Investigating pesticide transport in the León-Chinandega aquifer, Nicaragua

Josh E. Moncrieff · Laurence R. Bentley · Heyddy Calderón Palma

Abstract Nicaragua's León-Chinandega aquifer has seen extensive contamination by persistent organochlorine pesticides applied over decades of intensive agricultural activity. Models of flow and transport of a 330 km^2 sub-region of the aquifer were developed to test conceptual models of contaminant transport, to constrain the value of certain key transport parameters, and to investigate contamination-related concerns raised in past studies. To support these models, a variety of hydrogeologic and geochemical data were collected. It was concluded that the organochlorine pesticides seen in groundwater originate in soils, and are transported to the water table through widespread preferential flow, through shortcutting around wells, or through wind-blown particles delivered to poorly protected hand-dug wells. The distribution coefficient (K_d) of these pesticides is estimated to be between 0.1 and 100 ml/g and the concentration of pesticides being delivered to the water table is estimated to be between 10^2 and 10^5 ng/L. It was found that the distribution and concentration of pesticides in the aquifer would be affected by an increase in groundwater abstraction within the region.

Résumé L'aquifère du León-Chinandega au Nicaragua a été soumis à des contaminations étendues par des pesticides organochlorés persistants qui ont été répandus pendant des dizaines d'années d'agriculture intensive. Des modèles d'écoulement et de transport d'une sous-région de 330 km^2 de l'aquifère ont été construits pour tester des modèles conceptuels du transport de contaminants, pour contraindre la valeur de certains paramètres de transport clefs, et pour étudier les problèmes liés au transport de contaminations qui avaient été rencontrés lors d'études précédentes. Différents types de données hydrogéologiques et géochimiques ont été collectés pour réaliser ces modèles. La conclusion montre que les pesticides organochlorés rencontrés dans les eaux souterraines proviennent des sols et qu'ils sont transportés jusqu'à l'aquifère via de grandes zones d'écoulement préférentiel, via des raccourcis hydrauliques autour des puits, ou encore via des particules emportées par le vent et tombées dans des puits creusés à la main et mal protégés. Le coefficient de distribution (K_d) de ces pesticides est estimé entre 0.1 et 100 ml/g et la concentration de pesticides arrivant dans la nappe est estimée entre 10^2 et 10^5 ng/L. L'étude a montré qu'une augmentation des prélèvements d'eau souterraine dans la région aurait un impact sur la distribution et la concentration des pesticides dans l'aquifère.

Resumen El acuifero León-Chinandega (Nicaragua) presenta una contaminación extensiva por la aplicación de plaguicidas organoclorados en la agricultura intensiva durante décadas. Se han desarrollado modelos de flujo y transporte de una subregión del acuífero de 330 km^2 para comprobar los modelos conceptuales del transporte de contaminantes, estimar los valores de ciertos parámetros clave de transporte e investigar los temas relacionados con la contaminación en estudios anteriores. Para elaborar estos modelos, se recogió una variedad de datos hidrogeológicos y geoquímicos. Se ha concluido que los plaguicidas organoclorados presentes en el agua subterránea se originan en el suelo y son transportados hacia el nivel freático a través de flujos preferentes generalizados, a través de cortes alrededor de los pozos o a través de partículas transportadas por el viento depositadas en pozos excavados a mano con poca protección. El coeficiente de distribución (K_d) de estos plaguicidas se ha estimado entre 0.1 y 100 ml/g y la concentración de plaguicidas que sale hacia el nivel freático se estima que puede estar entre 10^2 y 10^5 ng/L. Se ha concluido que el incremento de la extracción de aguas subterráneas dentro de la región afectaría la distribución y concentración de plaguicidas en el acuífero.

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J. E. Moncrieff (✉) · L. R. Bentley
Department of Geology and Geophysics,
University of Calgary,
2500 University Dr. NW, Calgary, AB, Canada T2N 1N4
e-mail: josh.moncrieff@gmail.com
Tel.: +1-778-8375631
Fax: +1-403-2840074

H. C. Palma
Centro para la Investigación en Recursos Acuáticos de Nicaragua,
La Universidad Nacional Autónoma de Nicaragua,
ENITEL Villa Fontana 500m norte, Managua, Nicaragua

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Introduction

Despite being widely outlawed over 20 years ago, DDT and other organochlorine pesticides (OCPs) continue to be found at detectable concentrations throughout the hydrologic system in many regions of the world. The low rate of degradation of these chemicals, their hydrophobic nature, and their propensity for bioaccumulation indicate that they will continue to persist in the environment and pose potential health problems for years to come. While much progress has been made in understanding the physicochemical properties of these chemicals in various media, it remains exceedingly difficult to make predictions of their fate and transport in hydrologic systems at regional scales.

The Departments of León and Chinandega in the northwest of Nicaragua are areas where the effects of extensive OCP application over many years continue to be widely observed. From the 1950s through the 1980s, intensive cotton cultivation was widespread in the region, leading to the application of hundreds of thousands of tonnes of OCPs. The use of most of these chemicals was made illegal in Nicaragua in the 1970s and 1980s, though many recent studies have shown that their residues continue to persist in soils (Carvalho et al. 2003; Cuadra 2002), sediments (Carvalho et al. 1999; Castilho et al. 2000), surface water (Castilho et al. 2000), and groundwater (Alvarez 1994; Briemberg 1994; Castilho et al. 2000; CIRA-UNAN 1999; Dahlberg and Odebjer 2002; Delgado 2003) in the León-Chinandega region. As groundwater constitutes the entirety of the region's drinking-water supply, the prevalence of these highly toxic chemicals is of great concern.

Despite the extensive scientific attention paid to this contamination, the fate and transport of OCPs in the area is not well understood. In particular, little is known about how, despite their low solubility, significant quantities of these chemicals have been transported to groundwater. The objective of this study is to investigate quantitatively the transport of OCPs in the León-Chinandega region. Data are presented on concentrations of OCPs in groundwater, river water, and soils in the area around the municipality of Posoltega. These data are utilized alongside the findings of past studies to develop numerical and analytical models of groundwater flow and transport, with the aim of evaluating the nature of contaminants and their distribution in groundwater, of constraining the value of certain transport-related parameters, and of investigating the possible effects of increased pumping upon groundwater contamination. VisualMODFLOW is used for all numerical modeling.

The study area

Much of the León-Chinandega region is underlain by a shallow, unconfined aquifer, supplying the majority of the water consumed by agriculture, industry, and domestic users in the two departments. While this aquifer extends over an area of 1,300 km^2, modeling such an area would have prohibitively large data and computational requirements, so this study models only a 330 km^2 sub-basin of the aquifer. The aquifer as a whole and this sub-basin are illustrated in Fig. 1. Of particular focus in this study is an area of roughly 30 km^2 around the municipality of Posoltega. This region was selected because it is the primary population center within the modeled area and it is relatively easily accessed, facilitating data acquisition.

The León-Chinandega region is classified under the Köppen system (Köppen 1923) as having a tropical savannah climate, characterized by distinct wet and dry seasons. The wet season lasts roughly from May to November, and the dry season from roughly December to April. The wet season is generally interrupted in July and August by a 4–5-week dry spell referred to as the *canícula*. Mean annual precipitation in Posoltega is around 1,850 mm, but can vary greatly from year to year. The region is hot and humid year-round, with an annual mean temperature of 27°C, and annual mean humidity of 78%. Mean annual potential evaporation is around 2,100 mm (INETER 2000; UN 1974).

Most soils in the area are coarse-grained and loamy, loamy-clayey, or loamy-sandy, with higher elevation soils generally being coarser than those at lower elevations (Corriols 2003). This soil structure allows for relatively fast infiltration rates. Infiltration rates in cultivated soils are up to four times greater than in uncultivated soils (UN 1974). Organic matter content in soils varies between 0.8 and 3.7% within the first meter, with topsoil generally having a much higher organic content than soil at one meter depth (CIRA 1999).

Geomorphologically, the area of the study comprises three zones: the plains (0–100 m elevation), the volcanic foothills (100–300 m), and the Maribios Cordillera Mountains (above 300 m). Stratigraphically, the region is comprised of three formations: near-surface pyroclastic and alluvial deposits, the Las Sierras Formation, and the Tamarindo Formation. The aquifer comprises the first two of these formations, and can reach a thickness of over 340 m inland, generally thinning towards the Pacific coast. The pyroclastic materials have grain sizes from fine sand to fine gravel, while the alluvial materials consist of silt, gravel, and pebbles, and become increasingly fine with distance from the Cordillera. The Las Sierras Formation consists of older pyroclastic materials, and varies in grain size from silts to fine gravels. The Tamarindo Formation is a low-permeability unit formed by pyroclastic deposits and lava flows, and forms the aquifer basement (UN 1974).

