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Article *in* Environmental Science and Technology · July 2020

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Environmental Tradeoffs between Nutrient Recycling and Greenhouse Gases Emissions in an Integrated Aquaculture–Agriculture System

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Cite This: https://dx.doi.org/10.1021/acs.est.0c00869

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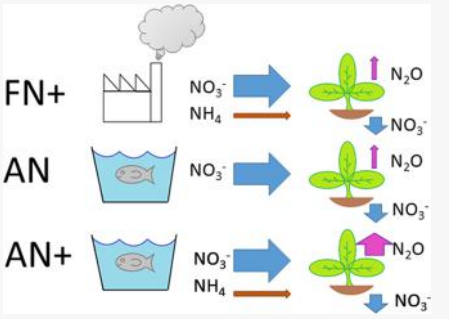
ABSTRACT: The unlimited nitrogen (N) availability that has characterized crop production in the last few decades is accompanied by environmental burdens, including the greenhouse gas (GHG) emissions associated with fertilizer production, post-application nitrate (NO_3^-) pollution of water bodies, and emissions of reactive gaseous N forms into the atmosphere. Here, we quantified the environmental tradeoffs of replacing mineral N fertilizer with NO_3^- and ammonium (NH_4^+) originating from effluent water of aquaculture in a cucumber (*Cucumis sativus*) cultivation system. While the yield, nitrogen use efficiency (NUE), and NO_3^- leaching were similar between the cucumbers fertilized and irrigated (fertigated) by aquaculture effluent water containing 100 mg of NO_3^--N L^{-1} (AN), by aquaculture effluent water supplemented with NH_4^+ (AN+), or by tap water with NO_3^- and NH_4^+ added (FN+), there were significant differences in the nitrous oxide (N_2O) emissions between the systems. The N_2O emissions peaked after each irrigation event followed by an exponential decline. The cumulative N_2O emissions were between 60 and 600 g N_2O-N ha^{-1}, smaller than predicted based on a fertilizer application rate of 600 kg N ha^{-1} and were in the order AN+ ≫ FN+ > AN.



INTRODUCTION

With the growing global population, worldwide food demand is increasing along with concerns about the environmental impact of food production.[1] A nutritionally balanced diet cannot depend only on cereals and meat consumption but must include vegetables,[2,3] resulting in an increased demand for vegetable production. In 2018, ~9.4×10^8 Mg of fresh vegetables were produced globally on an area of ~6.4×10^7 ha, and this production is predicted to expand further.[4] One promising method that can increase the sustainability of vegetable production is to integrate it with intensive fish production.[5,6] Integration of aquaculture and agriculture allows the recycling of nutrients excreted by fish due to nonefficient nutrient use (i.e., nitrogen (N) and phosphorus (P)[7]), which otherwise require removal at a centralized wastewater treatment plant or are discharged into the environment, leading to contamination of surface and ground waters.[8,9] An additional benefit is that the use of effluent water for vegetable fertigation (fertilization through irrigation) allows the recycling of otherwise wasted water for secondary crop production, increasing the water use efficiency (WUE) of the integrated system. This is especially appealing in semi-arid countries with limited water availability.

Continuous depletion of the wild fish supply due to overfishing has led to an increase in aquaculture, including the use of recirculating aquaculture systems (RASs) around the world,[10] potentially ensuring an uninterrupted supply of effluent water for vegetable fertigation. RASs have the potential to reach a wide range of nitrate (NO_3^-) concentrations suitable for fertigation,[11] and the modularity of such systems makes it possible to install them in series with fertigated crop production systems.[5] While water reuse benefits of RASs are well described,[12] fertigation with N originating from fish production is not well understood as the addition of large quantities of readily available N and carbon (C) sources[13] may enhance denitrification and nitrous oxide (N_2O) emissions from the integrated system,[14] contributing to their negative environmental impact. Furthermore, the reuse of effluent N from RASs allows for a reduction in dependence on mineral fertilizers and the carbon dioxide (CO_2) emissions associated with their production.[15] The N cycle in agriculture is inherently leaky with ~50−70% of applied N lost to the environment.[15] Two major forms of N pollution from horticulture are N_2O[16] and NO_3^-.[17] The NO_3^- leaches out of the root zone and contaminates groundwater and water bodies,[8] while N_2O is the third major greenhouse gas (GHG) and the agent of ozone (O_3) destruction in the stratosphere.[18]

Received: February 13, 2020
Revised: June 29, 2020
Accepted: July 8, 2020

Agriculture is responsible for the majority of new reactive N in the environment,[19,20] and mineral fertilizers are the biggest input of new N into the agricultural system.[21]

In general, microbial nitrification and denitrification are responsible for most N_2O production in agroecosystems,[22] including aquaculture.[23] However, a range of other microbial metabolic pathways that produce or consume N_2O complicates the determination of the specific sources of N_2O emissions measured in the field and their controlling factors.[24] Due to the similarity of the required environmental conditions, some pathways can only be determined with a degree of certainty through isotopic studies.[25,26] Nitrous oxide emission due to heterotrophic nitrification and the aerobic denitrification associated with it are examples of pathways that are often grouped together with others (heterotrophic denitrification and autotrophic nitrification) and are frequently neglected from overviews of N_2O production pathways,[24,27−29] though included in others.[30−33] Finally, traces of anammox bacteria have been found in aquaculture systems,[34] but a properly designed biofilter is required to use this pathway for efficient N removal.[35]

