1	Soil-Aquifer Phenomena Affecting Groundwater under Vertisols: a Review
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- 20 Abstract
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22 Vertisols are cracking clayey soils that: i) usually form in alluvial lowlands where 23 normally, groundwater pools into aquifers; ii) have different types of voids (due to 24 cracking) which make flow and transport of water, solutes and gas complex, and iii) are 25 regarded as fertile soils in many areas. The combination of these characteristics results in 26 the unique soil-aquifer phenomena that are highlighted and summarized in this review. 27 The review is divided into the following four sections: 1) Soil cracks as preferential 28 pathways for water and contaminants; in this section lysimeter- to basin-scale 29 observations that show the significance of cracks as preferential flow paths in vertisols 30 which bypass matrix blocks in the unsaturated zone are summarized. Relatively fresh-31 water recharge and groundwater contamination from these fluxes and their modeling are 32 reviewed; 2) Soil cracks as deep evaporators and unsaturated-zone salinity; deep 33 sediment samples under uncultivated vertisols in semiarid regions reveal a dry 34 (immobile), saline matrix, partly due to enhanced evaporation through soil cracks. 35 Observations of this phenomenon are compiled in this section and the mechanism of 36 evapoconcentration due to air flow in the cracks is discussed; 3) Impact of cultivation on 37 flushing of the unsaturated zone and aquifer salinization; the third section examines 38 studies reporting that land-use change of vertisols from native land to cropland promotes 39 greater fluxes through the saline unsaturated-zone matrix, eventually flushing salts to the 40 aquifer. Different degrees of salt flushing are assessed as well as aquifer salinization on 41 different scales, and a comparison is made with aquifers under other soils; 4) Relatively 42 little nitrate contamination in aquifers under vertisols; In this section we turn the light on

43	observations showing that aquifers under cultivated vertisols are somewhat resistant to
44	groundwater contamination by nitrate (the major agriculturally related groundwater
45	problem). Denitrification is probably the main mechanism supporting this resistance,
46	whereas a certain degree of anion-exchange capacity may have a retarding effect as well.
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48	Keywords: groundwater under vertisols; cracking clays; groundwater salinization
49	
50	1. Introduction
51	
52	Vertisols can be briefly defined as soils with 30% or more clay to a depth of 50 cm that
53	have shrinking/swelling properties (Brady and Weil, 2002). More detailed definitions
54	require the existence of a subsurface vertic horizon in which slickenside features are
55	formed by the shrink/swell dynamics (FAO Corporate Document Repository, 2014; IUSS
56	Working Group WRB, 2014). Other names used for these types of soils are: vertosols
57	(common in Australian studies, e.g. Radford et al., 2009; Silburn et al., 2009;
58	Gunawardena et al., 2011; Ringrose-Voase and Nadelko, 2013), and the more general
59	'cracking clays' (e.g. Bronswijk, 1991; Liu et al., 2010). This latter generic term
60	emphasizes both the hydrological complexity of these soils due to the inherent
61	discontinuities (cracks) and their relevance for agriculture, being heavy, relatively fertile
62	soils in many semiarid regions (good water-holding capacity, relatively higher organic
63	content, etc.). Vertisols usually form in lowlands (Yaalon, 1997) where, typically,
64	groundwater pools into alluvial aquifers. Hence, the interface between agricultural
65	activity on these soils and the underlying groundwater resources is both complex and

relevant. This review focuses on vertisol studies that have implications for the underlying groundwater resources; it does not cover the substantial body of literature concerning shrinking/swelling dynamics and its modeling (e.g. Bronswijk, 1988; Chertkov et al., 2004; te Brake et al., 2013), the purely agricultural and mineralogical aspects of vertisols (e.g. Bhattacharyya et al., 1993; Ahmad and Mermut 1996; Hati et al., 2007) or environmental topics like the capacity of vertisols to sequester carbon (Hua et al., 2014).

72 Vertisols cover 335 million hectares out of a total earth land area of 14.8 billion 73 hectares (2.3%). The largest areas covered with vertisols are in Eastern Australia, India, 74 Sudan–Ethiopia and Argentina–Uruguay (FAO Corporate Document Repository, 2014). 75 Smaller areas of vertisols are found in various countries (e.g. China, Israel, Mexico, Spain, Tunisia, USA, and many more). Although vertisols are very hard when dry, and 76 77 very sticky when wet (making them difficult to till), in semiarid regions, irrigated crops 78 such as cotton, corn, wheat, soybeans and others are grown on this soil. We acknowledge 79 the dominant contribution of Australia to the literature on agro-hydrological aspects of 80 vertisols. Conclusions from those studies have strengthened and generalized some of the 81 findings obtained by the authors of this review in vertisol–groundwater studies in Israel, 82 and have motivated this review (e.g. Arnon et al., 2008; Kurtzman and Scanlon, 2011; 83 Baram et al. 2012a, 2012b, 2013, Kurtzman et al., 2013).