Transmissivity in the aquifer shows a decreasing trend from the Maribios Cordillera towards the Pacific coast. Pumping tests have indicated that transmissivity varies in the aquifer from 33 to 7,100 m^2/day, and the storage

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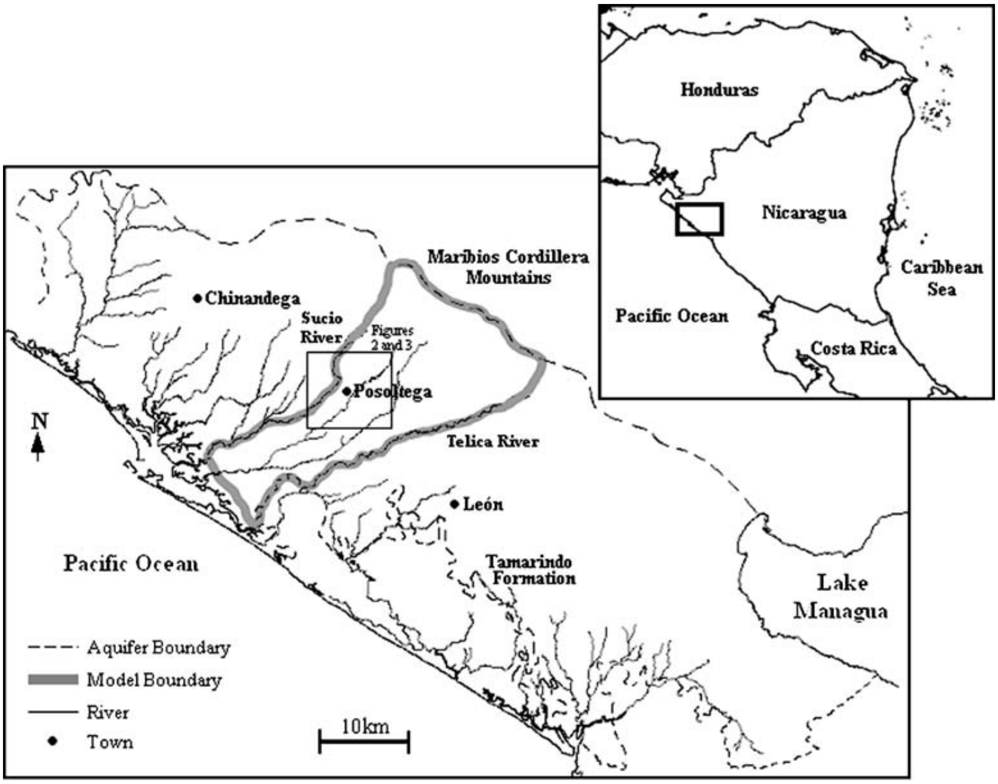

Fig. 1 The study area

coefficient has been estimated to fall in a range from 0.01 to 0.35 (INETER 2000; UN 1974). Hydraulic conductivity also decreases with depth in the aquifer due to greater compaction. Hydraulic conductivity of the Tamarindo Formation is expected to be at least two orders of magnitude less than that in the overlying aquifer (UN 1974). The water table in coastal areas can be as shallow as a few tens of centimetres, whereas at higher elevations it can be deeper than 100 m. It is believed that the aquifer has semi-distinct deep and shallow systems of flow, with anthropogenic contamination significantly more prominent in the shallow system (CIRA 1999; Dahlberg and Odebjer 2002; Calderón Palma 2003; Delgado 2003).

Isotopic studies have shown that most recharge to the aquifer originates above 280 m elevation, with infiltration in the plains recharging the shallow aquifer, and recharge at higher elevations traveling through a deeper flow system and discharging in rivers, wells, and the Pacific Ocean (Payne and Yurtsever 1974; UN 1974). Most stretches of the rivers running through the modelled region are gaining (Calderón Palma 2003). The majority of discharge from the aquifer occurs as baseflow in local rivers, or is extracted by pumping. In 1999, 234×10^{6} m^3 of water were extracted from the aquifer by pumping (INETER 2000). Most extraction occurs through relatively deep drilled wells, though there still exist many hundreds of shallow, hand-dug wells in the area.

Due largely to its favorable soil conditions and climate, much of the area to be modeled is highly developed agriculturally. Cotton production in the region is now minimal, though for many years cotton was the most widespread crop in the area. Its cultivation was dependant upon the extensive use of OCPs and other agrochemicals. The application of OCPs came into widespread use in the 1950s, increased through the 1960s and 1970s, and was greatly reduced in the early 1980s when they were outlawed, heavily regulated, or their use became economically unfeasible.

Data collection

Hydraulic and chemical data were collected in the area immediately surrounding the municipality of Posoltega. These data complement data from past studies, helping in defining the conceptual models of flow and transport, and serving as model calibration targets.

Weekly water-level measurements were made at 14 wells from August of 2004 to July of 2005 to provide a high-frequency recording of water-table fluctuations in the area. Measurements were collected manually with a water-level meter. These well locations are illustrated in Fig. 2. In addition, sporadic measurements were made at an additional 20 wells to provide a broader picture of static water levels. Water levels are generally highest near the end of the rainy season and lowest near the end of the dry season, consistent with the unconfined, shallow nature of the aquifer. The water table fluctuates by between 0.4 and 1.9 m between the wet season and the dry season, and generally fluctuates more at lower elevations than at higher elevations.

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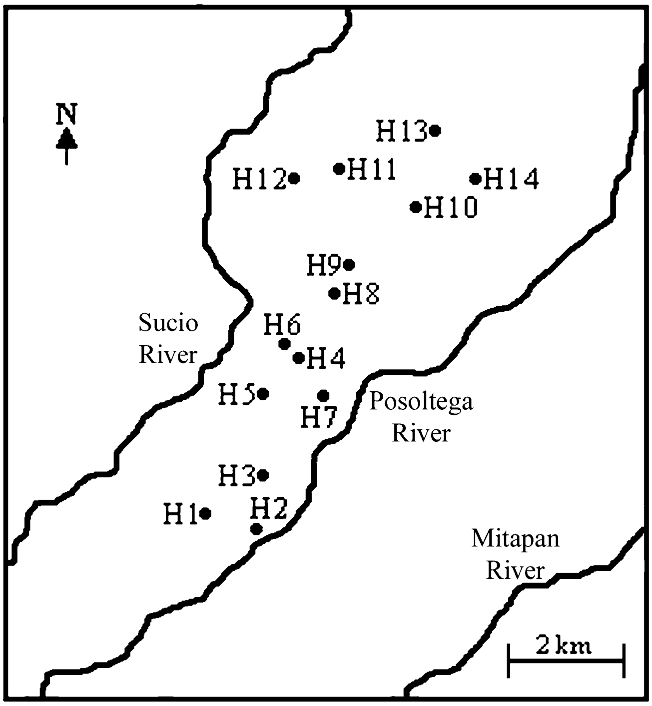

Fig. 2 Locations of head observation wells (area delineated in Fig. 1)

Water samples were collected for OCP analysis from 15 wells and each of the Sucio and Posoltega Rivers in October 2004, and from 19 wells and the two rivers in March 2005. These locations are illustrated in Fig. 3. Groundwater samples were extracted using electric or hand pumps and PVC tubing. Ideally, each well was purged of three well volumes prior to sample collection, although in a small number of situations this was impractical and wells were purged for as long as possible, or low-flow sampling was used. River water samples were collected directly from surficial river-flow. All samples were collected in 5-L glass jars, to which 50 ml of hexane was immediately added. The jars were then put on ice and protected from light and transported within 8 h to a laboratory for gas-chromatography analysis, following the technique outlined in Villeneuve (1995). Analysis was performed to determine concentrations of the following: alpha-BHC, beta-BHC, lindane, delta-BHC, heptachlor, aldrin, heptachlor epoxide, alpha-endosulphan, dieldrin, *p,p′*-DDE, endrin, beta-endosulphan, *p,p′*-DDD, *p,p′*-DDT, and toxaphene. Detection limits range from 0.02 ng/L to 1.04 ng/L for all pesticides other than toxaphene, which has a detection limit of 8.70 ng/L. It has been found that the analysis method used has a mean recovery of between 60 and 90% for all pesticides other than DDT (35%), dieldrin (192%), and aldrin (216%). The repeatability relative standard deviation has been found to be less than 15% for all pesticides analyzed (Jarquín 2005). This level of analytical precision is sufficient for achieving the objectives of this study.