Regardless of the specific N_2O production pathway, the emission factors (EFs) or the fertilizer N-induced N_2O emissions can be estimated for aquaculture, as has been done for agriculture.[36] The EF estimation for aquaculture, however, is not done often, and knowledge about GHG emissions from the aquaculture and integrated systems is lacking. Recently, Hu et al.[23] estimated that ∼6% of all anthropogenic N_2O emissions come from aquaculture using an EF of 1.8% taken from a study of activated sludge wastewater treatment plants.[37] The United Nations Environment Programme (UNEP) report[38] citing the same publication used an EF of 1%. Such an inconsistency in estimations of EF for aquaculture is due to data scarcity. A handful of EF estimations for aquaculture have been published, generally reporting low values between 0.5 and 0.8%.[39−41] Because the optimal conditions for fish and plant cultivation are different (pH,[42] nutrient concentrations,[43] and temperature[11]) and recycling drainage water of plants back to the fish (i.e., aquaponic system) requires a much more expensive infrastructure,[6] the majority of future integrated aquaculture−agriculture systems (IAAS) are expected to be constructed with unidirectional water and nutrient flow.[5,11] Furthermore, such a system allows for the integration of the sludge treatment (feces and food waste removed from the RAS) into the IAAS, which will further increase the NUE of the integrated system. It has been suggested that, after undergoing anaerobic digestion, this sludge is a valuable potential source of NH_4^+ for plant cultivation.[11,44]

We are not aware of any published research assessing direct N_2O emissions from the plant production module of an IAAS where the plants were grown in soil. Thus, the overall objective of this study was to understand the N balance and cycle within the vegetable production component of an IAAS with an emphasis on sources of gaseous N_2O emissions. To this end, an IAAS[11] was modified to inject the supernatant from anaerobically digested sludge with a high NH_4^+ concentration into irrigation water to change the NH_4^+:NO_3^- ratio to that found in the mineral fertilizer used as the control treatment. The effect of the fertigation by the aquaculture effluent with and without NH_4^+ addition, on yield, NUE, NO_3^- leaching, and N_2O emissions from a cucumber cultivation was assessed in comparison to cultivation using a mineral fertilizer with the same nutrient composition. The specific aim was to quantify the N_2O emissions and to compare the overall environmental impact of the fertigation in the integrated and conventional systems with an emphasis on N pollution pathways.

MATERIALS AND METHODS

Recirculating Aquaculture System. The experimental setup was based on the one used by Groenveld et al.[11] In brief, cucumbers (*Cucumis sativus*) were grown in 18 lysimeters filled with a local sandy soil and were fertigated from three different sources: aquaculture wastewater with NO_3^- (AN), aquaculture wastewater with NO_3^- and NH_4^+ added from the anaerobic digester (AN+), and fresh water with fertilizer (MOR+, ICL, Israel) containing NO_3^- and NH_4^+ (FN+).

The two RASs used to produce AN and AN+ irrigation water each consisted of a polypropylene fish tank (800 L), a clarifier (300 L), and a moving bed bioreactor (MBBR; 400 L) with a flow rate of 0.5 m^3 h^{-1}. Barramundi fish (*Lates calcarifer*) were grown in the RASs for 6 months prior to the start of the experiment at which time they were weighed. Each system was provided with 500 g of food daily (45% protein, Raanan Fish Feed, Israel) for the duration of the experiment, which was 2% of the initial fish biomass and was the sole source of N into the system. Sludge was removed by flushing the clarifier every 2 h (resulting in about 30 L of sludge removed per kg feed) into an anaerobic sludge collection tank (500 L). The organic matter sunk to the bottom of the tank where it was anaerobically digested, and the reduced N remained in the supernatant as NH_4^+.

Nitrification of the NH_4^+ excreted by the fish took place in the MBBR on plastic biobeads (Aridal Ltd., Israel) where dissolved oxygen (DO) was maintained at about 90% saturation (∼7.5 mg L^{-1}). A heating coil in the reactor kept water in the fish tank between 28 and 30 °C. The MBBRs of the two RASs were connected with a pump that exchanged water between them so that their NO_3^--N concentration would be the same at around 100 mg L^{-1}. Once a day, 150 L of water was transferred from the MBBR through a 0.13 mm screen filter (with continual backwash to the biofilter) to an intermediary irrigation tank and was replaced with fresh water. The AN irrigation water was left as is, and the supernatant from the sludge tank, which had been diluted to a NH_4^+-N concentration of 100 mg L^{-1}, was added to AN+ irrigation water to bring the NH_4^+: NO_3^- ratio to about 1:10, similar to that of the synthetic fertilizer. The irrigation tanks were emptied before refilling each day.

