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Spatio-temporal impacts of dairy lagoon water reuse on soil: Heavy metals and salinity

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## Abstract

Diminishing freshwater resources have brought attention to the reuse of degraded water as a water resource rather than a disposal problem. The spatial impact and sustainability of dairy lagoon water reuse from concentrated animal feeding operations (CAFOs) has not been evaluated at field scale. The objective of this study is to monitor the impact of dairy lagoon water blended with recycled water on a 32-ha field near San Jacinto, CA from 2007 to 2011. Spatial monitoring was based on soil samples collected at locations identified from apparent soil electrical conductivity (EC<sub>a</sub>) directed sampling. Soil samples were taken at depth increments of 0-0.15, 0.15-0.3, 0.3-0.6, 0.6-0.9, 0.9-1.2, 1.2-1.5, and 1.5-1.8 m at 28 sample sites on 7-11 May 2007 and again on 31 May – 2 June 2011 after 4 years of irrigation with the blended waters. Chemical analyses included salinity (electrical conductivity of the saturation extract,  $EC_e$ ), pH<sub>e</sub> (pH of the saturation extract), SAR (sodium adsorption ratio), trace elements (As, B, Mo, Se), and heavy metals (Cd, Cu, Mn, Ni, Zn). Results indicate a decrease in mean values of pH<sub>e</sub> at all depth increments; a decrease in EC<sub>e</sub> and SAR above a depth of 0.15 m, but an increase below 0.15 m; a decrease in all trace elements except B, which increased throughout the 1.8-m profile; and the accumulation of Cd, Mn, and Ni at all depth increments, while Cu was readily leached from the 1.8-m profile. Zinc showed little change. The results focused concern on the potential long-term agronomic effect of salinity, SAR, and B, and the long-term environmental threat of salinity and Cu to detrimentally impact groundwater. The accumulation of Cd, Mn, and Ni in the soil profile raised concern since it provided a potential future source of metals for leaching. The long-term sustainability of dairy lagoon water reuse hinges on regular monitoring to provide spatial feedback for site-specific management.

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22	9	the saturation extract (dS m <sup>-1</sup> ); EMI, electromagnetic induction; EM <sub>h</sub> , electromagnetic induction
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## 1. Introduction

Water scarcity is a global problem. Finite water resources are stressed by increased urban demands, increased water needs by agriculture to meet growing food demands from a growing world population, increased frequency of drought due to erratic weather patterns stemming from climate change, and continued degradation from point and non-point sources of pollution. Concomitantly, as demand for freshwater resources has increased the volumes of degraded waters have increased and their disposal has become a concern due to regulations that restrict the conditions under which disposal is permitted stemming from the heightened awareness of the public to detrimental environmental impacts. The reuse of degraded water is viewed as a means of ameliorating water scarcity and reducing degraded water volumes for disposal. 

Degraded waters include agricultural drainage water, municipal wastewater, confined animal feeding operations (CAFO) wastewater, food processing wastewater, urban and agricultural runoff, and industrial wastewater. Each type of degraded water contains contaminants that make its reuse problematic. For instance, drainage water from the west side of California's San Joaquin Valley has elevated levels of salinity and trace elements (Corwin et al., 2008)<sup>1</sup>, whereas CAFO wastewater contains pathogens, nutrients, antibiotics, hormones, heavy metals, and salinity (Bradford et al., 2008).<sup>2</sup>

Even though the physical, chemical, and biological characteristics of degraded waters are known, the documented impact of degraded water reuse at field scale on agricultural lands over several years to establish the sustainability of degraded water reuse is limited (Corwin and Bradford, 2008).<sup>3</sup> For instance, there have been reuse studies for drainage water (Goyal et al., 1999; Corwin et al., 2008)<sup>1,4</sup>, CAFO wastewater (Evans et al., 1984; Burns et al., 1985; King et al., 1985)<sup>5-7</sup>, and food-processing wastewater (Kroyer, 1995; Wersin et al., 2001; Zvomuya et al.,

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2005; Johns and Bauder; 2007)<sup>8-11</sup> that have looked at the fate of particular associated contaminants but the majority of these have been over limited time periods of generally 1 to 2 years, which makes an evaluation of the sustainability of their reuse untenable. The reuse of municipal wastewater (e.g., Schmidt et al., 1975; Chakrabarti, 1995; Kivaisi, 2001; Lubello et al., 2004; Bixio et al., 2006; Chen et al., 2008)<sup>12-17</sup> and drainage water (e.g., Corwin, 2012)<sup>18</sup> are the most intensively studied degraded waters, with monitoring studies conducted under realworld conditions over extended time periods of a decade or longer.

The review by Bradford et al.  $(2008)^2$  on reuse of CAFO wastewater on agricultural lands concludes that additional studies on CAFO wastewater reuse are needed to test the hypothesis that when applied according to the current regulatory framework, which is based on nutrient management plans at agronomic rates to meet nutrient demands of crops, lagoon water contaminants are implicitly assumed to be retained, inactivated, or degraded in the root zone. Bradford et al. (2008)<sup>2</sup> warn that "potential environmental problems may occur due to accumulation of salts and heavy metals, incomplete degradation of anitbiotics or hormones, and/or survival of pathogens." 

Producers throughout the world use dairy lagoon wastewater as an alternate water resource for raising agricultural crops (Bradford et al., 2008).<sup>2</sup> Producers consider it a source of water, organic matter, and nutrients while resource specialists consider it a viable option for disposal of marginal waters. However, application of dairy lagoon wastewater poses a possible environmental peril to soil and water resources. The benefits of using dairy lagoon wastewater are partially offset due to accumulation of salts and heavy metals in the root zone, with potential deleterious effects on plant quality and yield (Alloway, 1995).<sup>19</sup> Heavy metals are of great concern because of their non-degradation, non-biodeterioration, and their bioaccumulation in the

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food chain (Kamari et al., 2011; Wu et al., 2011).<sup>20-21</sup> Heavy metals such as zinc (Zn), copper (Cu), Nickel (Ni), arsenic (As), cadmium (Cd), and lead (Pb) are potential bioaccumulative toxins from the dairy production system (Alloway, 1995; Thomas et al., 2006).<sup>19-22</sup> Soils tend to act as long-term sinks for these heavy metals due to their sorption onto metal oxides, particularly iron and manganese oxides, clay minerals, and soil organic matter (Kumpiene et al., 2008).<sup>23</sup> Heavy metal application to soils from dairy lagoon wastewater may result in their accumulation in soils, where they can be taken up by crops, perhaps exacerbating heavy metal exposure to animals and humans over the long term through the food chain. Therefore, heavy metal accumulation in soil from dairy lagoon water application is a concern due to its potential negative impacts on soil, plants, and animals. 

Corwin and Bradford (2008)<sup>3</sup> indicate that "to most efficiently manage degraded water reuse with the least detrimental environmental impact, it will be necessary to account for the spatial variability of sites where degraded waters are applied" thereby making "site-specific management of degraded water reuse possible, which will optimize the reuse of degraded waters and minimize detrimental impacts." This level of mangement requires a knowledge of the spatial and temporal distribution of contaminants associated with degraded water reuse. The need for site-specific management of degraded water reuse is reaffirmed by Vance et al.  $(2008)^{24}$  and Ganjegunte et al. (2008)<sup>25</sup>, whose work stress the need for alternative site-specific management practices and monitoring. Corwin and Bradford  $(2008)^3$  point out that future research is needed that will not only monitor long-term environmental impacts of degraded water reuse, but will monitor the impacts spatially to evaluate the true potential of degraded waters as a viable alternative water resource. Spatially and temporally monitoring contaminants from degraded water reuse will provide their fate and distribution, the threats they pose to the different

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1 environmental compartments, and useful information for management and mitigation.

In a series of papers by Corwin and Lesch (2003, 2005a, 2013)<sup>26-28</sup> the protocols and guidelines have been developed for characterizing the spatial variability of a range of soil properties using geophysical techniques (i.e., electromagnetic induction or EMI and electrical resistivity or ER) to measure apparent soil electrical conductivity (EC<sub>a</sub>) with an approach referred to as EC<sub>a</sub>-directed soil sampling. Because the geospatial measurement of EC<sub>a</sub> is a complex spatially measured property of soil that reflects the influence of several soil physical and chemical properties (including soil salinity, texture, water content, bulk density, organic matter, and cation exchange capacity) it is rarely used to map a single property, but rather  $EC_{a}$ serves as a surrogate to characterize the spatial variation of those soil properties that are found to influence EC<sub>a</sub> within a field. Characterizing spatial variability with EC<sub>a</sub>-directed soil sampling is based on the notion that when EC<sub>a</sub> correlates with a soil property or properties, then EC<sub>a</sub> information can be used to identify sites that reflect the range and variability of the property or properties. It has been repeatedly shown that in instances where EC<sub>a</sub> correlates with a particular soil property, an EC<sub>a</sub>-directed soil sampling approach will establish the spatial distribution of that property (Corwin and Lesch, 2005b; Corwin et al., 2010).<sup>29-30</sup> This holds true even for soil properties not directly measured by  $EC_a$  as shown by Corwin (2012)<sup>18</sup> in the characterization of the spatial variability of the trace elements B and Se. 

The spatio-temporal impact of drainage water reuse has been monitored using  $EC_a$ -directed soil sampling (Corwin et al., 2008; Corwin, 2012)<sup>1,18</sup>, but similar studies have not been conducted for other degraded waters. It is the objective of this study (i) to utilize  $EC_a$ -directed soil sampling to characterize and monitor at field scale the impact of dairy lagoon water reuse on salinity, pH, trace elements, and heavy metals over a 4-year period and (ii) to evaluate its

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sustainability in light of the potential environmental risks the reuse of dairy lagoon water may pose.

## 2. Methods and materials

A dairy lagoon water reuse study was initiated in June 2006 on a 32-ha field in San Jacinto, CA. Spatial variability of soil chemical and physical properties was determined using the EC<sub>a</sub>-directed soil sampling approach developed by Corwin and Lesch (2003, 2005a).<sup>26-27</sup> Assessments of spatio-temporal change focused on impacts on soil salinity, pH, trace elements, and heavy metals, which were characterizing from soil samples taken in 2007 and 2011 at locations in the field established from an initial EC<sub>a</sub> survey. 

**Study site** 

The on-farm research study site (lat. 33° 50' 25.43" N, long. 117° 00' 14.93" W) is located on Scott Brothers' Dairy Farm in San Jacinto in southern California's Riverside County. The 32-ha field site provided an extensive range of spatial variability in soil properties to make a real-world evaluation of the impact of dairy lagoon water reuse with respect to the fate and distribution of salinity, pH, trace elements, and heavy metals. The USDA National Resource Conservation Service classifies the western half of the field as a Fluvaquentic Haploxerolls, coarse-loamy, mixed, thermic soil and the eastern half as a Typic Xerofluvents, coarse-loamy, mixed (calcareous), thermic soil. These are well drained soils with slopes of generally 0-2% and textures ranging from loam in the western portion of the field to fine sandy loam in the middle to a sandy loam in the eastern portion of the field.

