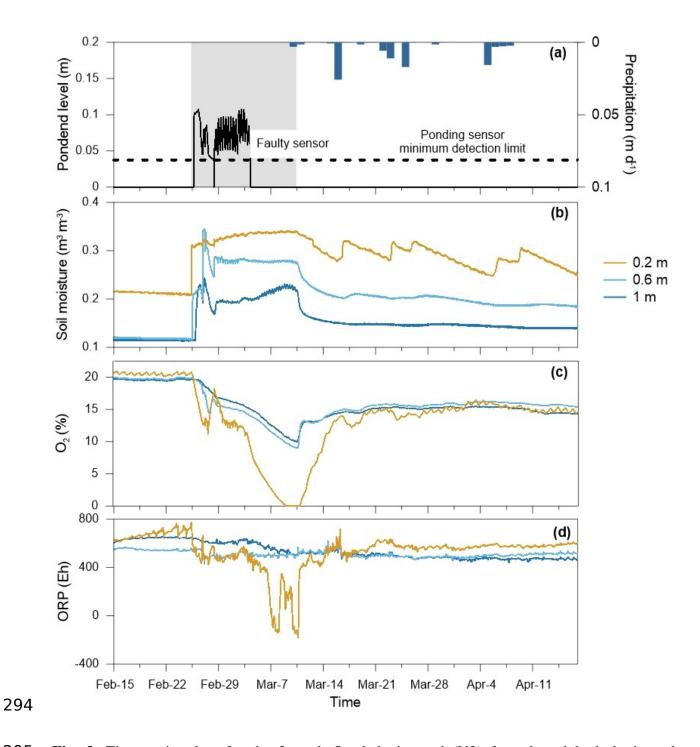


Fig. 2. Time series data for the 4-week flooded vineyard (V1) for selected hydrologic and
physico-chemical parameters from profile 1 (see Fig. 1a for profiles location). ORP – oxidationreduction potential. The gray shaded area in (a) shows the time of water application. Blue



precipitation bars show the daily totals taken from a meteorological station located 400 m fromthe experimental site.

Fig. 3. Time series data for the 2-week flooded vineyard (V2) for selected hydrologic andphysico-chemical parameters from profile 7 (see Fig. 1a for profiles location). ORP – oxidation-

reduction potential. Gray shaded area represents the flooding duration. Blue bars in plot a showdaily precipitation totals.

299 *3.2. Infiltration rates and recharge*

300 The experimental site received little precipitation prior and during the experiment (Fig. 2a), thus 301 providing an opportunity to study infiltration processes and water balance changes associated 302 with Ag-MAR more closely. The infiltration rate in V1 was 0.088 ± 0.031 m d⁻¹. Estimated 303 groundwater recharge was $83\% \pm 1.2\%$ of the applied water using Eq. 3. The V2 site had a 304 higher infiltration rate of 0.171 \pm 0.025 m d⁻¹ with 86% \pm 0.7% of the applied water percolating 305 below 1 m. The higher V2 recharge rate is also reflected by the soil suitability ranking developed 306 by O'Geen et al. (2015) for California soils, which rates V2 as "excellent" for Ag-MAR whereas 307 the ranking of V1 is "moderately good" as supported by soil textural analysis, which showed a 308 higher sand fraction in V2 compared to V1 (Fig. S1). Upscaling the above values to each 309 vineyard indicates that 1.31 and 1.32 m³ m⁻² of water was recharged in the flooded areas in V1 310 and V2, respectively. Although V2 was only flooded for two weeks, the higher infiltration rate 311 (0.171 m d⁻¹ in V2 compared to 0.088 m d⁻¹ in V1) resulted in higher total recharge amounts.

312 3.3. Pore water and residual soil chemistry

Pore water NO_3^- and NH_4^+ data from both vineyards are presented in **Fig. 4**. During the first week of flooding, NO_3^- concentrations in both vineyards decreased to zero at all three depths (**Figs. 4b** and **4e**). Pore water NH_4^+ showed near-zero concentrations in the deeper soil profile (0.6 and 1 m) before flooding and a clear reduction at 0.2 m depth (**Figs. 4c** and **4f**). DOC concentrations in pore water remained approximately 20-60 mg DOC L⁻¹ in both vineyards and

318 at all depths during flooding (Figs. 4a and 4d). Nitrite (NO_2) was not measured directly and

319 assumed to be negligible following Bachand et al. (2014), which reported nitrite levels lower

320 than $0.1 \text{ mg NO}_2^{-1} \text{ N L}^{-1}$ during their Ag-MAR study.