Liquid NPK (4:2.5:6) fertilizer with microelements (MOR+ (NH_4^+:NO_3^- 1:10), ICL, Israel) was used to prepare fertilized water for the FN+ treatment. This solution was remade twice a week to eliminate NH_4^+ oxidation within the irrigation tank. The prepared solutions were continually mixed and cooled to 24 °C but not aerated. Potassium sulfate (K_2SO_4, Solucros, Belgium), potassium phosphate (MKP, Haifa Chemicals, Israel), and a cocktail of micronutrients (Super Koratin, ICL, Israel) were added to all the irrigation treatments to keep their levels similar (Table S2).

The pH in the RASs was kept at 6.5 by dosing potassium hydroxide (KOH), while tap water had a pH of about 8.2 and was brought down to 6.5 for the FN+ treatment by dosing hydrochloric acid (HCl); both processes were done by means of automated dispensers (Profilux, GHL, Germany).

Irrigation water was sampled every 2 days to measure the electrical conductivity (EC) (DDS 120 W, Bante Instruments,

https://dx.doi.org/10.1021/acs.est.0c00869
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China), NO_3^--N (Reflectoquant nitrate test 116971, Merck KGaA, Germany), NH_4^+-N (ammonium test 11117, Merck KGaA, Germany), NO_2^--N (nitrite test 14658, Merck KGaA, Germany), and pH (HI 9126, Hanna Instruments, RI USA). The DO levels were measured in the irrigation water tank itself (Handy Polaris, OxyGuard, Denmark). Drainage water of each lysimeter was sampled every 2 days and tested for NO_3^--N, NH_4^+-N, pH, and EC by the same methods listed for irrigation water. Total organic C (TOC) was measured in irrigation water every 5 days and three times in drainage water at periodic intervals (multi N/C 2100S; Analytik Jena, Germany). N as a dissolved organic material (DOM-N) was assumed to be about 6% of N in the aquaculture effluent as this was measured in earlier experiments with the same feeding and water exchange rates.[11] The supernatant from the sludge digestion tanks was tested twice a week for NH_4^+ (Reflectoquant ammonium test 116977, Merck KGaA, Germany). Samples from all of the irrigation water treatments were tested twice for other nutrients essential for plant growth (K, Ca, Mg, Na, Mn, Fe, Zn, Cu, and B) by means of atomic absorption (AA240FF, Varian, USA); P and K by means of spectrophotometry, (Lambda 25, Perkin Elmer, USA); and Cl by means of titration, (665 Dosimat, Metrohm, Switzerland)) to confirm that these were being added in the right proportion (Table S2).

Plant Cultivation. Cucumbers (Sanyal, Soli, Israel) were grown in round 50 L lysimeters of 40 cm in diameter fertigated from each of the three treatments in six replicates (total of 18 lysimeters). The growth medium was sandy soil (3% 1−2 mm, 29% 0.5−1 mm, 52% 0.25−0.5 mm, 15% 0.125−0.25 mm, and 2% <0.125 mm) packed at an initial bulk density of 1.54 kg L^{-1}. Below the sand, a layer of highly permeable geotextile (polyester fibers, Perlon, Germany) separated the sand from a 5 cm layer of plastic biobeads (Aridal Ltd., Israel) to ensure that the bottom boundary condition was equal for the whole container. The 35 cm sand above this layer ensured that the upper root zone was sufficiently aerated. The soil−air oxygen concentration was measured continuously (KE-50, Figaro, Japan) at a 5 cm depth next to the base of the plant. From the water collection layer, the drainage flowed into a separate container from which it was pumped out, weighed, and sampled to perform water and N balances. The daily transpiration was calculated for each lysimeter by subtracting the amount of drainage from the amount of irrigation water.

One cucumber seedling was transplanted into each lysimeter on 5/5/19; from here on, dates are referred to as days after transplant (DAT). The lysimeters were irrigated with water from their respective treatments 3 weeks prior to the start of the experiment. After the transplantation, each replicate was irrigated five times per day with 1 L of water till the measured transpiration reached 2 L per day per lysimeter after which each lysimeter was irrigated at 2.5 times the measured transpiration, rounded up to 1 L increments.[11] The soil surface was covered with white geotextile (polyester fibers, Perlon, Germany) to reduce evaporation while allowing free gas movement. One runner of each plant was trained up a single string, and all side branches were removed by cutting after the first node of each side branch. Cucumbers were harvested daily from 21 DAT till the end of the experiment at 51 DAT. From 21 DAT, one replicate of each treatment was destructively sampled weekly to determine the dry biomass and N content of the plant; therefore, the number of replicates decreased over time from six at the beginning of the experiment to one for the final measurements. Leaves, shoots, and fruits were dried separately and were tested for the C and N content every week by combustion (OEA-CHNS Flash, 2000, Thermo Fisher Scientific, MA, USA) after grinding to pass through a 200 μm screen. Lysimeters of replicates that were destructively sampled were repacked with fresh soil (to an initial bulk density of 1.54 kg L^{-1}), returned to their former location in the greenhouse, and continued to be fertigated according to their respective treatment.