OCPs detected in well water samples were DDT, DDE, DDD, toxaphene, heptachlor, dieldrin, and endrin. DDE and dieldrin were detected most commonly, appearing in 52 and 48% of the wells sampled, respectively, though generally in low concentrations. The median DDE concentration found in positive samples was 0.65 ng/L, while the minimum was 0.37 ng/L and the maximum was 3.37 ng/L. The median dieldrin concentration found in positive samples was 1.4 ng/L, while the minimum was 0.51 ng/L and the maximum was 38 ng/L. Toxaphene was detected in 14% of the wells sampled, though was generally at much higher concentrations than the other pesticides—ranging from 71 to 140 ng/L. Full analysis results are illustrated in Fig. 4. It is clear that pesticide contamination is less common and generally of lower concentration in drilled wells than in dug wells. The results of the analyses are consistent with the hypothesis that two flow systems exist in the region—one shallow, contaminated system recharged in the plains, and one deeper, cleaner system recharged in the foothills and cordillera/mountains. Dug wells are often poorly constructed and are rarely enclosed, so they may receive direct contamination from windblown materials or contaminated surface waters delivered by shortcutting of infiltrating water around the wellbore. Figure 4 also indicates that there is little consistency in observed pesticide concentrations between seasons. Some wells exhibited high levels of contamination during one sampling period, and no contamination during the other. Consequently, the distribution of pesticides through the subsurface may be quite complex, suggesting that the distribution of pesticide sources from the surface is also quite complex.

Total OCPs detected in the Posoltega River (R1) were 2 ng/L in the wet season and 1 ng/L in the dry season. In

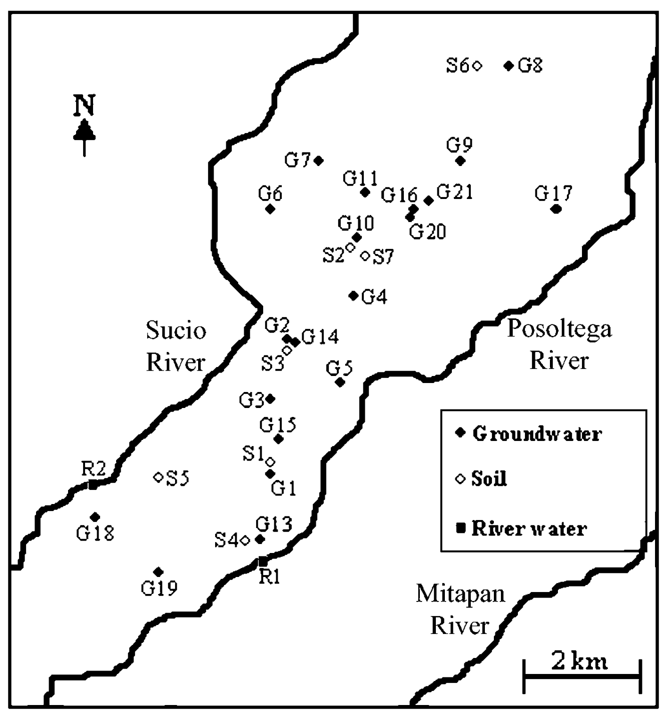

Fig. 3 Locations of samples collected for OCP analysis (area delineated in Fig. 1)

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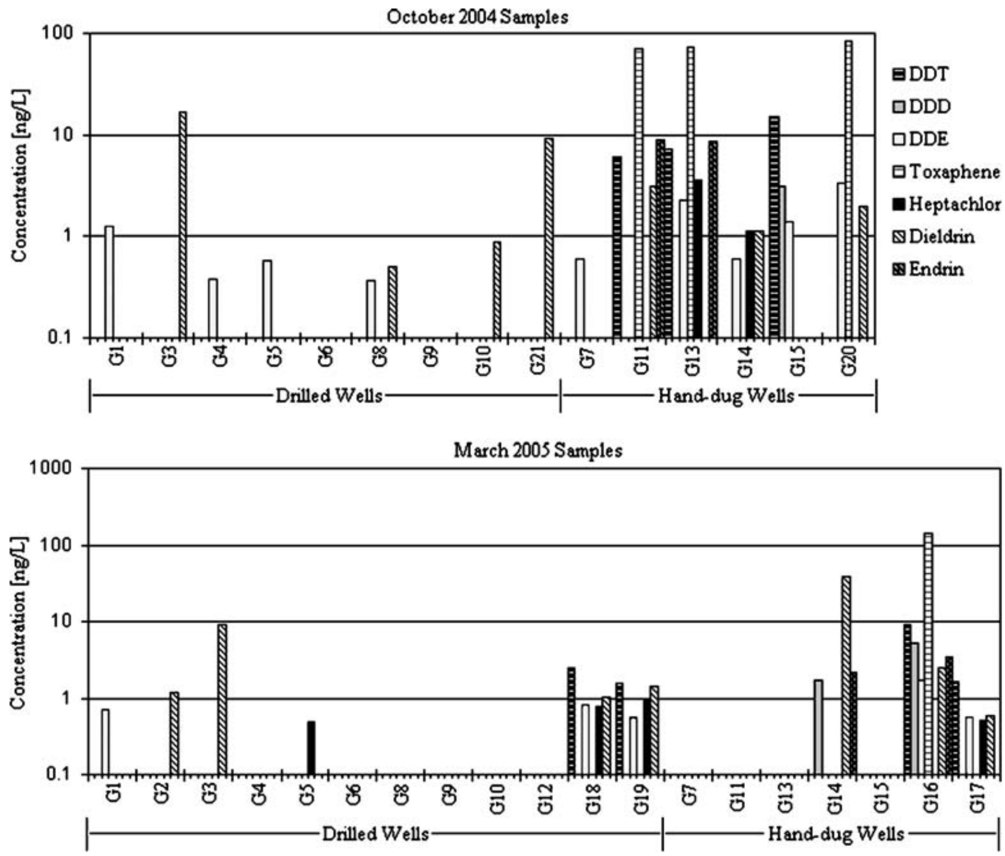

Fig. 4 Results of analysis for OCPs in groundwater samples

the Sucio River (R2), the wet-season concentration was 114 ng/L and the dry-season concentration was 1 ng/L.

Soil samples were collected for pesticide analysis from seven sites during 2004 and 2005. These locations are illustrated in Fig. 3. At three of these locations, a horizon from 0–15 cm depth and a horizon from 40–50 cm depth were collected, while at the other four locations, only one sample from 0–15 cm depth was collected. Samples were collected in 500-ml glass jars and transported on ice and protected from sunlight within 6 h to a laboratory for gas-chromatography analysis. Analysis targeted the same OCPs as for the analysis of water samples.

A large range of pesticide concentrations was found in the soil samples collected (Table 1). Toxaphene is often found in much higher concentrations than the other pesticides. Among samples of the surficial horizon (0–15 cm), total OCP concentrations range from over 10,000 to 2.2 μg/kg. The latter sample was collected from beside a road in the heart of Posoltega town, which is likely to never have seen direct pesticide application. From the three locations where both surficial and deeper samples were collected, pesticide concentrations appear to decrease rapidly with depth in the soil column. In the few tens of centimetres between sampled depths, concentrations at these three locations drop from 10,000 to 21 μg/kg, from 71 to 0.19 μg/kg, and from 4,100 to 60 μg/kg. This decreasing concentration with depth is consistent with the findings of past studies (Carvalho et al. 2003; Cuadra 2002) and indicates that infiltration of water does not effectively transport pesticides through the entire soil column.

Groundwater flow modeling

While this study's primary objective involves the investigation of contaminant transport, numerical modeling of transport requires a model of groundwater flow. The groundwater flow model utilized here was based upon the model presented in Calderón Palma and Bentley (2007). Only a basic description of the model is given here, highlighting those areas where the flow model differs from that of the preceding model. For the purposes of this study, only transient modeling was performed.

Model construction

The study area grid system is finest around the municipality of Posoltega, and becomes increasingly coarse with distance from this area (Fig. 5). Around Posoltega, grid cells are 133 m on each side, while distant cells are as large as 266 m on each side. Vertical model layers are illustrated in Fig. 6. The layer of alluvial and pyroclastic deposits is split into three layers, while the Upper and Lower Las Sierras Formations are split into two layers each. The Tamarindo Formation is represented with a single layer.

