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Phreatic zone wastewater irrigation: Sensitivity analysis of contaminant fate

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ABSTRACT

Subsurface irrigation by recharging shallow phreatic aquifers to raise the water table allows treated wastewater and other marginal water to be used in irrigation, without directly exposing crops to contaminants of emerging concern (CECs). The effects of soil and aquifer properties, environmental hydrological fluxes, irrigation parameters, and CEC biogeochemical reaction parameters, on crop and environmental contamination risks, are studied through numerical modeling. Non-biodegraded CEC solutes leave the agricultural field mostly by lateral discharge within the phreatic zone. The solute mass discharged beneath the simulated domain (potentially into deeper confined groundwater) typically represents the smallest portion of solute fate, but varied by orders of magnitude across scenarios. In contrast, other components of solute fate: the solute mass recovered by the subsurface drains, crop solute uptake, and solute mass discharged laterally within the phreatic zone are larger (in ascending order), but varied across scenarios mostly within one order of magnitude. Furthermore, solute biogeochemical reaction parameters most greatly (by orders of magnitude) affected crop solute uptake and solute discharge into the environment, followed by the hydrogeological parameters, atmospheric fluxes, and finally irrigation parameters. Hence, unfavorable biogeochemical or hydrogeological conditions cannot be mitigated by optimizing irrigation parameters. Although biogeochemical parameters affect only the partitioning of irrigated solute fate across the possible outcomes, hydrogeological parameters may also affect the irrigated solute mass, as more irrigation is needed to maintain target groundwater levels in phreatic aquifers with higher hydraulic conductivities or deeper confining layers. The irrigated solute mass strongly determines contaminant discharge to the environment, but has less effect on crop solute uptake, which is limited by crop water uptake. This study also shows that phreatic zone wastewater irrigation has crop contamination risks that are sensitive to factors different than (near-)surface irrigation techniques, and therefore contributes a meaningful alternative technique for reusing marginal water in irrigation.

1. Introduction

Subsurface irrigation (subirrigation henceforth) and drainage in the phreatic zone is a new method of agricultural water management, that prevents crop water stress caused by insufficient and excessive soil moisture (Tang et al, 2023; Narain-Ford et al., 2022). During dry periods, water is fed directly into the soil through subsurface drains buried in the phreatic zone. This raises the water table, which increases capillary fluxes to the root zone and helps fulfil crop water requirements. During wet periods, the drains can be used to drain away excess water to prevent the waterlogging of crops (de Wit et al., 2022). Furthermore, in the context of wastewater irrigation, subirrigation through the phreatic zone may minimize the risks of crop and environmental contamination associated with irrigation methods that directly introduce irrigated water into the root zone, such as sprinkler or drip irrigation (Narain-Ford et al., 2021).

The soil between the drains and the root zone, and the background groundwater and vadose zone moisture naturally present in the subsurface, act as a buffer separating the crops from the effluent. Many contaminants in the effluent are organic contaminants of emerging concern (CEC) that would undergo adsorption and microbe-induced biodegradation within this buffer zone (Narain-Ford et al., 2022).

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Therefore, the contaminant load of the water that rises to the root zone is smaller than if the wastewater were to be directly applied to the root zone through conventional irrigation techniques such as sprinkler irrigation and (sub)surface drip irrigation (Tang et al., 2024). Furthermore, if the wastewater is rich in nutrients, then it will contribute towards soil fertigation (Ofori et al., 2021), while soil microbes may potentially transform the irrigated nutrients into more bioavailable forms (Melia et al., 2017). After the crop season, the buried drains remove soil water during wet periods and minimizes groundwater level fluctuations, which reduces nutrient leaching from the root zone during the non-crop season (Chen et al., 2022). Although soil microbes biodegrade some of the CECs introduced into the soil through the irrigated wastewater, they are unable to remove non-biodegradable contaminants such as salts and metals. Therefore, drainage after the crop season may also prevent the accumulation of persistent contaminants in the soil (Skaggs et al., 1994). Altogether, the use of subsurface drains placed within the phreatic zone, for irrigation and drainage, may be highly synergistic with the use of wastewater for irrigation.

Treated wastewater is typically discharged to surface waters, which may allow CECs to be transported across the environment by surface water fluxes (Pronk et al., 2021; Jones et al., 2021; Drewes et al., 2017). In dry areas or periods, the treated wastewater may comprise a substantial proportion of surface water fluxes (Luthy et al., 2015; Drewes et al., 2017). In addition to the direct ecotoxicological impacts of discharging treated wastewater to surface water (Hamdhani et al., 2020; Nieto-Juárez et al., 2021), the wastewater-polluted surface water may be harvested by farmers for irrigation (at nearly undiluted concentrations during dry periods) using conventional irrigation techniques that directly expose crops to CECs (Beard et al., 2019; Thebo et al., 2017). Hence, the intentional harvesting and reuse of treated wastewater may help reduce the discharge of wastewater into the environment, and the associated adverse environmental impacts. Furthermore, the dependence of agriculture on alternative water sources such as treated wastewater is expected to increase, yet treated wastewater remains undervalued and underutilized in agriculture, due to (perceived) risks of crop and environmental contamination (Jones et al., 2021; Mesa-Pérez and Berbel, 2020). The ability of the vadose zone soil to bioremediate CECs, and to shield crop roots from CECs due to the physical distance between the irrigated wastewater (in the phreatic zone) and the crop roots (Tang et al., 2024), allows phreatic zone irrigation to directly address one of the main barriers to using reclaimed water for irrigation: the risks of crop contamination (Mesa-Pérez and Berbel, 2020).

Subsurface irrigation and controlled drainage has been part of scientific research for decades (Singh et al. 2022, De Wit et al. 2022, and references therein). It is increasingly being considered as a measure for discharging, retaining and recharging groundwater. Around 35 % of agricultural land in the Netherlands is already underlain by buried drains, originally laid for excess water drainage (de Wit et al., 2022). Phreatic zone irrigation has been found to increase water availability for crop growth (de Wit et al., 2022), with a water-use efficiency that is sensitive to the hydrogeological properties of the subsurface (de Wit et al., 2024). Given the increased pressure on the use of groundwater and surface water resources, and policies to stimulate the use of alternative water resources such as treated wastewater for irrigation purposes (Rizzo et al., 2018), phreatic zone irrigation with treated wastewater is being considered. Although it has not been addressed much by the scientific literature, existing field evidence suggests that most CECs are either immobilized or biodegraded at short distances from the irrigation drains, which suggests low risks of crop and environmental pollution (Tang et al., 2024; Narain-Ford et al., 2022). In the Netherlands, the precipitation surplus during the non-crop season satisfies the leaching requirement for irrigation with water of marginal quality (Tang et al., 2023), thus it is not necessary to intentionally apply freshwater to leach contaminants from the root zone soil (e.g. Letey et al., 2011; Hanson et al., 2008).

Despite results from the preliminary (field) studies discussed above, the environmental risks of phreatic zone irrigation with treated wastewater in other geographical locations with different hydroclimatic conditions and hydrogeological properties have yet to be sufficiently characterized. There is still no general study on the environmental fate of CECs irrigated into the phreatic zone, and its sensitivity to various hydroclimatic and hydrogeological parameters. Therefore, the objective of this study is to perform a sensitivity analysis of effluent solute fate, under varying hydrogeological properties, irrigation parameters, atmospheric fluxes, and contaminant biogeochemical behavior, using a field scale physical model developed in a previous study (Tang et al., 2023). This would yield further insight on the circumstances under which phreatic zone irrigation with treated wastewater is associated with acceptable risks of crop and environmental contamination, so that its potential for implementation beyond the experimental plot may be evaluated.

2. Methods

2.1. Numerical model

In a recent study (Tang et al, 2023), we described an experimental plot of the phreatic zone irrigation system in the Netherlands, that was irrigated with treated domestic wastewater. Based on the experiment, we constructed a numerical model to characterize the system, calibrated the model to a generic conceptualization of the experimental scenario, and described the fate of tracers and solutes that the effluent contains. The solute transport model for the subsurface irrigation system was created with HYDRUS-2D (Šimůnek et al., 2016). The model domain (Fig. 1a), finite-element mesh (Fig. 1b), and the parameter values used in the base configuration of the model, are identical to that in Tang et al (2023). A brief description of the model setup is given below.

An atmospheric boundary that represents precipitation is situated at the top of the model domain. If the precipitation rate is larger than the soil's infiltration capacity, then the difference is assumed to be surface runoff and removed from the model (Šimůnek et al., 2016). During the crop season, the daily potential evapotranspiration rates were computed by multiplying the reference evapotranspiration rates with weekly crop factors for maize crops (LAGO, 1984). During the non-cropping season, the potential evapotranspiration rates were set equal to the reference evapotranspiration rates. The spatial distribution of crop roots has a maximum intensity at depth $z = 0.05$ m, and thereafter an exponentially decreasing intensity with depth until the bottom of the root zone, which is at a depth of 0.6 m. The potential evapotranspiration rate is partitioned across each node in the root zone proportionally to the root density at each node (Šimůnek and Hopmans, 2009). Then, the actual evapotranspiration at each node is determined with the Feddes reduction function, with parameter values corresponding to maize (Wesseling et al, 1991). Two irrigation drains are present in the simulated subsurface at a depth of 1.2 m, 6 m apart from each other. The irrigation drains are modelled as circular openings with 4 cm internal diameters and 8 cm external diameters. The modelled crop seasons span 150 days starting from the first of May each year. The irrigation drains are imposed pressure head boundaries during the crop season, and 'seepage face' boundary conditions otherwise. The bottom boundary has a deep drainage boundary condition, which represents an imposed flux whose magnitude depends on the groundwater level (Hopmans and Stricker, 1989) and is calculated as $q = A\exp(B|h - Z|)$ m/day, where the variable h is the hydraulic head at the boundary, and Z,A,B are constant parameters. The direction of regional groundwater flow is from left to right. The left (upstream) boundary for groundwater flow has an imposed head gradient. The right (downstream) boundary is a Cauchy boundary condition comprised of a prescribed groundwater level and a column of aquifer material with a prescribed hydraulic conductivity that can be varied independently from the rest of the aquifer. The initial conditions for moisture content were set to hydrostatic equilibrium

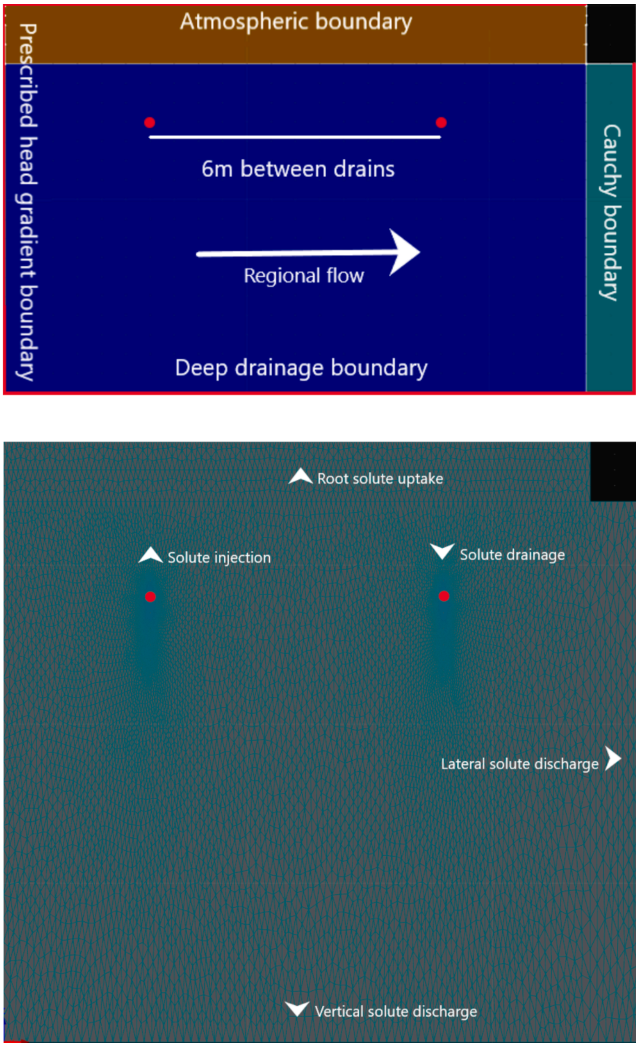

Fig. 1. (a) The two-dimensional numerical model domain showing the locations of the drains and the boundary conditions used. The root zone is contained within the brown region. The green region is part of the downstream Cauchy boundary, and has a different saturated conductivity than the rest of the aquifer (blue region). Regional groundwater flow flows from left to right. (b) The finite-element mesh of the numerical model. The various possible solute fluxes into and out of the simulated domain are indicated. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

relative to a water table depth of 1 m. In the simulations, the background concentration of the injected solute is 0 mmol/m^2, and the irrigated concentration is set to 1 mmol/m^2 as an example, but results apply to other input concentrations as the solute transport processes simulated are not concentration-dependent. Note that volumes are in m^2 because the model is two-dimensional.