The review is divided into four sections which are partially connected and together deal with major issues concerning aquifers under agricultural land: recharge, salinization and nitrate contamination (other contaminants are mentioned as well). The following four sections cover the most general and relevant issues concerning soil-aquifers phenomena under vertisols:

89	• Soil cracks as preferential pathways for water and contaminants (section 2)
90	• Soil cracks as deep evaporators and unsaturated-zone salinity (section 3)
91	• Impact of cultivation on flushing of the unsaturated zone and aquifer salinization
92	(section 4)
93	• The relatively little nitrate contamination in aquifers under vertisols (section 5)
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95	2. Soil cracks as preferential pathways for water and contaminants
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97	There are probably hundreds of studies that acknowledge preferential flow and
98	transport through cracks in clays-too many to be mentioned here. This section aims to
99	review works from the soil-column and lysimeter scale to the basin and aquifer scales
100	that show the relations between preferential flow via cracks, deep drainage, and aquifer
101	recharge and contamination. It also provides a short description of the development of
102	models of preferential flow and transport through soil cracks.
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104	2.1. Preferential flow of water in vertisols – evidence from the lysimeter to aquifer scale
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106	On a small scale, Kosmas et al. (1991) observed bypass flow through cracks in clayey
107	soils from Greece using undisturbed soil columns (authors' terminology) with a diameter
108	of 23 cm. Ringrose-Voase and Nadelko (2013) measured flow in preferential paths
109	directly using a field lysimeter that was installed 2 m below the surface of a furrow-
110	irrigated cotton field without disturbing the overlying soil. Significant drainage was
111	collected in this study when the hydraulic gradient in the matrix was in the upward

112 direction, advocating drainage through preferential pathways that bypasses the matrix. In 113 a paragraph on tension-lysimeters measurements, Silburn et al. (2013) acknowledge that: 114 "deep drainage measured at 1m depth was dominated by matrix flow, with only 10% of 115 drainage attributed to preferential flow (note that the soil was never dry enough to 116 crack)", pointing that under well-irrigated vertisols matrix deep-drainage and recharge 117 may be of importance despite the low saturated hydraulic conductivity of the clay. A 118 weighing-lysimeter experiment in irrigated vertisols in eastern Australia revealed a 119 complex drainage mechanism following spray irrigation, where only deep parts of the 120 cracks act as preferential pathways for the drainage when the top soil is moist and 121 uncracked (Greve et al., 2010).

On the field scale (~100–1000 m²), a similar phenomenon—i.e. open cracks at depth when surface cracks are mostly sealed—was reported by Baram et al. (2012b) throughout the rainy season in Israel. These authors compared transient deep (up to 12 m) water-content data collected by vadose zone-monitoring systems (VMSs, Dahan et al., 2009) at various sites, including very sandy soils; the comparison showed that by far, the fastest propagation of wetting fronts in deep vadose zones is observed in cracking clays.

Ben-Hur and Assouline (2002) conducted measurements of runoff in a vertisol cotton field in Israel that was irrigated with a moving sprinkler irrigation system. They observed that the high infiltration of runoff through soil cracks limited the overall surface runoff from the field. Other field-scale vadose-zone studies reported that preferential flow through cracked clay enhances infiltration from rice paddies (Liu et al., 2004). Losses of up to 83% of the water to deep drainage (including preferential and\or matrix flows) during furrow irrigation of cotton and sugar cane in vertisols were reported (Raine and

135 Bakker, 1996; Dalton et al., 2001; Moss et al., 2001; Smith et al., 2005). Losses to deep 136 drainage averaged 42.5 mm per irrigation (Smith et al., 2005), ranging from 50 to 300 137 mm/year (Silburn and Montgomery, 2001). Chen et al. (2002) and Bandyopadhyay et al. 138 (2010) showed that the transition from flood to micro-sprinkler irrigation and careful 139 scheduling of water-application rates can dramatically reduce water losses and 140 contaminant transport due to deep drainage. Observation from groundwater supported 141 this phenomenon: Acworth and Timms (2009) used nested piezometers and automated 142 logging of groundwater levels and electrical conductivity to show evidence of shallow-143 aquifer (16 m depth) freshening (decrease in salinity) due to fast deep-drainage of 144 irrigation water during the irrigation season.

