

# A Combined Salt Transport-Chemical Equilibrium Model for Calcareous and Gypsiferous Soils<sup>1</sup>

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## ABSTRACT

Chemical precipitation-dissolution and cation exchange sub-routines were interfaced with an existing water movement-salt transport model. Three model options available for testing the prediction of salt transport and storage were (i) individual ion transport without soil interaction, (ii) precipitation and dissolution of lime and gypsum during transport, and (iii) cation exchange in addition to the precipitation-dissolution reactions. The transport model also predicts relative crop growth and water uptake as affected by soil moisture and salinity. The chemical subroutine used by the second and third options calculated ionic activities, corrected for ionic strength and ion pair formation, and was used to calculate lime and gypsum precipitation and dissolution. Cation activities were also used to calculate Ca, Mg, Na, and K exchange equilibria by a method that allows for addition of any number of exchangeable cations. Values predicted by the three options for EC, SAR and Ca, Mg, Na, K, Cl, SO<sub>4</sub>, and HCO<sub>3</sub> concentrations were compared to experimental data obtained from a lysimeter study and were only satisfactorily predicted when both chemical precipitations and cation exchange were considered for a gypsiferous and a nongypsiferous soil irrigated with a high, medium, and low CaSO<sub>4</sub> water at two leaching fractions.

**Additional Index Words:** electrical conductivity, soluble salts, lime, gypsum, calcium carbonate, calcium sulfate, saline soils.

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SALT MOVEMENT AND STORAGE SIMULATION in soils is at least a two-step process. First, transient water flow coupled with soluble salt transport must be considered, followed by calculation of slightly soluble salt dissolution and precipitation. If the irrigation water quality or quantity applied is variable, or if steady-state exchange has not been established, reasonable salt transport and storage prediction with depth and time also requires cation exchange consideration (Jury and Pratt, 1980). Several investigators have developed models that include several of these processes, but to date a transient water flow model that includes soil water-soil salinity-plant-atmospheric interactions has not been coupled to a mechanistic soil chemistry and cation exchange description.

Chemical models using CO<sub>2</sub>, CO<sub>3</sub>, HCO<sub>3</sub>, Ca, and pH interactions to calculate lime precipitation or dissolution (Langelier, 1936) and its effects on the sodium adsorption ratio (SAR) and the exchangeable sodium percentage (ESP) have been developed (Bower et al., 1968; Pratt and Bair, 1969). Gypsum models (Dutt et al., 1972b; Tanji, 1969) have also been developed

to predict solution composition and concentration effects on CaSO<sub>4</sub> solubility. A detailed description of Ca-SO<sub>4</sub>-HCO<sub>3</sub>-CO<sub>3</sub> interactions in solution is given by Nakayama (1969) and provides an invaluable overview of the combined lime and gypsum interactions in soil-water systems.

Oster and Rhoades (1975) predicted irrigation drainage water chemical composition for long-term irrigation management with a steady-state chemistry and water flow model that assumes the presence of lime and gypsum.

Using a plate model for water flow and assuming that leaching takes place at field capacity, Tanji et al. (1972) simulated the reclamation of soils high in soluble salts and soluble boron. Cation exchange and gypsum solubility were considered, whereas lime was not.

Using a mixing cell plate model for salt calculations, Dutt et al. (1972a) predicted salt, water flow, and solute changes due to (i) cation exchange, lime, and gypsum solubility; and (ii) N movement, transformation and uptake.

Jury and Pratt (1980) compared predicted Ca, Mg, and Cl and SO<sub>4</sub> values by a proportional model, a lime and gypsum model, and a chemistry plus cation exchange model. These models did not consider K exchange nor did they include salinity and soil water influence on plant growth.

Childs and Hanks (1975) model used a finite difference solution to the diffusion-convection equation to simulate noninteractive soluble soil salt transport. This model incorporated a dynamic root growth function, hourly evaporation and transpiration rates, relative crop yield, and considered salinity effects on crop growth and water uptake. Melamed et al. (1977) added a source-sink salt term to the diffusion convection equation to simulate chemical precipitation and dissolution during salt transport. This approach was empirical wherein the salt source-sink term was adjusted by trial and error to predict the measured soil solution parameters. Simulation of field leaching trials by this method was not satisfactory.

The present study was directed toward chemical equilibrium and cation exchange description during transient water and salt movement in soil, coupled with a relative plant growth model affected by soil water and salinity.

## METHODS

Childs and Hanks (1975) salt flow model was modified to transport solution Ca, Mg, Na, K, Cl, and SO<sub>4</sub> as nonreactive ions. The model then either prints the salt profile without considering chemical precipitation or dissolution (SALTFLOWI), calls the chemistry subroutines and brings the solution salts into chemical equilibrium with lime and gypsum, and prints the soil water and salt profile description (SALTFLOWII), or calls the chemistry and the cation exchange subroutines and prints the soil water, salt, and exchangeable ion profile description (SALTFLOWIII).

The transport model initial and boundary water and salt values are flexible and the depth increment number and spacing is variable. The bottom boundary for water flow can be established as no flux, constant soil-water potential, unit hydrau-

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lic gradient, or a water table. The soil physical properties are characterized by soil water potential and hydraulic conductivity vs. volumetric water content data. The initial soil water content can be established at any value equal to or below saturation. The surface water flux can be positive or negative to simulate rainfall and irrigation, or evaporation. The surface salt flux is specified on an individual ion basis to simulate changes in irrigation water quality or the difference in quality between irrigation and rainfall. An actively transpiring plant is simulated in which cover growth and root growth are calculated, and potential evapotranspiration is partitioned between potential transpiration and potential evaporation.