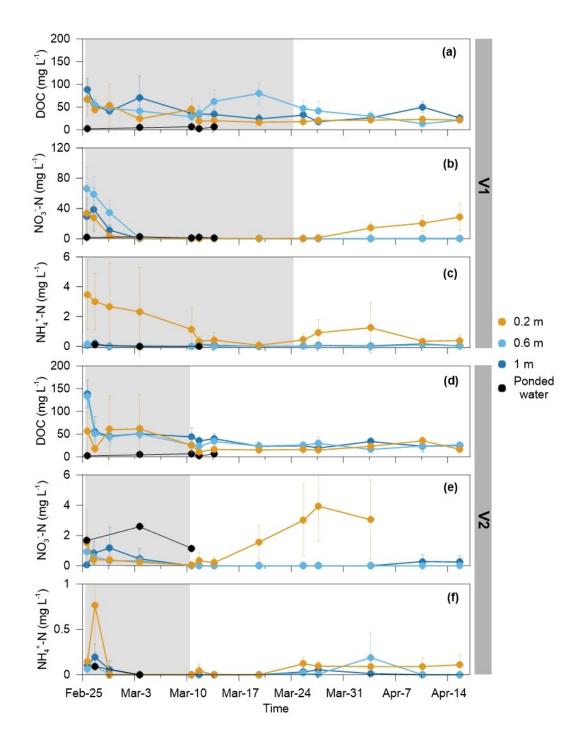


Fig. 4. Pore water results for the 4-week flooded vineyard (V1) (a-c) and the 2-week flooded
vineyard (V2) (d-f). Values are averages of the four flooded profiles in each vineyard with error
bars representing the standard deviation. Gray shaded areas represent the flooding duration.

325 Residual soil NH₄⁺ in V1 depleted from 1-2 to near 0 mg NH₄⁺-N kg⁻¹ soil at all depths during 326 the flooding (Fig. 5a). In V2, pre-flooding NH_4^+ concentrations were already low (e.g., ~0 mg 327 NH₄⁺-N kg soil⁻¹ below 0.1 m), with no significant change during flooding (**Figs. 5c** and **5d**). 328 Two weeks after the flooding ended, NH_4^+ levels in both vineyards increased uniformly at all 329 measured depths to ~1 and ~3 mg NH₄⁺-N kg soil⁻¹ for V1 and V2, respectively (Figs. 5a and 5c). 330 Similar post-flooding trends of increasing concentrations were also observed for NO_3^- in both 331 vineyards, however, only within the upper soil profile (0-0.2 m) where NO_3^{-1} increased to 20-60 332 mg $NO_3^{-}N$ kg⁻¹ soil (**Figs. 5b** and **5d**).

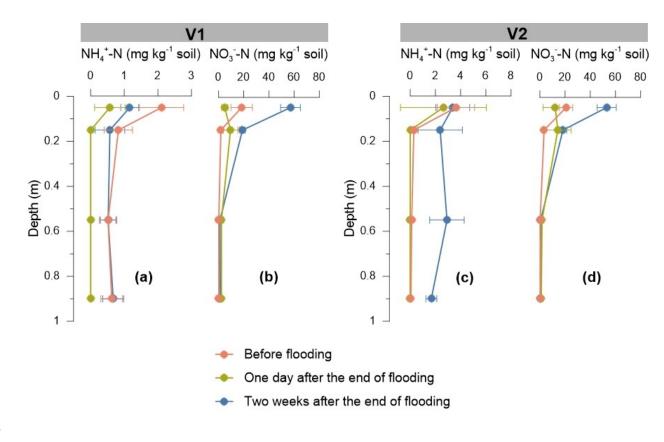


Fig. 5. Residual NO_3^- and NH_4^+ concentration in soil prior and after flooding for the 4-week flooded vineyard (V1) (a-b) and the 2-week flooded vineyard (V2) (c-d). Values are averages of the four flooded profiles in each vineyard with error bars representing the standard deviation.