Contaminant transport outcomes are quantified in terms of solute mass balances across the various environmental compartments, including crop solute uptake, leaching to deeper depths beneath the simulated domain, drainage into the subsurface drains, and lateral advection through the phreatic zone, as illustrated in Fig. 1b. In addition, given the limited size of the simulated domain, the travel directions of solutes beneath the agricultural field are characterized with a proxy indicator, the ratio of total horizontal to vertical solute discharge (H/V ratio) from the model domain. Horizontal solute discharge implies lateral advection out of the simulated domain, while vertical solute discharge implies transport deeper into the subsurface. A scenario with a larger H/V ratio has a plume that travels in a more horizontal direction, and a scenario with a smaller H/V ratio has a plume that travels in a more vertically downwards direction. In this way, we investigate the general sensitivity of the mean direction of solute advection to variations in model parameters, in a manner that generally holds for all field sizes.

2.2. Sensitivity analyses

The base model scenario we use for the sensitivity analysis characterizes essentially the subirrigation of a sandy-loamy soil under a temperate climate with an annual precipitation excess but temporary

Table 1
List of parameter values for the best model. Parameter values for the base model are obtained from Tang et al (2023).

Parameter	Base model value	Comment
Precipitation [mm/day]	Daily actual at field site, 2016 – 2020, total = 3152 mm	Atmospheric boundary (top)
Evapotranspiration [mm/day]	Daily reference evapotranspiration at field site, 2016 – 2020, modified with crop factor for maize (LAGO, 1984) total = 1916 mm	Atmospheric boundary (top)
Regional head gradient [m/m]	0.0014	Prescribed head gradient boundary (left)
θr [–] Residual saturation	0.01	
θs [–] Maximum saturation	0.42	
α[1/m] van Genuchten soil water retention parameter	2	
n [–] van Genuchten soil water retention parameter	1.5	
Saturated conductivity of soil shallower than 0.6 m $K_{s,s}$ [m/day]	2	Root zone soil
Saturated conductivity of aquifer $K_{s,a}$ [m/day]	5	Non-root zone soil
L [–] Tortuosity parameter	0.5	
Z[m] (Deep drainage parameter)	0	Deep drainage boundary (bottom)
A[m/day] (Deep drainage parameter)	0.0025	Deep drainage boundary (bottom)
B[1/m] (Deep drainage parameter)	−1.250	Deep drainage boundary (bottom)
Water table depth at downstream boundary [m]	1.6	Cauchy boundary (right)
Conductivity of downstream boundary [m/day]	0.02	Cauchy boundary (right)
Irrigation drain conductivity [m/day]	0.025	
Irrigation pressure [m]	0.3	
Drainage backpressure [m]	0.3	
Longitudinal dispersivity D_l [m]	0.2	
Transverse dispersivity D_t [m]	0.02	
Soil bulk density ρ [kg/m^3]	1.5	
Adsorption coefficient [m^3/kg]	0	
Biodegradation rate in the aqueous phase [1/day]	0	
Biodegradation rate in the adsorbed phase [1/day]	0	

precipitation shortage during the crop season, with moderate regional groundwater fluxes (see Table 1 for parameter values). Sensitivity analyses are performed on the base model by varying 1 – 3 thematically related parameters simultaneously. We perform sensitivity analyses of the model parameters, localized around the base model parameter values (Table 1), on various sets of simultaneously varied parameters belonging to common themes (Table 2). The parameter ranges encompass a wide range of soil types, geological compositions, and hydrogeological situations. In the sensitivity analyses where multiple parameters are simultaneously varied, all possible combinations are simulated.

Daily precipitation and reference evapotranspiration data from the Twenthe weather station of the Dutch meteorological institute (KNMI) are used to model the atmospheric fluxes. Since the base model achieves annual periodic steady-state after four years, most of the variation in CEC fate arising from atmospheric variability is expected to materialize during the four-year start-up phase (Tang et al., 2023). Hence, we simulate every possible period of four years (i.e. four crop – drainage cycles) that occurs between 1990 and 2020, for a total of 27 simulations. This is sufficient to characterize long-term average behavior, while also revealing the variability that may occur during the start-up period where most variability is expected to occur.

Table 2
List of sensitivity analysis themes where one or multiple model parameters are varied in combination.

Sensitivity analysis theme (Shortened theme name in brackets)	Parameters varied:	Range
Atmospheric fluxes (RainET)	Precipitation [mm/day] and evapotranspiration [mm/day]	Daily timeseries from 1990 to 2020.
Lateral inwards flux (Grad)	Regional head gradient [m/m]	0 – 0.0035
Downstream boundary (Right)	Downstream boundary conductivity [m/day]	(0.005, 0.01, 0.02, 0.04, 0.08, 0.16, 0.32, 0.64, 1.25, 2.5, 5, 10)
Deep drainage boundary (Deep)	A [m/day]	(−0.00125, −0.0025, −0.005)
	B [1/m]	(−1, −1.25, −2, −5)
Root zone soil hydraulic properties (Soil)	$K_{s,s}$ [m/day]	(0.05, 0.5, 5)
	α[1/m]	(0.5, 1, 2, 3)
	n[−]	(1, 1.5, 2.5)
Aquifer hydraulic properties (Cond)	$K_{s,a}$ [m/day]	(1, 5, 25)
	α[1/m]	(0.5, 1, 2, 3)
Mechanical dispersivity (Disp)	D_l[m]	(0.02, 0.2, 2)
	D_t[m]	(0.002, 0.02, 0.2, 2)
Biogeochemical parameters $\lambda_a = 0$ (Chem)	K_d [L/kg]	10^(−2, −1, 0, 1, 2, 3)
	λ_s [1/day]	10^(−3.5, −3, −2.5, −2, −1.5, −1)
Biogeochemical parameters with $\lambda_a = \lambda_s$ (ChemS)	K_d [L/kg]	10^(−2, −1, 0, 1, 2, 3)
	λ_s [1/day]	10^(−3.5, −3, −2.5, −2, −1.5, −1)
Irrigation pressure (Irr)	Irrigation pressure [m]	0.1 – 0.6
Drainage backpressure (Drain)	Drainage backpressure [m]	0 – 0.5
Irrigation pipe conductivity (Pipe)	Irrigation pipe conductivity [m/day]	(0.005 0.025 0.125 0.625 3.125 15.625)

The range of adsorption coefficients K_d and solute phase biodegradation rates λ_s used in the analysis on biogeochemical parameters are representative of a wide variety of CECs (Williams et al., 2009; Kodesova et al 2016; Nham et al., 2015). However, the ability for biodegradation to occur in the adsorbed phase is uncertain (Poeton et al., 1999; Gamerdinger et al., 1997; Scow and Johnson, 1996; Woo et al., 2001). Therefore, we perform the analysis of biogeochemical parameters in two separate sets of simulations: one without and one with adsorbed phase biodegradation, setting the adsorbed phase biodegradation rate to $\lambda_a = \lambda_s$. The simulated solute transport scenarios represent a wide range of possible contaminants, because the biogeochemical properties of the solute (adsorption coefficient and biodegradation rate) are varied across multiple orders of magnitude in the sensitivity analyses. For the base model of the sensitivity analysis, we model the irrigated solute as a biogeochemically non-reactive tracer, as tracers conservatively reflect the maximal spatial extent of environmental contamination by wastewater irrigation.

3. Results

3.1. Atmospheric fluxes

Across the simulations with different atmospheric flux timeseries, root solute uptake is strongly positively correlated with the net solute mass injected (total injected minus drained through the subirrigation drains) (Fig. 2a) and strongly negatively correlated with the excess precipitation (rainfall minus actual evapotranspiration for each four year period) (Fig. 2b). This is because the net solute mass injected is strongly negatively correlated with the excess precipitation. Note that the total solute mass injected scales proportionally with the irrigation water use, because the irrigated solute concentration is constant. Therefore, the larger the precipitation excess (i.e. more precipitation, planting crops with lower water requirements), the lower the crop contamination risk. Fig. 2c shows that the solute mass drained away by the irrigation drains is a very small fraction of the total solute influx, thus the total and net solute mass injected are very similar in all simulations. This close relationship between solute mass injected and root solute uptake enables the prediction of relative crop contamination risks over multiple years based on irrigation volume data.

Unlike the root solute uptake, the H/V ratio is weakly correlated with the solute mass injected, and essentially uncorrelated with the excess precipitation implying that wetter conditions increase the horizontal and vertical solute discharge in the saturated zone by similar extents. Fig. 2d shows that the horizontal and vertical solute discharges in the saturated zone, and the H/V ratio, are relatively insensitive to the atmospheric flux timeseries. The horizontal solute discharge remained within 5 % of the mean of around 15000 mmol in all 27 four-year simulations, while little vertical solute discharge occurred in all cases. We note that the peak in solute injected and root solute uptake in Fig. 2c in the simulation beginning in 2016 reflects the influence of the extremely dry year 2018.

A correlation matrix of outcomes across the 27 starting years, shows that the drained solute mass is positively correlated with the irrigated solute mass (Fig. 3): more solute is available to be drained away if more solute is irrigated. Indeed, the drained solute mass increases more than linearly in proportion to the increase in irrigated solute mass, similarly to other forms of managed aquifer recharge (Tang and van der Zee, 2021). In contrast, the drained water volume is uncorrelated with the irrigated water volume, because the irrigated water volume accounts for a small proportion of the overall water balance of the system.

The correlation matrix (Fig. 3) also shows that the root solute uptake and lateral solute discharge is strongly positively correlated to the

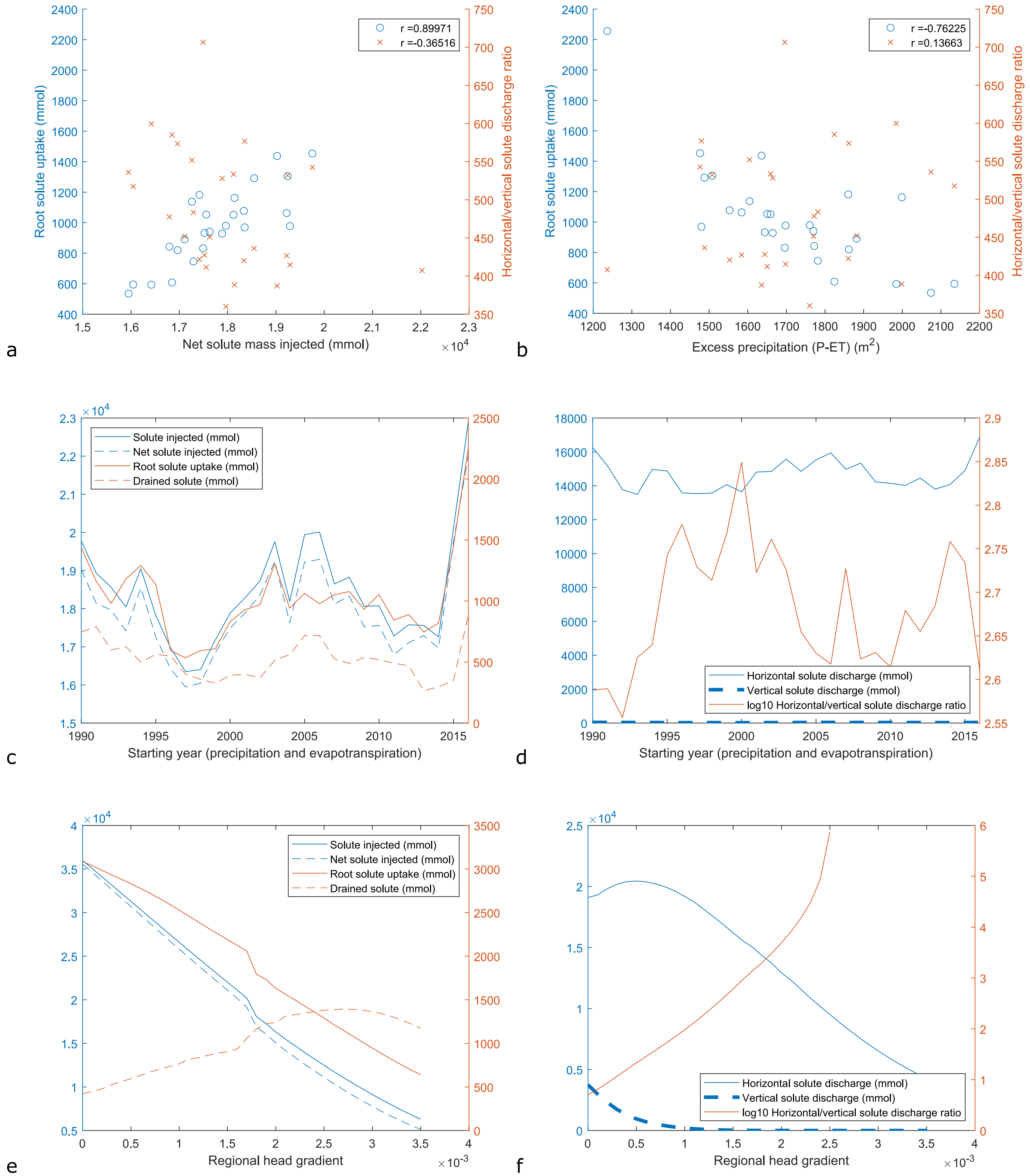

Fig. 2. Scatter plot of root solute uptake and the H/V ratio against the (a) net solute mass injected and (b) excess precipitation, in the sensitivity analysis of atmospheric fluxes. (c) Root solute uptake and drainage, and (d) saturated zone solute discharge outcomes as a function of the starting year of the atmospheric flux timeseries. (e) Root solute uptake and drainage, and (f) saturated zone solute discharge outcomes as a function of the regional head gradient.