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Table 1 Results of analysis for OCPs in soil samples

Location	Samples collected over depth range 0–15 cm (μg/kg)					
	DDT	DDD	DDE	Toxaphene	Dieldrin	Endrin
S1	19	6.0	92	B	28	4.0
S2	43	B	97	10,000	8.4	5.9
S3	6.3	1.4	6.3	63	0.68	0.18
S4	6.6	2.3	13	49	B	B
S5	1.6	0.39	2.9	B	0.10	B
S6	180	56	71	3,800	3.0	B
S7	0.70	0.46	0.99	B	B	B
	Samples collected over depth range 40–50 cm (μg/kg)					
S1	N	N	N	N	N	N
S2	8.7	B	8.4	B	3.6	B
S3	N	N	N	N	N	N
S4	B	B	0.05	B	0.14	B
S5	N	N	N	N	N	N
S6	B	1.5	2.6	56	0.29	B
S7	N	N	N	N	N	N

B below detection limit

N no sample collected

Six types of boundaries are utilized in the model, including constant head, river (third type condition), drain (third type), general (third type), no-flow, and recharge (second type) (Anderson and Woessner 1992; Waterloo Hydrogeologic Inc. 2002). The 5 m topographic contour is assigned as the southwestern limit of the model, and a constant head boundary is imposed along this limit. The four rivers in the study area are represented by river boundary conditions. This condition is only applied to the top model layer, as it is believed that the rivers' strong control over hydraulic heads extends only into the shallow aquifer. The ephemerally flowing reaches of the four rivers are represented by drain boundary conditions in the top layer of the model. Along one stretch of the Sucio River, a general head boundary condition was used to simulate flow passing under the riverbed. The Maribios Cordillera, the stretches between the Cordillera and the ends of the Sucio and Telica Rivers, and an outcropping of the Tamarindo Formation in the south of the model are all represented by no-flow boundaries. At these locations, such a boundary is imposed in all model layers. No-flow boundaries also exist in all model layers below the river and drain boundaries of the Sucio and Telica Rivers, under the assumption that that these boundaries are groundwater divides. These boundaries are illustrated in Fig. 7.

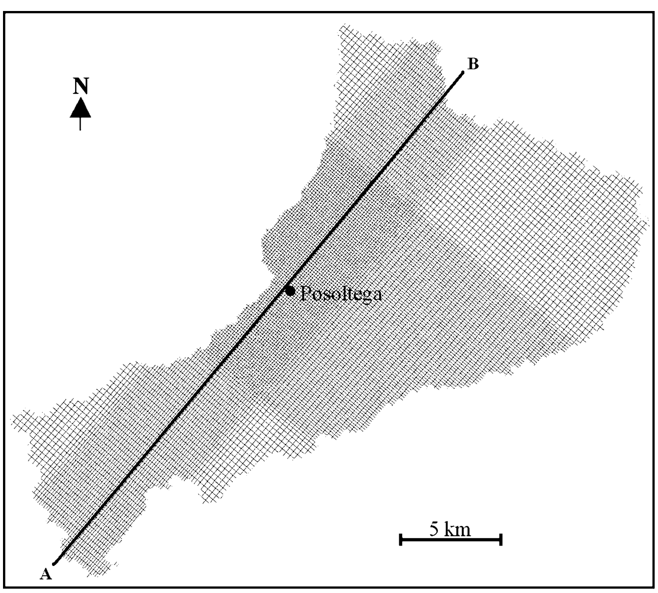

Fig. 5 Model grid in plan view

To simulate the increase in recharge with increasing elevation, recharge over the study area is separated into four zones (Fig. 7). The lower limits of these four zones correspond to 5, 25, 100, and 140 m elevation (meters above sea level), respectively. Recharge in the four zones corresponds to 10, 26, 26, and 38% of precipitation, respectively. To account for the fact that the unsaturated zone is thinner at lower elevations and thus infiltration occurs more quickly here, recharge in zone 1 and zone 2 varies with monthly variations in precipitation, whereas recharge in zone 3 and zone 4 is constant throughout the year.

Hydraulic conductivity values were determined using pumping test analysis results. Horizontal hydraulic conductivity in the aquifer varies between 6 and 270 m/day, and vertical conductivity is set to one-fifth of hydraulic conductivity in all cells. Throughout the model, specific yield is set to 0.1, specific storage to 0.0001 m^{-1}, and effective porosity to 0.3. These values were selected as they are standard values for unsaturated aquifers of this geological type, and they were found to be acceptable in the model of Calderón Palma and Bentley (2007).

Pumping plays an important role in the aquifer's modern-day water balance, and thus must be properly represented in the model to achieve an accurate calibration and allow reasonable model simulations. All known drilled wells in the model area are represented. Only drilled wells are modeled, as withdrawals from dug wells are negligible in comparison.

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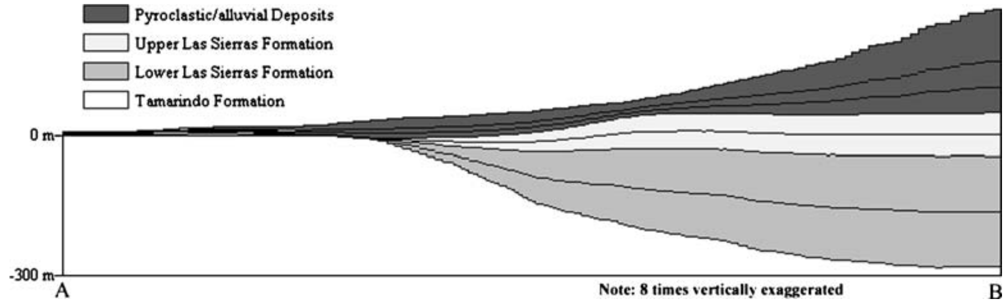

Fig. 6 Representative horizontal model grid (modified from Calderón Palma and Bentley 2007)

Model calibration

The model presented in Calderón Palma and Bentley (2007) was successfully calibrated using a variety of legacy water table and river-stage data. This indicates that the conceptual model employed is likely a reasonable representation of the region's flow system. As this study utilized a different model grid and a slightly different set of boundary conditions, the model was recalibrated using the newly collected data from around the municipality of Posoltega. Calibration targets include hydraulic head at 14 wells over a 12-month period in 2004 and 2005 (wells shown in Fig. 2), and river baseflow in the Posoltega River over a 10-month period in 2004 and 2005. Given the fact that the preceding model was well calibrated, it was expected that few changes in parameter values would be needed to calibrate this model using new data.

Figure 8 illustrates the results of calibration to hydraulic heads over the course of one year. Over this period, the mean residual between observed and predicted heads in all wells is around 3 m. The transient behaviour of the water table is predicted more accurately at lower elevations than at higher elevations. This may be the result of multi-year water-table fluctuations that the model was unable to capture. The hydraulic gradient is generally under-predicted by the model at higher elevations and over-predicted at lower elevations. Between most wells, this mismatch is within a factor of 1.7, though between a few pairs of wells it is greater than 2.

Figure 9 illustrates observed flow in the Posoltega River at El Trianón versus baseflow predictions at the corresponding grid cell. These observed data are total river flow, including contributions from both baseflow and overland flow in the wet season. Taking this uncertainty into consideration, the difference between model predictions and observations is acceptable. Residuals in the dry season are within 10% at all times, and baseflow is predicted to be around 20% less than total flow during the wet season. This value is a reasonable estimate of the expected magnitude of overland flow. Overall, the model calibration with respect to both hydraulic heads and baseflow is considered acceptable given the methods and objectives of this study.

The discrepancy in the model's mass balance was 0.9% in the wet season and 0.5% in the dry season, which was deemed acceptable. The relative magnitude of the terms in the mass balance are also reasonable, given the conceptual understanding of the region's hydrologic system.

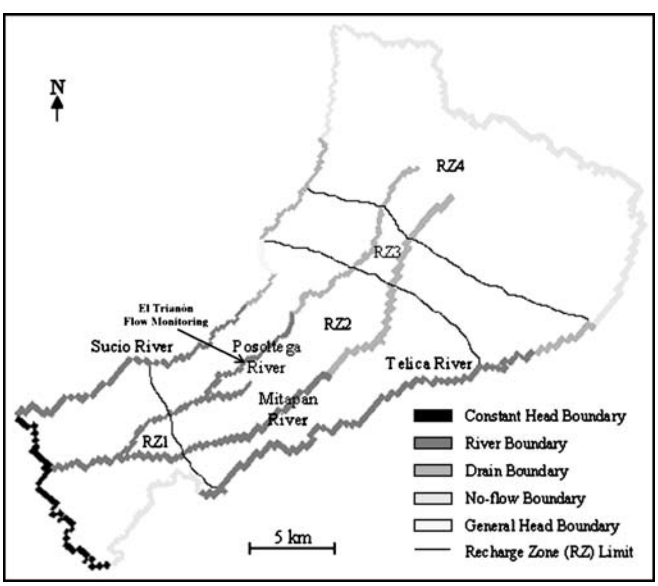

Fig. 7 Modeled boundary conditions

Sensitivity analysis

A sensitivity analysis of the calibrated model was performed to quantitatively asses the level of uncertainty in model predictions resulting from the uncertainty in model parameters. While all parameters yield some level of control over model predictions, certain parameters are expected to play a greater role than others. The parameters tested as a part of this sensitivity analysis were hydraulic conductivity, recharge, the combination of conductivity and recharge, and streambed conductance.