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Geospatial EC<sub>a</sub> measurements were obtained with the Geonics EM38 dual-dipole electrical conductivity meter.<sup>†</sup> The EC<sub>a</sub> survey followed the detailed survey protocols outlined by Corwin and Lesch (2005a).<sup>27</sup> The  $EC_a$  survey was conducted on 22-23 April 2007. The survey consisted of geospatial EC<sub>a</sub> measurements taken with mobile EMI equipment where measurements were simultaneously taken both in the horizontal  $(EM_h)$  and vertical coil configurations  $(EM_v)$  at time intervals resulting in measurements approximately every 5 m. Measurements were taken at 16,122 locations on transects running in a north-south direction resulting in the EC<sub>a</sub> survey maps of EM<sub>v</sub> and EM<sub>h</sub>. However, EM<sub>v</sub> provided the most useful information for sample site selection; consequently, only the  $EM_v EC_a$  survey is provided in Fig. 1. 

## 12 Soil core and plant yield sampling

Soil core sample sites were selected using the geo-referenced EC<sub>a</sub> survey data as a surrogate for the spatial variation of soil properties correlating with EC<sub>a</sub>. Based on the variation in EC<sub>a</sub>, 30 soil sample sites were selected that reflected the range and variation in EC<sub>a</sub> using a design-based sampling strategy, i.e., stratified random sampling design (SRSD). The design-based sampling plan (i.e., SRSD) was developed using the ESAP software program, version 2.35 (Lesch et al., 2000).<sup>31</sup> Figure 1 shows the location of the selected soil sampling sites. Soil samples were collected at each site for the following depth increments: 0-0.15, 0.15-0.30, 0.30-0.60, 0.60-0.90, 0.90-1.20, 1.20-1.50, 1.50-1.80 m. At each of the 30 sites, soil-core samples were taken at 2 points (i.e., two sets of soil cores per site) roughly 5 centimeters apart. One set of soil cores was designated for soil chemical property analysis and the other set for soil physical property analysis. A total of 420 soil samples were taken (210 soil chemical property samples, 210 soil

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physical property samples). Unfortunately, 2 sampling sites were located at positions where
center-pivot tracks overlapped the sites; consequently, the 2 sites were dropped resulting in 28
total sites.

To observe temporal changes resulting from the application of dairy lagoon water, soil core samples were taken at the same selected sample site locations on 7-11 May 2007 and again after 4 years of irrigation with dairy lagoon water on 31 May – 2 June 2011. Soil samples were taken when the field was at field capacity. Soil samples were also taken when the crop would have little short-term influence on spatial variability. In 2007 and 2011 soil samples were taken a few days after planting of a crop and just following irrigation. These precautions were taken to minimize short-term temporal influences, particularly at shallow depths. All soil cores were kept in refrigerated storage prior to being air-dried and sieved (2-mm sieve), which occurred within a few days after their collection.

Saturation extracts of the soil sample depths were prepared and the electrical conductivity of the saturation extracts (ECe, dS m<sup>-1</sup>) were measured using the method presented in Rhoades (1996).<sup>32</sup> The depth of measurement of the EM38 in the vertical coil configuration (EM<sub>v</sub>) most closely corresponds to the root zone; consequently, it was used to direct the soil sampling following the protocols of Corwin and Lesch (2005a).<sup>27</sup> The depth-weighted average EC<sub>e</sub> at each sample site was calculated over the 0-0.15, 0-0.30, 0-0.60, 0-0.90, 0-1.20, 0-1.50, and 0-1.80 m depth increments to establish the actual depth of measurement of the  $EM_{y}$  according to the protocols of Corwin and Lesch (2005a).<sup>27</sup> The depth increment that provided the best correlation between EC<sub>e</sub> and EM<sub>v</sub> EC<sub>a</sub> indicated the depth of measurement. The depth increment of 0-1.80 m was found to correlate best with a correlation coefficient of 0.91; consequently, the sample design for the 0-1.8 m composite depth and all depth increments down to 1.8 m was verified.

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Sorghum (Sorghum bicolor (L.) Moench) was grown in 2007. Plant yield samples were taken
 within a 1 m<sup>2</sup> area at each of the 28 soil sampling sites. Plant yield was determined on a total wet
 weight basis. No crop yield monitoring was conducted in 2011.

5 Soil physical and chemical analyses

The soil cores were analyzed for a range of physical and chemical properties that could be potentially impacted by the reuse of dairy lagoon water. The soil chemical properties included:  $EC_e$ , pH<sub>e</sub>, sodium adsorption ratio (SAR is a ratio of Na<sup>+</sup> to Ca<sup>2+</sup> and Mg<sup>2+</sup> measured in meq L<sup>-1</sup> where SAR=  $Na^{+}/[0.5(Ca^{2+} + Mg^{2+})]^{1/2}$ , anions (HCO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>) and cations (NH4<sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>) in the saturation extract, trace elements (B, Se, As, Mo) in the saturation extract, heavy metals (Cd, Cu, Mn, Ni, Zn) in the saturation extract, heavy metals (Cd, Cu, Mn, Ni, Zn) in the Mehlich-I (double acid) extract, organic C (OC), inorganic C (IC), total C (TC), total N (TN), and exchangeable cations ( $NH_4^+$ ,  $Na^+$ ,  $K^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ). Saturation percentage (SP) was determined during the process of obtaining the saturation extracts to perform the chemical analyses. The soil samples designated for analysis of soil physical properties were analyzed for gravimetric water content ( $\theta_{o}$ ) and bulk density ( $\rho_{b}$ ). Volumetric water content was calculated from  $\theta_g$  and  $\rho_b$ . Bulk density was measured from undisturbed 5-cm diameter soil core samples taken over the depth increments of 0-0.15, 0.15-0.30, 0.30-0.60, 0.60-0.90, 0.90-1.20, 1.20-1.50, 1.50-1.80 m. The soil sample preparation, and chemical and physical methods used for each analysis were from SSSA Book Series No. 5 Methods of Soil Analysis Parts 3 and 4 (Sparks, 1996; Dane and Topp, 2002; respectively)<sup>33-34</sup> except for total C and N, which were analyzed with a Leco C-N 2000 Analyzer<sup>†</sup> (Leco Corporation, St. Joseph, MI). 

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A geographic information system (GIS) was used to display and manipulate the spatial data. All spatial data were entered into the GIS with the commercial GIS software ArcGIS 9.0. Interpolated maps of the soil chemical properties most significantly impacted by dairy lagoon water reuse (i.e., EC<sub>e</sub>, SAR, pH<sub>e</sub>, Cd, Cu, Mn, Ni, and Zn) or influencing the impact (i.e., texture as reflected by the SP) were prepared using ordinary inverse-distance-weighting (IDW) interpolation. A comparison of IDW interpolation to kriging for all sampling times using jackknifing showed a general improvement in prediction of the interpolated values using IDW; consequently, all interpolations were done using IDW.

11 Irrigation with dairy lagoon wastewater

GIS and map preparation

The study site was irrigated using a center-pivot irrigation system (Reinke, Nebraska). Initial irrigation uniformity studies indicated a coefficient of uniformity of 82% during the study period, which reflected a cyclic pattern in the irrigation water application as a function of radial distance. Three water sources were used: treated dairy lagoon wastewater (DLWW), recycled water, and well water. The DLWW was treated in two stages: (1) a series of notched weirs along the canal, which channeled the DLWW from the dairy to the lagoon, removed most of the solids before entering the lagoon and (2) a self-cleaning filter (SAF-3000, Amiad Filtration Systems, Israel) with 200 mesh screen was installed before blending with the well or recycled waters to remove solids that could clog the irrigation system emitters. The recycled water originated from a nearby wastewater treatment plant. As an indication of their general composition, the average concentrations of salts, macro-nutrients, and micro-nutrients of treated DLWW, recycled water, and well water are presented in Table 1 for the time periods winter 2007 and summer 2009. From

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May 2007 to May 2008 treated DLWW was blended with well water and from May 2008 to June 2 2011 treated DLWW was blended with recycled water. The total dissolved solids (TDS) of the 3 recycled water was roughly double that of the local well water (600 versus 320 mg·L<sup>-1</sup>; see Table 4 1), whereas the considerable inorganic *N* in the recycled water needed to be taken into account in 5 the *N* mass balance.

The blend ratio for each irrigation application was based on the projected inorganic nitrogen uptake by the plant. The projected plant uptake for each time interval was determined from potential nitrogen uptake curves for the crops (i.e., triticale was the winter crop and sorghum was the summer crop) under optimum growth conditions.<sup>35-36</sup> The blend ratio for each application was determined by meeting the nitrogen need from the equation  $N_{Application} = N_{Well/Recycled}$ .  $I_{Well/Recycled} + N_{DLWW} \cdot I_{DLWW}$ , where the total applied irrigation water ( $I_{Application}$ ) is comprised of the total applied well or recycled water ( $I_{Well/Recycled}$ ) and total applied dairy lagoon wastewater  $(I_{DLWW})$  and N<sub>Application</sub> is the total applied inorganic nitrogen in  $I_{Application}$  comprised of the inorganic nitrogen supplied by the well or recycled water ( $N_{Well/Recycled}$ ) and dairy lagoon wastewater ( $N_{DLWW}$ ). The inorganic N level of each water was measured prior to each irrigation event. 

Table 2 shows an analysis of the chemical composition of the blended water that actually reached the soil surface from the center-pivot irrigation system. The detailed chemical analysis was conducted on water samples taken from collection pans placed at 3 of the 28 soil sample locations. Water samples were collected for approximately 2/3 of the irrigations. All chemical constituents of the blended water reaching the soil surface shown in Table 2, except SAR, are below the maximum concentration levels in irrigation water for continuous agricultural use on all soils as recommended by the National Academy of Sciences-National Academy of Engineering

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Committee on Water Quality (NAS-NAE Committee on Water Quality, 1972).<sup>37</sup> The
recommended limits for As, B, Mo, Se, Cu, Cd, Mn, Ni, and Zn in mg L<sup>-1</sup> are 0.1, 0.75, 0.01,
0.02, 0.20, 0.01, 0.20, 0.20, and 2, respectively.<sup>37</sup> Table 2 indicates that most of the center-pivot
irrigation water reaching the soil surface over the four years of the study had SAR over 4.
Bernstein (1967)<sup>38</sup> indicates that irrigation water with a SAR from 4-8 poses a potential threat to
crop yields. With EC of the blended irrigation water hovering around 1 dS m<sup>-1</sup> and SARs in the
vicinity of 4, slight to moderate reduction in infiltration can be expected over time.

## 3. Results and discussion

Even though only SAR exceeded the recommended maximum concentration level in irrigation water for continuous agricultural use on all soils, the levels of salinity (i.e., EC of the irrigation water), B, and Cu in the blended irrigation water are sufficiently high to be of potential long-term concern (Table 2).