N_2O Emissions. *Measurements.* A slightly modified static chamber method was used for estimating N_2O emissions after Gelfand et al.[45] The 10 cm tall permanent collar was placed adjacent to the plant and irrigation water emitter, inserted ∼2−3 cm into the soil, and left in place for the duration of the experiment in every lysimeter. During the measurements, the collar was covered with a lid, which had inlet and outlet sampling ports and a pressure stabilization coil made out of a ∼1 mm diameter tube.[46] The total static chamber volume during sampling was ∼3 L. The chamber was connected to an ICOS N_2O/CH_4 analyzer (M1-919; ABB-LGR, Cary, NC, USA), and air from the chamber was continuously circulated through the analyzer, which measured the N_2O concentration inside the chamber and recorded the average every 5 s. The N_2O fluxes were calculated as the linear change of the gas concentration in the chamber headspace over an incubation period of ∼1 min, corrected for ambient air temperature. Measurements of N_2O started from DAT 7 after plant acclimation and were carried out at least once a week, usually for two consecutive days, so that each lysimeter was sampled multiple times throughout the day. Due to the high irrigation frequency, emission measurements of each individual lysimeter were performed at different time intervals from the fertigation, which is reflected in high flux variability.

Dissolved N_2O. Nitrous oxide dissolved in irrigation water was measured by placing a 250 mL water sample into a glass container of a 1 L volume with two pipes inserted into the sealing lid, bringing the headspace air to and from the N_2O analyzer. The pipe with the returning air extended to below the surface of the sample so that it bubbled up through it. After measuring the temperature of water, the container was closed, the sample was shaken profusely for 1 min, and then remained still till the N_2O reading stabilized, which was considered the point at which the air space concentration was at equilibrium with that of water. The difference between the atmospheric and equilibrium concentration and the water temperature was used to calculate the amount of N_2O dissolved in the 250 mL sample, according to Weiss and Price.[47]

Degassing Estimation. To test whether the irrigation water degassing was responsible for part of the observed N_2O emissions, the emission rates over time after fertigating with water of different dissolved N_2O concentrations were compared. Different dissolved N_2O concentrations were prepared from the same AN+ treatment irrigation water tank by heating water to 90 °C, transferring it between two containers to ensure a large gas exchange area, and finally cooling it to ∼30 °C. The NO_3^-, NH_4^+, and nitrite (NO_2^-) concentrations of heated and untreated water remained the same, but the heated sample had no dissolved N_2O. After measuring the dissolved N_2O concentration in the two water samples, 1 L of each was used to irrigate a lysimeter without plants, which had been fertigated with water from the AN+ treatment for more than a month. The N_2O emissions were

https://dx.doi.org/10.1021/acs.est.0c00869
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measured repeatedly (>five times) in the 1.5 h period following the irrigation, and the entire process was repeated three times.

Carbon Addition Experiments. To test whether the N_2O production was limited by available C in the FN+ treatment, dextrose ($C_6H_{12}O_6$) was added to water from this treatment at 2.5 g L^{-1}. Water was incubated at room temperature overnight, and concentrations of dissolved inorganic N (NO_3^-, NH_4^+, NO_2^-, and N_2O) were measured before and after the incubation and were identical. Two lysimeters of the FN+ treatment without plants that had been fertigated with water from the FN+ treatment for more than a month were irrigated with 1 L of water with added C each. One similar control lysimeter was irrigated with FN+ water without the added C. The N_2O emissions were measured repeatedly following the irrigation; after 7 and 18 h, this cycle was repeated in the same lysimeters.

N_2O Emissions Model. As the biggest factor influencing the N_2O emissions was the time after irrigation (Figure S1), an exponential model was fit to all the emission data measured during the experiment for each treatment

$$E_t = E_0 e^{-kt} \quad (1)$$

where E is the rate of N_2O emission (μg N m^{-2} min^{-1}), t is time after the irrigation event (min), E_0 is the rate of N_2O emission at $t = 0$ (μg m^{-2} min^{-1}), and k is the rate of decline (min^{-1}). The E_0 and k parameters were determined for each set of measurement data by minimizing the sum of square errors of modeled to measured data by means of the generalized reduced gradient (GRG) nonlinear solving method in the solver function of MS Excel (Microsoft Inc.) without any additional constraints. This model of N_2O emission with time after an irrigation event was used to calculate the cumulative N_2O emissions for each treatment by integrating the emission rates between irrigation events. The use of linear interpolation between the measurements, as is commonly done to calculate cumulative flux for field measurements,[48] was not possible due to pulse behavior of the post-irrigation flux (Figure S1). The methodology for the estimation of the pathways involved in the N_2O production is reported in Table S1.