irrigation volume and negatively correlated to the excess precipitation, while the solute mass drained is positively correlated with the volume of water drained. Hence, these important solute fate outcomes are strongly correlated to easily measurable quantities. However, the vertical solute discharge and H/V ratio, which are strongly correlated with each other, are not strongly correlated to any easily observable quantities. The H/V ratio and vertical solute discharge are not correlated with the amount of water drained, but is moderately correlated with the solute mass drained. This is because when the average direction of solute transport is more strongly vertical and less strongly horizontal, the drains are more

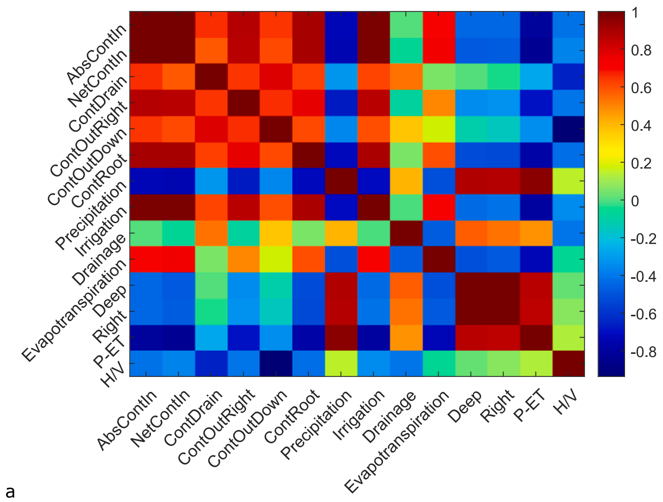

Fig. 3. Correlation matrix of simulation outcomes for the sensitivity analyses over the 27 four-year atmospheric flux time series. AbsContIn = absolute injected solute mass; NetContIn = net injected solute mass (injected – drained), ContDrain = drained solute mass, ContOutRight = horizontal solute discharge, ContOutDown = vertical solute discharge, ContRoot = total root solute uptake, Precipitation = total rainfall volume, Irrigation = total irrigation volume, Drainage = total drainage volume, Evapotranspiration = total evapotranspiration volume, Deep = vertical water discharge, Right = horizontal water discharge, P-ET = excess precipitation, H/V = H/V ratio (ContOutRight/ContOutDown).

likely to intercept the solute plumes that previously rose to and subsequently descend from the unsaturated zone.

3.2. Regional head gradient

Increasing the regional head gradient decreases the volume of water and solute mass injected (Fig. 2e). It also increases the mass of solutes recovered by the drains, except at very high regional head gradients where injected solutes are immediately advected away from the drains. Although it might be expected that a larger regional head gradient predominantly leads to a smaller recovered solute fraction due to solutes being displaced horizontally away from the drains, the larger regional head gradient also raises the water table higher, thereby pushing effluent upwards. Therefore, the effluent may be more easily intercepted by the drains as it subsequently moves downwards due to gravity and excess precipitation, thereby increasing the recovery fraction. Thus, the solute fraction taken up by crop roots and drained away is higher with high regional head gradient, but the absolute mass of root solute uptake is higher with low regional head gradient, because the total irrigation volume is much larger with a low regional head gradient. The lateral and horizontal solute discharge both decrease as the regional head gradient increase, because less effluent is irrigated. As the regional head gradient increases, the vertical solute discharge decreases faster than the lateral solute discharge, causing the H/V ratio to greatly increase (Fig. 2f).

3.3. Boundary parameters

3.3.1. Lateral discharge resistance

Increasing the conductivity of the Cauchy boundary condition at the right boundary greatly increases the amount of solutes injected, root solute uptake (Fig. 4a), and the amount of horizontal and vertical solute discharge (Fig. 4b). This is because higher irrigation fluxes are necessary to maintain the groundwater level at the target position, as the high boundary conductivity causes irrigated water to be quickly removed from the domain. When the right boundary conductivity is large, further increases do not increase the amount of solute injected and discharged any further, because then solute injection is limited by the irrigation drain's conductivity.

3.3.2. Vertical discharge resistance

As the two deep drainage boundary parameters are varied, the amount of vertical water discharge changes. The most direct effect of increasing the vertical water discharge is that the solute mass injected and root solute uptake increases because more irrigation is required to maintain the groundwater level at its target position (Fig. 4c). However, the amount of solutes drained decreases, because the natural equilibrium groundwater level decreases, resulting in less water drained, and also causing solutes to sink downwards more quickly. Furthermore, the vertical solute discharge increases, while the H/V ratio decreases (Fig. 4d).

3.4. Irrigation parameters

3.4.1. Irrigation pressure

As the irrigation pressure increases, the injected water, injected solute mass, drained solute mass, and root solute uptake increases (Fig. 5a). As the irrigation pressure increases, the horizontal and vertical saturated zone solute discharge increase and the H/V ratio decreases (Fig. 5b). Thus, as more water is irrigated, the fraction of contaminant that sinks downwards increases more, compared to contaminant that is advected laterally.

3.4.2. Drainage backpressure

Imposing a higher drainage backpressure (by lowering the drainage crest in the SSI system, or due to clogging of the drains) greatly reduces the amount of solutes drained, thereby greatly increasing the net injected solutes. While the drained solute mass decreases from 3600 mmol to 400 mmol when the drainage backpressure increases from 0 m to 0.5 m, the root solute uptake increases by merely 250 mmol or 10 % (Fig. 5c). The drainage backpressure hardly affects root solute uptake because most of the solutes that come within the capture zone of the drains do so during the drainage season, on their way downwards from the soil above drain level. Hence, the decrease in solutes drained due to higher drainage backpressures is mostly compensated by the increase in saturated zone solute discharge (Fig. 5d). The H/V ratio appears to be independent of the drainage backpressure, because the drainage volume is in all scenarios an order of magnitude smaller than the environmental flow or irrigation fluxes.

3.4.3. Drain conductivity

At low drain conductivities, an increase in drain conductivity increases root solute uptake because more effluent is injected. However, at high drain conductivities, an increase in drain conductivity does not cause more effluent to be injected because lower drain conductivities are already sufficient to maintain the target groundwater level (Fig. 5e). At higher drain conductivities, further increasing drain conductivity decreases root solute uptake. When the excess precipitation is large, highly conductive drains quickly remove large volumes of effluent from the root zone, thus effluent residence times in the root zone become shorter. As drain conductivity increases, the saturated zone horizontal and vertical solute discharges also increase (Fig. 5f).

3.4.4. Drain depth and spacing

In addition to the analyses of irrigation parameters listed in Table 2, the 27 simulations with different atmospheric flux time series were repeated for irrigation drains placed at a depth of 1.8 m, which is 0.6 m lower than in the base model, and separately, also for drains spaced 12 m apart, which is double the spacing of the base model. With drains located deeper than in the base model, the maintained groundwater level, and the amount of water and solute irrigated and drained remain similar to the base model. Root solute uptake is approximately halved, because when the irrigation drains are buried deeper, a larger portion of water reaching the root zone originates from the regional groundwater flow.

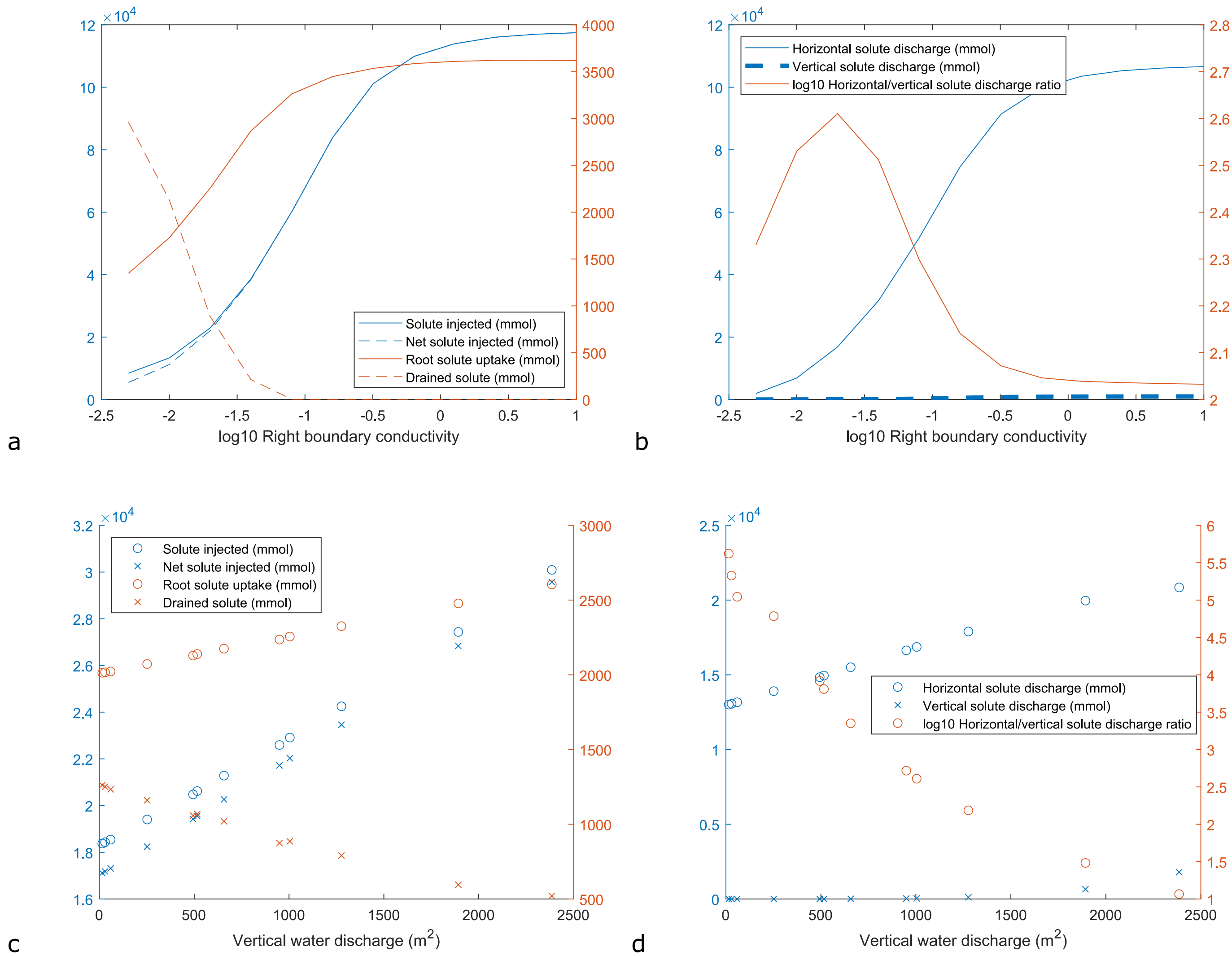

Fig. 4. (a) Root solute uptake and drainage, and (b) saturated zone solute discharge outcomes as a function of the right boundary conductivity. (c) Root solute uptake and drainage, and (d) saturated zone solute discharge outcomes as a function of the vertical water discharge (due to variations in the deep drainage boundary parameters).