The sensitivity analysis indicates clearly that model predictions have a significant level of sensitivity to uncertainty in all four of the parameters considered. Considering the range of uncertainty of these parameters, heads are most sensitive to riverbed conductance, recharge in all recharge zones, and hydraulic conductivity of the alluvial and pyroclastic deposits. River baseflow is most sensitive to riverbed conductance, but also to conductivity in the upper two hydrostratigraphic units. Recharge in zone 4 and zone 2 also exert a significant level of control over baseflow.

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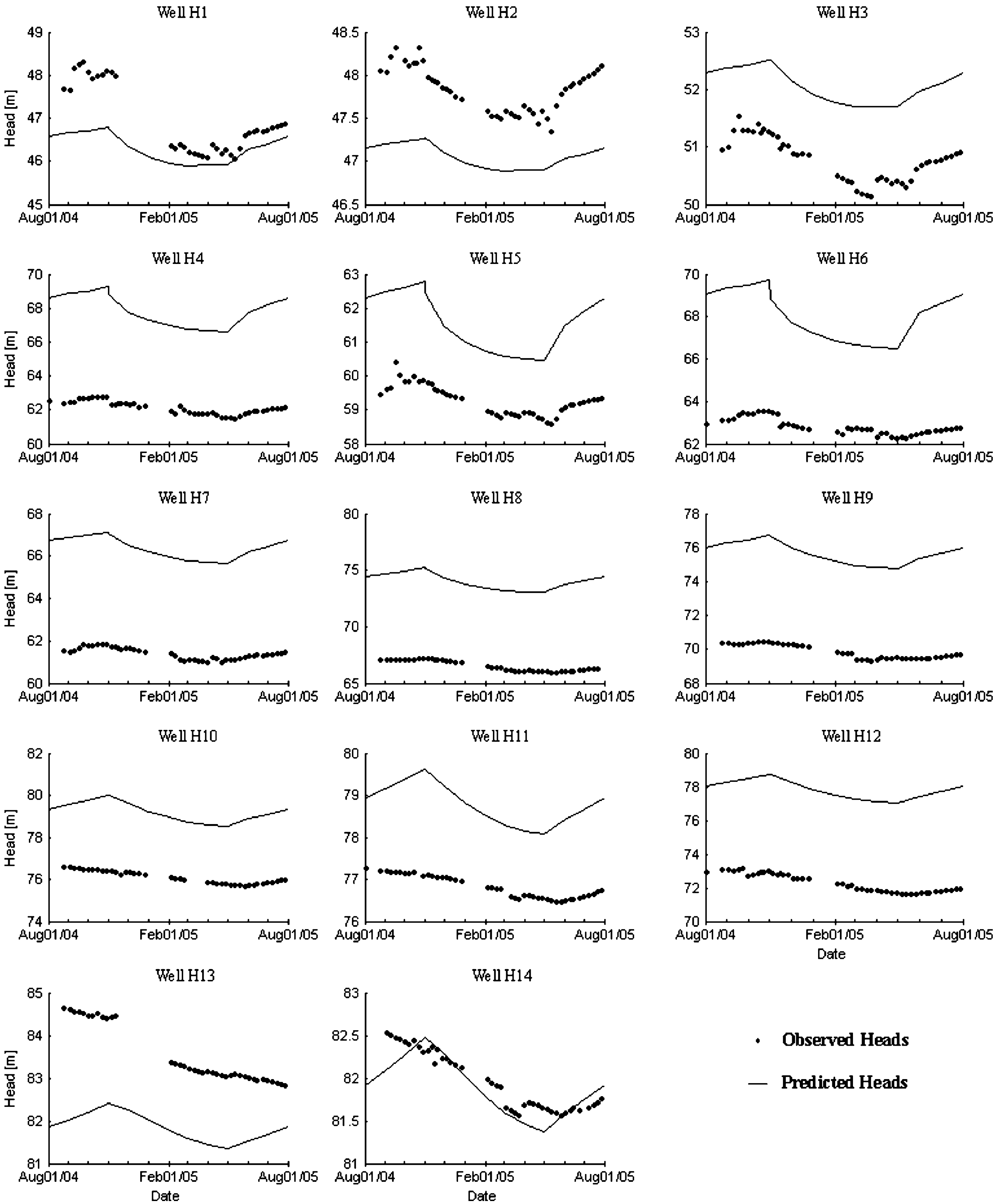

Fig. 8 Predicted versus observed hydraulic heads at 14 wells

Model simulations

While a comprehensive approach to contaminant transport modeling requires that chemical reactions and dispersion are accounted for, particle tracking allows simple transport-related predictions to be made without considering these processes. A number of concerns regarding the adverse effects of increases in pumping extractions in León-Chinandega have been raised in past hydrogeologic

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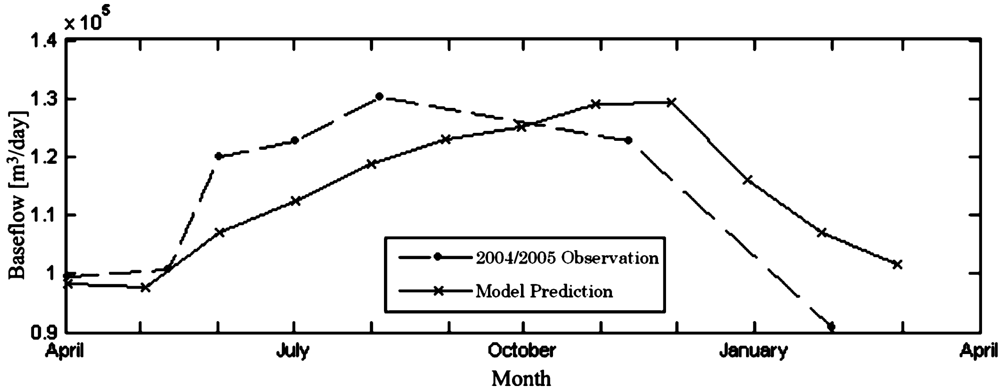

Fig. 9 Predicted Posoltega river baseflow versus observed total flow. From Moncrieff (2006)

investigations. These include the potential mixing of the deep and shallow aquifer, and the induction of flow from rivers, which are highly susceptible to contamination, into the aquifer (Calderón Palma 2003; Delgado 2003).

Data from this and past studies have shown that the shallow aquifer is much more contaminated by OCPs than the deep aquifer, and that the same may be true of nitrate contamination. As a result, water extracted from shallow wells is often of questionable quality, while water from deep wells generally conforms to drinking-water standards. To investigate this situation, particle transport through the aquifer is simulated under 2005 pumping conditions, and under a 50% increase in pumping. Ten particles are placed near the town of Posoltega at varying depths from the water table to the aquifer bottom, and are tracked backwards in time. The results of this particle tracking are illustrated in cross-section in Fig. 10. Under the increased pumping regime wells extract particles that have followed a deeper path through the aquifer than under 2005 pumping conditions. This indicates that increased pumping does indeed produce vertical gradients in the aquifer, potentially leading to mixing between shallow and deep water. Such a scenario could presumably lead both to contamination of the deep aquifer, and to dilution of contaminants in the shallow aquifer.