Table 3 shows the correlation between EC<sub>a</sub> for EM<sub>v</sub> for the various soil properties measured at the study site in 2007 and 2011. The properties that are significantly correlated at the p < 0.05level indicate those properties that are spatially well characterized by the EC<sub>a</sub>-directed stratified random sampling design. The significantly correlated properties (significant at p<0.05 level) include:  $\theta_g$ ,  $\theta_v$ ,  $\rho_b$ , SP, EC<sub>e</sub>, pH<sub>e</sub>, Cl<sup>-</sup>, HCO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, SAR, trace elements (As, B, Mo) and heavy metals (Cu, Ni, Zn) in the saturation extract, OC, IC, TC, exchangeable Na (Ex-Na<sup>+</sup>), exchangeable Mg (Ex-Mg<sup>2+</sup>), and double acid heavy metals (Cd, Cu, Mn, Ni, Zn). Of these, many are significant at the p $\leq 0.01$  level:  $\theta_g$ ,  $\theta_v$ ,  $\rho_b$ , SP, EC<sub>e</sub>, pH<sub>e</sub>, Cl<sup>-</sup>, HCO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, SO<sub>4</sub><sup>2-</sup>, Na<sup>+</sup>, SAR, trace elements (As, B, Mo) and heavy metals (Cu, Ni, Zn) in the saturation extract, Ex-Na<sup>+</sup>, Ex-Mg<sup>2+</sup>, and double acid heavy metals (Cd, Ni). Any properties that are not 

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significant at p≤0.05 level are properties not spatially characterized by the EC<sub>a</sub>-directed stratified
 random sampling design.

Table 3 also provides the correlation between sorghum yield on a wet weight basis in 2007 and various soil properties. Sorghum yield is significantly correlated at the p<0.05 level to  $EM_{y}$ EC<sub>a</sub>, EC<sub>e</sub>, SAR, the trace elements As and B in the saturation extract, and Cl<sup>-</sup> and Na<sup>+</sup> in the saturation extract. The correlation coefficients are moderate in value (r between -0.4 to -0.5). The correlation coefficient between EM<sub>v</sub> EC<sub>a</sub> and sorghum yield is -0.470. As indicated by Corwin et al.  $(2003)^{39}$ , when correlation exists between EM<sub>v</sub> EC<sub>a</sub> and crop yield, then it is likely that EC<sub>a</sub> is measuring some edaphic property(ies) that influences crop yield. Sorghum yield is significantly correlated (at the p<0.05 level) to salinity (EC<sub>e</sub>), SAR, Cl<sup>-</sup>, Na<sup>+</sup>, As, and B, with correlation coefficients of -0.435, -0.429, -0.431, -0.453, -0.435, and -0.434, respectively. However, multi-collinearity exists between EC<sub>e</sub> and Cl<sup>-</sup>, and between SAR and Na<sup>+</sup>. Furthermore, Cl<sup>-</sup>, Na<sup>+</sup>, As, and B are not at sufficiently high levels to cause yield decrements due to toxicity. For instance, the B threshold for sorghum is 7.4 g m<sup>-3</sup>. Nowhere does the composite profile (0-1.8 m) exceed the B threshold nor is the threshold exceeded within any single depth increment (i.e., 0-0.15, 0.15-0.30, 0.30-0.60, 0.60-0.90, 0.90-1.20, 1.20-1.50, 1.50-1.80 m) anywhere in the field. The same can also be said for  $Cl^{-}$ ,  $Na^{+}$ , and As. The double acid extract heavy metals had no statistically significant correlation with sorghum yield. Subsequently, ECe and SAR are the edaphic properties most likely affecting yield in 2007. 

The salinity threshold for sorghum is  $6.8 \text{ dS m}^{-1}$ . Even though there are no locations within the study site where the composite profile (0-1.8 m) exceeds this threshold, there are 8 soil core locations where the salinity threshold is exceeded within the 0-0.15 m depth increment. This is significant since the crop germination stage is particularly sensitive to salinity. The 8 locations

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are found in the northwest corner and mid-section of the field. Correlating sorghum yield to EC. for the 0-0.15 m depth increment at these 8 locations shows a strong negative correlation (r = -0.91). In addition, indirect influences on yield are likely occurring from permeability problems created from the irrigation water quality, moderately high SARs, and fine-textured soil present in the northwest corner and mid-section of the study site. Permeability is lowered due to the dispersion of clay by Na<sup>+</sup>. Low permeability influences salt accumulation by decreasing the leaching of salts, which explains why salt accumulation occurred in the western two-thirds of the field where higher SARs and finer textured soil occurred. 

## 10 Temporal trends of EC<sub>e</sub>, SAR, pH<sub>e</sub>, B, and heavy metals from 2007-2011

Table 4 provides the means and associated standard deviations of the soil properties of potential concern related to dairy lagoon water reuse for 2007 and 2011 over the entire 0-1.8 m soil profile. These statistics show the extent of change in the field means of soil properties over the 4-year period of dairy lagoon water reuse (i.e., 2007-2011). The most significant chemical impacts of dairy lagoon water reuse over the entire 0–1.8 m soil profile are for EC<sub>e</sub>, pH<sub>e</sub>, SAR, B, and the heavy metals Cd, Cu, Mn, and Ni. For each of these properties (i.e., EC<sub>e</sub>, pH<sub>e</sub>, SAR, B, and the heavy metals Cd, Cu, Mn, and Ni) the paired t-test indicates a statistically significant difference between 2007 and 2011 at the p<0.05 level. The trend from 2007 to 2011 over the 0-1.8 m soil profile is a general increase in the field means for  $EC_{e}$ , SAR, B, and double acid extract Cd, Mn, and Ni and decrease for pHe and double acid extract Cu; double acid extract Zn shows little change. From 2007 to 2011 EC<sub>e</sub> increased by 42%, SAR by 29%, B by 132%, and double acid extract Cd, Mn, and Ni increased by 482%, 44%, and 928%, respectively, while pHe and double acid extract Cu decreased by 7% and 85%, respectively. For both 2007 and 2011

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1	ECe, SAR, pHe, B, and double acid extracts of Cd, Cu, Mn, Ni, and Zn were significantly
2	correlated with $EC_a$ (Table 3); consequently, it can be concluded that the sampling design
3	generated from the $EC_a$ survey in 2007 characterized the spatial distribution of $EC_e$ , SAR, pH <sub>e</sub> ,
4	B, and double acid extracts of Cd, Cu, Mn, Ni, and Zn for both sample times and reliably
5	determined their field means.
6	Table 5 shows that in 2007 the salinity (ECe) was the highest at the soil surface (i.e., 0-0.15
7	m) with an EC <sub>e</sub> of 3.54 dS m <sup>-1</sup> and below 0.15 m fluctuated between 1.42 and 1.79 dS m <sup>-1</sup> . Table
8	5 shows that in 2007 the SAR was lower in the top 0.6 m, ranging from 4.32 to 6.15, and was
9	higher below 0.6 m, ranging from 9.94 to 21.43. From 2007 to 2011 (see Tables 5 and 6) $\text{EC}_{e}$
10	and SAR increased at all depth increments, except at the surface (i.e., 0-0.15 m). The leaching
11	fraction (LF) of 0.24, which was estimated from the ratio of average chloride concentration in
12	the irrigation water (5.86 meq $L^{-1}$ ) to average chloride concentration in the soil solution below
13	the root zone (average Cl <sup>-</sup> concentration at field capacity for a soil depth of 1.8 m is 24.12 meq
14	$L^{-1}$ ), and the volume-weighted blended irrigation water EC of 1.60 dS m <sup>-1</sup> were the reasons EC <sub>e</sub>
15	and SAR decreased at the surface from 2007 to 2011 but increased at all depths below 0.15 m. It
16	is no surprise that $EC_e$ is strongly correlated with LF, having correlation coefficients of -0.84
17	(p $\leq$ 0.01 level) for 2007 and -0.81 (p $\leq$ 0.01 level) for 2011, and correlated with SP, having
18	correlation coefficients of 0.63 for 2007 and 0.54 for 2011. The correlations between SAR and
19	LF are less predictable. SAR is strongly correlated with LF in 2007, with a correlation
20	coefficient of 0.78 (p $\leq$ 0.01 level), but unexpectedly there is little correlation in 2011, with a
21	correlation coefficient of 0.29. However, SAR correlates with SP with r=0.62 for 2007 and
22	r=0.47 for 2011, much like $EC_e$ correlated with SP.
23	The lack of correlation between SAR and LF in 2011 reflects a deviation in spatial patterns

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1	between $EC_e$ and SAR from 2007 to 2011. In 2007 and 2011 the general east to west spatial
2	patterns for the entire 0-1.8 m profile were similar for $EC_e$ and SAR with higher $EC_e$ and SAR in
3	the west and decreasing from west to east (Fig. 2A, 2B and 3A, 3B). Even though the general
4	trends for $EC_e$ and SAR were similar for 2007 and 2011, the spatial patterns for change in $EC_e$
5	and change in SAR from 2007 to 2011 as shown in Fig. 2C and 3C are less similar than would be
6	expected and reflect the lack of correlation of SAR to LF in 2011. The deviation in spatial
7	patterns is particularly evident in the 0-1.8 m profile depth in Fig. 2C and 3C. SAR distinctly
8	shows an increase from 2007 to 2011 in the eastern half of the field (Fig. 3C.h), while $EC_e$
9	clearly shows a decrease in $EC_e$ in a narrow north-south band in the middle of the field (Fig.
10	2C.h), which was the consequence of a management decision by the producer to leach salts from
11	the middle section of the field where higher salinity levels were present, causing yield
12	decrements. Since SP reflects texture (i.e., low SP indicates coarse-textured soils and high SP
13	indicates fine-textured soils), the correlation of SAR and SP indicates low Na levels associated
14	with coarse-textured soil due to greater leaching and high Na levels associated with fine-textured
15	soils where Na resides on the exchange sites of clays causing the soil to disperse thereby limiting
16	the leaching of Na and other major cations (e.g., $NH_4^+$ , $Na^+$ , $K^+$ , $Ca^{2+}$ , $Mg^{2+}$ ) and anions (e.g.,
17	$NH_4^+$ , $Na^+$ , $K^+$ , $Ca^{2+}$ , $Mg^{2+}$ ). The fine-textured soils with higher SPs, higher SARs, and higher
18	ECes were located in the western half of the field (Fig. 2A, 2B and 3A, 3B). The failure to leach
19	$Na^+$ from the middle section of the field along with the other salts indicates that there were
20	insufficient divalent cations in the irrigation water to remove $Na^+$ from the exchange sites. In
21	fact, $Na^+$ is accumulating in the middle of the field (Fig. 3C.h), particularly at the 0.9-1.2 and
22	1.2-1.5 m depths (Fig. 3C.e and Fig. 3C.f). Presumably the composition of the irrigation water
23	was the cause, resulting in an increase in SAR from 2007 to 2011 in the eastern half of the field

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In 2007 pH<sub>e</sub> was lowest at the surface with a pH<sub>e</sub> of 7.57 for 0-0.15 m and gradually increased with depth to 8.08 at 1.5-1.8 m (Table 5). In 2011  $pH_e$  was more erratic with depth, decreasing from 7.34 at the surface (i.e., 0-0.15 m) to 7.13 at 0.3-0.6 m and then increasing to 7.57 at 1.5-1.8 m (Table 6). Tables 5 and 6 indicate that from 2007 to 2011 pH<sub>e</sub> decreased at all depth increments. The greatest decrease in pH<sub>e</sub> occurred on the western half of the field (Fig.4C). The presence of  $NO_3^-$  and  $NH_4^+$  in the dairy lagoon water (see Table 2) and decomposition of organic matter in the top 15 cm of the soil (see % organic carbon, i.e., %OC=4.56 in Table 5) that had been applied to the soil as manure from 2004-2006, prior to the initiation of this study, were the causes for the decrease in  $pH_e$  over time. Organic matter decay produces  $H^+$ , which lowers the pH, but acidic soil development from decaying organic matter is generally less significant in the short term. Rather, the harvest of high-yielding crops plays a more significant role in increasing soil acidity. Ammonium in the dairy lagoon water also increases soil acidity, but the increased soil acidity is primarily the consequence of the increase in crop yields due to  $NO_3^-$  and  $NH_4^+$  from the dairy lagoon water. During growth, crops absorb basic elements such as  $Ca^{2+}$ ,  $Mg^{2+}$ , and  $K^+$  to satisfy their nutritional requirements. As crop yields increased (yield monitoring of triticale and sorghum showed a 13% and 15% increase in yield, respectively, from 2007 to 2011), more of these basic elements were removed (Tables 5 and 6 show the decrease in Ca<sup>2+</sup>, Mg<sup>2+</sup>, and K<sup>+</sup> from 2007 to 2011), resulting in increased soil acidity. Higher levels of ammonium and organic matter (particularly in the top 60 cm), higher crop yields, and greater decreases in Ca<sup>2+</sup>, Mg<sup>2+</sup>, and K<sup>+</sup> were found on the west side of the field, causing increased soil acidity in the western half of the field. 