However, as a tradeoff, the amount of downwards vertical solute discharge increased by about 10 %, as the irrigation drains are now closer to the bottom boundary.

With doubly wide spacing between drains, the amount of water and solute irrigated per unit area of the agricultural plot is about 25 % smaller than in the base model, because the irrigation flux at each drain is limited by the drain conductivity. This causes the maintained groundwater level to be lower than in the base model, which may reduce crop yields, unless the irrigation pressure is increased to compensate for this. The amount of root solute uptake per unit field area decreased by 30–50 % with doubly wide spaced drains, depending on the starting year of the atmospheric flux timeseries. Similarly to the base model (Tang et al., 2023), most root solute uptake occurs immediately above the irrigation drains.

3.5. Hydrogeological parameters

3.5.1. Root zone soil hydraulic properties

Root zone soil hydraulic parameters were found to weakly affect outcomes. A distinct trend in increasing vertical solute discharge is observed as the soil texture moves from clayey to loamy to sandy, based on variations in the root zone soil hydraulic parameters. This trend is associated with increasing horizontal water discharge, decreasing drained solute mass, and slightly increasing root solute uptake (Fig. 6a; Fig. 6b). This is because sandy soils are less water retentive, thereby allowing quicker downwards discharge of water from the root zone, consequently requiring more irrigation.

Three outlier soil types are visible in the data: these are soils with large α, large n, and low $K_{s,s}$. The van Genuchten parameter α is an increasing function of the modal pore size, while n is related to the pore size distribution variance (Ghezzehei et al., 2007). These outlier soils have hydraulic properties that are characteristic of (hydrophobic) coarse sandy soils (e.g. Lamparter et al., 2006). Very little capillary rise occurs in these soils, which reduces actual evapotranspiration (i.e. crop yield), and consequently also reduces crop solute uptake. The large reduction of actual evapotranspiration decreases the total volume of water and solute mass injected and taken up by crops. As actual evapotranspiration is reduced, the average downwards vertical velocity increasing, thereby decreasing the vertical solute discharge and increasing the H/V ratio.

3.5.2. Aquifer hydraulic properties

Varying the aquifer water retention parameter α has minimal effect on contaminant fate, as its influence is mainly confined to the unsaturated zone, where total fluxes are much smaller than the saturated zone. As the hydraulic conductivity of the aquifer increases, the natural groundwater level rises because the incoming regional flow flux from

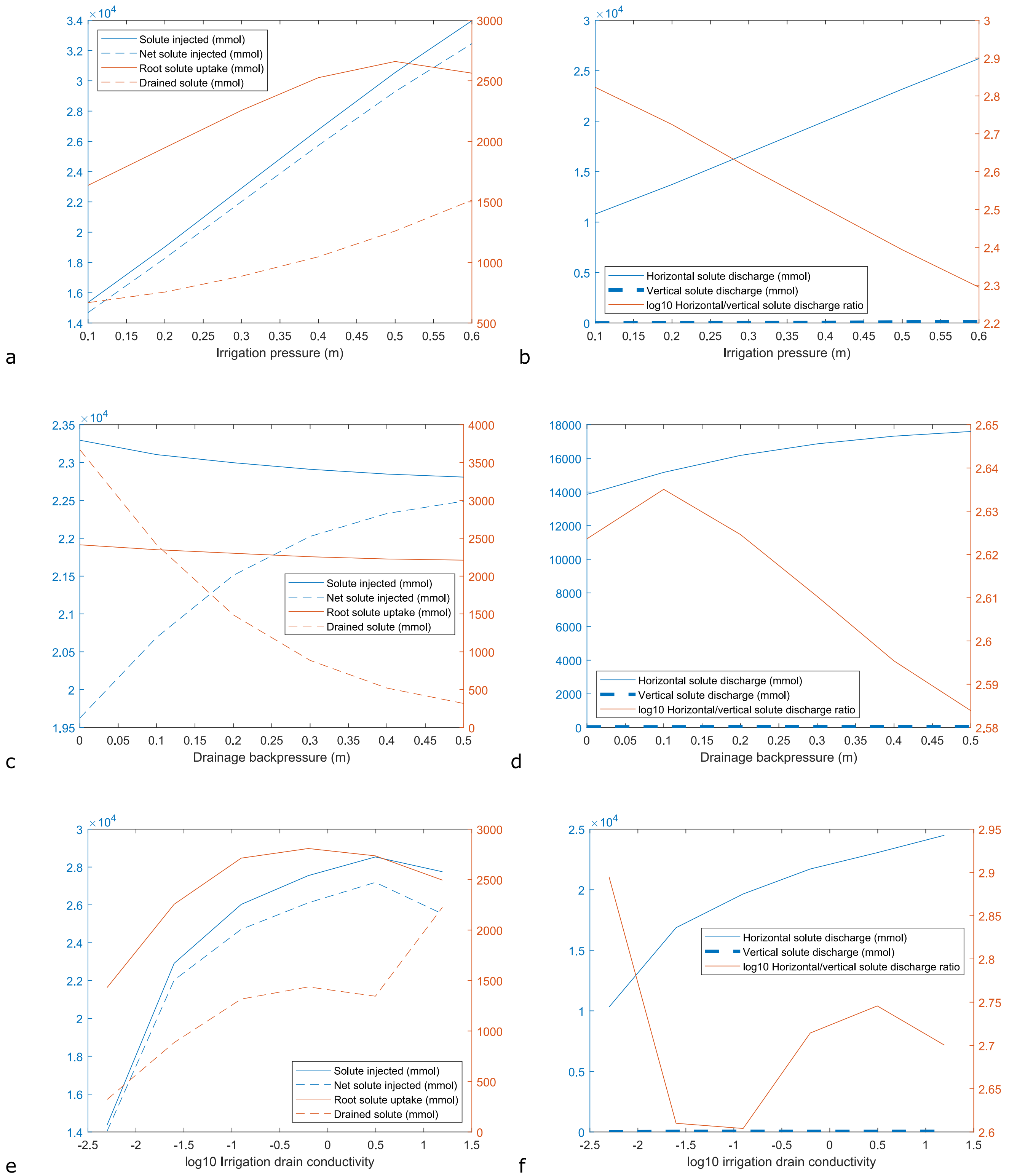

Fig. 5. Root solute uptake and drainage as a function of (a) irrigation pressure, (c) drainage backpressure, and (e) irrigation drain conductivity. Saturated zone solute discharge outcomes as a function of (b) irrigation pressure, (d) drainage backpressure, and (f) irrigation drain conductivity.

the imposed head gradient boundary increases. Therefore, as the aquifer hydraulic conductivity increases, the amount of solute injected and root solute uptake decreases significantly (Fig. 6c). The vertical solute discharge decreases while the H/V ratio increases as the conductivity increases (Fig. 6d), because more horizontal groundwater flow occurs. The downwards flux however is not significantly affected due to the presence of the deep drainage (i.e. head-dependent imposed flux) bottom boundary. At a conductivity of 25 m/day, the natural groundwater level is high enough that no irrigation flux occurs for most of the simulated period, resulting in negligible solute injection.

3.5.3. Mechanical dispersivity

Capillary fluxes bring irrigated effluent upwards to the root zone, alternating with periods where precipitation excesses push root zone

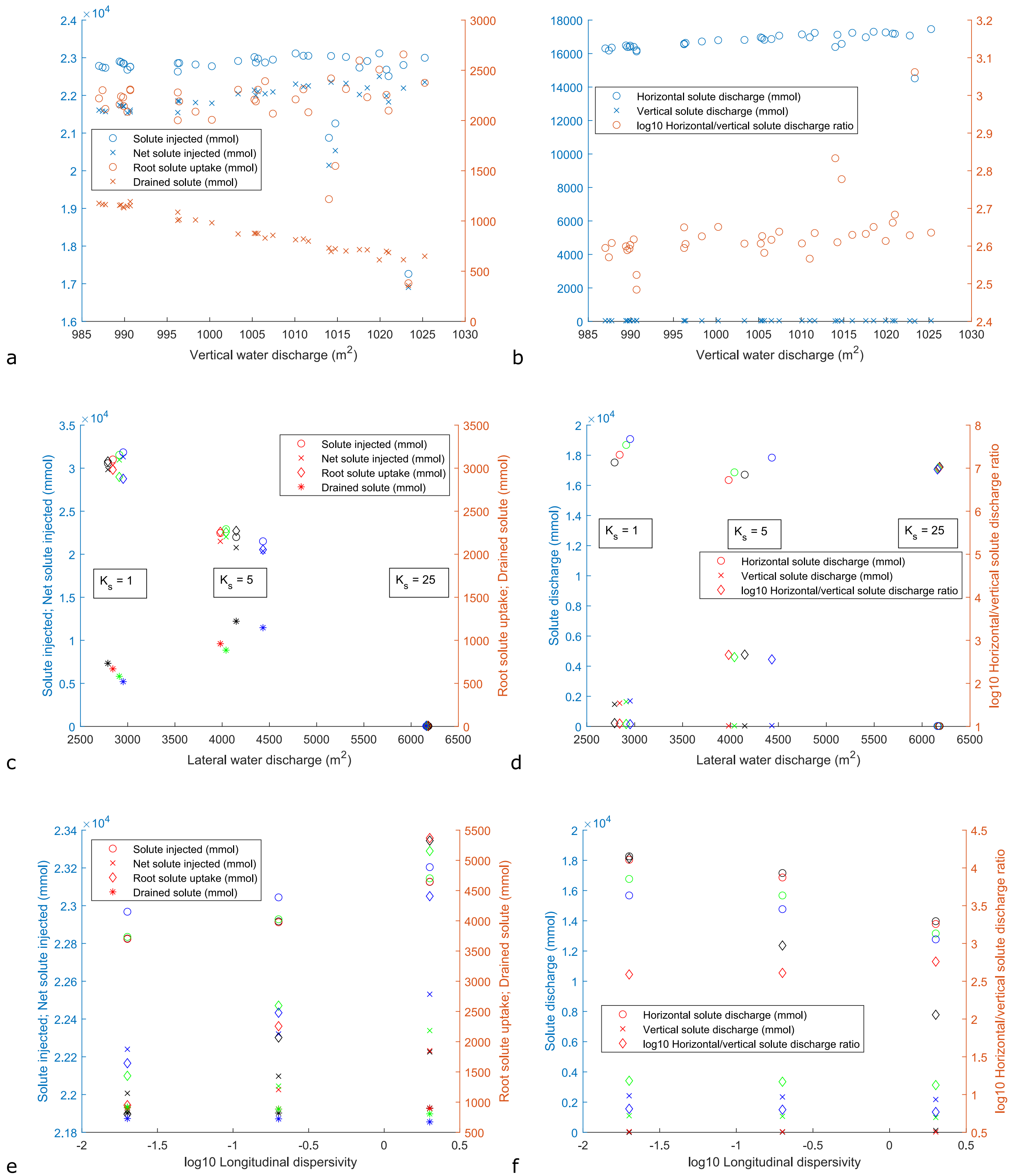

Fig. 6. (a) Root solute uptake and drainage, and (b) saturated zone solute discharge outcomes as a function of the vertical water discharge (due to variations in the root zone soil hydraulic parameters). (c) Root solute uptake and drainage, and (d) saturated zone solute discharge outcomes as a function of the horizontal water discharge (due to variations in the aquifer hydraulic properties) [Colors black, red, green, blue correspond to $\alpha = 0.5$, 1, 2, 3 respectively]. (e) Root solute uptake and drainage, and (f) saturated zone solute discharge outcomes as a function of the mechanical dispersivity [Colors black, red, green, blue correspond to $D_t = 0.002$, 0.02, 0.2, 2 respectively]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

effluent downwards in a largely one-dimensional manner. Therefore, an increase in longitudinal dispersivity significantly increases solute dispersion to the root zone and hence root solute uptake (Fig. 6e). In contrast, transverse dispersivity does not have such a significant impact on solute fate.