While most sections of the rivers passing through the study area are gaining reaches, there is some concern that increased pumping in the future will induce a significant flow of river water into the aquifer. If this surface water is highly contaminated, this will presumably lead to further contamination of groundwater. To investigate this scenario, 24 particles are placed in the flow model either directly in river cells, or within 200 m of the Sucio and Posoltega Rivers around the town of Posoltega. Forward tracking of these particles in time is simulated over a period of 5 years under 2005 pumping conditions, and under a 50% increase in pumping conditions. These pathlines are illustrated in Fig. 11. Particles placed in river cells only leave the cells if the net flux to a cell is negative. The total calculated fluxes into and out of all river cells is listed in Table 2. This simulation predicts that under both pumping scenarios, no particles originating in the rivers enter the aquifer within a 5-year period. This signifies that no net gaining reaches are transformed into net losing reaches given the 50% increase in pumping extractions. Table 2 indicates that fluxes out of the rivers given these two pumping scenarios is similar during the wet season, though during the dry season, up to 48% more water exits the river under the increased pumping scenario. Increased pumping significantly decreases dry-season baseflow, and may draw river water into the aquifer, but the river water stays near the rivers and is transported back to them during the wet season. This simulation also predicts that increasing pumping can change the flow paths of particles originating near the rivers. Particles that quickly discharge to the rivers given 2005 pumping conditions are minimally diverted by increased pumping, but particles that would travel for some distance given 2005 pumping conditions are diverted away from the rivers under increased pumping. These findings indicate that while increased pumping can seriously affect river baseflow, it is unlikely to lead to extensive contamination of the aquifer. It is, however, likely that increased pumping could lead to contamination of the areas near the rivers, as river water could seasonally enter the aquifer in these regions. This should be a consideration when planning the location of drinking-water wells.

Contaminant transport modeling

The primary objective of modeling contaminant transport in this study is the evaluation of the nature of contaminants and their distribution in groundwater. To do this, it is necessary to consider their movement through both the unsaturated and saturated zones. Here, analytical modeling is used to represent unsaturated-zone transport, while saturated-zone transport is modeled numerically.

Conceptual OCP source model

A number of pathways exist through which OCPs could conceivably be transported to groundwater in León-Chinandega. The first involves their travel through the unsaturated zone, either at the site of application, or elsewhere, following transport by overland flow or wind drift. The second entails contaminants being delivered to wells while sorbed to wind-blown particles. The third

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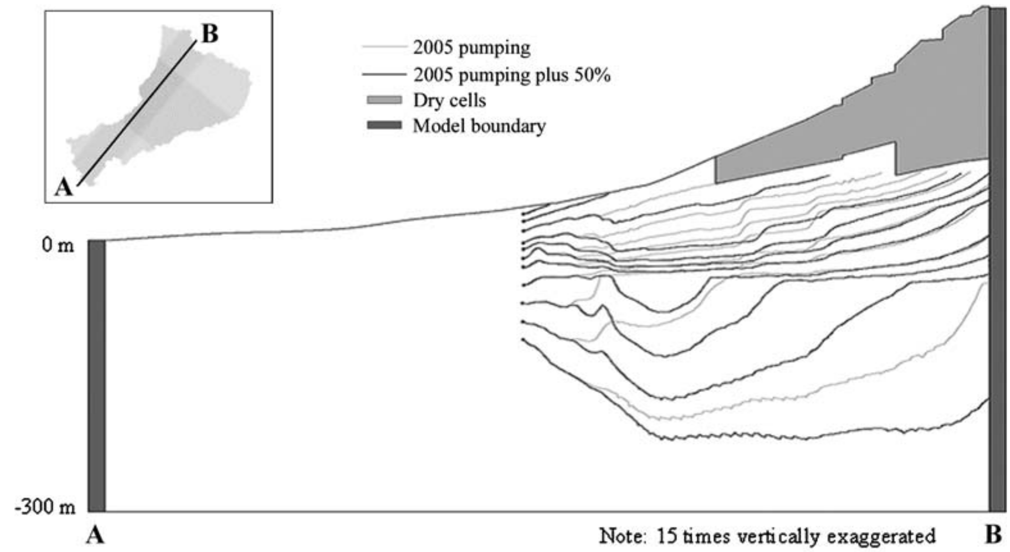

Fig. 10 Flow pathways through the aquifer under 2005 pumping conditions, and increased pumping conditions

involves their transport to rivers by overland flow, and their subsequent transfer into groundwater through the riverbed. This study aims to better understand the relative importance of each of these pathways, and to investigate the nature of each pathway.

Considering first the case, where contaminants travel through soils to the water table, the hydrophobic nature of OCPs suggests that contaminated soils may act as a long-term source of contamination to infiltrating water. There are a number of ways that pesticides could potentially be transported through the unsaturated zone to the water table. The first entails transport in the dissolved phase through matrix flow infiltration of recharging water. This process could cause contaminants to enter groundwater either at a specific location, or spread over a large area, depending on whether the source was a single point, or diffuse. The nature of this flow suggests that much of the infiltrating water comes into contact with the soil. As these chemicals readily sorb to organic materials and certain mineral grains, this method of transport would leave pesticide residues sorbed to soils over most or all of the distance between the surface and the water table. The soil contamination data presented in this and past studies indicate that this is not that case, because little to no pesticides are found in soils beyond a depth of one meter. Consequently, it is considered unlikely that transport by matrix flow plays a major role in the movement of OCPs to groundwater in the area of this study.

Another possible method of transfer through the unsaturated zone entails transport in the dissolved phase through preferential flow of infiltrating water. This process can also cause contaminants to enter groundwater either at a specific location, or spread over a large area, depending on the source. Preferential flow occurs when water and solutes move primarily through macropores or fractures, largely bypassing the soil matrix. A simple, commonly used model of preferential flow involves a mixing layer that extends from the surface to less than one meter depth, where matrix flow occurs (Steenhuis et al. 1994). Below this layer exists another layer that extends to the water table where preferential flow is dominant. The soil data collected in this and other studies shows that little contamination exists below one meter depth in the study area. This distribution may indicate that transport by matrix flow essentially terminates at this depth, either because matrix flow does not occur beyond this depth, or because almost all pesticides have been sorbed to soils by the time infiltrating water reaches this depth.

An additional possible transport mechanism entails transport in the dissolved phase through shortcutting. In other words, transport carried by the relatively fast flow of water around the margins of poorly constructed wells. This mechanism would deliver water quickly to the water table, thus sharing many of the same transport properties as preferential flow, but could only introduce contaminants to groundwater at point sources.

All of the above-mentioned mechanisms of infiltration could also occur while pesticides are sorbed to particulate matter, or colloids, rather than in the dissolved phase. Depending upon the properties of the colloids, hydrophobic contaminants such as the OCPs considered in this study may be readily adsorbed to them. As these particles are mobile, the transport of sorbed pesticides is enhanced.

As discussed above, transport through the unsaturated zone is just one possible explanation for the observed transfer of pesticides to groundwater. It is likely that a high proportion of the wind-blown particles in the area hold some sorbed pesticides. Any such particles delivered to dug wells are effectively carrying contaminants to the water table, and these contaminants can subsequently be transported in the subsurface. While this mechanism of transport can only occur where wellheads are unprotected, very few dug wells in the area are covered, and almost none are consistently covered effectively.

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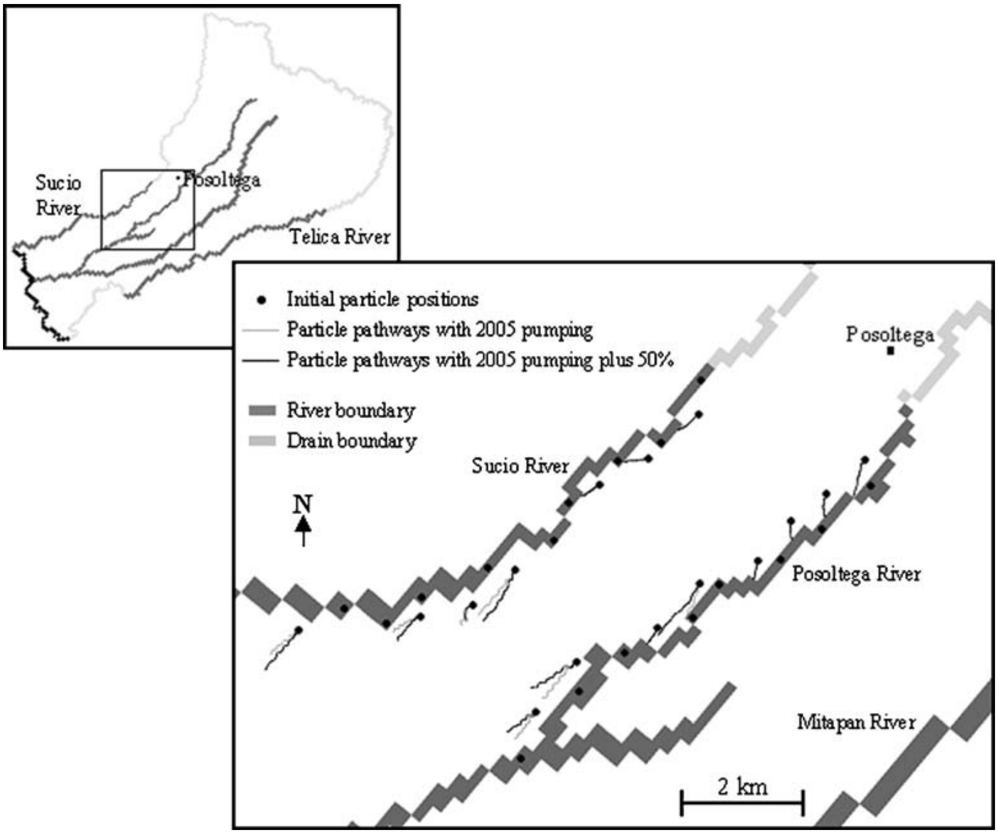