Carbon Footprint, Nitrogen Use Efficiency, and Yield-Scaled N_2O Emission Calculations. For estimation of the C footprint, we calculated the global warming impact (GWI), including interpolated N_2O emissions and the C cost of the N fertilizer (after Gelfand and Robertson[49]) using a 100 year time horizon for all GWI calculations.[50] We used a factor of 298 g of a carbon dioxide equivalent (CO_2e) per g N_2O for N_2O GWI estimation and 8 g of CO_2e per g N of ammonium nitrate fertilizer production.[51] The GWI was normalized to Mg of fresh yield to facilitate comparison with other studies.[52]

As other works have been done concerning the GWI of RASs[44,53,54] and given that the focus of the current study was the environmental impact of integrating such systems with horticulture, only the GWI of the vegetable production module was calculated, assuming that water would be discharged into the sewage system if not used for vegetable production. The GWI was calculated as the sum of the GWIs of N_2O emissions and fertilizer production. Finally, the NUE was calculated as the N retained in the harvested plant biomass over the N applied (g N in biomass g^{-1} N applied);[52] the fact that the N in aquaculture was already used once for fish production (within the fish feed) was not taken into consideration in this calculation.

Data Analysis. The irrigation treatment data was tested for normality with the Kolmogorov–Smirnov test, and for homogeneity of variance with Levene's test, a one-way analysis of variance (ANOVA) with a post hoc Tukey test was used to determine significantly different interactions between tested groups when the ANOVA p value was below 0.05. The data was analyzed using on-line resources.[55,56]

RESULTS AND DISCUSSION

Irrigation water's NO_3^- and NH_4^+ concentrations were steady over time, and their average values are shown in Table 1. The

Table 1. Average Concentrations of Different N Forms, Total Organic Carbon (TOC), and Dissolved Oxygen (DO) that Were Measured in Irrigation Water (the Standard Deviation in Parentheses, n Varies between 12 and 50)[a]

	mg L^{-1}		
system/parameter	AN	AN+	FN+
TOC	40.0 (5.0) [a]	39.0 (5.0) [a]	9.0 (3.0) [b]
DO	8.4 (1.3) [a]	6.7 (0.6) [b]	7.8 (0.5) [a]
NO_3^-	100.0 (17.0) [a]	99.0 (10.0) [a]	96.0 (11.0) [a]
NH_4^+	1.2 (0.5) [a]	8.3 (2.1) [b]	13.9 (4.3) [c]
NO_2^-	0.6 (0.3) [a]	3.4 (1.2) [b]	3.6 (4.6) [b]
N_2O	0.0018 (0.0028) [a]	0.0082 (0.0069) [b]	0.0003 (0.0000) [a]
total N	102 [a]	111 [a]	114 [a]

[a]Lowercase letters identify significant differences between the treatments for the individual parameter.

NH_4^+ concentration was slightly lower for AN+ and higher for FN+ than the target, but the total amounts of N fertigated were not significantly different. The cumulative transpiration (Figure 1A) showed a linear increase over time, indicating continual healthy plant growth. Transpiration reached 2 L per plant at 20 DAT; the resulting irrigation over transpiration ratio (I/T) average for all treatments after that time is shown on the same graph, and it remained close to the target I/T of 2.5.

The average cumulative cucumber yields per treatment (Figure 1B) were similar for all treatments. The NUE (Figure 1B) was calculated for one lysimeter per treatment per week on the day that it was destructively sampled and was ~40%. The slight increase of the NUE over time is due to the fact that the N was supplied in relation to the transpiration level, while an increasing proportion of the N was incorporated into the fruit, which transpires less per unit of N taken up than the vegetative parts of the plant.[57] This caused the N application/N uptake ratio to decrease over time despite the steady I/T regime.

On average, we accounted for 87% of applied N in measured nongaseous N forms (Figure 2). As N_2O emissions were calculated to not exceed 0.1% (see below), the 13% of N missing from the balance can be assumed to be N_2 and NO but was unquantified in our study. The allocation of the N fertigated to different plant parts and to drainage water was calculated by linear interpolation of the treatment-averaged dry matter N concentration. After 21 DAT, the N allocation to the fruits increased rapidly in relation to the leaves and stems. The difference between the fertigated N and what was accounted for in drainage water and plant uptake was assumed to be the amount of total gaseous N lost and was similar between treatments. A growth curve of the different parts of the cucumber plants per treatment is presented in Figure S2.

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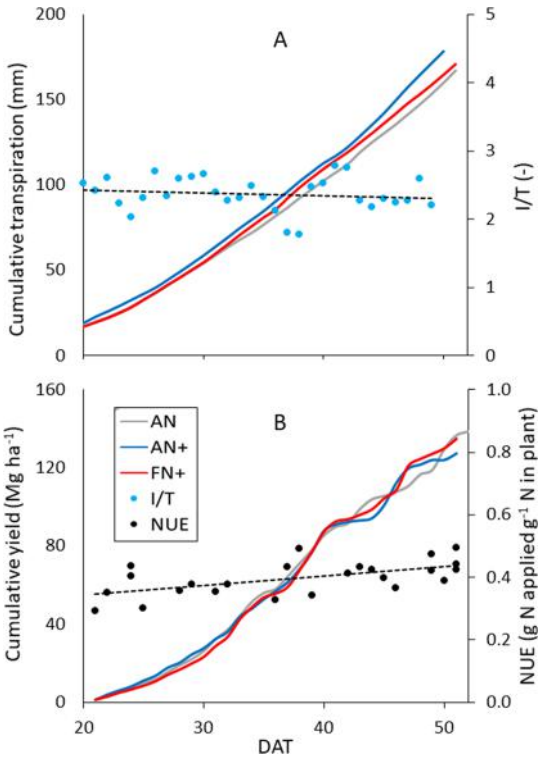

Figure 1. (A) Average cumulative transpiration and the average irrigation over transpiration rate (I/T; L L^{-1}) and (B) cucumber yield (fresh weight) and N use efficiency (NUE; g N applied g^{-1} N in plant) for all treatments.