An increase in longitudinal dispersivity decreases the horizontal solute discharge but appears to have little effect on the vertical solute discharge (Fig. 6f). Note that the decrease in horizontal solute discharge resulting from an increase in longitudinal dispersivity is of a similar magnitude as the resulting increase in crop solute uptake. Generally, increasing the transverse dispersivity decreases the horizontal solute discharge but increases the vertical solute discharge, thereby decreasing the H/V ratio. This is because the primary direction of advection in the saturated zone is horizontal. The solute mass injected increases very slightly as either of the dispersivities increase, because of the concentration-flux solute boundary condition at the irrigation drains that allows solutes to not only be advected, but also be dispersed from the drains into the soil.

3.5.4. *Confining layer depth*

In this study, we have used a deep drainage boundary condition (DDBC) at the bottom of the simulated domain, which may represent the presence of a confining layer at that position (Hopmans and Stricker, 1989). In order to explicitly investigate how the depth of the confining layer could affect solute fate, the 27 simulations with different atmospheric flux time series were repeated in scenarios where the DDBC is placed at 2 m, 6 m, 8 m depths, for comparison with the base scenario with the DDBC at 4 m depth. When the DDBC is shallow, it retains water in the shallow aquifer, as it is relatively hydraulically non-conductive. When the DDBC is deeper, there is more space for the irrigated water to flow out of the subsurface of the agricultural field by horizontal discharge through the phreatic zone, which limits the extent to which the confining layer can help retain water in the phreatic zone, therefore requiring more irrigation to maintain the water table at target levels (Fig. 7a). The result is that as the DDBC becomes deeper, increased contaminant loading in the phreatic zone causes the root solute uptake to increase (Fig. 7b). In addition, the vertical solute discharge in the saturated zone decreases (Fig. 7c), the solute mass drained by the irrigation pipes decreases (Fig. 7c), and the horizontal solute discharge in the saturated zone increases (Fig. 7c) as the DDBC becomes deeper. For scenarios where the DDBC were to be even deeper than 8 m depth, the outcomes can be qualitatively extrapolated from our findings: more wastewater will have to be irrigated to raise the water table, leading to more crop solute uptake as a result, but solutes leaching through the confining layer will remain the same (i.e., essentially zero) (Fig. 7b).

In the base scenario, vertical solute discharge through the 4 m deep DDBC accounts for 0.1–0.2 % of injected solute mass. When the DDBC is moved to 2 m depth, the vertical solute discharge fraction increases hundredfold (Fig. 7c) to around 22–25 % of injected solute mass. When the DDBC is lowered from 4 m depth to 6 m or 8 m depth, the vertical solute discharge decreases 4–8 orders of magnitude to essentially zero (Fig. 7c). Hence, root solute uptake increases linearly as the confining layer becomes deeper (Fig. 7b). In contrast, solute mass leached through the confining layer decreases exponentially as the confining layer becomes deeper, and vice-versa. For scenarios where the DDBC were to be even deeper than 8 m depth, the outcomes can be qualitatively extrapolated from our findings: more wastewater will have to be irrigated to raise the water table, leading to more crop solute uptake as a result, but solutes leaching through the confining layer will remain the same (i.e., essentially zero). Therefore, with a confining layer deeper than 4 m, essentially all the solutes that are not taken up by crop roots exit the simulated domain by horizontal discharge through the saturated zone. Hence, the depth of the confining layer plays an instrumental role in the tradeoff between crop contamination and confined aquifer contamination. Given that one of the aims of phreatic zone wastewater irrigation is to facilitate contaminant bioremediation in the phreatic and vadose

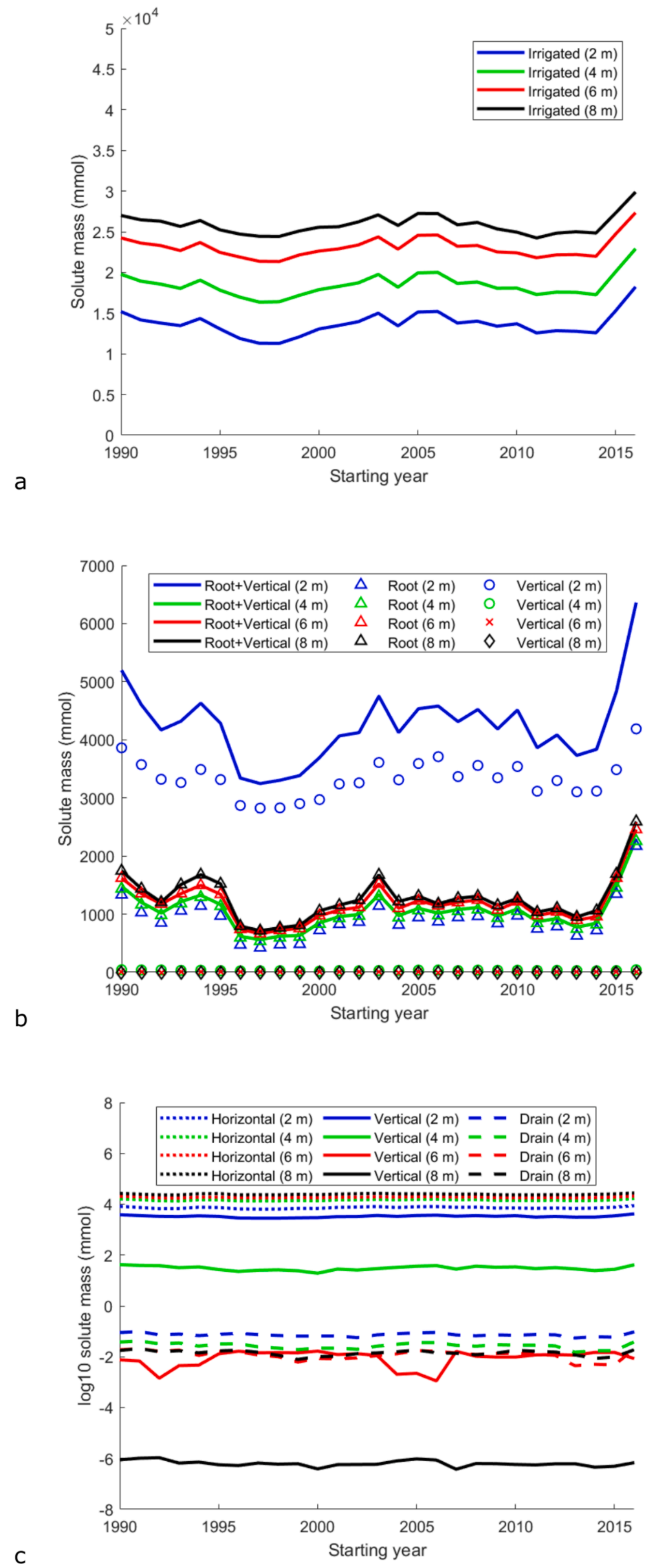

Fig. 7. Effects of moving the deep drainage boundary condition (DDBC) from 4 m depth (the base scenario) to 2, 6, or 8 m depth, on solute fate. (a) The irrigated solute mass. (b) The absolute root solute uptake, solute mass leached vertically through the confining layer, and the combined total root solute uptake and leached solute mass. (c) Logarithm of the solute mass discharged horizontally through the phreatic zone, leached vertically through the confining layer, and drained by the irrigation pipes.

zones, while minimizing crop and confined groundwater contamination, this tradeoff could be an important design parameter for phreatic zone wastewater irrigation systems. We find that for the scenarios we simulated, the combined total (of root solute uptake and solute leached through the confining layer) may be minimized at some intermediate confining layer depth, which is at approximately 4 m depth (Fig. 7b).

3.6. Biogeochemical parameters

Solute fate is highly sensitive to the biogeochemical parameters, regardless of whether the solute is able to biodegrade in the adsorbed phase. The fraction of injected solute drained by the irrigation drains is a non-monotonic function of the adsorption coefficient, with a maximum around $K_d = 10$ (Fig. 8a, b). When K_d increases from 0.01 to 10, the drained fraction increases because the effluent plume remains closer to

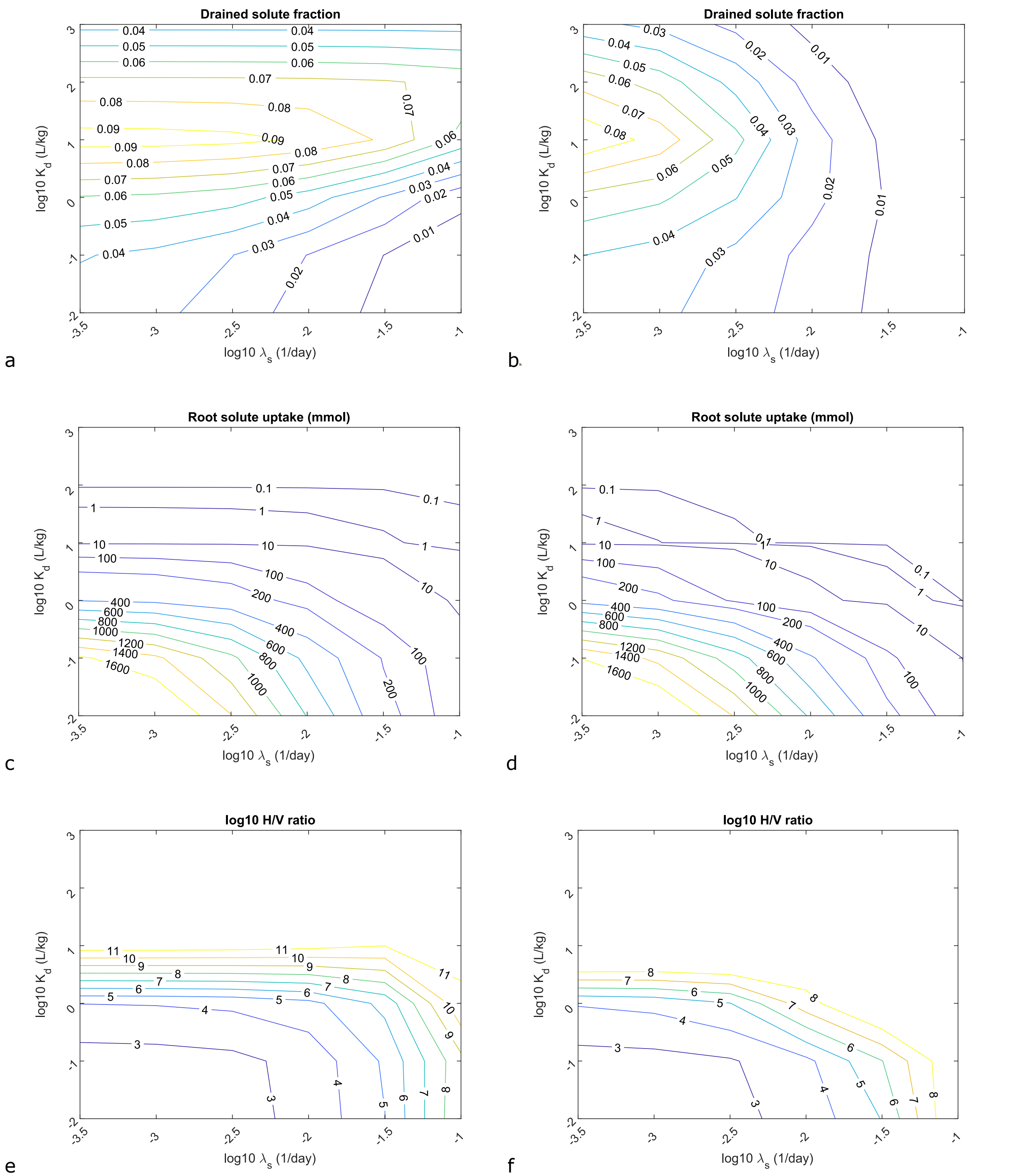

Fig. 8. Contour plots of the (a) drained solute fraction, (c) root solute uptake, and (e) H/V ratio as a function of the biodegradation rate and adsorption coefficient, for scenarios with biodegradation only in the solute phase. The same is illustrated in (b,d,f) for scenarios with biodegradation in the solute and adsorbed phases.

the irrigation drains, such that solute-rich water is preferentially drained whenever drainage occurs. When K_d increases beyond 10, the drained water contains very little contaminant, as most of the contaminant although situated near the drains are in the adsorbed phase, thereby decreasing the drained solute fraction.

Root solute uptake is a monotonically decreasing function of both K_d and λ_s (Fig. 8c, d), which is a straightforward consequence of the fact that less mobility and increased biodegradability decreases the solute mass reaching the root zone. The H/V ratio is also a monotonically decreasing function of both K_d and λ_s (Fig. 8e, 8f). The H/V ratio increases as λ_s increases because the contaminant travel time from the drains to the right boundary is shorter than the travel time to the bottom boundary, hence solute particles moving downwards have more time to biodegrade.