Fig. 11 Flow pathways near modeled rivers under 2005 pumping conditions and increased pumping conditions

Another possible explanation for the transport of OCPs to groundwater involves the transport of pesticides to rivers by overland flow or wind drift, and their subsequent movement through the riverbed. Legacy data show that riverbed sediments hold significant concentrations of OCPs, indicating that transport of contaminants from river water to groundwater may be occurring. Nevertheless, current understanding of groundwater flow in the area of this study is not consistent with this hypothesis. Firstly, most stretches of the rivers in the study area are gaining. Secondly, extensive contamination has been observed to the northeast of where any rivers in the study area begin. Thus, for the rivers to lead to extensive groundwater contamination, transport of pesticides against the predominant hydraulic gradients would have to be prevalent.

In summary, the most plausible transport mechanisms for sources of OCP contamination to groundwater are preferential flow (from point or diffuse sources), shortcutting around wells, and transport of wind-blown particles to unprotected wells.

Modeling approach

As the current understanding of the transport of contaminants in groundwater and the unsaturated zone is relatively poor, transport modeling aims simply to test

Table 2 Total flux into and out of river cells under 2005 pumping conditions and increased pumping conditions

Month	2005 pumping			2005 pumping plus 50%		
	Into rivers (m^3/day)	Out of rivers (m^3/day)	Net flux (m^3/day)	Into rivers (m^3/day)	Out of rivers (m^3/day)	Net flux (m^3/day)
May 2005	484,050	86,162	397,888	427,399	103,519	323,880
June	502,506	83,209	419,297	456,644	92,639	364,005
July	579,076	78,509	500,567	547,346	80,872	466,474
August	620,574	76,337	544,237	600,656	78,813	521,843
September	631,062	75,548	555,514	617,868	77,705	540,163
October	690,047	72,379	617,668	679,907	75,110	604,797
November	448,594	121,581	327,013	412,804	166,745	246,059
December	363,587	146,221	217,366	321,652	204,736	116,916
January 2006	331,787	157,216	174,571	286,260	222,852	63,408
February	316,766	162,876	153,890	268,459	232,274	36,185
March	325,457	160,065	165,392	274,320	230,602	43,718
April	337,154	153,930	183,224	282,825	227,647	55,178

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the various conceptual models discussed above, rather than to make predictions of future contaminant concentrations. This will be achieved by comparing observed contamination levels to modeled predictions given different transport mechanisms and parameter values.

While the OCPs under consideration all have somewhat different chemical properties, these properties are not well enough constrained to make the modeling of each individual pesticide a worthwhile venture. Instead, this study considers a single 'generic' chemical that exhibits transport-related properties within the range of the pesticides under consideration. While this approach is a highly simplified method of representing these pesticides, it can help constrain the source distribution and the range of transport parameter values. The key properties for modeling transport include the organic carbon partition coefficient (K_{oc}), and degradation half-life. Given the range over which K_{oc} of these chemicals is most commonly reported, this parameter will be assigned a value between 10^3 and 10^5 ml/g. Although degradation half-life in soils and groundwater has been observed to range from days to decades, it is most commonly reported to be on the order of 10 years (MacKay et al. 1997). Thus, this parameter will be assigned a value of 10 years in all media. This is a gross simplification, as half-life undoubtedly varies spatially and temporally in the study area and between different media. However, this simplification is deemed acceptable, as the uncertainty in this parameter is significantly less than the uncertainty in other model parameters.

Unsaturated zone modeling

As discussed above, an effective method of quantifying the concentrations of contaminants transported through preferential flow involves a relatively simple two-layer model. In this model, the first layer is a mixing zone of between 5 and 25 cm, where matrix flow occurs, and where contaminants reach an equilibrium between infiltrating water and soils. In the second layer, flow occurs predominantly through preferential flowpaths. Preferential flow passes quickly through this zone to the water table with little chemical modification (Steenhuis et al. 1994).

Using this two-layer model the dissolved-phase concentration (C_d) in the mixing zone can be calculated analytically given a pair of assumptions. The first assumption is that the sorbed- and dissolved-phase contaminants in the mixing zone are at equilibrium. The second assumption is that the distribution coefficient—the ratio of sorbed contaminants (C_s) to dissolved contaminants (C_d)—is equal to the product of the organic carbon partition coefficient (K_{oc}) and the fraction of organic carbon in soils (f_{oc}). While both organic matter and clay content can in certain instances play an important role in the sorption of organic compounds (Means et al. 1982; Väisänen 2004), it is generally accepted that unless the ratio of f_{oc} to clay content is very low, sorption to organic matter is much more prevalent than sorption to clays (Karickhoff 1984; Kleineidam et al. 1999; Means et al. 1982). As clay content in soils of the León-Chinandega region is relatively low (Cuadra 2002; Väisänen 2004), this second assumption has been deemed acceptable. Given these two assumptions, a simple relation can be used to calculate the concentration of dissolved contaminants in the mixing zone:

$$C_d = \frac{C_s}{K_{oc} \cdot f_{oc}} \qquad (1)$$

As little chemical modification occurs during preferential transport through the unsaturated zone, this equation, along with some knowledge of its parameter values, can be used to define a reasonable range of contaminant concentrations reaching the water table. It has been observed that concentrations sorbed to soils in the mixing zone currently are between 10 and 1,000 μg/kg in most regions of the study area. This value varies over time as pesticide degradation and leaching occurs, so it will be assumed that this value may have been as high as 10,000 μg/kg in the past. The fraction of organic carbon in this mixing zone is widely seen to be between 1 and 5%, so this range will be adopted for this parameter. As discussed above, the organic carbon partition coefficient is assumed to be between 10^3 and 10^5 ml/g.

By applying combinations of these parameters in Eq. (1), a plausible range of concentrations of dissolved-phase pesticides transported to the water table through preferential flow spans from 2×10^{-3} to 100 μg/L. These calculated values will be utilized in the following section to represent the range of pesticide concentrations in recharging waters.

While these values represent the dissolved-phase transport of contaminants, facilitated transport by colloidal materials also must be considered as a mechanism for delivering contaminants to the water table. Also worth consideration is the possibility that wind-blown particles deliver sorbed contaminants to the water table when they enter dug wells. As no data exist regarding colloidal or wind-blown transport in the area, it is not possible to quantitatively estimate what role these transport mechanisms could be playing.

Saturated zone modeling

Developing a precise model of contaminant transport in groundwater requires a well-defined source term, in addition to well-defined transport parameters. As neither of these is available, a highly simplified approach to modeling is adopted here. Quantitative analysis for OCPs was performed on samples from 21 wells as part of this study The mean total concentration between all samples at all of these wells is 19.6 ng/L. While this value does not represent the concentration at any given well, it is a good order-of-magnitude estimate of the concentrations seen throughout the study area. The transport models developed here aim to vary the distribution and concentration of the simulated pesticide source, as well as the value of the distribution coefficient, in order to establish the range of

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parameter values that yield concentration distributions that are reasonably close to the observed distribution.

As discussed in the previous section, the modeled concentration of the contaminant source, regardless of distribution, ranges from 2 to 10^5 ng/L. Other parameters of importance in groundwater transport models include the distribution coefficient, which is the product of the organic carbon partition coefficient and organic matter content, degradation rate, bulk density of geologic materials, and dispersion. K_{oc} is expected to be somewhere between 10^3 and 10^5 ml/g. The value of f_{oc} is assumed to be between 0.01 and 5%, the range commonly seen in groundwater studies. Thus, the possible range of the distribution coefficient is from 0.1 to 5,000 ml/g. The degradation half-life is set to 10 years for both the dissolved and sorbed phases, for the reasons discussed previously. Bulk density is set to 1.7 g/cm^3, longitudinal dispersivity to 15 m^{-1}, horizontal transverse dispersivity to 1.5 m^{-1}, vertical transverse dispersivity to 0.15 m^{-1}, and diffusion coefficient to zero. These values result in a Peclet number varying from 9 to 18, depending upon cell dimensions, indicating that transport is advection-dominated. These parameter values are simply rough estimates of what is expected in this scenario, but it is believed that they are within one order of magnitude. This level of uncertainty is less than the level of uncertainty in the other parameters discussed above.