The ratio of drainage NO_3^- to that taken up by the plant was significantly higher in the first 3 weeks than during the rest of the short growing season; at DAT 20, already 45% of the total NO_3^- drainage had leached out, while only 20% of the N uptake by the plant had taken place. Such excessive fertigation to ensure proper seedling establishment is a common practice in horticulture (personal communication with local farmers), and addressing this issue could potentially prevent a significant part of NO_3^- leaching. A technical solution could be to increase the irrigation frequency while decreasing the irrigation amount. Some potential problems with this solution include the magnification of any lack of homogeneity in the flow rate between drippers, and in desert climates, the temperature of water remaining in the dripline between irrigation events can get very hot, exposing the sensitive plant root zone to frequent pulses of scalding water.

On average, 38% of the N applied was leached as NO_3^--N, or 1 g kg^{-1} fresh weight of cucumber harvested, as opposed to 19% by Groenveld et al.[11] in which the same variety of cucumbers was grown on a shallow bed of perlite at the same I/T but at a 3 times higher irrigation frequency. Grewal et al.[58] reported that 59% of the applied N was removed with the drainage in a hydroponic cucumber production system that had a lower normalized yield; this, however, may include gaseous N emissions. Yao et al.[59] did not measure the NO_3^- leached, but a mass balance estimate would put the NO_3^- leaching at around 10 g of NO_3^--N kg^{-1} cucumber.

The average of N_2O emission measurements was higher for AN+ than for the other treatments, but the variability was very large (Figure 3B). Measurements of the N_2O emissions from a single lysimeter over time after an irrigation event show that there was a strong decline in emissions over time after the fertigation event (Figure 3A). The exponential model (eq 1) fit the data well, as can be seen from the R^2 values reported in Table 2, and was subsequently used to describe the change in N_2O emissions with time after irrigation per treatment for all data collected (parameters reported in Table 2). The fit to all the data was not as good as it was for the single event (Table 2 and Figure 3B) due to the overall variability of gaseous N emissions in time and space, although no trend was observed in the change in N_2O emissions throughout the growing season. Nonetheless, as it takes into consideration the observed reduction in emissions with time after the fertigation event, the exponential model was a more reliable tool to use in approximating the cumulative N_2O emissions from all studied

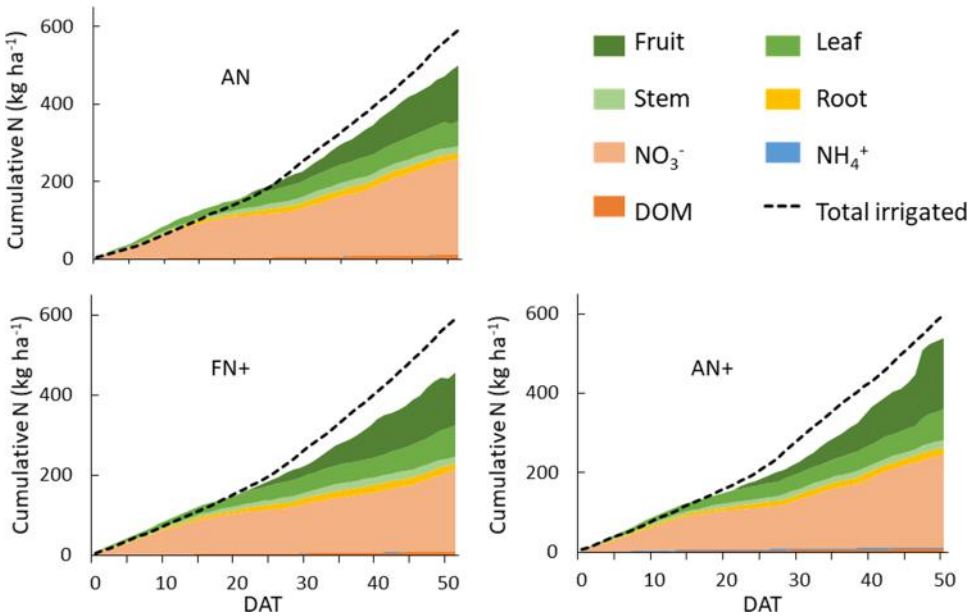

Figure 2. Average allocation of the total applied N in fertigation (dashed line) for each of the treatments. NH_4^+ and N-DOM levels in the drainage are too small to be seen.