337 3.4. Greenhouse gas emissions

During flooding, no changes in N_2O and CO_2 emissions were observed in the flood or control plots of V1 and V2 (**Fig. S2**). Post-flooding, N_2O emissions increased steadily for two weeks in the flooded plots in V1 before they declined again (**Fig. S2b**, blue line). However, peaks in N_2O and CO_2 emissions following the flooding were not as high as the spike observed two months after flooding, shortly after both vineyards received 336 kg ha⁻¹ of N fertilizer (ammonium sulfate 21-0-0 with 24% sulfur; end of Apr-2020), which was the only fertilizer application during that year. We note that CH₄ emissions were not detected during the experiments.

345 *3.5. Yield data*

Both vineyards were harvested in early Sep 2020. In V1, a significant 29% decrease in yield was observed compared to the control (t-test, p < 0.05), whereas in V2, there was no evidence of a significant difference in yield although yield was 14% lower in the flooded plots (p = 0.24) (**Fig. S3**).

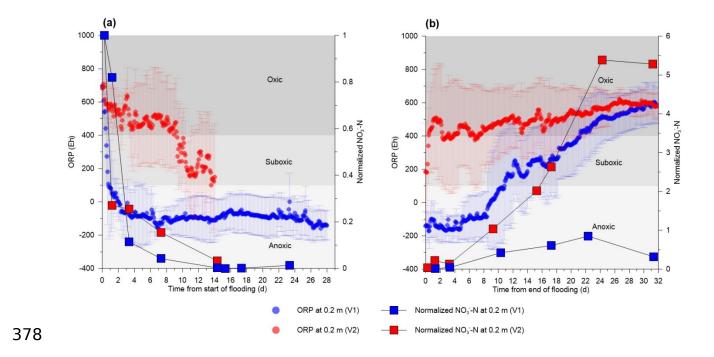
350 4. Discussion

351 *4.1. Nitrate leaching vs. denitrification during flooding*

We observed a rapid decline in pore water NO_3^- concentrations within the first days of flooding for Ag-MAR. Two possible processes can be attributed to the observed NO_3^- decrease: NO_3^- 354 removal due to denitrification, a microbial-controlled process, and/or leaching of NO_3^{-} , a solute 355 transport process. The former would be preferable during Ag-MAR (or any other MAR type) 356 since it can transform NO_3^- into the inert gas N_2 , thereby preventing NO_3^- from reaching the 357 groundwater (Levintal et al., 2022). Denitrification rates measured through incubation 358 experiments (Petersen et al., 2012; Verchot et al., 2001), showed the highest denitrification 359 activities in the top 0.1 m of the soil profile, decreasing to approximately zero below 0.6 m (Fig. 360 S4). Denitrification rates in the top 0.1 m were 1.4 and 1.35 mg NO₃⁻-N kg soil⁻¹ d⁻¹ for V1 and 361 V2, respectively. These rates are considered maximum rates, since they were obtained under 362 optimal conditions (25 °C, $O_2 \sim 0\%$, C substrate addition), favoring denitrification over 363 nitrification. In both vineyards, oxic conditions dominated the root zone before flooding started 364 and during imbibition. Thus, it is expected that denitrification rates were lower than the rates 365 derived from the incubation experiments, at least until the root zone became fully 366 suboxic/anoxic.

367 Infiltration rates not only determined the transition from oxic to suboxic conditions in each 368 vineyard, but also the rate at which NO_3^- was transported below the active denitrification zone in 369 the topsoil (i.e., 0-0.2 m). Fig. 6 describes the tradeoff between the development of 370 suboxic/anoxic conditions and the depletion of NO_3^- at 0.2 m. To allow comparison between the 371 vineyards, NO₃⁻ concentrations in each vineyard were normalized to the initial concentrations 372 observed in each vineyard prior to flooding. V1 reached suboxic conditions at 0.2 m (ORP < 400 373 Eh) after one day of flooding, compared to V2 which developed suboxic conditions only after 10 374 days. When the transition from oxic to suboxic conditions occurred in V1, more than 80% of the 375 initial NO_3^- was still available at 0.2 m depth (Fig. 6a, blue squares). In contrast, by the time V2

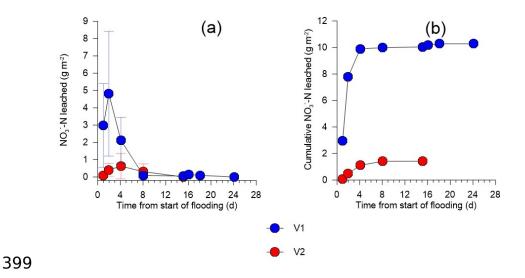
376 reached suboxic conditions only $\sim 10\%$ of the initial NO₃⁻ was available for denitrification (**Fig.**



377 6a, red squares), indicating that 90% was leached.