The trace element composition of the blended irrigation water indicates that only B was high

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enough to be of potential concern (Table 2). Table 4 indicates that from 2007 to 2011 the 0-1.8
m soil profile decreased in As by 15%, increased in B by 132%, decreased in Mo by 50%, and
decreased in Se by 88% in the saturation extract. Only B showed accumulation while leaching
removed As, Mo, and Se due to their low levels in the blended irrigation water. The leaching of
As, Mo, and Se could pose potential detrimental impacts to the groundwater, but the levels are
sufficient low so that this is not a concern.

Tables 5 and 6 show that B increased at all depth increments from 2007 to 2011. Greater B was found on the west side of the field associated with finer-textured, higher organic matter content soil, which predominated the west side (Fig. 5). The clay and organic matter surfaces provided adsorption sites for B, which retained B and slowed its leaching through the soil profile. Boron was more readily leached from the east side of the field where coarser-textured, lower organic matter soil did not adsorb or retard the movement of B. The change in B levels at individual depth increments (Fig. 5C.a-g) shows a general accumulation in B at each depth increment except 1.5-1.8 m (Fig. 5C.g) in the northwest corner. A closer analysis of the northwest corner at 1.5-1.8 m (Fig. 5C.g) shows that of the 3 sites mapped as having a decrease in B from 2007-2011 two sites actually showed little change in B level (change in B of -0.001 and -0.002 mg  $L^{-1}$ ); therefore, it is surmised that B levels essentially increased at all depths across the field. 

Boron is of concern since plants require a narrow range of B between B deficiency and toxicity. Even though B levels did not reach toxic levels for the crops grown at the study site, the increasing average B levels in the saturation extract at all depths (Tables 5 and 6) raise potential concern about the long-term sustainability of dairy lagoon water reuse at the site and points to the need for close and regular monitoring.

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Table 5 shows that in general all the heavy metals studied decreased with depth for the double acid extract down to 1.2 m and then increased at the bottom two depths (i.e., 1.2-1.5 and 1.5-1.8 m). From 2007 to 2011 the distribution of the heavy metals varied for the entire 0-1.8 m profile, with Cu decreasing, while Cd, Mn, and Ni increased and Zn remained largely unchanged (Table 4). This held true for both the saturation extract and double acid extract. Since the double acid extract is considered to be more representative of what is available to the plant, the focus of the discussion will be on the double acid extract. From 2007 to 2011 Cu decreased within the entire 0-1.8 m profile by 85% for the double acid extract, whereas Cd, Mn, and Ni increased by 482%, 44%, and 930%, respectively. Tables 5 and 6 show that Cu decreased from 2007 to 2011 for all depth increments, while Cd, Mn, and Ni increased for all depth increments. Figure 6A shows that in 2007 the highest concentrations of plant available Cd were found in the western half of the field, but by 2011 the greatest concentration had shifted to the eastern half of the field (Fig. 6B). The Cd concentration correlated strongly with SP (r=0.64) and OC% (r=0.55) in 2007, suggesting that the decrease in plant available Cd from west to east was the consequence of greater leaching in the east due to more porous soil and less leaching in the west due to greater retention of Cd from adsorption on organic matter and reactive clay surfaces. The shift of higher Cd concentration to the eastern half of the field by 2011 is likely the consequence of the greater decrease in pH in the west over time (see Fig. 4), which increased its mobility in the west; consequently, even though Cd accumulated throughout the field, less accumulated in the west where Cd was more mobile due to lower pH, resulting in its removal by leaching. The correlation coefficient between change in Cd (double acid extract) and change in pH<sub>e</sub> from 2007 to 2011 of r=0.43 supports this assertion. 

Figures 6D-F shows that plant available Cu was substantially reduced from 2007 to 2011,

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particularly in the eastern half of the field where the more porous coarser-textured soil was
found. The decrease in Cu in the east is the result of greater leaching. When using the sequential
extraction technique to determine the concentration and partitioning of different heavy metals in
soil, the highest concentration of Cu was found in the organic matter fraction, whereas Cd was
found in the carbonate fraction, and Zn in the Fe-Mn oxide fraction. Ostensibly, the higher
organic matter in the western half of the field slowed the leaching of Cu.

Over time Mn and Ni behaved similar to Cd. From 2007 to 2011 plant available Mn increased particularly in the east (Figs. 6G-I). As was the case for Cd, the increased Mn concentration to the eastern half of the field by 2011 is likely the consequence of the greater decrease in pH in the west over time (see Fig. 4), which increased its mobility in the west. The pH effect overshadowed any textural influences. The negative correlation of Mn with SP both in 2007 (r = -0.69) and 2011 (r = -0.75) suggests that the finer the soil texture, the lower the concentration of plant available Mn; consequently, plant available Mn was higher where the soil was coarser textured, which is where it would be expected to be leached more and subsequently lower in concentration. Ostensibly, the lower pH in the west increased Mn mobility beyond what was leached from the coarser textured soil in the east, causing greater removal of Mn from the west than the east. Nickel shows similar patterns in distribution as Mn from 2007 to 2011, suggesting a similar relationship with pH (Figs. 6J-L). 

In Zinc showed erratic and complex spatial patterns (Figs. 6M-O), which did not correlate with any soil properties that would provide insight into the mechanism that most influenced the spatial distribution. The only discernible spatial feature was the accumulation of Zn at the surface (0-0.15 m) where the organic matter was the highest.

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4. Conclusions and implications

Expectedly, there are some overlaps in the results presented in this paper and the work of Corwin et al.  $(2008)^1$  and Corwin  $(2012)^{18}$ , who introduced the use of EC<sub>a</sub>-directed soil sampling to monitor the spatio-temporal impact of degraded water reuse. However, there are distinct contributions that distinguish this dairy lagoon water reuse study from the previous evaluation of the impacts of drainage water reuse on a saline-sodic soil. The most obvious is the demonstration that EC<sub>a</sub>-directed soil sampling can be used to monitor spatial and temporal changes in heavy metals due to degraded water reuse. This study also demonstrated the short-term (4 years) viability of dairy lagoon water reuse as an alternative water resource for agriculture with potential broad geographic application on irrigated agricultural lands throughout the world where water scarcity is a concern.

The reuse of dairy lagoon water blended with recycled or well water on agricultural land presented no detrimental environmental impacts over the 4 years of the study. From the perspective of impacts on soil chemical properties within the depth of 0-1.8 m, there were few detrimental agronomic impacts of concern from dairy lagoon water reuse based on the results of this study. However, there are potential long-term concerns. Long-term sustainability as a viable alternative agricultural water resource depends on regular monitoring of soil properties of potential concern as determined from the chemical composition of the source dairy lagoon water and from trends revealed over the 4 years of study regarding potential agronomic and environmental impacts. As a precaution, it is advisable to monitor certain soil chemical properties beyond the length of this study. In this study the properties of salinity (EC<sub>e</sub>), SAR, B, and Cu were identified as properties that would require regular future monitoring.

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associated with monitoring are costly. A sampling design is essential that minimizes the number of monitoring (i.e., sampling) locations to monitor cost effectively at field scale and that provides a characterization of the spatial variability of impacted soil properties from which management decisions can be made to optimize yield with minimal detrimental impacts on soil and water. Corwin *et al.*  $(2008)^1$  and Corwin  $(2012)^{18}$  demonstrated that EC<sub>a</sub>-directed soil sampling is a viable means of monitoring drainage water reuse impacts on salinity, SAR, and trace elements to manage its long-term sustainability. This study further demonstrates the utility of ECa-directed soil sampling through its expanded application of monitoring impacts of dairy lagoon water reuse on heavy metal accumulation in soil. Even though  $EC_a$  does not directly measure heavy metals, it measures properties that influence the distribution and fate of heavy metals in soil, e.g., texture, organic matter, cation exchange capacity, and water content; consequently, the spatial and temporal measurements of these directly measured properties with EC<sub>a</sub> provide a means of spatially sampling properties (e.g., heavy metals) that are influenced by these directly measured properties, but not directly measured by EC<sub>a</sub>. 

Figures 2-6 provide maps of the spatial distribution of properties impacted by dairy lagoon water reuse. These maps provide the producer with detailed spatial and temporal information for the site-specific management of dairy lagoon water reuse with the goal of optimizing yield and mitigating detrimental environmental impacts. The properties of salinity, SAR, B, and Cu are of potential long-term concern to the sustainability of dairy lagoon water reuse at the San Jacinto site. From an agronomic perspective salinity, SAR, and B are of greatest concern, whereas from an environmental perspective salinity and Cu present the greatest potential effect upon groundwater. To optimize crop yield and minimize detrimental environmental impacts it is necessary (1) to sufficiently leach salinity and B from the root zone (i.e., top 1.5 m of soil) to

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levels below the salinity and B tolerance thresholds, (2) to add a calcium amendment to maintain
 SAR below 4, and (3) to minimize the leaching of salinity and copper into the groundwater.

Soil cores taken to a depth of 10 m at the study site before the commencement of the study revealed no evidence of a water table. Estimates from nearby wells indicate that the groundwater is greater than 60-70 m from the soil surface. Sufficient leaching of salinity and B without pushing too much salinity and Cu beyond the root zone to detrimentally impact groundwater for a triticale-sorghum-barley crop rotation can be achieved with a LF=0.23 as estimated following the approach outlined by Corwin et al. (2007)<sup>40</sup> for determining leaching requirement. The LF along with the maps in Figs. 2-6 provides the information to manage site specifically dairy lagoon water reuse at San Jacinto to achieve long-term sustainability. If a LF of 0.23 or less is maintained, then deleterious levels of salinity, Na, B, and Cu will not likely penetrate below 60-70 m unless preferential flow paths are present to provide a direct conduit to the groundwater. However, if a LF is maintained well above 0.23 for a sustained period of time (e.g., 10-20 years), then the leaching of salinity, Na, B, and Cu may become a threat to the groundwater and should be closely monitored by regularly obtaining soil samples or in situ extracts of soil solution below the root zone. 

The evaluation of the impact and sustainability of dairy lagoon water reuse has shown that dairy lagoon water reuse can be suitably managed (i) to transform it from an environmental burden into a water resource that produces crops and (ii) to reduce dramatically the volume of degraded water that is costly and difficult to store or dispose.