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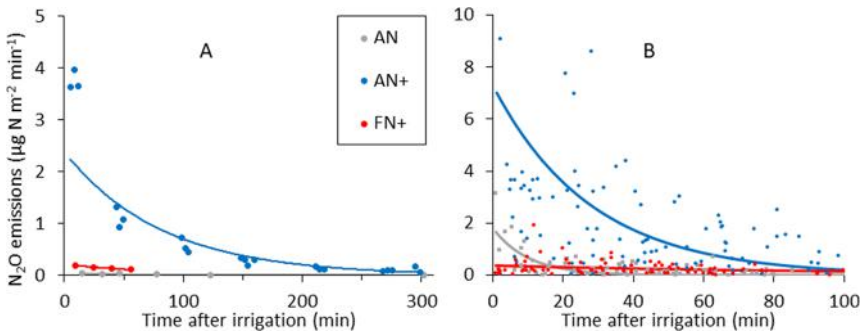

Figure 3. (A) N_2O emissions, measured over time after an irrigation event, of one lysimeter of each treatment; fit with the exponential model whose parameters and R^2 values are listed in Table 2. (B) All of the N_2O oxide measurements over time (note that the *Y*-axis scale changes) with the exponential model fit to each treatment whose parameters and R^2 values are listed in Table 2. The axes of panel (B) are limited to 10 μg N m^{-2} min^{-1} and 100 min to make it more readable.

Table 2. Number of Emission Measurements of the Total Dataset (n), the Parameters of the Exponential Model's (eq 1) Fit to the Whole Season Measurements, and the R^2 Values for this Model's Fit to the Emissions Measured after a Single Irrigation Event and the Whole Season Data.

		exponential model		R^2	
treatment	n	E_0	k	single event	all data
AN	104	1.77	0.10	0.70	0.48
AN+	134	7.30	0.04	0.91	0.31
FN+	103	0.38	0.01	0.98	0.12

[a]The parameters for the best fit were determined by means of the Solver.

systems than either average values or single values representing the time at which they were taken. Measurements of N_2O emissions throughout the growing season can be seen in Figure S1.

The amount of N_2O dissolved in the AN and AN+ water was higher than in the FN+ treatment, which was in equilibrium with the atmospheric concentration (Table 1). Part of the large initial post-irrigation peak in N_2O emissions was hypothesized to be due to irrigation water degassing. This was confirmed by measuring the emissions following the irrigation with water of different dissolved N_2O concentrations. Figure 4A shows that the N_2O emissions, following irrigation with degassed water, were lower and remained stable over time as opposed to water with higher dissolved N_2O concentrations with the E_0 and k parameters of eq 1 increasing and decreasing, respectively, with increasing irrigation water N_2O concentration. The concentration of the degassed water (~0.2 μg N_2O-N L^{-1}) was lower than what was reported to be at equilibrium with the atmospheric concentration (Table 1) because of the warmer water temperature (~30 °C).

The only difference between the AN+ and FN+ treatments was the absence of C in FN+ water, and it was hypothesized that N_2O production was C-limited in the FN+ treatment. This was tested by comparing the N_2O emissions of lysimeters fertigated with water from FN+ with and without C (Figure 4B). The N_2O emissions did not increase after the first irrigation event, but following the second and third events, there was a clear increase similar to Liang et al.[60] who demonstrated that C availability can alter the N_2O flux in laboratory incubations. Dependence of the N_2O flux on the C availability in a mostly aerobic environment suggests that heterotrophic N_2O production pathways other than denitrification occur in this system, a point that is discussed in-depth in the Supporting Information.

The cumulative N_2O emissions are shown in Figure 5, and it is possible that we slightly underestimated the cumulative N_2O

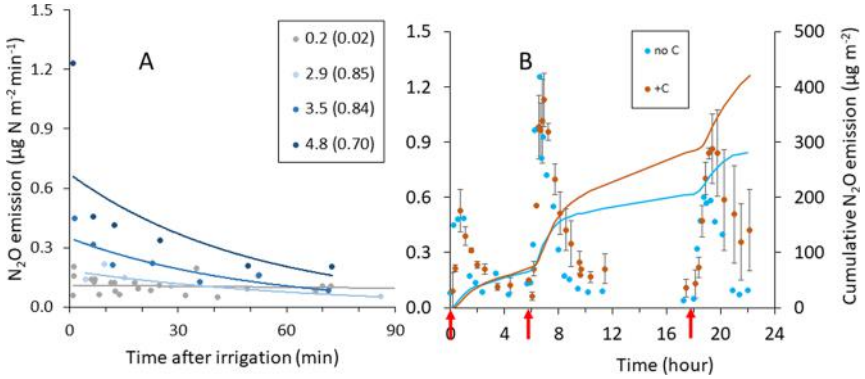

Figure 4. Experiments on lysimeters without plants to test hypotheses concerning N_2O emission pathways. (A) N_2O emissions over time after irrigation with water of different dissolved N_2O concentrations. The exponential model (eq 1) was fit to each dataset with the R^2 value of the modeled fit in parentheses after the dissolved N_2O (μg L^{-1}) concentration reported in the legend. (B) N_2O emissions following fertigation with synthetic fertilizer with and without added carbon (irrigation events marked with red arrows). The emissions over time are shown in dots with the standard deviation reported on the +C treatment (n = 2), and the cumulative emissions are shown with lines whose colors match those of the dots reported in the legend.