At the small-watershed scale ($\sim 10,000 \text{ m}^2$), Pathak et al. (2013) indicated that 145 146 runoff from vertisols is smaller than runoff from sandier soils (Alfisols) in an agricultural 147 watershed near Hyderabad, India. The smaller runoff from the vertisols was attributed to 148 preferential infiltration of local runoff into the soil cracks. Similar observations of 149 minimal drainage and rapid recharge of shallow groundwater (~3 m) below a vertisol-150 shale watershed in Texas following rainstorms were reported by Allen et al. (2005) and 151 Arnold et al. (2005). This process was most dominant during the first rainstorms when 152 the cracks were fully developed (at the end of the dry season).

On the aquifer scale (100+ km²), Kurtzman and Scanlon (2011) concluded that parts of the Israeli Coastal Aquifer overlaid by vertisols were fresh (before the influence of modern intensive cultivation) only due to recharge flow through preferential paths that bypassed the saline vadose-zone matrix. Dafny and Silburn (2014) reported that following the growing evidence of the feasibility of percolation through cracking clays, several recent studies have included a component of diffuse recharge in their assumptions
or models of the Condamine River Alluvial Aquifer in Australia. This diffused recharge
originates in deep drainage flowing through clay matrix and/or preferential paths.

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- 162 2.2. Preferential transport in vertisols
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164 In the last two decades, many transport studies with dyes and/or other 165 conservative tracers (e.g. bromide, Br) have indicated the pervasiveness of deep 166 preferential transport through cracks in vertisols. Bronswijk et al. (1995) sprayed a 167 bromide solution on cracking clays in the Netherlands that overlay a shallow water table (~1 m from ground surface). The authors reported rapid (on the order of days after rain 168 169 event) preferential transport of Br into the groundwater, and relatively fast (weeks to 170 months) propagation within the vadose zone. Bronswijk et al. (1995) concluded that large 171 cracks control the rapid transport of Br⁻ to the groundwater, and preferential paths made 172 up of tortuous "mesopores" control transport in the unsaturated zone (suggesting that 173 transport through vertisols could be described as a triple domain medium—macropores, 174 mesopores and matrix). Van Dam (2000) used the Crack module in SWAP to model the 175 aforementioned experiment. This effort improved fits to the observations (relative to a 176 single-domain model), but the variability of Br⁻ in the unsaturated zone still could not be 177 well reproduced. Lin and McInnes (1995) used dye to study and model flow in vertisols. 178 They showed that infiltrating water passes first through the soil cracks and then into the 179 soil matrix; they concluded that uniform flow through the soil cannot be used to describe 180 the dye transport. A dye experiment in a soil column consisting of a sandy A horizon and a vertic clay B horizon showed preferential downward flow through the cracks in horizon
B, bypassing more than 94% of the matrix (Hardie et al., 2011).

183 Kelly and Pomes (1998) estimated equivalent hydraulic conductivities from 184 arrival times of Br⁻ and ¹⁵N-labeled nitrate in gravity lysimeters installed above and under 185 a clay pan in Missouri (USA). They reported equivalent conductivities that were four 186 orders of magnitude higher than the saturated hydraulic conductivity of the clay matrix.

187 Unlike tracers used in experiments, fast transport of herbicides and pesticides is of 188 concern in aquifers and drainage systems down gradient from cultivated fields. Graham et 189 al. (1992) reported that in cultivated vertisols in California (USA), herbicides were only 190 found deep below the root zone in samples taken from the cracks' walls and not within 191 the matrix, suggesting rapid transport of herbicides through the cracks, either as solutes or 192 on colloids. Transport of pesticides in preferential flow paths absorbed on colloids was 193 also suggested for cotton fields on vertisols in Australia (Weaver et al., 2012). Early and 194 deep drainage of herbicides from a lysimeter in cracking clays in the UK (early = well 195 before reaching field capacity in the matrix) was reported by Harris et al. (1994). 196 Similarly, fast arrival of herbicides to drains in cultivated clays was observed by Tediosi 197 et al. (2013) on a larger scale (small catchment).