Fig. 6. Oxidation-reduction potential (ORP) and normalized pore water NO_3^- concentrations at 0.2 m depth during the wetting (a) and drying phase (b) of Ag-MAR. Values are averages of the flooded profiles in each vineyard with error bars representing the standard deviation. NO_3^- was normalized to the initial pore water NO_3^- concentrations in each vineyard; values above 1 indicate that NO_3^- exceeded the initial pre-flooding concentrations (i.e., V2 in subplot 'b').

The NO₃⁻ which was not consumed during the denitrification process in the topsoil was leached below the root zone and eventually transported towards the groundwater as denitrification is assumed to be negligible in the deep vadose zone (up to 3-5% per year) (Baram et al., 2016a). In both vineyards, the majority of NO₃⁻ was leached below 1 m during the first week of flooding, with a leaching peak of NO₃⁻ occurring during day 2 and 4 in V1 and V2, respectively (**Fig. 7a**). The cumulative amount of NO₃⁻ leached (using **Eqs. 1** and **2**) was 10.3 and 1.6 g NO₃⁻-N m⁻² for 390 V1 and V2, respectively (**Fig. 7b**), which equates to a NO_3^{-1} loading of 47.4 and 4.3 kg NO_3^{-1} -N 391 for V1 (flooded area of 4,613 m²) and V2 (flooded area of 3,035 m²) below 1 m, respectively. 392 Averaging the cumulative NO₃ leached per total applied water results in 8.6 and 1 g NO₃-N m² 393 for every 1 cm³ cm⁻² of water applied for V1 and V2, respectively (i.e., an average leaching 394 concentration of 8.6 and 1 mg L⁻¹ NO₃⁻-N for V1 and V2). These values are similar in magnitude 395 to NO₃⁻ leaching amounts estimated in a previous Ag-MAR field experiment (Bachand et al., 396 2014) and in an Ag-MAR column experiment (Murphy et al., 2021). Yet, it is also possible to 397 have different leaching amounts between sites due to site-specific factors influencing nitrate 398 leaching (e.g., soil texture and soil pre-flooding NO_3^- concentration).



400 Fig. 7. Observed and cumulative amounts of NO₃⁻ leached below 1 m for (a) the 4-week flooded
401 vineyard (V1) and (b) the 2-week flooded vineyard (V2).

402 The amount of NO_3^- leached differed by one-order of magnitude between the two vineyards.

403 These differences can be attributed to the initial pore water and soil NO_3^- concentrations prior to

404 flooding. In V1, the pre-flooding pore water NO₃⁻ concentrations ranged between 20-80 mg NO₃⁻-

405 N L⁻¹ (Fig. 4b), whereas in V2, concentrations were below 2 mg NO₃⁻⁻N L⁻¹ (Fig. 4e). Therefore, 406 estimating legacy NO_3^- levels prior to Ag-MAR is essential for assessing leaching risks. It is 407 emphasized that the low initial NO_3^- concentrations in V2's topsoil does not necessarily indicate 408 a lower risk of groundwater contamination. Both vineyards were fertilized and irrigated 409 following the same protocols in the years prior to the Ag-MAR experiment, and therefore, it is 410 likely that elevated NO_3^- levels were also present in V2. Yet, the surplus NO_3^- was pushed deeper 411 into the soil profile with each flood irrigation and precipitation event due to the higher infiltration 412 rate of V2 compared to V1.