The extensive spatio-temporal dataset of this study and demonstrated methodology for monitoring management-induced changes using EC<sub>a</sub>-directed soil sampling are additional significant assets of this study. There are no other known spatial datasets recording the impact of

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dairy lagoon water reuse over a time period of this extent. Furthermore, the methodology for mapping and monitoring degraded water reuse impacts provides a means of assessment that assists the producer in site-specific management to identify where, when, and the amounts of irrigation or soil amendments that are needed to maintain the sustainability of degraded water reuse as well as providing the information that is needed for crop selection.

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## 1 References

- D.L. Corwin, S.M. Lesch, J.D. Oster and S.R. Kaffka, Short-term sustainability of drainage
  water reuise: Spatio-temporal impacts on soil chemcial properties, *J. Environ. Qual.*, 2008,
  37, S-8-S-24.
- S.A. Bradford, E. Segal, W. Zhang, Q. Wang and S.R. Hutchins, Reuse of concetrated animal
   feeding operation wastewater on agricultural land, *J. Environ. Qual.*, 2008, 37, S-97-S-115.
- B 3 D.L. Corwin and S.A. Bradford, Environmental impacts and sustainability of degraded water
   reuse, J. Environ. Qual., 2008, 37, S-1-S-7.
- 4 S.S. Goyal, S.K. Sharma, D.W. Rains and A. Läuchli, Long-term reuse of drainage waters of
  varying salinities for crop irrigation in a cotton-safflower rotation system in the San Joaquin
  Valley of California: A nine year study: I. Cotton (*Gossypium hirsutum* L.), *J. Crop Prod.*,
  13 1999, 2(2), 181–213.
- R.O. Evans, P.W. Westerman and M.R. Overcash, Subsurface drainage water quality from
   land application of swine lagoon effluent, *Trans. ASAE*, 1984, 27, 473–480.
- 6 J.C. Burns, P.W. Westerman, L.D. King, G.A. Cummings, M.R. Overcash and L. Goode,
  Swine lagoon effluent applied to 'Coastal' Bermudagrass: I. Forage yield, quality, and
  element removal, *J. Environ. Qual.*, 1985, 14, 9–14.
  - I.D. King, P.W. Westerman, G.A. Cummings, M.R. Overcash and J.C. Burns, Swine lagoon
     effluent applied to 'Coastal' Bermudagrass: II. Effects on soil, *J. Environ. Qual.*, 1985, 14,
     14–21.
  - 8 G.T. Kroyer, Impact of food processing on the environment– An overview, *Lebensm. Wiss. Technol.*, 1995, 28, 547–552.
  - 9 P. Wersin, J. Abrecht and P. Höhener, Large-scale redox plume in glaciofluvial deposits due
    to sugar-factory wastes and wastewater at Aarberg, Switzerland, *Hydrogeol. J.*, 2001, 9, 282–
    26 296.
  - 10 F. Zvomuya, S.C. Gupta and C.J. Rosen, Phosphorus leaching in sandy outwash soils
     following potato-processing wastewater application, *J. Environ. Qual.*, 2005, 34, 1277–1285.
- 11 M.M. Johns and J.W. Bauder, Root zone leachate from high chemical oxygen demand
  cannery water irrigation, *Soil Sci. Soc. Am. J.*, 2007, **71**, 1893–1901.
  - 12 C.J. Schmidt, I. Kugelman and E. Clements, Municipal wastewater reuse in the US, *J. Water Pollut. Control Fed.*, 1975, 47(9), 2229–2245.
  - 13 C. Chakrabarti, Residual effects of long-term land application of domestic wastewater,
     *Environ. Int.*, 1995, 21(3), 333–339.
  - 14 A.K. Kivaisi, The potential for constructed wetlands for wastewater treatment and reuse in
     developing countries: A review, *Ecol. Eng.*, 2001, 16, 545–560.
  - 15 C. Lubello, R. Gori, F.P. Nicese and F. Ferrini, Municipal-treated wastewater reuse for plant
     nurseries irrigation, *Water Res.*, 2004, 38, 2939–2947.
- <sup>48</sup>
  <sup>49</sup>
  <sup>40</sup>
  <sup>40</sup></l
  - 41 17 W. Chen, L. Wu, W.T. Frankenberger, Jr. and A.C. Chang. 2008. Soil enzyme activities of
    42 long-term reclaimed wastewater irrigated soils, *J. Environ. Qual.*, 2008, **37**, S-36-S-42.
- <sup>53</sup>
  <sup>43</sup>
  <sup>43</sup>
  <sup>43</sup>
  <sup>44</sup>
  <sup>55</sup>
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  <sup>43</sup>
  <sup>44</sup>
  <sup>44</sup>
  <sup>44</sup>
  <sup>45</sup>
  <sup>45</sup></l
- 46 19 B.J. Alloway, Soil processes and the behavior of metals, *In* B.J. Alloway (ed.) *Heavy Metals*

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in Soils, 2nd edition, Kluwer Academic Publ., Dordrecht, The Netherlands, 1995, pp. 11-37. 20 A. Kamari, I.D. Pulford and J.S.J. Hargreaves, Binding of heavy metal contaminants onto chitosans--an evaluation for remediation of metal contaminated soil and water, J. Environ. Manage., 2011, 92, 2675-2682. 21 S. Wu, S. Zhou and X. Li, Determining the anthropogenic contribution of heavy metal accumulations around a typical industrial town: Xushe, China, J. Geochemical Exploration, 2011, 110(2), 92–97. 22 C.N. Thomas, W.L. Bauerle, J.P. Chastain, T.O. Owino, K.P. Moore and S.J. Klaine, Effects of scrubber by-product-stabilized dairy lagoon sludge on growth and physiological responses of sunflower (Helianthus annuus L.), Chemosphere, 2006, 64, 152-160. 23 J. Kumpiene, A. Lagerkvist and C. Maurice, Stabilization of As, Cr, Cu, Pb and Zn in soil using amendments - A review. Waste Manage., 2008, 28, 215-225. 24 G.F. Vance, L.A. King and G.K. Ganjegunte, Soil and plant responses from land application of saline-sodic waters: Implications of management, J. Environ. Qual., 2008, 37, S-139-S-148. 25 G.K. Ganjegunte, L.A. King and G.F. Vance, Land application with saline-sodic coalbed natural gas water: Cumulative effects on soil chemcial properties, J. Environ. Qual., 2008, 37, S-128-S-138. 26 D.L. Corwin and S.M. Lesch, Application of soil electrical conductivity to precision agriculture: Theory, principles, and guidelines, Agron. J., 2003, 95(3), 455-471. 27 D.L. Corwin and S.M. Lesch, Characterizing soil spatial variability with apparent soil electrical conductivity: I. Survey protocols, Comp. Electron. Agric., 2005a, 46(1-3), 103-133. 28 D.L. Corwin and S.M. Lesch, Protocols and guidelines for field-scale measurement of soil salinity distribution with EC<sub>a</sub>-directed soil sampling, J. Environ. Eng. Geophysics, 2013, (1), 1-25. 29 D.L. Corwin and S.M. Lesch, Apparent soil electrical conductivity measurements in agriculture, Comput. Electron. Agric., 2005b, 46, 11-43. 30 D.L. Corwin, S.M. Lesch, E. Segal, T.H. Skaggs and S.A. Bradford, Comparison of sampling strategies for characterizing spatial variability with apparent soil electrical conductivity direct soil sampling, J. Environ. Eng. Geophysics, 2010, 15(3), 147-162. 31 S.M. Lesch, J.D. Rhoades and D.L. Corwin, ESAP-95 version 2.10R: User manual and tutorial guide, Technical Research Rpt. # 146, USDA-ARS, U.S. Salinity Laboratory, Riverside, CA, 2000. 32 J.D. Rhoades, Salinity: Electrical conductivity and total dissolved solids, In D.L. Sparks (ed.) Methods of Soil Analysis: Part 3 – Chemical Methods, Book Series No. 5, SSSA, Madison, WI, 1996, pp. 417-435. 33 Methods of Soil Analysis: Part 3 – Chemical Analysis, ed. D. L. Sparks, SSSA Book Series No. 5, SSSA, Madison, WI, 1996. 34 Methods of Soil Analysis: Part 4 – Physical Methods, eds. J. H. Dane and G. C. Topp, SSSA Book Series No. 5, SSSA, Madison, WI, 2002. 35 R. Karow, E. Marx, K. Morrow, M. Bohle, R. Dovel, E. Eldredge, P. Hayes, S. James, J. Peterson, G. Reed, C. Shock and D. Smiley, Spring wheat varieties for 1999, Special Report 986, Oregon State University, Corvallis, OR, 1999. 36 M. Rahman, S. Fukai and F.P.C. Blamey, Forage production and nitrogen uptake of forage sorghum, grain sorghum and maize as affected by cutting under different nitrogen levels, In

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- Spatio-temporal impacts of dairy lagoon water reuse on soil: Heavy metals and salinity Proceedings of the 10<sup>th</sup> Australian Agronomy Conference, Hobart, Australia (available at http://www.regional.org.au/au/asa/2001/1/c/rahman.htm), 2001. 37 National Academy of Sciences-National Academy of Engineering (NAS-NAE) Committee on Water Quality, Water Quality Criteria 1972, Ecological Research Series, EPA, Washington D.C., 1972. 38 L. Bernstein, Quantitative assesment of irrigation water quality, Special Technical Publication 416, American Society for Testing and Materials, Philadelphia, PA, 1967, pp. 51-65.
  - 39 D.L. Corwin, S.M. Lesch, P.J. Shouse, R. Soppe, and J.E. Ayars. Identifying soil properties that influence cotton yield using soil sampling directed by apparent soil electrical conductivity, Agron. J., 2003, 95, 352-364.
  - 40 D.L. Corwin, J.D. Rhoades, and J. Simunek. Leaching requirement for soil salinity control: Steady-state vs. transient-state models, Agric. Water Manage., 2007, 90(2), 165-180.

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**Environmental Science: Processes** 

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Figure titles:

- Figure 1. Maps showing the apparent soil electrical conductivity  $(EC_a)$  survey taken with electromagnetic induction in the vertical  $(EM_v)$  coil configuration for a 32-ha study site on Scott Brothers' Dairy Farm in San Jacinto, CA. The locations of 28  $EC_a$ -directed soil sampling sites, which were selected using a stratified random sampling design, are identified by solid circles.
- Figure 2. Field-scale distribution of salinity (i.e., EC<sub>e</sub>, electrical conductivity of the saturation extract) at 0-1.8 m for the years (A.) 2007 and (B.) 2011, and (C.) change in salinity from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.
- Figure 3. Field-scale distribution of SAR (sodium adsorption ratio) at 0-1.8 m for the years
  (A.) 2007 and (B.) 2011, and (C.) change in SAR from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.
- Figure 4. Field-scale distribution of  $pH_e$  (saturation extract) at 0-1.8 m for the years (A.) 2007 and (B.) 2011, and (C.) change in  $pH_e$  from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.
- Figure 5. Field-scale distribution of B (saturation extract) at 0-1.8 m for the years (A.) 2007 and (B.) 2011, and (C.) change in B from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.

Figure 6. Distribution of metals (double acid extract) at 0-1.8 m for the years 2007, 2011,

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and change from 2007 to 2011: (A-C.) Cd, (D-F.) Cu, (G-I.) Mn, (J-L.) Ni, and

(M-O.) Zn.

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Table 1. Chemical composition of treated dairy lagoon wastewater (DLWW), recycled water, and well water.