Due to the fact that in semiarid regions, vertisols are arable, agriculture-oriented settlements have developed on these soils. In many cases, these settlements include concentrated animal feeding operations (CAFOs), such as dairy farms. Arnon et al. (2008) reported deep transport (>40 m) of estrogen and testosterone hormones into the unsaturated zone under an unlined dairy-waste lagoon constructed in a 6-m thick vertisol in Israel. They concluded that deep transport of such highly sorptive contaminants can

only occur by preferential transport. Baram et al. (2012a, 2012b) reported that preferential infiltration of dairy effluents through the cracks at the same site can transport water and solutes into the deep unsaturated zone. Locally, groundwater under dairy farm areas also shows relatively high concentrations of nitrate (Baram et al., 2014).

Fig. 1 provides a visual summary of sections 2.1 and 2.2. It shows the potential for matrix-bypassing groundwater recharge and pollution under vertisols. Passing the biogeo-

210 active matrix enables both freshwater recharge and transport of reactive substances.

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212 2.3. Development of flow and transport models in cracking clays

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214 The field evidence described above motivated the development of quantitative 215 methods to enable better predictions of flow and transport from ground surface to water 216 table under vertisols. Nevertheless, modeling of unsaturated flow and transport as a dual 217 (or multiple) domain in their different variants (e.g. mobile-immobile, dual-porosity, 218 dual-permeability) did not develop exclusively to deal with cracking clays. Macropores 219 such as voids between aggregates, or worm-holes, are the preferential-flow paths of 220 interest in many agricultural problems. Computer codes for modeling unsaturated 221 preferential flow include among others: MACRO (Jarvis et al., 1994) and nonequilibrium 222 flow and transport in HYDRUS (Šimůnek and van Genuchten, 2008). For further 223 information on the kinematic wave approach used in MACRO, the reader is referred to 224 German and Beven (1985); for comparative reviews of the different models and codes see 225 Šimůnek et al. (2003), Gerke (2006), Köhne et al. (2009) and Beven and Germann (2013). The latter is critical of the common use of the Richards (1931) formulation insingle- and multiple-domain unsaturated-flow simulators.

228 One of the earlier crack-specific unsaturated-flow models was developed by 229 Hoogmoed and Bouma (1980), who coupled vertical (crack) and horizontal (into the 230 matrix) 1D models using morphological data for parameterization of the linkage between 231 the two flows. Novák et al. (2000) attached a FRACTURE module to HYDRUS in which 232 a source term was added to the Richards Equation accounting for infiltration from the 233 bottom of the fractures, bypassing matrix bulks. Van Dam 2000, added a crack sub-model 234 to SWAP (van Dam et al., 2008) and Hendriks et al. 1999 used a code called 235 FLOCR\AMINO, to study flow and transport phenomenon in shallow and cracked 236 clayey unsaturated-zones in the Netherlands. A model of herbicide transport through the 237 preferential paths was fitted successfully with the improved MACRO version 5.1 (Larsbo 238 et al., 2005).

A more comprehensive dual-permeability module for 2D and 3D variably saturated models was introduced into HYDRUS much later (Šimůnek et al., 2012) following the formulations of Gerke and van Genuchten (1993). Coppola et al. (2012) took another step forward in modeling flow and transport in cracking clays by also introducing cracking dynamics (adopting formulation of Chertkov, 2005) into a dualpermeability flow and transport model.

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3. Soil cracks as deep evaporators and unsaturated-zone salinity

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248 Whereas during rain events or under irrigation, cracks are a concern in terms of 249 loss of water and fertilizers and/or contamination of groundwater (section 2), under dry 250 conditions, deep soil cracks are relevant for their evaporation capacity from deep parts of 251 the soil column. Kurtzman and Scanlon (2011), Baram et al. (2013) and others have 252 reported the low water content and high salinity typical of the sediment matrix under 253 uncultivated vertisols. Deep chloride profiles under native-land vertisols often show an 254 increase in salinity down to 1–3 m and a relatively constant concentration in deeper parts 255 of the vadose zone (e.g. Radford et al., 2009; Kurtzman and Scanlon, 2011; Silburn et al., 256 2011). In the reported cases from Israel, water uptake by roots was limited to the upper 1 257 m of the soil profile and to the rainy season, therefore, could not fully explain the 258 increase in salinization in the deeper layers. Deep cracks form an additional mechanism 259 of deep evaporation that supports the chloride profiles and low water content in the 260 matrix under vertisols.