The H/V ratio increases as K_d increases partly because solute retardation magnifies the aforementioned travel time difference between rightwards and downwards travelling solute particles, therefore giving solutes moving downwards more time to biodegrade. However, the same pattern is observed in test simulations with no biodegradation, which warrants an additional explanation. Increased solute residence times due to adsorption causes a higher order effect that further increases solute residence times. During the drainage season, water and solutes in the subsurface generally travel downwards due to low potential evapotranspiration rates. If injected solute has not exited the domain by the start of the following year's crop season, then the water and solutes within the subsurface begin to move upwards again, due to large precipitation shortages that result in net upwards flow. This causes the position of the solute plume to oscillate up and down, thereby significantly reducing downwards solute discharge. Conversely, the solute plume's direction of travel along the horizontal axis is always rightwards along with regional flow. In summary, solute retardation causes an increase in solute residence times, during which its average downwards velocity decreases more significantly than its average rightwards velocity, thereby increasing the H/V ratio.

The ability of solutes to biodegrade in the adsorbed phase leads to more biodegradation, less solute drainage, less root solute uptake, and a higher H/V ratio. The increase in total biodegradation due to adsorbed phase biodegradation is largest for large K_d, because more contaminant exists in the adsorbed phase, and small λ_s, because the biodegraded mass as a function of time spent biodegrading has diminishing returns. Root solute uptake and the drained solute mass are reduced by biodegradation in the adsorbed phase, especially for large K_d and large λ_s, relative to the scenario with no adsorbed phase biodegradation. The H/V ratio is increased by adsorbed phase biodegradation, especially for large K_d and large λ_s, because the additional biodegradation amplifies the reduction in vertical solute discharge discussed in the previous two paragraphs.

4. Discussion

4.1. Field scale solute fate sensitivity

The analyses, which were conducted with realistic ranges of model parameters, may be summarized to classify the parameters that important outcomes are more sensitive to. Typically, solute fate was sensitive to model parameters in the following order: 1) biogeochemical parameters, 2) physical parameters (hydrogeological parameters and environmental fluxes) and mechanical dispersivity, and 3) irrigation parameters. The range of outcomes associated with each of the sensitivity analysis themes listed in Table 2 are shown in Fig. 9. Data on the effects of spatial heterogeneity in soil properties, taken from Tang et al (2023), are also included in Fig. 9 for comparison. Fig. 9a shows that the irrigated solute mass essentially depends only on the physical parameters. This is because the amount of irrigated water required to maintain target groundwater levels depends strongly on the physical parameters. Fig. 9b shows that root solute uptake varies across a large range only due to varying biogeochemical parameters, and strong soil spatial

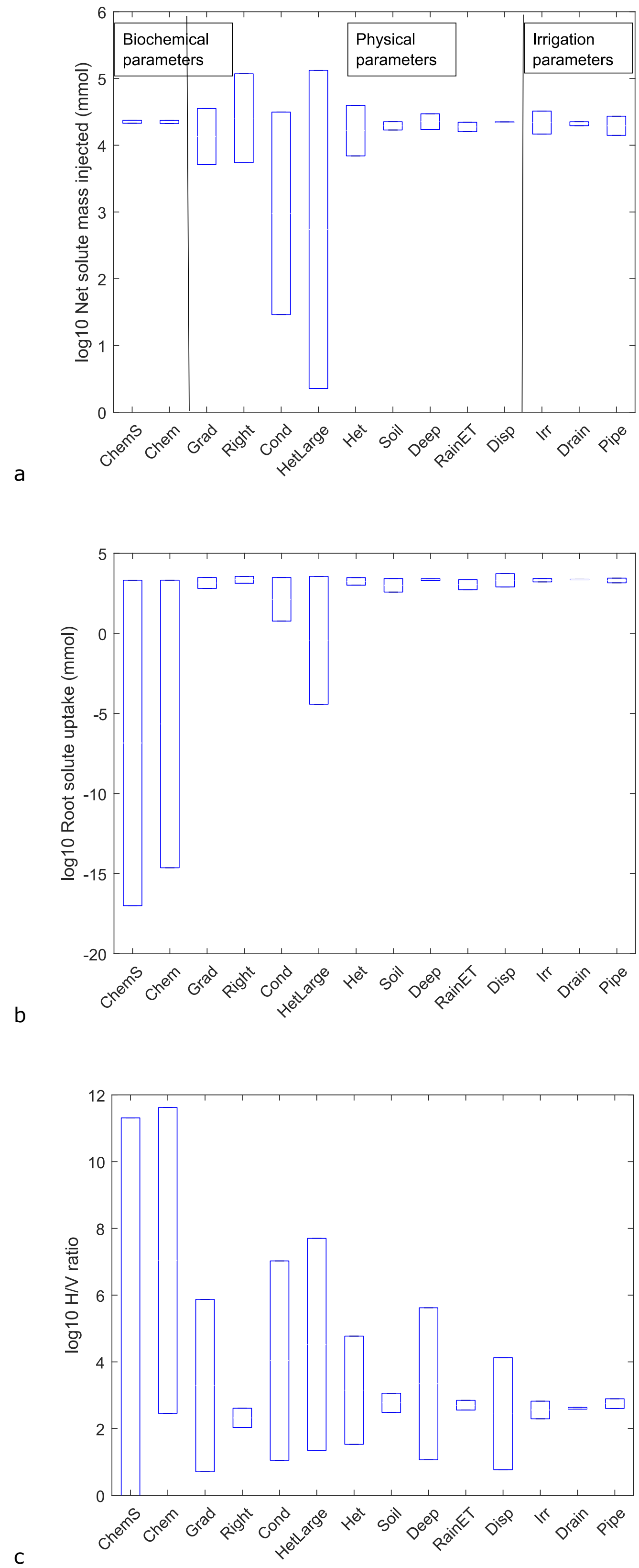

Fig. 9. Range of outcomes for (a) net contaminant injected, (b) root solute uptake, and (c) the H/V ratio for each individual sensitivity analysis. Shortened parameter names in the x-axis are explained in Table 2. The parameters "Het" and "HetLarge" refers to weak soil heterogeneity and strong soil heterogeneity respectively; these were not simulated in this study, and the data was obtained from Tang et al (2023).

heterogeneity. The root solute uptake does not vary too much as the physical parameters vary because it is constrained by the water requirements of the crop, whereas the biogeochemical parameters determine how much solute is present in the water taken up by the crops. Fig. 9c shows that the H/V ratio is strongly affected by varying both the biogeochemical and physical parameters. Unlike the root solute uptake, the H/V ratio varies greatly in response to varying physical parameters as the relative resistances of the various boundary conditions strongly control the direction of solute advection.

The analysis of Fig. 9 shows that biogeochemical parameters are the primary determinant of solute fate. Uncertainties arising in the field also follow this hierarchy: the biogeochemical parameters of individual solutes in the field may be uncertain by orders of magnitude (Nham et al., 2015) as they depend not only on the chemical compound but also a wide range of environmental factors (e.g. Denora et al., 2023) that are not feasible to account for during laboratory characterization of biogeochemical reaction rates. Aquifer properties and environmental fluxes also present a significant source of uncertainty, though these uncertainties should not span multiple orders of magnitude if some measurement of subsurface properties is performed. Uncertainties in irrigation parameters are the smallest, as they can be directly controlled, but the irrigation parameters also have the smallest relative impacts on solute fate. Hence, the parameters that solute fate are more sensitive to, are also parameters that tend to be more uncertain in field conditions. This may be one of the primary challenges in achieving a wider adoption of the studied irrigation technique.

As previously discussed, the irrigation parameters only minimally affect the hazards of subirrigation with treated wastewater. Nevertheless, varying the irrigation parameters still allows for several aspects of contaminant fate to be meaningfully controlled. Placing the irrigation drains deeper allows one to reduce the risk of crop contamination at the cost of increasing the contaminant mass seeping to deeper aquifers, or vice-versa. The optimal drain depth is therefore determined by the relative subjective hazard posed by root solute uptake and solute discharge to deeper aquifers. Additionally, this aspect of the subsurface irrigation system could be optimized to simultaneously bring CEC concentrations beneath guideline threshold concentrations in crops and in groundwater. Note that we modelled biodegradation as a spatially homogeneous first-order process here. In practice, the biodegradation rate may decrease with depth because microbial populations and oxygen (for aerobic biodegradation processes) tend to be concentrated near the topsoil (Hickman and Novak, 1989). Thus, to maximize the biodegradation of CECs in effluent with high organic matter content (e.g. domestic wastewater), it might be advantageous to place the irrigation drains shallower in the soil. On the contrary, effluent with primarily non-biodegradable contaminants such as metal ions (e.g. industrial wastewater or brackish water) might be better irrigated from deeper depths, to minimize crop contamination and salinity stress.

Spreading the drains further apart but increasing irrigation fluxes per drain will increase the spatial heterogeneity in crop contamination risks (i.e. elevated risks directly above drains, as discussed in Tang et al (2023)), and may be synergistic with intercropping. Future research could investigate the feasibility of growing non-food crops such as crops for textile fibres and biofuels directly above drains, to reduce the risks of CECs entering crops intended for human consumption. Bioenergy crops present an interesting choice for intercropping here, as incineration and pyrolysis are both highly effective at degrading and removing organic contaminants (Buss, 2021).

Since our sensitivity analysis of the precipitation and evapotranspiration timeseries (section 3.1) shows that the H/V ratio and vertical solute discharge are substantially variable but have little correlation with the total precipitation, evapotranspiration, or excess precipitation volumes, it follows that these outcomes must be primarily affected by the actual sequences, magnitudes, and variances of individual rainfall events within those timeseries. This is also a logical consequence of the fact that the H/V ratio is determined primarily by the vertical solute discharge, and that the vertical discharge of water is a nonlinear function of the groundwater level due to the deep drainage boundary condition. The sensitivity of the vertical discharge of solutes to the statistics of the precipitation timeseries may appear to be, but is not inconsistent with the notion that the effects of atmospheric variability on soil water fluxes are primarily concentrated in the shallow unsaturated zone (Salvucci and Entekhabi, 1994). As the solute mass fraction leaching vertically downwards is a very small component of the solute mass balance, stochastic perturbations to the general direction of solute transport would be reflected most readily here. Hence, although the effects of atmospheric variability on water fluxes are mostly confined to the shallow soil, the effects on solute fluxes may be substantial in both shallower and deeper soil.

Given that adopting phreatic zone irrigation with treated wastewater may also be of interest in dryer or warmer climates with greater precipitation shortages, in particular considering the possible effects of future climate change, it would be interesting to briefly consider how the irrigation technique could perform in such cases. Using the numerical model, we simulate an extreme hypothetical situation with zero precipitation, for illustration. With zero precipitation, the water and solute mass injected was approximately 50 % higher and the root solute uptake was seven times that of the base model, but the horizontal solute discharge was a quarter less and the vertical solute discharge was unchanged. Hence, even under such extreme circumstances, one of our main findings that lateral solute discharge is much greater than vertical solute discharge continues to apply for this site. The outcomes of this extreme scenario are also consistent with the correlation matrix of the sensitivity analyses (Fig. 3), which shows that the excess precipitation (P-ET) is more strongly correlated with the root solute uptake and lateral solute discharge than the vertical solute discharge. Therefore, the negative effects of a greater precipitation shortage are primarily confined to an increased risk in crop contamination, as the crops fulfil a greater proportion of their transpiration water requirements from capillary fluxes. In the context of phreatic zone irrigation with treated wastewater in very dry climatic conditions, one should consider whether the quantitative benefits outweigh the disadvantages with respect to water quality.

Aside from the quantitative insight on solute fate provided by our numerical models, several examples of interesting qualitative insights from this numerical modelling study include: 1) the total (crop + confined aquifer) pollution is minimized at some optimal confining layer depth, 2) the fraction of irrigated solutes recovered by the drains is a non-monotonic function of the solute retardation rate, 3) optimizing the irrigation parameters cannot compensate for the effect of unfavorable hydrogeological conditions on crop and environmental pollution.