413 Once pre-flooding pore water NO_3^- depleted, concentrations in both vineyards and at all three 414 measured depths (0.2, 0.6, and 1 m) stayed zero for the remainder of the flooding even though 415 NO_3^- was added to the system with the floodwater (2-3 mg NO_3^- -N L⁻¹; Figs. 4b and e, black 416 circles). The source water NO_3^{-1} was depleted by denitrification in the first 0.1 m of the soil 417 profile. This conclusion is supported by the denitrification rates estimated through lab incubation 418 experiments. For example, the denitrification rate estimated for the top 0.1 m soil profile in V1 419 was 1.4 mg NO₃⁻-N kg soil⁻¹ d⁻¹ (Fig. S4), which translates to a potential *in-situ* soil 420 denitrification of 10 mg NO₃-N per day during flooding. This rate is \sim 5-fold higher than the 421 floodwater NO_3^- concentration, thus can explain the NO_3^- depletion during infiltration. At this 422 stage, water percolating below 1 m contributed to the dilution of the already leached residual soil 423 NO_3^{-} . This process is also referred as the dilution effect (Bachand et al., 2014). While Ag-MAR 424 with low-contaminant water (e.g., rainfall-runoff or snowmelt) can cause not only NO₃⁻ leaching 425 from the root zone, but also dilution of NO_3^- in pore water, its final impact on groundwater 426 quality is a long-term process, which depends on several factors including the legacy NO₃⁻ load 427 in the vadose zone, the NO_3^- concentration of groundwater, geogenic sources, and groundwater 428 flow velocity (Dahan et al., 2014; Levintal et al. 2022). Therefore, quantifying the dilution effect 429 from a single year is not feasible; further discussion is given below in section 4.3.

430 4.2. Post-flooding nitrogen dynamics

431 Owing to the legacy of N in agricultural plant systems, we investigated the post-flooding N 432 cycling processes during the growing season. In V1, soil NH₄⁺ levels recovered to pre-flooding 433 levels in the first two weeks after the flooding ended (Fig. 5a). An increase was also observed for 434 NO_3^- , however, only in the topsoil (Fig. 5b, 0-0.2 m). Similar post-flooding NH_4^+/NO_3^- 435 concentrations increases were observed in V2 (Figs. 5c and 5d), while NH_4^+ increased at all 436 depths, NO_3^{-1} increased only in the topsoil, indicating that mineralization occurred throughout the 437 1 m profile, whereas nitrification was mainly limited to the topsoil. NH_4^+ concentrations were an 438 order-of magnitude lower than NO₃⁻ for both pore water and soil. The accumulation of NO₃⁻ in 439 the top of the soil profile (0-0.2 m) rather than at deeper layers is consistent with previous studies 440 (e.g., Dahan et al., 2014) who concluded that this accumulation pattern is preferable for both 441 crop uptake and reducing leaching risks.

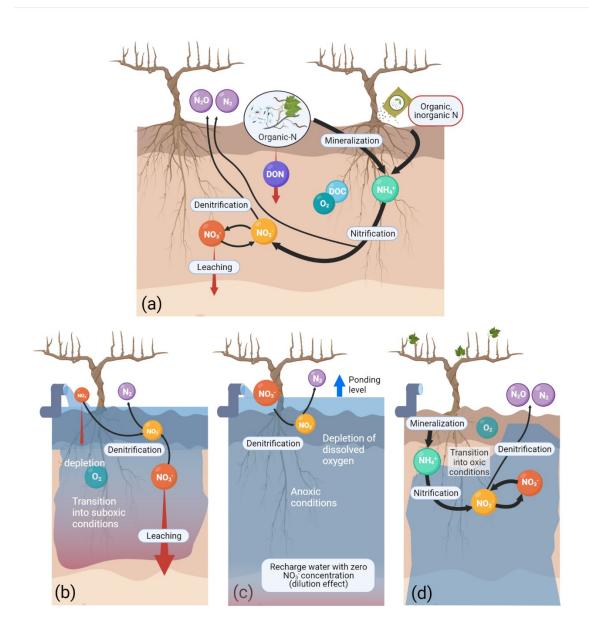
442 NO_3^{-} in pore water is considered to be more mobile in the vadose zone compared to the 443 immobile NH_4^+ cations attached to soil particles and organic matter (Subbarao and Searchinger, 444 2021). In terms of NO_3^{-} leaching risk, the chemical composition of the pore water has a more 445 immediate and significant effect on groundwater, rather than contaminants adsorbed onto 446 sediment in the immobile phase. Thus, prioritization of pore water data over soil data is 447 potentially recommended when assessing NO_3^{-} leaching risk to groundwater. We emphasize that 448 additional research is needed to validate this statement as previous studies also suggested that 449 nitrogen can be released from the soil to the pore water at later stages of the flooding and in the 450 deep vadose zone (Mienis and Arye, 2018; Xin et al., 2019). In general, the relationship between 451 pore water and soil samples is complex (Darrouzet-Nardi and Weintraub, 2014; Rimon et al., 452 2011). Studies suggested equations to convert between the two (Zhu et al., 2021). Yet, to date, no 453 empirical relationship has been validated for different soils or soil moisture variations as 454 expected during flooding.