261 Sun and Cornish (2005) used SWAT to model runoff and groundwater recharge at 262 the catchment scale ($\sim 500 \text{ km}^2$) in a vertisolic catchment in eastern Australia. 263 Considering water balances at this scale, they concluded that recharge models need to 264 have a component that enables taking moisture out of the lower soil profile or 265 groundwater during dry periods. Trees with roots in groundwater and deep soil cracks can 266 maintain deep evaporation in long dry periods. Another, indirect observation that 267 supports evaporation through cracks in vertisol was reported by Liu et al. (2010). In this 268 work discrepancies between satellite and model estimates of soil water content in dry 269 seasons in vertisols are assumed to be related to the extra evaporation through the cracks.

Both local- and higher-scale observations and analyses point to possible significant roleof soil cracks as deep evaporators in dry periods.

272 Baram et al. (2013) suggested a conceptual model termed 'Desiccation Crack-Induced 273 Salinization' (DCIS) based on previous work on subsurface evaporation and salinization 274 in rock fractures (e.g. Weisbrod and Dragila, 2006; Nachshon et al., 2008; Kamai et al., 275 2009; Weisbrod et al., 2009). In DCIS, vertical convective flow of air in the cracks is 276 driven by instability due to cold (and dense) air in the crack near the surface and warmer 277 air down in deeper parts of the crack at night or other surface-cooling periods. The 278 difference in the relative humidity between the invading surface air (low humidity) and 279 the escaping air (high humidity) leads to subsurface evaporation and salt buildup (Fig. 2). 280 Earlier studies that support the significance of evaporation via cracks in vertisols through 281 field and laboratory observations include: Selim and Kirkham (1970), Chan and Hodgson 282 (1981) and Adams and Hanks (1964). The latter showed enhanced evaporation from 283 crack walls due to increase in surface wind velocity, this is another mechanism (in 284 addition to surface cooling described before) causing instability in the crack's air, hence 285 convection, evaporation and salt build up.

Leaching of salts from horizontal flow through the crack-network evident in salinity-rise in tail water of furrow-irrigated fields in cracking clays in California was reported by Rhoades et al. (1997). This Californian study acknowledge that this phenomenon was not observed in similar fields (crop and irrigation technique) in lighter soils with no cracks.

In many semiarid regions, deep matrix percolation under non-cultivated vertisols is very small due to the clay's high retention capacity and low hydraulic conductivity, root uptake of the natural vegetation in the rainy season, and further evaporation through

293 cracks in dry periods. Low water content in the deeper unsaturated zone results in low 294 hydraulic conductivities and makes aquifer recharge through matrix flow very small year-295 round. Matrix fluxes in the order of 1 mm/yr under the root/crack zone were reported in a 296 number of studies (e.g. Silburn et al. 2009, Kurtzman and Scanlon 2011; Timms et al. 297 2012). These very low water fluxes contain the conservative ions (e.g. chloride) 298 originating from 200-600 mm/yr of precipitation (with salts from wet and dry fallout) that 299 enter the matrix at soil surface. Therefore a dry (relatively immobile) and salty deep 300 unsaturated matrix, developed for centuries-millennia under these non-cultivated 301 vertisols. Nevertheless, some fresh recharge to the underlying aquifer through preferential 302 paths related to cracks during heavy rain events creates an anomaly whereby relatively 303 fresh water in the aquifer (e.g. ~250 mg/l chloride, Kurtzman and Scanlon, 2011) lies 304 beneath a salty and immobile unsaturated zone with pore-water chloride concentration of 305 a few thousands of milligrams per liter (O'Leary, 1996; Kurtzman and Scanlon, 2011; 306 Tolmie et al., 2011; Baram et al., 2013). River, mountain-front, paleo or other types of 307 recharge may contribute, as well, to a situation where relatively fresh aquifer exists under 308 a saline vadose zone.

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4. Impact of cultivation on flushing of the unsaturated zone and aquifer salinization

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The anomalous situation of fresh groundwater under a saline unsaturated zone found in some native-land vertisols in semiarid regions exists due to the efficient evapotranspiration by natural vegetation and cracks (making deep unsaturated matrix immobile and saline) and fresh groundwater recharge through preferential flow in cracks or other types of recharge. However, what happens when natural conditions are changed to less favorable for native-vegetation and soil cracks (e.g. cultivated land and more ever irrigated intensive cropping)? The answer is obvious: higher fluxes may develop in the unsaturated matrix which will flush salts and ultimately cause salinization of the underlying aquifer.