4.2. Groundwater mobility controls field scale environmental risks

Despite the presence of multiple physical transport processes (including transient environmental fluxes, climate-adaptive transient irrigation fluxes, and solute dispersion) that spatio-temporally interact across the simulated model to determine solute fate, the results of the study enable us to make some general conclusions about the controlling physical processes that predominantly determine solute fate. Note in this subsection (section 4.2), we do not consider the effect of the contaminant-specific biogeochemical reaction rates, as the discussion here is meant to aid in the selection of suitable physical locations for the implementation of phreatic zone wastewater irrigation. Denote the aggregated subsurface resistance as the sensitivity of the subsurface pressure heads and water table level to changes in boundary fluxes, such as irrigation, precipitation and evapotranspiration. This definition follows directly from Darcy's law, as the net flux in or out of the subsurface domain is proportional to the spatial head gradient divided by the resistance. This sensitivity is controlled by the various hydrogeological parameters, such as the hydraulic conductivity and water retention of the soil, the conductances of the boundaries, the depth of the confining layer, and also the design parameters of the subsurface irrigation system such as drain depth, the spacing between drains, drain conductance, drain material, and drain pressure heads (Siyal and Skaggs, 2009; Naglic et al., 2014; Saefuddin et al., 2019; Cai et al., 2017).

Amongst the pressure head timeseries illustrated in Fig. 10a and Fig. 10b, the simulations with large subsurface resistances are those that have large amplitude fluctuations in pressure head over time, in response to transient variability in atmospheric fluxes and irrigation fluxes. Aside from higher amplitudes, pressure head fluctuations in scenarios with larger subsurface resistances are also of lower frequencies, as the subsurface water excesses or deficits (relative to equilibrium levels) are retained for a longer period of time, allowing excesses or deficits associated with separate atmospheric events to accumulate. In contrast, scenarios with smaller subsurface resistances have nearly constant pressure heads, because the boundary fluxes rapidly dampen pressure fluctuations originating from infiltration rate variations. Fig. 10c shows that all simulations with different drain conductances have similar pressure head timeseries, thus the drain conductivity does not significantly affect the subsurface resistance. Simulations with large subsurface resistances are the scenarios with a shallower confining layer, and small aquifer and boundary conductances.

As the subsurface resistance increases, the volume of irrigation water needed to maintain the water table at the target level decreases, which reduces the total solute mass injected into the soil. This is because a larger subsurface resistance retains irrigated water in the vadose and phreatic zones beneath the agricultural field for a longer time. Suitable locations for the implementation of the studied irrigation system can be identified accordingly. For example, if a low conductivity substratum underlies the irrigation drains, then the resistance of the system is increased, and smaller irrigation fluxes (and less effluent injection) are sufficient to maintain the groundwater level at a specified height (Mohammad et al., 2014).

The parameters that affect the subsurface resistance more significantly are the same parameters that affect solute mass balances more significantly, and vice-versa. These parameters are the hydrogeological parameters of the simulated domain, the regional head gradient, and the depth of the confining layer (if present), which within a single (climactic or land-use) zone is to a large degree determined by the hydrogeological parameters of the wider region. Note that if the confining layer is shallower, then more solutes will leach vertically downwards through the confining layer because not only because the subsurface resistance decreases and more irrigation must occur in order to maintain the water table at target levels (Fig. 7a), but also because the irrigated solutes have less distance to travel from the irrigation pipes to the confining layer, allowing them less time and space to be retained by adsorption, biodegraded, or carried away by lateral flow.

The hydrogeological parameters of the simulated domain and of the wider region would typically be strongly correlated due to their interrelated geological origins. Therefore, the hydrogeology of the region is the general underlying factor that determines solute fate. These parameters affect groundwater and solute mobility most strongly amongst the physical parameters for the same fundamental reason: they strongly determine the amount of irrigation water required to bring the groundwater level to the target level, and to maintain the elevated groundwater level brought about by subsurface irrigation. This suggests that the sensitivity of subsurface pressure heads to perturbations in hydrological fluxes, which is easier to test and observe than tracer experiments involving solute concentrations and mass balances, may serve as a proxy observable for evaluating the suitability of a particular location or region for implementing phreatic zone wastewater irrigation.

4.3. Limitations of the numerical model

We have studied contaminant fate in the shallow subsurface beneath the agricultural plot through sensitivity analyses of a numerical model. The numerical model is limited to the shallow subsurface, and thus does not explicitly show how the contaminants may behave in the shallow subsurface beyond the agricultural plot, or in the deeper subsurface. In the deep subsurface, the behavior of the solute plumes becomes less dependent on whether they infiltrated from the soil surface or whether they were injected into the shallow subsurface, due to dispersive processes that smooth out concentration gradients. Hence, in the context of contaminant transport in the deep subsurface, the large body of existing literature on surface irrigation and other soil contaminant infiltration problems may be used to characterize the consequences of phreatic zone wastewater irrigation. For example, van der Zee and Boesten (1991) studied how the thickness of the unsaturated zone, and spatial heterogeneity in soil properties, may affect reactive contaminant leaching to groundwater. Malaguerra et al (2013) modelled solute leaching through a thick phreatic aquifer to an underlying confined aquifer, and performed sensitivity analyses that explicitly varied the phreatic aquifer thickness (between 1 – 30 m), confining layer thickness, and confining layer conductivity, unlike the deep drainage boundary condition used in this study. They found that the thickness of the unconfined aquifer, and the conductivity of the confining layer, had substantial impacts on the amount of contaminants that leach into the underlying confined aquifer. Hantush et al (2000) derived an analytical solution for the lateral transport of solutes in phreatic aquifers, which may be used to estimate the lateral evolution of a solute plume within a deep phreatic zone under varying regional groundwater fluxes.

Note that our sensitivity analysis of the aquifer hydraulic conductivity corresponds to the hypothetical situation where the hydraulic conductivity is changed only within the irrigated field and not across an entire catchment, and where the size of the field is small compared to the catchment. In practice, a hypothetical increase in the hydraulic conductivity across a substantial portion of a hydrological catchment may cause the regional head gradient to decrease. This limitation of separately considering these two parameters, and not accounting for possible correlations, should be taken into account when analyzing the parameter sensitivity of contaminant fate within a single catchment.

We have studied how the various parameters of our numerical model affects the total root solute uptake in the irrigated plot, but our sensitivity analysis is limited to one set of root water uptake parameters, and the Feddes model parameters were not studied. Changing the Feddes model parameters would likely result in some changes to the model results, but the impacts would likely be small. The root solute uptake is highly correlated with the net solute mass injected (Fig. 2a) and the actual precipitation excess (P-ET) (Fig. 2b), where ET here refers to the actual evapotranspiration. The correlation coefficient between root solute uptake and (P-ET) is 0.762, and the correlation coefficient between root solute uptake and the net solute mass injected is 0.9. This suggests that any plant water stress that follows from the Feddes model would affect root solute uptake mostly insofar as it decreases (P-ET). Furthermore, the effects of root water uptake compensation and the spatial distribution of plant roots on crop and environmental contamination risks were not studied, but would not substantially change the total crop solute uptake to an extent large enough to change the conclusions of our study. This is because the root solute uptake is primarily constrained by the volume of crop water uptake in excess of rainfall (as evident in the correlation matrix in Fig. 3), and thus the effects would primarily materialize as heterogeneity in the spatial distribution of crop solute uptake.

Accordingly, an aspect of crop contamination risk that may be addressed in more detail in future studies is the spatial distribution of crop solute uptake across the field, and how this interacts with various spatially important aspects of the system. Such spatially important aspects include not only the location of the irrigation drains, as previously discussed in section 4.1, but also spatial heterogeneity in soil properties, and crop rooting characteristics such as the depth of the root zone, root profile characteristics, the spatial distribution of roots, crop solute stress that depends on solute concentrations, and compensated root water and

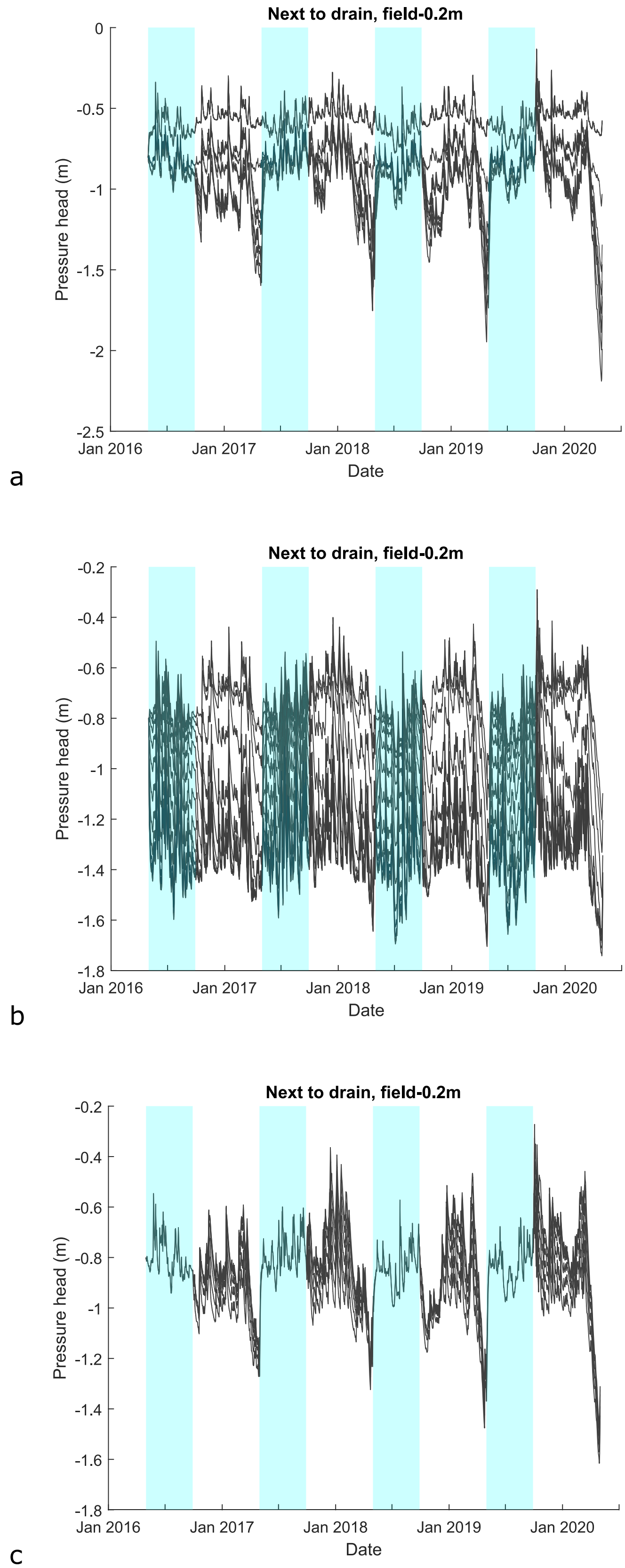

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Fig. 10. Pressure heads over time in the root zone for the sensitivity analysis simulations with (a) varying aquifer conductivity, and (b) varying right boundary conductivity, and (c) varying irrigation drain conductivity. Each line represents an individual simulation within the sensitivity analysis, and many lines may overlap, as in Fig. 10c. The periods indicated with cyan highlights indicate the crop (i.e. irrigated) season, whereas the non-highlighted periods are the non-crop season. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

solute uptake (Šimůnek and Hopmans, 2009). These factors may influence to some extent the risks and spatial distribution of crop contamination. For example, a deeper root zone may increase spatial heterogeneity in crop solute uptake, as irrigated solutes experience less dispersion before reaching the root zone, whereas crop solute stress and compensation processes may reduce this spatial heterogeneity (Šimůnek and Hopmans, 2009). Such spatial aspects should be studied using three-dimensional models, as solute spreading close to radial sources and sinks (i.e. the irrigation drains and root clusters respectively) is rather sensitive to the spatial dimensionality of the model, particularly in heterogeneous soils (Tang and van der Zee, 2022).

4.4. Broader implications for phreatic zone wastewater irrigation

The insights of this study, and other studies related to phreatic zone irrigation with wastewater (see introduction section), suggest that phreatic zone irrigation with wastewater may be an appropriate combined solution for irrigation and wastewater reuse. Regarding the former, the use of treated wastewater for irrigation contributes towards freshwater conservation, in line with recent government incentives in Europe and across the world (Ballesteros-Olza et al., 2022). On the latter, phreatic zone irrigation with treated wastewater causes some of the wastewater contaminants to be attenuated in the soil through biodegradation and adsorption. However, as we have shown in this study, the extent of contaminant attenuation and the risks of crop and environmental contamination under phreatic zone wastewater irrigation depend substantially on the hydrogeological properties and hydroclimatic conditions. Therefore, it is important to identify appropriate soil types, and areas with environmental characteristics and wastewater properties that are suitable for environmentally responsible implementation of this irrigation technique.