455 Anoxic conditions continued in V1 for about 3 weeks after the flooding ended, while V2 stayed 456 mostly in oxic conditions throughout and after the flooding (Fig. 7b). The topsoil (0.2 m) in V1's 457 flooded area stayed in anoxic/suboxic conditions for 20 consecutive days once flooding ended, 458 promoting denitrification. We observed elevated N_2O emissions, which can be a byproduct of 459 denitrification, in the 20 days of anoxic/suboxic conditions following the flooding of V1 (Fig. 460 **S2b**, blue circles during the start of April). We note that N_2O emissions can also be attributed to 461 nitrification, however, under near-saturated conditions denitrification dominates N₂O production 462 (Bateman and Baggs, 2005; Zhu et al., 2013). N₂O emissions started to decrease in V1 after the 463 20 days due to developing oxic conditions following flooding. N₂O emissions started to increase 464 again after the vineyard's fertilization and first irrigation of the growing season on 4/29/2021. 465 The measured N₂O emissions were similar in magnitude to reported fertilization-related 466 emissions from vineyards under a similar Mediterranean climate (Garland et al., 2014, 2011).

467 4.3. Conceptual model – coupling nitrogen and water dynamics under Ag-MAR

We developed a conceptual model for N fate and transport in agricultural soils flooded for extended periods of time with low-contaminant water for Ag-MAR (**Fig. 8**). The model was developed based on data from the two vineyards investigated in this study and from recent Ag-

MAR studies conducted on alfalfa (Dahlke et al., 2018), almonds (Ganot and Dahlke, 2021b,
2021a), and in controlled soil column experiments (Murphy et al., 2021). We identify four stages
that determine N fate during Ag-MAR: pre-flooding, start of flooding, quasi-steady flooding, and
post-flooding. In each stage, the N fate is determined by the soil physico-chemical characteristics
and the initial N concentration.



477 Fig. 8. Conceptual model for N fate under long-term flooding conditions at the topsoil. (a) pre478 flooding, (b) start of flooding, (c) quasi-steady flooding, and (d) post-flooding. Detailed
479 description for each stage is given in section 4.3. Figure created with Biorender.

Pre-flooding stage: In the pre-flooding stage (**Fig. 8a**), the root zone is unsaturated and predominantly oxic. The soil matrix and pore water contain variable amounts of legacy inorganic N species (i.e., NH_4^+ and NO_3^-) from previous fertilization events and from mineralization of organic-N, which are either transported with the infiltrating water or transformed through various microbial processes.

485 Start of flooding stage: With the start of flooding (Fig. 8b) the root zone saturates with water 486 thereby decreasing oxygen and ORP levels. This is the most dynamic stage during the flooding 487 event, yet with high environmental significance as this stage will define the magnitude of the 488 NO_3^{-1} leached. The amount of NO_3^{-1} leached below the root zone and the amount of NO_3^{-1} 489 denitrified in the topsoil is dependent on site-specific physico-chemical parameters (K_{sat}, 490 infiltration rate, ORP, available NO_3^- and carbon, and temperature) and the abundance and 491 activity of both nitrifying and denitrifying microbial communities in the topsoil. NO₃⁻ leaching is 492 the dominating loss process in soils with high K_{sat} (or infiltration rate), while denitrification plays 493 a larger role in soils with lower K_{sat} where the residence time of water in the topsoil is longer. Of 494 course, denitrification is the preferred loss pathway for NO_3^- to reduce NO_3^- contamination to 495 groundwater, however, in the time it takes to reach suboxic conditions that promote 496 denitrification, high K_{sat} soil might already leach > 80% of the legacy NO₃⁻ present in the profile.