This transport modeling process aims to identify possible combinations of K_d and C_d that could lead to contaminant concentrations similar to those seen today. Models are run over a 40-year period, as the height of pesticide application began around this long ago. If at any time over this 40-year period, the mean predicted concentration at the 21 observation wells is within one order of magnitude of the observed mean concentration of 19.6 ng/L, the model is considered reasonable. This criterion is arbitrary, though given the uncertainty in model parameters and the poorly understood historical progression of the contaminant source, it is considered appropriate.

Sorption is simulated using a linear isotherm, as concentrations are low, and degradation is represented by a first-order irreversible decay process. Advective transport is solved using the hybrid method of characteristics (HMOC) with a Courant number of 0.75. Time steps are calculated automatically by the solver.

Table 3 Predicted plausible combinations of distribution coefficient (K_d) and source concentration (C_d)

K_d (ml/g)	C_d (ng/L)
0.1	100
0.1	1,000
1	100
1	1,000
10	1,000
10	10,000
100	10,000
100	100,000

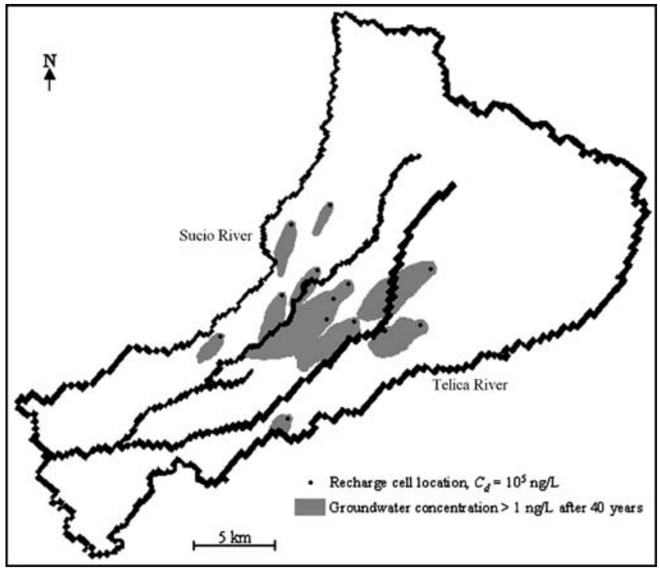

Fig. 12 40-year distribution of OCP concentrations over 1 ng/L from 12 point sources

The first source distribution to be simulated covers the entire modeled region between the coast and the 180 m elevation contour, with the exception of the town of Posoltega. This is meant to represent a highly diffuse source resulting from widespread agriculture, as much of this area has seen some level of intensive agriculture in the past, and soil contamination has been observed throughout the area. The contaminant source is simulated by applying a specified concentration to the boundary condition to all recharging water over the area of the source.

A number of plausible combinations of K_d and C_d lead to reasonable model predictions (Table 3). Thus, it is concluded that preferential flow operating from a diffuse source could have led to the observed groundwater contamination by OCPs in the area.

The second source distribution to be simulated consists of a small number of point sources distributed throughout the study area. This is meant to represent a source that is focused around areas where soil contamination is highly concentrated such as at airstrips and manufacturing and storage facilities. It is conceivable that only highly contaminated soils such as these lead to significant concentrations of pesticides being transported to the water table. A single point source is simulated by imposing a recharge boundary condition at a single cell. Twelve such point sources were distributed around the model area. Seven of these locations correspond with the locations of airstrips, while five were chosen at random, as the location of other highly contaminated sites are not known. Models were run using the same range of parameters discussed in the previous sections.

Using the lowest plausible dispersion coefficient and the highest plausible source concentration, the areas predicted to exhibit concentrations over 1 ng/L extended less than 5 km downstream of the original sources after 40 years (Fig. 12). It was also found that over the distances traveled over the 40-year period, the contaminants did not exhibit a large amount of transverse

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dispersion, so were not distributed over a large area. Given these predictions, it was concluded that it is highly unlikely that a small number of point sources in soils could possibly lead to the widespread distribution of groundwater contamination observed in the area.

The third source distribution to be simulated involves point sources at the 21 observation wells. This is meant to represent groundwater contamination directly at these well locations through shortcutting or through contamination by wind-blown particles. In the case of shortcutting, soil contamination would likely be widespread, but contaminants would only be transported to the water table at the wells.

This source distribution is simulated by imposing recharge boundary conditions at each of the concentration observation well locations. Models were run using the same range of parameters discussed in the previous sections. These parameter values are considered to be reasonable for shortcutting, but it is unknown whether this is the case for contamination by wind-blown particles. It was found that the same parameter combinations found to be plausible using a diffuse source are also plausible using this source distribution. This was taken to indicate that contamination by one or both of these transport mechanisms may be playing a significant role in groundwater contamination in the area.

Discussion and conclusions

A numerical model of groundwater flow in the León-Chinandega region of Nicaragua was developed and calibrated using hydraulic head and river-flow data. Using this calibrated model, a model of saturated-zone contaminant transport was developed. In addition, an analytic model of transport in the unsaturated zone was developed, with the aim of identifying the concentration of pesticides reaching the water table. As transport processes in groundwater and the unsaturated zone are not well understood, a highly simplified modeling approach was adopted. While this approach is not appropriate for making highly accurate concentration predictions, it is considered suitable for testing conceptual models of transport.

Flow-model calibration was performed using hydraulic head and baseflow data as calibration targets. While the overall match between observations and model predictions was good, the model was unable to accurately predict hydraulic gradients in some areas of the model, including some locations where the ratio of predicted to observed gradient was greater than 2. Nevertheless, given the highly simplified and conservative approach taken to transport modelling, and the qualitative nature of flow simulation conclusions, this mismatch is not considered significant. Thus, the model calibration was deemed acceptable.

The calibrated flow model was used to perform particle-tracking simulations designed to assess the possible consequences of increases in pumping extractions upon aquifer contamination. It was found that increasing pumping by 50% over 2005 pumping levels could induce vertical hydraulic gradients in the aquifer, possibly leading to mixing of the shallow, contaminated aquifer with the deep, relatively clean aquifer. This would act to dilute contaminants in the shallow aquifer, but would increase the depth to which contamination extends, making it more difficult to access water completely free of contaminants. It was also predicted that increasing pumping extractions by 50% would greatly increase the volume of water flowing from rivers into the aquifer during the dry-season, and would greatly decrease dry-season baseflow. However, the majority of this water would return to the rivers during the wet season. Thus, even if this water is extensively contaminated, it likely does not pose a major risk to the aquifer as a whole, though may pose a risk to wells located near a river.

Analytical modeling of the unsaturated zone indicated that preferential flow or shortcutting around wells could deliver OCP concentrations of between 2×10^{-3} and 100 μg/L to the water table. The large uncertainty in this value results from the fact that soil contamination is seen to vary greatly, and from the fact that the values of the distribution coefficients of the pesticides being considered are not well constrained.

Using this range of possible source magnitudes, it was determined that the observed groundwater contamination distribution could not have resulted solely from a small number of point sources in soils. The nature of the flow system and the high distribution coefficients of the pesticides in question do not allow point source contamination to be widely distributed throughout the shallow aquifer within a reasonable period of time. Instead, either a diffuse source, or a source operating directly at wells is necessary. The presence of a possible diffuse source is seen in the widespread nature of soil contamination in the area. Transport from the soil to groundwater may occur from a diffuse source through preferential flow, or solely at wells, through shortcutting or wind-blown transport.

Modeling shows that it is possible for shortcutting alone to lead to the observed pesticide distribution in groundwater. While not explicitly modeled, this implies that the same may be true of contamination delivered to wells with wind-blown particles. However, it seems unlikely that either of these are the sole mechanism at play, as contamination is seen not only at shallow, hand-dug wells, but also occasionally at deeper, well-constructed, drilled wells. It is more likely that transport of contaminants through preferential flow is prevalent in the area, though it seems likely that both shortcutting and wind-blown transport are also active to some extent.

It is also plausible that colloidal transport plays an important role in the movement of OCPs in the unsaturated zone and in groundwater. If this were the case, it could prove that current understanding of contaminant transport in the area is greatly flawed, as such transport has the potential to move much greater quantities of hydrophobic contaminants than does dissolved-phase transport.

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