321 A large bulk of literature from eastern Australia has reported increased deep-322 drainage and leaching of salts, and in some cases, salinization of aquifers under cultivated 323 vertisols. A typical increase in deep drainage from < 1 mm/yr under native conditions to 324 10 - 20 mm/yr under rain-fed cropping were reported by Silburn et al., 2009; Timms et 325 al., 2012 and Young et al., 2014; whereas variable deep fluxes often in the 100's mm/yr 326 range were reported for irrigated fields (mostly furrow-irrigated cotton,. Gunwardena et 327 al., 2011; Silburn et al., 2013; Weaver et al., 2013). These deep fluxes desolate salts that 328 accumulated in the vadose zone in the native-vegetation period, moving them down 329 towards the water table (Fig. 3). Earlier studies reporting leaching of salts from the 330 vadose zone after clearing of natural eucalyptus trees for cropping include Allison and 331 Hughes (1983) and Jolly et al. (1989), who worked in semiarid zones in Southern 332 Australia. In those studies, neither vertisols nor the role of soil cracks was mentioned; 333 however, deep eucalyptus roots act similar to cracks to form a very saline and immobile 334 deep-unsaturated-zone matrix which becomes more mobile and less saline after the land-335 use change. Timms et al. (2012) inferred, from combined soil and groundwater data, deep 336 drainage and salt leaching after conversion to cropping under gray vertosols in the 337 Murray–Darling Basin. Fresh groundwater was found in that study under shallower saline 338 waters, strengthening the source of groundwater salinity from the vadose zone.

339 Scanlon et al. (2009) compared mobilization of solutes in the vadose zone after a 340 change in the natural landscape to cultivated fields in three semiarid regions: Amargosa 341 Desert (southwestern USA), Southern High Plains (central USA) and Murray Basin 342 (southeast Australia). Flushing of chloride from the top 6-10 m of the vadose zone after 343 cultivation was very clear (e.g. Fig. 3 in Scanlon et al., 2009). Flushing has been observed 344 in many arid and semiarid regions, and not exclusively related to vertisols (e.g., Oren et 345 al., 2004, in the arid Arava Valley, southern Israel). Nevertheless, salinization of an 346 aquifer due to cultivation and salt mobilization may be more pronounced under vertisols 347 due to: preferential flow paths related to soil cracks (enabling the native aquifer to be 348 relatively fresh) and the cracks evaporative capabilities (making the native deep vadose 349 zone more saline).

350 A good example of an aquifer in which vertisols made a difference is the 351 Mediterranean Coastal Aquifer in Israel (Fig. 4). Although known as a coastal aquifer the 352 phenomena discussed here are all a few km inland and are not related to seawater 353 intrusions. The parts of this aquifer overlain by vertisols were salinized a few decades 354 after intensive-cultivation, whereas the water in those parts of the aquifer overlain by 355 cultivated loamy-sand is still potable (Kurtzman, 2011; Kurtzman and Scanlon, 2011; 356 Fig. 4). Similar to the Murray–Darling Basin (Timms et al., 2012), the upper groundwater 357 under vertisols in this aquifer were more saline than the deep groundwater (e.g., Fig. 1 in 358 Baram et al., 2014). Identification of the source of the salt and the cause of the 359 salinization in the deep unsaturated zone under vertisols and land-use change, respectively, contradicted previous works which attributed the salinization of these parts 360 361 of the Israeli Coastal Aquifer to the intrusion of deep brines and intensive pumping (e.g., 362 Vengosh and Ben-Zvi, 1994; Avisar et al., 2004). A different and shorter temporal trend 363 that might also be interpreted in light of soils covering the recharge area is the response 364 of groundwater salinity of the Israeli Coastal Aquifer to extreme precipitation (e.g. winter 365 of 1991/1992): under vertisols, freshening of the aquifer (decrease in salinity) was 366 generally observed due to recharge of freshwater through preferential paths, mostly under 367 uncultivated parts; under loamy-sand soils, salinization of the aquifer was observed due 368 to piston-flow recharge pushing relatively saline vadose-zone pore-water down to the 369 water table (Goldenberg et al., 1996, interpreted by Kurtzman and Scanlon, 2011).

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5. Relatively little nitrate contamination in aquifers under vertisols

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373 Whereas the literature concerning salinization of aquifers and draining of salts 374 from the vadose zone under cultivated vertisols is abundant, much less has been written 375 about the contamination of groundwater by nitrate under these soils. Nitrate is the most 376 problematic groundwater contaminant associated with agriculture worldwide (Jalali, 2005; Erisman et al., 2008, Burow et al., 2010; Vitousek et al., 2010; Kourakos et al., 377 378 2012). Both mineral nitrogen fertilizers (e.g. Kurtzman et al., 2013) and organic forms of 379 nitrogen (e.g. Dahan et al., 2014) are often applied in excess with respect to the plants' 380 ability to take up the nitrogen, leaving significant quantities of nitrate as a potential 381 groundwater contaminant.