497 **Quasi-steady flooding stage:** After the highly dynamic NO_3 leaching and N transformation 498 phase at the start of flooding, the system transitions into the quasi-steady flooding stage (Fig. 8c), 499 which is characterized by saturated, suboxic/anoxic conditions and more stable soil physico-500 chemical parameters. If anoxic conditions are reached in the root zone, it can be assumed that 501 dissolved oxygen is likewise depleted as reported in other MAR studies (Turkeltaub et al., 2022). 502 Under these conditions, denitrifiers in the topsoil consume NO_3^{-1} in the infiltrating water. This 503 NO_3^{-1} removal will eventually contribute to the dilution of pore water NO_3^{-1} in the vadose zone. 504 Sustaining the quasi-steady flooding stage for as long as possible is preferred because it increases 505 the amount of 'clean' recharge while reducing NO_3^{-} levels in the pore water transported to 506 groundwater. However, the dilution effect only takes effect if NO_3^{-1} concentrations in the source 507 water are relatively low and the retention time of the infiltrating water in the denitrifying soil 508 layer is sufficiently long to allow complete NO_3^- consumption. For example, in this study, 509 infiltration rates of 0.1-0.2 m d⁻¹ were sufficient to allow denitrification of NO₃⁻ in the source 510 water (2-3 mg NO₃⁻⁻N L⁻¹) in the first 0.2 m of the soil profile. If recharge water is sourced from 511 pristine streamflow, it is typically low in NO_3^{-1} concentrations. Hence, there is a high probability 512 for the dilution effect to occur during the quasi-steady flooding stage when implementing Ag-513 MAR for periods of more than several days. However, flooding fields for long periods of time 514 might negatively impact crop performance, as shown for V1, which needs to be considered and 515 balanced (Levintal et al., 2022). A possible solution is to use a new model that predicts crop 516 damage as a function of the duration of saturated conditions in the soil root-zone, soil texture, 517 and crop tolerance to waterlogged conditions (Ganot and Dahlke, 2021a).

518 Post-flooding stage: In the post-flooding stage (Fig. 8d), oxygen and ORP will gradually 519 increase back to pre-flooding oxic levels. In soils with higher infiltration rates (sand or loamy-520 sand) this process can take several days compared to several weeks in soils with lower 521 infiltration rates (e.g., sandy loam or loam). Soil moisture will also decrease from near- or full-522 saturation to pre-flooding levels. However, this process is relatively slow (on the scale of weeks 523 to months) and controlled also by precipitation or irrigation events that will temporarily increase 524 soil moisture in the root zone. A post-flooding increase in NO₃⁻ and NH₄⁺ is also expected, 525 mainly in the topsoil (0-0.2 m) as a result of mineralization and nitrification. Denitrification is 526 another post-flooding process that can occur, yet it will be more pronounced in soils with slow 527 infiltration rates when soil moisture is still high during the first stage of drainage when 528 anoxic/suboxic conditions prevail (V1 in this study).

529 Ag-MAR includes three distinct differences compared to other MAR types (Goren et al., 2014; 530 Gorski et al., 2020, 2019; Mienis and Arye, 2018): (1) the nitrogen source is mainly in the soil 531 rather than in the recharge water, (2) recharge (i.e., the flooding) will be primarily seasonal 532 during wet periods and not yearly with continuous flooding and drying cycles, and (3) the 533 infiltration basin is not necessarily on high-infiltrating land, such as sandy soil. Therefore, 534 caution should be taken when adapting the four model's stages for long-term flooding of other 535 MAR types. For example, nitrogen fate under soil aquifer treatment (SAT; a MAR form where 536 treated wastewater is recharged) showed highly complex biogeochemical processes due to 537 changing nitrogen and carbon loading (e.g. NH_4^+) in the source water (Mienis and Arye, 2018). 538 This resulted in varying nitrate concentrations in the groundwater during the recharge, which our 539 model cannot adequately explain.

540 4.4. Implications for Ag-MAR operation

541 Understanding the N-related dynamics at each stage of Ag-MAR as presented in the conceptual 542 model (Fig. 8) is essential for the development of best management practices. Taking V1 as a 543 case study, a better practice to decrease the risk of NO_3^- leaching would be to irrigate the 544 vineyard initially for 1-2 days (before the continuous flooding for Ag-MAR) until anoxic 545 conditions are developed, which would promote denitrification and minimize NO_3^{-1} leaching. 546 Maintaining anoxic conditions is also important for the inhibition of the nitrification process that 547 can increase inorganic NO_3^{-1} concentrations between individual recharge (i.e., flooding) events. 548 The predicted outcome of the suggested recharge practice will be a decrease in the overall 549 amount of NO_3^{-1} leached. However, this practice is unlikely to be applicable in high infiltrating 550 soils in which nearly all the initial NO_3^- will be leached in the first few days before the 551 development of anoxic conditions (V2 in this study, infiltration rate of 0.171 m d⁻¹). A more 552 detailed discussion on the trade-offs between leaching and mineralization-denitrification 553 processes under repeated flooding events is provided by Murphy et al. (2021).