https://dx.doi.org/10.1021/acs.est.0c00869
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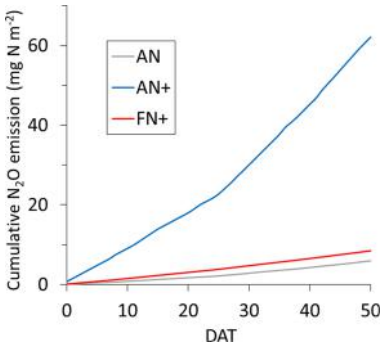

Figure 5. Cumulative N_2O emissions calculated by means of the exponential model (eq 1) with the parameters listed in Table 2.

emissions due to the model's (eq 1) failure to represent the sharp reduction of N_2O emissions in the first 5 min after irrigation (e.g., AN+; Figure 3A). The increase in rate of N_2O emission after DAT 25 is due to the increased amount of irrigation (i.e., irrigation events) on the basis of which the emissions were calculated.

The N_2O emissions in this study were lower than previously reported.[61,62] The calculated results indicate that the highest sum of emissions was from the AN+ treatment at 62 mg N m^{-2}, while emissions from AN and FN+ were similar (Figure 5). This amounts to 0.4, 4.5, and 0.6 g of N_2O-N emission per Mg of cucumbers produced for treatments AN, AN+, and FN+, respectively. The EFs were also very low, only 0.1% for the system with the highest emissions (AN+; Figure 5), compared to an average of 1% assumed for fertilized agriculture.[20] However, Hashida et al.[63] observed lower N_2O emissions of 9 mg m^{-2} in a tomato cultivation in fresh soil during the first season, which increased to 31 and 290 mg m^{-2} in the subsequent two half seasons (all data normalized to match the 50 day season length of the experiment described in this paper). They attributed this to increased N substrate availability over time, although an alternative explanation could be the development of microbial films in the soil. Daum and Schenk[16] reported an EF of about 0.9% in a cucumber cultivation on rock wool in Holland with an overall 25% lower yield compared to our results. Yao et al.[59] reported even higher N_2O emissions of 174 g Mg^{-1} for cucumbers grown in China with similar N application rates. However, their N application type and timing were very different from those reported here. They applied fertilizer in the form of urea or manure in a few large pulses, which potentially reduced the NUE and is reflected in 7 times lower yields, as compared to our study.

Fertigation with NO_3^- from the RAS effluent (AN), as opposed to a synthetic N source (FN+), prevents 37 kg of CO_2e emissions per Mg of fresh cucumbers produced. The reduction in the case of the AN+ was slightly lower at 32 kg per Mg of fresh cucumbers produced due to the increased N_2O emissions resulting from the sludge incorporation. As there was no apparent added benefit to the higher NH_4^+:NO_3^- ratio in terms of yield, an alternative way to use the sludge for fertigation would be to treat it in a second nitrifying biofilter.[64] Such systems have been shown to be capable of converting high NH_4^+ concentrations to NO_3^-[65] while potentially reducing total N_2O emissions.[66] Our estimations of the GWI reduction do not include the CO_2e emissions avoided by using the RAS effluent, which would otherwise require treatment by a wastewater treatment plant. Such a treatment is estimated to emit between 230 and 830 g of the CO_2e m^{-3} effluent.[67] In the IAAS system, 5865 m^3 ha^{-1} was used for irrigation, and inclusion of the CO_2e savings associated with the treatment of this water would increase climate change mitigation of the integrated systems by 30−100% (10−35 kg of CO_2e emissions per Mg of fresh cucumbers produced). Moreover, fish production and consumption accounts for 3.8% of the annual per capita impact for aquatic eutrophication and is a larger environmental load than the aquaculture contribution to the climate change.[68] The reuse of effluent water for horticulture thus will reduce not only the GWI of the integrated system but also its eutrophication potential.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c00869.

Supplementary methods, results and discussion all relate to the contribution of different pathways of N_2O production to the total N_2O emission from the studied systems (Figures S1−S3 and Table S1), and concentrations of nutrients other than nitrogen in water (Table S2) (PDF)

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Author Contributions

T.G., I.G., and N.L. designed the experiments, T.G. conducted the experiments and analyzed the data, and T.G. and I.G. wrote the paper with contributions from all co-authors.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank Matthias Overduin, Angelika Yurina, and Mais Dzhafarov for help with the field work at the Arava R&D station where the experiments were performed. Financial support for this work was provided by the Israel Ministry of Agriculture and Rural Development, Israel (grant number 3000013059), the Keren Kayemeth Le Israel-Jewish National Fund, Israel, the Goldinger Trust, USA, the Daniel Koshland Fund for Interdisciplinary Research, USA, and the JCA Foundation.

https://dx.doi.org/10.1021/acs.est.0c00869
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