While in the previous sections aquifers under vertisols were shown to be vulnerable to salinization, due to the agricultural practice above, there is an increasing number of observations that indicate lesser nitrate contamination in groundwater under 385 cultivated vertisols relatively to groundwater of the same aquifer located under cultivated 386 land of lighter soils. Kurtzman et al. (2013), dealing with nitrate contamination problems of the Israeli Coastal Aquifer, show that at the groundwater basin scale (~ 2000 km²) the 387 388 contamination plumes of nitrate are present in the aquifer only under cultivated sandy-389 loams, whereas under cultivated vertisols seldom sporadic wells produce water with 390 nitrate concentration above the drinking-water standard (Fig. 4). Dafny (2014), revealed, 391 by chi-square analysis that groundwater under cultivated vertisols and thick clayey-392 alluvial unsaturated zone, are less likely than groundwater under coarser sediments, to get 393 contaminated by nitrate in the Condamine Floodplain Aquifer in Australia.

In contrast to the relatively high capability of vertisols to reduce nitrate leaching from cultivated land both Baram et al. (2014) (Israel, Coastal Aquifer) and Dafny (2014) (Condamine Floodplain Eastern, Australia) acknowledge that concentrated animal feeding operations (CAFO) can be significant point sources of nitrate in vertisols as well. This might be due to incidental percolation of CAFO wastewater through the crack systems.

Silburn et al. (2013) indicate that modern deep drainage and any solutes are still migrating down through the unsaturated zone in vertisol-aluvial systems in Australia and the nitrate is accumulating in the unsaturated zone. Nevertheless in vertisols areas overlaying the Israeli Coastal Aquifer the rise in salinity and unsaturated flow and transport models, indicate that the cultivation effects reached the water table, yet nitrate contamination is not severe, suggesting other mechanism are responsible for the low levels of nitrate contamination.

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408 Denitrification in clayey soils is thought to be the major reason for the reduced deep
409 leaching of nitrate in semiarid climates; this reduction of nitrate to gaseous nitrogen is
410 less likely to be significant in lighter soils (Sigunga et al., 2002; Baram et al., 2012b;
411 Boy-Roura et al., 2013; He et al., 2013).

412 Jahangir et al. (2012) found that adding carbon to deeper soil horizons significantly 413 enhances denitrification in those layers. Profiles of dissolved organic carbon (DOC) in 414 deep vadose zones (down to 9 m below ground) under citrus orchards on thick vertisols 415 versus sandy-loam in Israel were compared. Whereas DOC in the lighter soils was higher 416 than 15 mg/kg_{drv soil} only in the top 1 meter, in the vertisols it was above 30 mg/kg_{drv soil} in 417 the entire 9 m profile (Shapira, 2012). These latter two studies support the notion that 418 denitrification in the root zone, and perhaps beyond, results in less nitrate problems in 419 aquifers under cultivated vertisols than under lighter soils. Thayalakumaran et al. (2014) 420 reported high DOC in shallow groundwater overlain by irrigated sugarcane corresponds with 421 absence of nitrate in this aquifer in northeast Australia.

Denitrification in the root zone and deeper in the soil profile explains the small amount of nitrate leached to the groundwater under rice fields in clayey soils in California, USA (Liang et al., 2014). Shallow groundwater (<1.5 m) under cultivated vertisols (e.g. Netherlands) showed large variability (spatial and temporal) in nitrate concentration, probably due to the highly variable oxygen concentrations and therefore variability in nitrogen transformations in these systems (Hendriks et al., 1999).