554 Our study was conducted in a Mediterranean climate, and therefore, adjustments are needed if 555 adapting the findings to other locations. For instance, lower ambient soil temperatures will 556 decrease the rate at which nitrification/denitrification occurs and to a lesser extent also influence 557 the infiltration rate (Grinshpan et al., 2022). Thus, Ag-MAR implementation in colder climates 558 will decrease denitrification rates during infiltration, potentially increasing the risk for leaching 559 of soil residual NO₃. Other considerations for Ag-MAR implementation are non-point source 560 contaminants with high leaching risks, such as salts, pesticides, and inorganic geogenic 561 contaminates. A comprehensive review of these contaminants with potential solutions is 562 provided in Levintal et al. (2022). Still, high NO_3^{-1} concentration in the aquifer is the main 563 concern at global scale (Beganskas et al., 2018; Dahan et al., 2014). With respect to yield, 564 several reasons can explain the yield difference between the vineyards investigated in this study, 565 such as differences in soil texture and the resulting K_{sat}, infiltration rates, O₂/redox levels, 566 duration of flooding, movement of available N below root zone, and timing of flooding with 567 respect to bud break in March. Therefore, possible effects on yield should be investigated in 568 more detail, and until then, caution is advised in using Ag-MAR in late winter-early spring, in 569 vineyards with less-than-ideal soil properties.

570 5. Conclusions

571 An agricultural managed aquifer recharge (Ag-MAR) experiment was conducted in the Central 572 Valley of California on two vineyards managed similarly but differing in soil texture and 573 hydraulic properties. One vineyard was flooded for four weeks (V1) and the other for two weeks 574 (V2). Although the flooding for groundwater recharge started on the same day, different soil 575 biogeochemical outcomes occurred. Suboxic conditions, favoring denitrification in the 576 microbially active topsoil (0-0.2 m), developed in the vineyard with the low-infiltration rate 577 (0.088 m d⁻¹; V1) in less than one day, compared to 10 days in the high-infiltration vineyard 578 (0.171 m d⁻¹; V2). Nevertheless, in both vineyards, NO₃⁻ leaching was the dominant N loss 579 process while denitrification played a lesser role in decreasing NO_3^{-1} in the root zone. The amount 580 of NO₃ leached below the 1 m root zone was mainly determined by the amount of residual NO₃ 581 present in the root zone prior to the flooding for groundwater recharge, showcasing the 582 importance of estimating legacy NO_3^- pools before establishing an Ag-MAR site.

583 A conceptual model for water-N dynamics under Ag-MAR was developed based on hydrologic, 584 geochemical, and microbial process analyses. Four stages were identified: pre-flooding, start of 585 flooding, quasi-steady flooding, and post-flooding. Out of the above, the start of flooding is the 586 most dynamic stage that will define the magnitude of NO_3^- leached. The proposed model can be 587 used to determine best Ag-MAR management practices. Adoption of Ag-MAR, as one approach 588 in a portfolio of MAR methods, is desirable as population growth and expansion of irrigated 589 agriculture contribute to unsustainable groundwater extraction and a growing need to replenish 590 groundwater resources to buffer growing water supply variability.

591 CRediT authorship contribution statement

Elad Levintal: Methodology, Writing – original draft, Formal analysis. Laibin Huang:
Writing - review& editing, Formal analysis. Cristina Prieto García: Methodology, Writing review& editing, Formal analysis. Adolfo Coyotl: Writing - review& editing, Formal analysis.
Matthew W. Fidelibus: Writing - review& editing, Formal analysis. William R. Horwath:
Writing - review& editing, Funding acquisition. Jorge L. Mazza Rodrigues: Writing - review&
editing, Funding acquisition. Helen E. Dahlke: Supervision, Methodology, Writing - review&
editing, Funding acquisition.

599 Declaration of competing interest

600 The authors declare that they have no known competing financial interests or personal601 relationships that could have appeared to influence the work reported in this paper.

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