Chemical Property	Treated DLWW	Recycled Water	Well Water
	(Summer 2009)	(Summer 2009)	(Winter 2007)
EC (dS $m^{-1}$ )	2.6	1.1	0.5
TDS (mg $L^{-1}$ )	NA	600	320
pH	8.6	7.38	8.7
Na (meq $L^{-1}$ )	12.8	8.7	2.7
K (meq $L^{-1}$ )	7.4	0.5	0.06
Ca (meq $L^{-1}$ )	6.7	5.8	2.5
Mg (meq $L^{-1}$ )	5.7	1.8	0.2
$Cl (meq L^{-1})$	8.6	5.3	0.5
$S-SO_4 (meq L^{-1})$	3.4	2.1	1.4
$HCO_3 (meq L^{-1})$	8.1	1.7	1.0
$NH_4-N \& NO_2 + NO_3-N (mg L^{-1})$	37.9	17.9	1.15
Organic N (mg $L^{-1}$ )	7.05	6.9	na
Total P (mg $L^{-1}$ )	29.1	4.3	0.06
B ( $\mu$ g L <sup>-1</sup> )	64.4	17.1	40
Fe ( $\mu g L^{-1}$ )	43.8	12.8	82
$Cu (ug L^{-1})$	13.5	1.7	na

EC=electrical conductivity, TDS=total dissolved solids, na=data not available.

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Table 2. Chemical composition of the blended irrigation water (treated DLWW +well water or treated DLWW+ recycled water) over the 4-year study. DLWW=dairy lagoon wastewater.

Chamical Analysis						Da	te (month/	'day/year)						
	7/19/07	3/23/08	7/28/09	8-4-09	8/12/09	8/19/09	8/28/09	9/1/09	9/8/09	11/15/09	1/16/10	3/2/11	5/24/11	6/28/11
$EC_e (dS m^{-1})$	1.33	1.01	2.14	2.04	1.18	1.32	1.08	1.09	1.09	1.08	3.00	1.61	0.96	1.51
$pH_e$	8.6	8.6	8.33	8.08	7.81	7.75	6.26	7.75	7.60	7.81	7.95	8.64	7.90	7.54
SAR	2.33	2.06	4.74	4.69	4.08	4.03	4.41	4.41	4.24	3.98	na	1.35	4.00	3.79
$Cl^{-}$ (meq $L^{-1}$ )	4.40	3.77	7.95	7.31	5.70	6.14	5.28	4.94	5.18	4.58	9.03	6.04	na	na
$HCO_3^{-1}$ (meq L <sup>-1</sup> )	3.03	2.78	9.15	10.08	2.29	2.46	3.03	3.81	3.69	3.41	3.75	7.43	na	na
$PO_4^{3-}(meq L^{-1})$	0.10	0.10	0.68	0.65	0.11	0.09	0.15	0.03	0.39	0.07	na	0.04	0.01	0.72
$NO_3$ (meq L <sup>-1</sup> )	0.67	0.61	0.98	1.01	0.59	0.65	0.89	0.42	0.34	0.53	1.31	na	na	na
$SO_4^{2-}(meq L^{-1})$	2.00	1.82	2.30	2.13	2.40	2.98	1.90	1.80	1.90	1.73	na	0.19	2.09	1.66
$N-NH_4^+ (meq L^{-1})$	0.39	0.38	1.05	1.52	0.19	0.09	0.85	1.54	1.29	0.71	1.44	na	na	2.53
$Na^+$ (meq L <sup>-1</sup> )	5.03	5.02	7.96	7.54	5.82	6.37	5.94	5.79	5.67	5.05	na	0.65	5.12	5.58
$K^+$ (meq L <sup>-1</sup> )	0.25	0.22	5.09	4.39	0.43	0.48	0.76	0.42	0.47	0.44	na	0.38	0.41	2.81
$Ca^{2+}$ (meq L <sup>-1</sup> )	2.24	2.06	3.32	3.09	2.84	3.51	2.92	2.85	2.84	2.66	na	0.34	2.66	2.88
$Mg^{2+}(meq L^{-1})$	0.57	0.38	2.30	2.09	1.25	1.48	0.71	0.60	0.73	0.57	na	0.13	0.62	1.45
As $(\text{mg } \text{L}^{-1})^{\dagger}$	0.033	0.029	0.023	0.004	0.012	0.006	0.000	0.000	0.000	0.007	na	0.000	0.005	0.000
B (mg $L^{-1}$ ) <sup>†</sup>	0.711	0.697	0.554	0.456	0.261	0.295	0.839	0.881	0.714	0.423	na	0.000	0.441	0.367
Mo (mg $L^{-1}$ ) <sup>†</sup>	0.005	0.003	0.000	0.000	0.000	0.000	0.005	0.007	0.006	0.009	na	0.007	0.007	0.002
Se (mg $L^{-1}$ ) <sup>†</sup>	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.002	na	0.000	0.000	0.000
Cd (mg $L^{-1})^{\dagger}$	0.012	0.010	0.000	0.000	Trace	Trace	0.000	0.000	0.002	0.003	na	0.001	0.000	0.001
Cu (mg $L^{-1})^{\dagger}$	0.220	0.140	0.147	0.129	0.022	0.024	0.075	0.038	0.072	0.005	na	0.002	0.003	0.124
Mn (mg $L^{-1}$ )	na	na	na	na	na	na	na	na	na	na	na	0.000	0.052	0.002
Ni (mg $L^{-1}$ ) <sup>†</sup>	na	na	na	na	na	na	na	na	na	na	na	0.002	0.000	0.002
$Zn (mg L^{-1})^{\dagger}$	0.000	0.000	0.000	0.000	0.000	0.053	0.099	0.029	0.032	0.114	na	0.001	0.014	0.065

<sup>†</sup>The recommended limits for As, B, Mo, Se, Cu, Cd, Mn, Ni, and Zn in mg  $L^{-1}$  are 0.1, 0.75, 0.01, 0.02, 0.20, 0.01, 0.20, 0.20, and 2, respectively.<sup>37</sup> na=data not available.

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Table 3. Correlation coefficients between apparent soil electrical conductivity (EC <sub>a</sub> ; dS m <sup>-1</sup> ) measured	b
with electromagnetic induction in the vertical coil configuration (EM <sub>v</sub> ) and soil properties from 0 to 1	8
m for 2007 and 2011 and between sorghum yield and soil properties from 0 to 1.8 m for 2007. (N=28	5).

_	Correlation	Coefficients	Correlation Coefficients			
Property	for E	$M_v EC_a$	for Yield of Sorghum'			
	2007	2011	2007			
$EM_v EC_a (dS m^{-1})$	- 	- 	-0.470 <i>°</i>			
Gravimetric water content ( $\theta_g$ )	$0.717^{\circ}$	$0.816^{\circ}$	-0.091			
Volumetric water content ( $\theta_v$ )	$0.770^{\circ}$	$0.782^{\circ}$	-0.088			
Bulk density ( $\rho_b$ )	-0.505	-0.595 <sup>b</sup>	-0.191			
Saturation percentage (SP)	$0.784^{\nu}$	$0.655^{v}$	-0.064			
Electrical conductivity of the saturation extract (EC)	0.906 <sup><i>b</i></sup>	0.713 <sup><i>b</i></sup>	-0.435 <sup>a</sup>			
pH of the saturation extract $(pH_{a})$	$0.461^{b}$	$0.477^{b}$	-0.176			
Sodium adsorption ratio (SAR)	$0.883^{b}$	$0.778^{b}$	-0.429 <sup><i>a</i></sup>			
Organic carbon (OC)	$0.372^{a}$	na	0.222			
Inorganic carbon (IC)	$0.831^{b}$	na	-0.121			
Total carbon (TC)	$0.611^{b}$	na	0.107			
Total nitrogen (TN)	0.106	na	0.185			
Exchangeable $NH_4^+$ (Ex- $NH_4^+$ )	0.084	na	-0.141			
Exchangeable $Na^+$ (Ex-Na <sup>+</sup> )	$0.859^{b}$	na	-0.371 <sup>a</sup>			
Exchangeable $K^+$ (Ex- $K^+$ )	-0.024	na	0.201			
Exchangeable $Ca^{2+}$ (Ex- $Ca^{2+}$	0.226	na	0.249			
Exchangeable $Mg^{2+}$ (Ex-Mg <sup>2+</sup> )	$0.469^{b}$	na	0.136			
laturation extract:	0.109	nu	0.120			
<u>cl</u>	$0.717^{b}$	$0.790^{b}$	$-0.431^{a}$			
HCO <sub>2</sub> <sup>-</sup>	$0.624^{b}$	$0.453^{b}$	-0.366			
$PO_4^{3-}$	$0.639^{b}$	$0.525^{b}$	$-0.418^{a}$			
$NO_2^-$	$0.416^{a}$	$0.529^{b}$	-0 294			
$SO_{4}^{2}$	$0.896^{b}$	$0.691^{b}$	$-0.385^{a}$			
$\mathbf{NH}_4^+$	$0.417^{a}$	na	-0 111			
Na <sup>+</sup>	$0.885^{b}$	$0.768^{b}$	$-0.453^{a}$			
K <sup>+</sup>	-0.082	0.090	0.036			
$Ca^{2+}$	$0.401^{a}$	0.048	-0.038			
$Mg^{2+}$	$0.570^{b}$	0.187	-0.116			
As	$0.570^{b}$	0.107	$-0.435^{a}$			
B	$0.000^{b}$	$0.33^{b}$	$-0.434^{a}$			
Mo	$0.583^{b}$	0.722	-0.160			
Se	$0.303^{b}$	0.081	-0.188			
Cd	0.197	$0.570^{b}$	na			
Cu	$0.19^{b}$	$0.370^{b}$	-0.276			
Mn	$0.01^{\circ}$	0 394	-0.270 na			
Ni	$0.630^{b}$	0.594	na			
Zn	0.030	0.530	-0.057			
Double acid extract:	0.517	0.511	-0.057			
Cd	0 559 b	-0 482 <sup>b</sup>	-0 102			
Cu	0.337	0.402	-0.102			
Cu Mn	-0.309	-0.370	-0.070			
1V111 N:	-0.309	-0.062	-0.122			
INI Zn	$0.307^{a}$	-0.525	-0.189			

<sup>*a*</sup> Significance test for  $|\mathbf{r}|$  at p $\leq$ 0.05 level. <sup>*b*</sup> Significance test for  $|\mathbf{r}|$  at p $\leq$ 0.01 level. na=data not available.

<sup>†</sup> Sorghum yield is based on total wet weight (Mg ha<sup>-1</sup>).

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Table 4. Means and standard deviations of soil properties in 2007 and 2011 for the composite depth increment of 0-180 cm. N=28.