A more speculative mechanism that might explain the relatively lower occurrence of groundwater nitrate contamination involves the anion-exchange capacity of the clay. Harmand et al. (2010) observed very significant adsorption of nitrate to kaolinite and oxyhydroxides under a fertilized coffee plantation growing on an acrisol in Costa Rica. In 432 vertisols, montmorillonite is usually the dominant clay mineral; nevertheless, some 433 kaolinite is found in most vertisols (e.g. Singh and Heffernan, 2002; Krull and Skjemstad, 434 2003; Baram et al., 2012b). Another drawback of this mechanism as dominant in vertisols 435 is: adsorption of anions to positively charged surface is more efficient at low pH, while 436 vertisols in semiarid regions are usually neutral to alkaline. Retardation of nitrate in the 437 vadose zone due to adsorption to positively charged sites within the clay might slow 438 down groundwater contamination under cultivated vertisols. Nevertheless, if significant, 439 this mechanism would only retard groundwater contamination, whereas denitrification 440 removes the nitrogen from the soil-unsaturated zone-aquifer system. The idea of nitrate 441 adsorption has been tested as an engineered solution for reducing deep nitrate percolation. Artificially synthesized materials that have nitrate-sorption capacity (e.g. $[Mg^{2+}_{0.82} Al^{3+}]$ 442 $_{0.18}$ (OH)₂]^{0.18+}[(Cl-)_{0.18} · 0.5(H2O)]^{0.18-}) are being tested as soil additives to buffer nitrate 443 444 leaching (Torres-Dorante et al., 2009).

445

446 **6.** Conclusions

447

448 Vertisols are considered arable soils in semiarid climates, and are intensively 449 cultivated. Located in lowlands, vertisols often overlie aquifers. Flow and transport 450 through the cracking clays is complex and results in unique land-aquifer phenomena. 451 Observations from the lysimeter to basin scale have shown (directly and indirectly) the 452 significance of cracks as preferential flow paths in vertisols that bypass matrix blocks in 453 the unsaturated zone. These preferential paths support recharge with relatively fresh 454 water in uncultivated vertisols, and groundwater contamination from point sources such 455 as CAFOs and under some conditions, from crop fields. Deep soil samples under 456 uncultivated vertisols in semiarid regions reveal a dry (immobile), saline matrix, partly 457 due to enhanced evaporation through the soil cracks. This evaporation is related to 458 convective instability due to colder air at ground surface and warmer air deep in the crack 459 during the night. In some aquifers lying beneath vertisols in these regions, relatively fresh 460 groundwater exists under the saline unsaturated zone. Land-use change to cropland 461 promotes greater fluxes through the saline matrix which flush salts into the aquifer and 462 eventually cause groundwater salinization. In contrast to the vulnerability of groundwater 463 under vertisols to salinization, observations show that this soil-aquifer setting has some 464 resistance to groundwater contamination by nitrate (the major agriculturally related 465 groundwater contamination). Denitrification is probably the main mechanism supporting 466 this resistance, whereas anion-exchange capacity may have a retarding effect as well. 467 468 Acknowledgment 469 470 The study was supported by the Agricultural Research Organization (ARO), Israel. 471 472 References 473 474 Acworth, R. I. and Timms, W.A.: Evidence for connected water processes through 475 smectite-dominated clays at Breeza, New South Wales, Aust. J. Earth Sci., 56, 81–96, 476 2009. 477

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904 Figure captions

905

906 Fig. 1. Illustration of potential fluxes of water and pollutants that bypass the matrix,907 which is typical of vertisols.

908

Fig. 2. Desiccation Crack-Induced Salinization (DCIS), Baram et al. (2013). Convective
instability of air in soil cracks, occurring mainly at night, leads to drying and salinization
of the unsaturated zone.

912

Fig. 3. Flushing of chloride down through the unsaturated zone under cultivated vertisols:
(a) Silburn et al. (2009) – 19 years of flushing; (b) Radford et al. (2009) - full diamond,
native vegetation; empty, annual cropping; flushing from the top 3 m (c) Kurtzman and
Scanlon, (2011) – red, natural land; blue, irrigated cropping; flushing from 2 -10 m depth;
(d) Tolmie et al. (2011) –flushing from the top 1.5 m. (e) Timms et al. (2012) - black,
cropping; empty - grass; flushing from the top 2 m.

919

920 Fig. 4. Plan views of the Israeli Coastal Aquifer. (a) Soil type (black polygons and red 921 ellipses for spatial comparisons with panels d and e, respectively). (b) Location map. (c) 922 Cultivated land in the year 2000. (d) Difference in chloride concentrations between 2007 923 and 1935 (modified from Livshitz and Zentner, 2009). Black polygons are characteristic 924 cultivated areas that were severely salinized (southern polygons) and barely salinized 925 (northern polygon) relative to soil type (panel a). (e) Nitrate concentration in groundwater 926 wells in 2007 (modified from Hydrological Service, 2008). Red ellipses - areas with 927 many wells contaminated with nitrate relative to soil type (panel a) (modified from 928 Kurtzman et al., 2013).