We found that the hydrogeological properties of the saturated zone were more important than the root zone soil properties in determining crop and environmental contamination risks. Aside from having a confining layer that is not too shallow, to prevent contaminant leaching to confined aquifers, the soil type of the phreatic zone also has to be appropriate for phreatic zone wastewater irrigation. As the hydraulic resistance of the saturated zone increases, less wastewater (and contaminants) have to be irrigated to maintain optimal water table levels. Nevertheless, the hydraulic resistance of the phreatic zone must also not be too high, so that the irrigated water can enter the subsurface at an acceptably fast rate (Yu et al., 2020). The hydraulic resistance of soils tends to increase in the order sand, silt, to clay. Furthermore, while sands, silts, and clays are able to support similar magnitudes of capillary fluxes, the effective range of capillary fluxes increases in the order sand, silt, to clay (Salvucci, 1993). Therefore, if the water-use efficiency of phreatic zone irrigation is considered, coarse sandy soils would perform least favorably because of their low hydraulic resistance and low capillary rise range. Nevertheless, sandy soils may nevertheless be suitable for phreatic zone irrigation if they have characteristics that enhance soil water retention, such as smaller particle sizes (Zeiliguer et al., 2000), or high organic matter contents (Rawls et al., 2004). Substantial organic matter content in the soils may reduce contaminant spreading risks, as organic matter may facilitate water retention (Rawls et al., 2004) and contaminant adsorption (Ukalska-Jaruga et al., 2023);

further research is needed to investigate this in more detail, in the context of phreatic zone wastewater irrigation.

In addition to their good water retention capabilities (Zeiliguer et al., 2020), soil types with smaller particle sizes, such as clays, silts and fine sands, also have larger specific surface areas, which may facilitate the attenuation of contaminants by adsorption (Petersen et al., 1996) and biodegradation (Cui et al., 2011). However, wastewater may often be somewhat saline, even if treated (Muyen et al., 2011). Repeated cycles of irrigation with saline water, and then flushing with freshwater (rainfall) during the non-crop season, may cause clay soils to become sodic and swell due to cation exchange processes (van de Craats et al., 2020). The sodicity-induced structural degradation of clay soils leads to a severe reduction in hydraulic conductivity, and is mostly irreversible (Sou et al., 2013). Therefore, clay and clayey soils are unlikely to be optimal for phreatic zone wastewater irrigation. Altogether, we have argued that soils with intermediate textures; not too fine and not too coarse, have the highest potential for phreatic zone irrigation, in agreement with Singh et al (2022).

We have also shown that phreatic zone irrigation can be considered in areas with (1) a temperate climate with a precipitation shortage during the crop season but a precipitation surplus on an annual average, so that contaminants in the vadose zone may be flushed away by the precipitation surplus, and (2) shallow water tables, so that minimal wastewater (and contaminants) have to be irrigated to raise the water table to an appropriate level. A population density sufficiently large to supply the necessary wastewater would also be useful (Narain-Ford et al., 2021). A map of water table depth shows that the low-lying areas of Europe with shallow water tables (Fan et al., 2013) tend to coincide with areas with large population densities, for example the Netherlands, Flanders, Denmark, (i.e., the North European Plain), and the Po valley among others. Amongst these regions, the North European Plain may have more potential for phreatic zone wastewater irrigation than the Po valley, as the Po valley has soils with larger clay contents (Poggio et al., 2021) and existing soil salinity problems (Ungaro et al., 2021). The Po valley also has a larger crop season precipitation shortage of around 200 – 400 mm (Nana et al., 2014; Bocchiola et al., 2013; Perego et al., 2012) as opposed to 40 – 200 mm in the Netherlands (de Wit et al., 2024), thereby increasing the required wastewater irrigation volume, and the risks of crop contamination and contaminant accumulation in soils. Hence, within Europe, the potential for adopting phreatic zone wastewater irrigation may be concentrated within the North European Plain. Outside of Europe, the literature suggests that phreatic zone irrigation may be suitable for parts of the United States Midwest (Singh et al., 2022; Yu et al., 2020), though there are no existing studies that specifically investigate wastewater irrigation potential. While parts of the Central Valley of California are also hydrologically suitable for phreatic zone irrigation, it may be less feasible in practice due to high salt levels (Singh et al., 2022).

Our sensitivity analyses have also shown that the physical parameters that have the greatest effects on solute mass balances notably belong to the hydrogeological parameters: the regional head gradient, aquifer conductivity, and right boundary conductivity (Fig. 9). The water-use efficiency of phreatic zone irrigation was also found to be controlled by hydrogeological parameters (de Wit et al., 2024). These findings contrast with those for surface irrigation and shallow drip irrigation, where amongst the physical parameters, the atmospheric flux timeseries and root zone soil properties are the primary determinants of solute fate, such as residence times in the root zone, root solute uptake, and leaching to groundwater (Schotanus et al., 2013; van der Zee and Boesten, 1991; Rakonjac et al., 2023). Furthermore, (near-)surface irrigation methods directly expose crops to contaminants, whereas under phreatic zone irrigation the risk of contaminating crops is smaller but the risk of contaminating groundwater is larger. This implies that the new method of phreatic zone subirrigation is associated with risks that are determined by different controlling factors compared to other existing irrigation methods. Therefore, phreatic zone subirrigation provides a complementary alternative for areas where wastewater irrigation using other irrigation methods may lead to unacceptably high crop or environmental contamination risks, and vice-versa.

The simplest example of this complementarity is that if water tables are very deep, then excessive amounts of wastewater (and contaminants) are needed to raise the water table to target levels, in order to support sufficient capillary fluxes to the root zone under phreatic zone irrigation. In contrast, if water tables are very deep, then contaminants in wastewater irrigated into the (near-)surface would have a large vadose zone to travel through, adsorb within, and biodegrade in, before leaching to groundwater (Rakonjac et al., 2023; Urbina et al., 2020). Another example is that if the confining layer is very shallow, and close to the irrigation pipe depth, then large amounts of solutes may leach through the confining layer under phreatic zone wastewater irrigation. In contrast, under (near-)surface wastewater irrigation, the contaminants may be partially attenuated in the vadose zone and phreatic zone before the wastewater leaches through the confining layer (Malaguerra et al., 2013). Another example is that in areas with high atmospheric flux variabilities, (near-)surface wastewater irrigation may lead to excessive contaminant concentrations in the root zone during dry periods, as there is less water to dilute and transport contaminants (van der Zee et al., 2010), and also because contaminant biodegradation may be suppressed under very dry conditions (Chow et al., 2023). In contrast, under phreatic zone irrigation such drying out (of the soil at irrigation pipe depth) is less likely to occur, as regional groundwater fluxes and vadose zone soil moisture are less affected by atmospheric flux variabilities than topsoil moisture (Salvucci and Entekhabi, 1994). Yet another example is that crop contamination risks might be considered less important when irrigating crops used for biofuel, as some organic contaminants are destroyed when the fuel is burned (Buss, 2021). Conversely, confined aquifer contamination risks may be perceived as less serious if the wastewater primarily consists of contaminants that efficiently degrade under anaerobic conditions (Farhadian et al., 2008).

Another key finding of the numerical sensitivity analysis is that larger regional groundwater fluxes lead to lower crop (Fig. 2e) and groundwater (Fig. 2f) contamination risks. Assuming that the hydrogeological properties of the entire catchment are fixed, the occurrence of larger regional fluxes within the agricultural plot may occur due to larger excess precipitation and groundwater recharge fluxes upstream. Therefore, we have shown that the overall crop and environmental contamination risks of the subirrigation system likely decreases when the upstream precipitation excess and groundwater recharge are larger. Accordingly, a remarkable aspect of phreatic zone irrigation (with treated wastewater) is that the required irrigation volume (and crop and environmental contamination risks) is decreased by increases in groundwater recharge upstream of the agricultural plot. In contrast, when using other irrigation techniques (with treated wastewater) that do not raise the water table, increased groundwater recharge upstream of the agricultural plot is less likely to decrease the required irrigation volume (and crop contamination risks), because the water table is more likely to be situated beneath the capillary flux extinction depth (Shah et al., 2007). Hence, in addition to its roles as an irrigation technique and as a nature-based solution for additional wastewater treatment, the subirrigation system and resulting higher groundwater levels could also harvest rainwater for agriculture, using the soil as a vessel.

Accordingly, although phreatic zone subirrigation is less water-use efficient than existing irrigation techniques in terms of irrigated water volume (de Wit et al., 2024), the quality of the crop-transpired water is much better than the irrigated effluent, except under very dry conditions where most crop water uptake originates directly from the irrigated effluent. This distinction between the irrigated and crop-transpired water quality is a unique characteristic of phreatic zone subirrigation with treated wastewater. Given the increasing need for reusing wastewater in agriculture to combat freshwater scarcity and fertigate crops (Mainardis et al., 2022), and the key role of wastewater reuse in future water security and the circular economy (Voulvoulis, 2018), methods to quantify irrigation water-use efficiency that separately account for irrigated and crop-transpired water quality could be a potential topic for future research. Such a quantitative indicator would be useful for comparing irrigation techniques for marginal water reuse, but no such indicator appears to be in common usage currently (Fernández et al., 2020). With such indicators, the benefits of wastewater irrigation over freshwater irrigation, through various irrigation techniques, could be better quantified and evaluated.

5. Conclusions

In this study, we performed an extensive sensitivity analysis on contaminant fate under phreatic zone irrigation. Key outcomes we have considered are root solute uptake, solute mass drained by the drainage system, and the amount and direction of solute discharge in the saturated zone. We have identified several implications of phreatic zone irrigation with treated wastewater, that apply in general throughout the sensitivity analyses:

(1) Regarding solute fate, the biogeochemical parameters are the most impactful, followed by hydrogeological parameters that directly affect groundwater mobility, then the other physical parameters, and finally the irrigation parameters. The most impactful parameters also tend to be the most uncertain and difficult to estimate in the field.
(2) Although the hydrogeological parameters do not affect the partitioning of irrigated solute fate across the possible outcomes as much as the biogeochemical parameters do, the hydrogeological parameters (e.g., subsurface hydraulic conductivity, confining layer depth and aquifer thickness) additionally affect the irrigated solute mass. This is an important difference to consider when evaluating the sensitivity of crop and environmental contamination risks to the biogeochemical and hydrogeological parameters.
(3) The drains may extract significant amounts of water from the subsurface, but not CECs, even if they do not biodegrade. This is because in many cases the CEC plumes do not intercept the drains during the drainage season, due to the complex and transient two-dimensional flow fields.
(4) Since crop solute uptake, solute drainage, and solute seepage through the aquifer bed comprise small proportions of the solute mass balance across the range of simulated scenarios, most irrigated solutes will be discharged laterally to the phreatic aquifer, if not biodegraded along the way.
(5) Crop solute uptake varies less substantially across scenarios, compared to solute discharge to the wider environment, because the total crop solute uptake is limited by crop water requirements. In other words, even if the root zone water becomes highly contaminated due to drought or large contaminant inputs, the total crop solute uptake would be upper-bounded by the crop water uptake volume, unlike the environmental solute discharge, which increases unboundedly with increasing wastewater irrigation.

Altogether, the fate of solutes in the effluent is primarily determined by factors that are mostly determined by the geography, geology, and ecology (i.e. effluent type and quality, soil and hydrogeological properties, microbiome) of the agricultural plot and region. Effluent type and quality are geographically determined because it is dependent on the type of wastewater available (domestic vs industrial), which is a function of land-use, and the ability of existing treatment plants to purify it. Since the availability of effluent in agricultural areas with more favourable soil and aquifer properties is limited by the cost efficiency of transporting effluent, and by the geography of human settlement (Narain-Ford et al., 2021), the placement of treatment plants and agricultural zones is therefore an important factor in the implementation of subirrigation systems. In summary, overcoming barriers towards wider adoption of phreatic zone subirrigation systems may ultimately be a challenge in land-use planning. Our discussion also shows that the crop contamination risks associated with phreatic zone wastewater irrigation are different from those associated with (near-)surface irrigation techniques. Therefore, phreatic zone irrigation may complement other existing irrigation techniques for expanding wastewater reuse in agriculture.

CRediT authorship contribution statement

Darrell W.S. Tang: Writing – original draft, Software, Methodology, Formal analysis, Conceptualization. **Sjoerd E.A.T.M. Van der Zee:** Supervision, Funding acquisition, Conceptualization. **Ruud P. Bartholomeus:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

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

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jhydrol.2024.132263.

Data availability

Data will be made available on request.

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