Chemical Analysis	20	07	2	Paired	
	Mean	Std. Dev.	Mean	Std. Dev.	t-test
$\theta_{\rm g} ({\rm g g}^{-1})$	0.15	0.06	0.18	0.08	а
$\theta_{\rm v}$ (cm <sup>3</sup> cm <sup>-3</sup> )	0.19	0.06	0.13	0.05	а
$\rho_b (g \text{ cm}^{-3})$	1.42	0.16	1.44	0.07	
SP	36.70	6.01	39.74	6.12	
$EC_e (dS m^{-1})$	1.76	0.67	2.50	0.79	а
$pH_e$	7.85	0.27	7.3	0.28	а
$Cl^{-}$ (meq $L^{-1}$ )	2.76	1.81	11.15	6.41	а
$\text{HCO}^{3-}$ (meq $\text{L}^{-1}$ )	4.41	1.86	2.89	1.09	а
$PO_4^{3-}$ (meq L <sup>-1</sup> )	0.20	0.24	0.15	0.11	
$NO_3^-$ (meq L <sup>-1</sup> )	2.31	1.81	1.28	1.01	а
$SO_4^{2-}$ (meq L <sup>-1</sup> )	10.71	10.73	11.83	9.93	
$NH_4^+$ (meq L <sup>-1</sup> )	0.15	0.08	na	na	
$Na^+$ (meq L <sup>-1</sup> )	12.62	12.79	19.01	13.18	а
$K^+$ (meq L <sup>-1</sup> )	0.56	0.41	0.37	0.19	
$Ca^{2+}$ (meq L <sup>-1</sup> )	4.01	1.62	4.89	3.38	
$Mg^{2+}$ (meq L <sup>-1</sup> )	1.32	0.73	1.66	1.32	
SAR	11.05	11.76	14.23	11.48	а
As (Sat. Ext.)( $\mu g L^{-1}$ )	34.33	65.09	29.27	44.56	а
B (Sat. Ext.)( $\mu g L^{-1}$ )	340.8	591.2	790.6	655.5	а
Mo (Sat. Ext.)( $\mu$ g L <sup>-1</sup> )	331.9	393.7	160.4	89.40	а
Se (Sat. Ext.)( $\mu g L^{-1}$ )	27.92	14.41	3.49	10.88	а
Cd (Sat. Ext.)( $\mu g L^{-1}$ )	0.13	0.10	0.92	1.58	а
Cu (Sat. Ext.)( $\mu g L^{-1}$ )	69.15	79.84	10.18	9.76	а
Mn (Sat. Ext.)( $\mu g L^{-1}$ )	na	na	6.94	14.12	
Ni (Sat. Ext.)( $\mu g L^{-1}$ )	na	na	13.96	12.78	
Zn (Sat. Ext.)( $\mu$ g L <sup>-1</sup> )	37.03	26.21	54.62	48.91	а
OC (%)	0.44	0.09	na	na	
IC (%)	0.20	0.06	na	na	
TC (%)	0.64	0.14	na	na	
TN (%)	0.03	0.01	na	na	
$Ex-NH_4^+$ (meg 100g <sup>-1</sup> )	0.02	0.01	na	na	
$Ex-Na^{+}$ (meg 100g <sup>-1</sup> )	1.89	2.45	na	na	
$Ex-K^{+}$ (meg 100g <sup>-1</sup> )	0.39	0.16	na	na	
$Ex-Ca^{2+}$ (meg 100g <sup>-1</sup> )	16.83	2.91	na	na	
$Ex-Mg^{2+}$ (meg 100g <sup>-1</sup> )	2.01	0.67	na	na	
Cd (Double Acid)( $\mu g kg^{-1}$ )	1.58	1.66	9.19	9.30	а
Cu (Double Acid)( $\mu$ g kg <sup>-1</sup> )	29.13	11.06	4.46	5.22	а
Mn (Double Acid)(mg kg <sup>-1</sup> )	3.36	0.89	4.85	1.97	а
Ni (Double Acid)(ug kg <sup>-1</sup> )	14.53	6.42	149.4	153.4	а
Zn (Double Acid)(mg kg <sup>-1</sup> )	0.23	0.20	0.20	0.16	

a=significantly different at the p $\leq$ 0.05 level.  $\theta_g$ =gravimetric water content,

 $\theta_v$ =volumetric water content,  $\rho_b$ =bulk density, SP=saturation percentage,

 $EC_e$ =electrical conductivity of the saturation extract, SAR=sodium adsorption

ratio, Sat. Ext.=saturation extract, OC(%)=percentage of organic carbon,

IC(%)=percentage of inorganic carbon, TC(%)=percentage of total carbon,

TN(%)=percentage of total carbon, Ex-NH<sub>4</sub><sup>+</sup>=exchangeable NH<sub>4</sub><sup>+</sup>, Ex-

Na<sup>+</sup>=exchangeable Na<sup>+</sup>, Ex-K<sup>+</sup>=exchangeable K<sup>+</sup>, Ex-Ca<sup>2+</sup>=exchangeable Ca<sup>2+</sup>, Ex-Mg<sup>2+</sup>=exchangeable Mg<sup>2+</sup>, Double Acid=double acid extract. na=data not available.

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Table 5. Mean and standard deviations of soil properties in 2007 for depth increments of 0-15, 15-30, 30-60, 60-90, 90-120, 120-150, and 150-180 cm. N=28.

	N								Standard Deviation								
Chemical Analysis				Me	an					Sta	indard I	Deviatio	n				
	0-15	15-30	30-60	60-90	90-120	120-150	150-180	0-15	15-30	30-60	60-90	90-120	120-150	150-180			
$\theta_{g} (g g^{-1})$	0.06	0.07	0.08	0.16	0.18	0.21	0.21	0.02	0.02	0.04	0.13	0.08	0.13	0.12			
$\theta_{\rm v} ({\rm cm}^3{\rm cm}^{-3})$	0.08	0.11	0.12	0.19	0.23	0.26	0.26	0.03	0.04	0.05	0.11	0.11	0.13	0.12			
$\rho_b (g \text{ cm}^{-3})$	1.34	1.66	1.51	1.37	1.38	1.35	1.40	0.31	0.20	0.11	0.27	0.31	0.25	0.30			
SP	42.65	38.35	36.75	36.40	37.77	34.92	33.87	3.39	5.10	7.02	9.22	8.28	9.37	9.67			
$EC_e (dS m^{-1})$	3.54	1.66	1.53	1.42	1.59	1.65	1.79	2.01	1.18	1.13	1.26	1.29	1.55	1.70			
$pH_e$	7.57	7.70	7.74	7.83	7.82	7.97	8.08	0.40	0.40	0.34	0.39	0.36	0.40	0.39			
$Cl^{-}$ (meq $L^{-1}$ )	11.38	2.79	1.67	1.53	2.08	2.08	1.66	7.06	1.86	1.75	1.65	2.35	2.32	1.44			
$HCO^{3-}$ (meq $L^{-1}$ )	9.87	5.23	3.81	3.29	3.14	3.92	4.48	3.17	1.49	1.28	1.36	2.06	3.03	4.18			
$PO_4^{3-}$ (meq L <sup>-1</sup> )	0.31	0.20	0.13	0.13	0.14	0.18	0.33	0.16	0.13	0.11	0.14	0.28	0.34	0.68			
$NO_3^-$ (meq L <sup>-1</sup> )	3.73	1.63	1.50	1.53	2.83	2.53	2.79	3.96	1.16	1.66	2.02	3.42	2.36	2.63			
$SO_4^{2-}$ (meq L <sup>-1</sup> )	17.74	10.06	10.82	9.42	9.31	9.77	11.02	19.77	12.80	12.07	10.51	9.65	12.83	13.17			
$NH_4^+$ (meq L <sup>-1</sup> )	0.41	0.19	0.12	0.12	0.12	0.13	0.12	0.31	0.12	0.07	0.07	0.08	0.09	0.09			
$Na^+$ (meq L <sup>-1</sup> )	17.04	8.49	9.89	11.10	11.83	13.77	16.36	18.70	9.35	10.43	12.80	13.38	16.81	18.79			
$K^{+}$ (meq L <sup>-1</sup> )	3.47	0.68	0.61	0.22	0.18	0.14	0.11	2.64	0.70	1.13	0.24	0.22	0.15	0.15			
$Ca^{2+}$ (meq L <sup>-1</sup> )	14.35	7.13	4.87	2.71	2.80	1.83	1.11	6.04	5.23	4.78	1.86	2.22	1.17	0.62			
$Mg^{2+}$ (meq L <sup>-1</sup> )	4.93	1.93	1.45	0.89	1.02	0.72	0.43	3.94	2.45	1.37	0.64	0.83	0.58	0.22			
SAR	5.43	4.32	6.15	10.44	9.94	13.47	21.43	5.62	5.13	7.53	15.34	13.11	16.02	23.58			
As (Sat. Ext.)(µg L <sup>-1</sup> )	27.97	24.88	24.62	31.67	38.64	50.28	114.8	16.49	26.09	20.61	36.24	70.97	81.57	210.0			
B (Sat. Ext.)(µg L <sup>-1</sup> )	316.9	204.9	226.4	330.9	400.3	485.5	831.9	278.0	224.5	253.9	470.4	677.2	729.9	1380.			
Mo (Sat. Ext.)(µg L <sup>-1</sup> )	216.5	207.6	278.4	317.3	397.5	454.2	477.8	230.1	233.1	247.3	291.3	650.1	541.5	658.9			
Se (Sat. Ext.)(µg L <sup>-1</sup> )	26.96	26.48	30.35	26.67	27.14	28.70	31.61	9.94	7.04	10.24	10.99	15.70	19.25	30.85			
Cd (Sat. Ext.)(µg L <sup>-1</sup> )	0.05	0.09	0.10	0.11	0.14	0.17	0.17	0.10	0.13	0.12	0.13	0.15	0.17	0.18			
Cu (Sat. Ext.)(µg L <sup>-1</sup> )	47.65	62.39	46.71	53.31	66.06	78.71	115.1	22.09	66.77	27.50	56.58	121.0	131.8	175.4			
Mn (Sat. Ext.)(µg L <sup>-1</sup> )	na	na	na	na	na	na	na	na	na	na	na	na	na	na			
Ni (Sat. Ext.)(µg L <sup>-1</sup> )	na	na	na	na	na	na	na	na	na	na	na	na	na	na			
Zn (Sat. Ext.)(µg L <sup>-1</sup> )	34.91	32.21	44.25	28.64	25.46	45.69	44.57	37.87	38.99	83.01	36.14	31.46	47.77	56.81			
OC (%)	4.56	0.81	0.46	0.28	0.31	0.24	0.17	0.35	0.27	0.13	0.16	0.20	0.17	0.12			
IC (%)	0.14	0.14	0.17	0.21	0.23	0.23	0.23	0.05	0.07	0.07	0.09	0.11	0.10	0.12			
TC (%)	4.70	0.95	0.63	0.48	0.54	0.47	0.40	0.34	0.27	0.18	0.24	0.28	0.26	0.22			
TN (%)	0.17	0.09	0.02	0.01	0.01	0.01	0.00	0.04	0.04	0.03	0.02	0.02	0.02	0.01			
Ex-NH <sub>4</sub> <sup>+</sup> (meq 100g <sup>-1</sup> )	0.07	0.03	0.02	0.01	0.01	0.01	0.01	0.03	0.02	0.02	0.01	0.01	0.01	0.01			
Ex-Na <sup>+</sup> (meq 100g <sup>-1</sup> )	0.71	0.53	0.82	1.43	1.75	2.58	4.14	1.07	0.50	0.92	2.45	2.80	4.46	5.42			
$Ex-K^{+}$ (meq 100g <sup>-1</sup> )	1.23	0.48	0.41	0.30	0.29	0.26	0.22	0.64	0.23	0.33	0.20	0.17	0.15	0.10			
$Ex-Ca^{2+}$ (meq 100g <sup>-1</sup> )	15.00	16.17	19.00	16.72	18.05	16.59	15.04	2.61	3.39	9.62	5.25	5.86	4.67	3.51			
$Ex-Mg^{2+}$ (meg 100g <sup>-1</sup> )	1.88	1.56	1.99	1.88	2.39	2.18	1.93	0.52	0.50	1.23	0.96	1.36	1.24	1.07			
Cd (Double Acid)( $\mu$ g kg <sup>-1</sup> )	2.34	2.07	1.72	1.36	1.16	1.71	1.33	2.86	2.36	2.11	1.51	1.42	2.04	1.73			
Cu (Double Acid)(µg kg <sup>-1</sup> )	36.32	34.35	31.02	24.38	25.99	28.66	29.39	9.01	13.71	17.36	15.13	17.42	18.09	24.19			
Mn (Double Acid)(mg kg <sup>-1</sup> )	6.39	4.73	3.14	2.92	2.66	3.02	2.83	2.07	2.37	2.01	1.69	1.83	1.65	1.54			
Ni (Double Acid)(µg kg <sup>-1</sup> )	17.44	23.60	17.35	13.73	9.80	13.11	12.66	14.81	22.22	12.95	7.89	9.89	10.60	8.20			
Zn (Double Acid)(mg kg <sup>-1</sup> )	0.70	0.46	0.18	0.09	0.29	0.16	0.06	0.47	0.47	0.27	0.23	0.78	0.40	0.10			

 $\theta_g$ =gravimetric water content,  $\theta_v$ =volumetric water content,  $\rho_b$ =bulk density, SP=saturation percentage, EC<sub>e</sub>=electrical conductivity of the saturation extract, SAR=sodium adsorption ratio, Sat. Ext.=saturation extract, OC(%)=percentage of organic carbon, IC(%)=percentage of inorganic carbon, TC(%)=percentage of total carbon, TN(%)=percentage of total carbon, Ex-NH<sub>4</sub><sup>+</sup>=exchangeable NH<sub>4</sub><sup>+</sup>, Ex-Na<sup>+</sup>=exchangeable Na<sup>+</sup>, Ex-K<sup>+</sup>=exchangeable K<sup>+</sup>, Ex-Ca<sup>2+</sup>=exchangeable Ca<sup>2+</sup>, Ex-Mg<sup>2+</sup>=exchangeable Mg<sup>2+</sup>, Double Acid=double acid extract. na=data not available.

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Table 6. Mean and standard deviations of soil properties in 2011 for depth increments of 0-15, 15-30, 30-60, 60-90, 90-120, 120-150, and 150-180 cm. N=28.

Chamical Analysis	Mean								Standard Deviation							
Chemical Analysis	0-15	15-30	30-60	60-90	90-120	120-150	150-180	0-15	15-30	30-60	60-90	90-120	120-150	150-180		
$\theta_{g} (g g^{-1})$	0.19	0.18	0.13	0.16	0.21	0.20	0.20	0.04	0.02	0.06	0.09	0.14	0.11	0.11		
$\theta_v (cm^3 cm^{-3})$	0.09	0.09	0.10	0.12	0.15	0.14	0.16	0.02	0.04	0.05	0.06	0.07	0.07	0.07		
$\rho_b (g \text{ cm}^{-3})$	0.48	0.64	0.77	0.77	0.78	0.75	0.84	0.09	0.14	0.07	0.14	0.16	0.14	0.20		
SP	45.94	43.56	40.56	39.52	38.92	38.74	35.93	5.06	8.07	7.97	8.46	10.38	9.44	9.07		
$EC_e (dS m^{-1})$	1.65	1.77	2.17	2.44	2.82	3.02	2.86	1.39	1.78	1.70	1.73	1.83	1.86	1.38		
$pH_e$	7.34	7.23	7.13	7.17	7.27	7.35	7.57	0.17	0.25	0.29	0.32	0.51	0.48	0.53		
$Cl^{-}$ (meq $L^{-1}$ )	6.67	6.55	9.11	11.62	14.30	13.27	12.06	5.84	6.88	7.77	9.57	11.94	8.70	7.14		
$HCO^{3-}$ (meq $L^{-1}$ )	4.43	3.20	2.87	2.31	2.43	2.53	3.39	0.61	1.06	0.87	0.80	1.64	1.85	2.78		
$PO_4^{3-}$ (meq L <sup>-1</sup> )	0.27	0.20	0.13	0.10	0.13	0.13	0.20	0.13	0.13	0.09	0.07	0.11	0.19	0.31		
$NO_3^-$ (meq L <sup>-1</sup> )	1.80	2.01	1.17	0.77	1.02	1.12	1.72	0.83	0.80	0.59	0.72	1.44	1.72	2.10		
$SO_4^{2-}$ (meq L <sup>-1</sup> )	6.28	7.96	10.98	11.43	13.09	14.64	13.72	11.26	14.52	14.19	11.28	10.62	14.87	9.16		
$NH_{4}^{+}$ (meq L <sup>-1</sup> )	na	na	na	na	na	na	na	na	na	na	na	na	na	na		
$Na^+$ (meq L <sup>-1</sup> )	8.37	12.52	16.50	18.62	21.72	22.92	23.89	9.68	13.80	13.53	14.84	16.24	21.56	15.50		
$\mathbf{K}^{+}$ (meq $\mathbf{L}^{-1}$ )	1.68	0.68	0.28	0.20	0.23	0.19	0.16	0.79	0.54	0.23	0.23	0.23	0.14	0.08		
$Ca^{2+}$ (meq L <sup>-1</sup> )	5.72	4.40	4.81	4.43	5.61	5.22	4.20	4.30	4.51	4.80	5.33	6.50	5.25	5.09		
$Mg^{2+}$ (meq L <sup>-1</sup> )	1.89	1.45	1.46	1.57	2.03	1.92	1.35	2.30	3.07	2.16	1.80	2.31	1.98	1.54		
SAR	4.10	7.28	10.02	13.26	15.99	17.42	22.98	2.45	4.05	5.55	13.10	17.21	22.43	20.79		
As (Sat. Ext.)(µg L <sup>-1</sup> )	7.51	13.95	15.62	31.99	21.13	30.65	65.53	13.58	12.87	25.25	94.77	35.74	72.73	130.4		
B (Sat. Ext.)(µg L <sup>-1</sup> )	678.9	663.8	674.0	668.1	881.0	839.7	1009.0	429.2	378.6	457.6	674.5	896.9	1029.	1128.		
Mo (Sat. Ext.)(µg L <sup>-1</sup> )	56.19	98.36	147.8	167.2	184.1	181.7	204.0	27.14	72.23	85.60	103.1	131.1	124.7	163.3		
Se (Sat. Ext.)(µg L <sup>-1</sup> )	14.53	6.56	7.65	0.46	1.38	0.66	0.23	34.36	24.49	28.09	2.46	7.31	2.85	0.87		
Cd (Sat. Ext.)(µg L <sup>-1</sup> )	0.29	0.63	0.73	0.95	1.68	0.63	1.05	0.25	0.32	0.48	2.22	5.41	0.80	2.77		
Cu (Sat. Ext.)(µg L <sup>-1</sup> )	113.3	102.5	124.8	71.55	79.63	86.42	140.6	29.29	34.45	158.1	55.27	105.5	167.7	201.1		
Mn (Sat. Ext.)( $\mu g L^{-1}$ )	7.99	9.42	3.10	4.27	4.88	7.03	13.68	7.29	24.81	5.61	11.57	17.45	30.95	31.10		
Ni (Sat. Ext.)(µg L <sup>-1</sup> )	13.57	19.93	18.44	13.15	11.04	11.02	13.34	5.16	7.61	11.09	13.74	19.23	21.11	23.48		
Zn (Sat. Ext.)(µg L <sup>-1</sup> )	45.59	59.34	39.41	50.88	44.29	60.06	80.60	26.29	83.40	47.24	81.76	70.58	76.90	133.3		
Cd (Double Acid)(µg kg <sup>-1</sup> )	7.22	6.62	12.39	8.36	12.42	6.86	8.16	6.10	5.14	28.69	8.45	21.16	4.77	6.91		
Cu (Double Acid)(µg kg <sup>-1</sup> )	15.28	0.90	0.60	3.36	4.78	7.06	2.86	41.10	3.50	3.06	9.28	11.83	16.56	8.56		
Mn (Double Acid)(mg kg <sup>-1</sup> )	8.08	4.80	3.60	4.50	4.52	5.13	4.90	3.72	3.17	2.66	2.88	2.93	3.43	3.16		
Ni (Double Acid)(µg kg <sup>-1</sup> )	159.9	114.4	98.90	169.6	145.9	150.3	194.6	286.2	138.6	105.3	295.4	190.9	257.5	313.2		
Zn (Double Acid)(mg kg <sup>-1</sup> )	0.98	0.48	0.14	0.06	0.07	0.07	0.13	1.05	0.68	0.21	0.06	0.12	0.08	0.22		

 $\theta_g$ =gravimetric water content,  $\theta_v$ =volumetric water content,  $\rho_b$ =bulk density, SP=saturation percentage, EC<sub>e</sub>=electrical conductivity of the saturation extract, SAR=sodium adsorption ratio, Sat. Ext.=saturation extract, Double Acid=double acid extract. na=data not available.

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Figure 1. Maps showing the apparent soil electrical conductivity ( $EC_a$ ) survey taken with electromagnetic induction in the vertical ( $EM_v$ ) coil configuration for a 32-ha study site on Scott Brothers' Dairy Farm in San Jacinto, CA. The locations of 28  $EC_a$ -directed soil sampling sites, which were selected using a stratified random sampling design, are identified by solid circles.

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0-180cm

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Figure 2. Field-scale distribution of salinity (i.e., EC<sub>e</sub>, electrical conductivity of the saturation extract) at 0-1.8 m for the years (A.) 2007 and (B.) 2011, and (C.) change in salinity from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.



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Figure 3. Field-scale distribution of SAR (sodium adsorption ratio) at 0-1.8 m for the years (A.) 2007 and (B.) 2011, and (C.) change in SAR from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.









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C

0-180cm

(b.) 15-30cm

(d.)

60-90cm

(f.) 120-150cm

(h.)

0-180cm

•

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Figure 4. Field-scale distribution of pH<sub>e</sub> (saturation extract) at 0-1.8 m for the years (A.) 2007 and (B.) 2011, and (C.) change in pH<sub>e</sub> from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.











<ul> <li>Sampling Sites</li> </ul>	
(ag.) Soil pHe	
-2.030 – -0.870	
-0.870 – -0.680	
-0.680 – -0.470	
-0.470 - 0.000	
0.000 - +1.460	



0-180cm

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Figure 5. Field-scale distribution of B (saturation extract) at 0-1.8 m for the years (A.) 2007 and (B.) 2011, and (C.) change in B from 2007 to 2011 at depth increments of (a.) 0-15, (b.) 15-30, (c.) 30-60, (d.) 60-90, (e.) 90-120, (f.) 120-150, (g.) 150-180 cm and composite depth of (h.) 0-180 cm.





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Figure 6(M-O; continued). Distribution of metals (double acid extract) at 0-1.8 m for the years 2007, 2011, and change from 2007 to 2011: (A-C.) Cd, (D-F.) Cu, (G-I.) Mn, (J-L.) Ni, and (M-