The chemical precipitation model used in this study assumes (i) that the soil contains lime, (ii) that the soil is sufficiently buffered that the pH of each depth increment is a constant, but can vary with depth, and (iii) that the soil solution for each depth increment is an open system with respect to  $\text{CO}_2$ , governed by the soil pH. An "open system" here meaning that  $\text{CO}_2$  can enter (as from roots or decomposing organic matter) or leave (as with moving water or soil air) the system, and does not mean the system is in equilibrium with atmospheric  $\text{CO}_2$ . Henry's Law constant ( $K_H$ ) for  $\text{CO}_2$  was assumed to be independent of temperature and salt concentration.

The soil solution electrical conductivity (EC, mmho/cm) was calculated from individual ion concentrations (McNeal et al., 1970).

Solution ionic strength ( $I$ , mol/liter) was calculated from the soil solution EC (Griffin and Jurinak, 1973) and the mono- and divalent ion activity coefficients ( $\gamma_1$  and  $\gamma_2$ ) were calculated from the Davies relationship (Stumm and Morgan, 1970).

The  $\text{CO}_2$  partial pressure ( $P_{\text{CO}_2}$ ) for each depth movement was calculated from pH and (Ca) data (where parentheses denote activity) using standard thermodynamic expressions.

Using the appropriate constants, ( $\text{CO}_2$ ) and ( $\text{HCO}_3^-$ ) were then calculated from (H) and  $P_{\text{CO}_2}$  values (Robbins, 1979).<sup>2</sup>

The ion pairs  $\text{CaCO}_3^0$ ,  $\text{CaHCO}_3^+$ ,  $\text{CaOH}^+$ ,  $\text{CaSO}_4^0$ ,  $\text{MgCO}_3^0$ ,  $\text{MgHCO}_3^+$ ,  $\text{MgOH}^+$ ,  $\text{MgSO}_4^0$ ,  $\text{NaSO}_4^-$ , and  $\text{NaCO}_3^-$  were accounted for in the ion activity calculations and the ion pair correction for (Ca) is given as an example.

The total analytical Ca concentration in solution,  $\text{Ca}_T$ , is defined as

$$\text{Ca}_T = \text{Ca}^{2+} + \text{CaHCO}_3^+ + \text{CaOH}^+ + \text{CaCO}_3^0 + \text{CaSO}_4^0 \quad [1]$$

where the concentration units are in mole/liter. By proper substitution and rearrangement Eq. [1] can be written as

$$(\text{Ca}) = \text{Ca}_T \div \left[ \frac{1}{\gamma_2} + \frac{K_{a1} K_H P_{\text{CO}_2}}{(\text{H}) K_{a2} \gamma_1} + \frac{K_w}{(\text{H}) K_{a2} \gamma_1} + \frac{K_{a1} K_{a2} K_H P_{\text{CO}_2}}{(\text{H})^2 K_{a2}} + \frac{(\text{SO}_4)}{K_{a4}} \right]$$

where  $K_{a4}$  represents the appropriate stability constant for a given ion pair and  $K_w = (\text{OH})(\text{H})$ . All other terms have been previously defined. Similar expressions were used for (Mg), (Na), and ( $\text{SO}_4$ ) (Robbins, 1979).

Using the corrected (Ca), and ( $\text{CO}_3$ ), or ( $\text{SO}_4$ ) values and the appropriate solubility product ( $K_{sp}$ ), the amount of slightly soluble salt that must be added or removed from solution to bring the system into equilibrium with solid phase  $\text{CaCO}_3$  and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  was calculated.

All constants were taken from Adams (1971) except the  $\text{CaCO}_3$  ion activity product value of  $1.13 \times 10^{-9}$ , measured in natural systems by Suarez (1977).

The chemical model required initial soil solution values for Ca, Mg, Na, K, Cl, and  $\text{SO}_4$ , and percent by weight gypsum and lime. Other parameters required in the model included soil

profile values of pH, CEC, bulk density, and moisture content. The calculated cation activity values were also used in the cation exchange calculations (Robbins et al., 1980).

The chemical and water movement data used for model validation were obtained by irrigating two soils with three different quality waters (Table 1) at 10 and 25% leaching fractions. The 12 treatments were randomly replicated three times in continuous weighing lysimeters. The lysimeter tanks were 0.30 m in diam and 1.18 m deep. Each contained 1.0 m of soil. Porous ceramic cups (1.0 bar) attached to sampling tubes in the sides of the lysimeters were inserted at 0.25, 0.50, and 0.75 m from the soil surface. Drains were provided in the bottom of the tanks. This system allowed for periodic nondestructive sampling of the soil solution and continuous monitoring of water movement into and out of the soil profile. (Robbins and Willardson, 1980).

Two Emery County, Utah soils were used. The Penoyer loam (coarse-silty, mixed, calcareous, mesic, Typic, Torrifluent) contained gypsum and the Hunting silty clay loam (fine-silty, mixed, calcareous, mesic Aquic Ustifluent) did not (Table 2).

The lysimeters were planted to alfalfa (*Medicago sativa* L. 'Ranger') and barley (*Hordeum vulgare* L. 'Stephoe') (day 1). Distilled water was used initially to irrigate the lysimeters and then once the plants were up and established (day 19), all lysimeters were irrigated according to their respective irrigation water and leaching fractions at 5- to 10-day intervals, depending on crop water use. The barley was harvested on day 140 and the alfalfa was cut on day 188, 226, and 258. The length of the lysimeter study was equivalent to two growing seasons where the barley crop would represent one growing season and the three alfalfa crops would represent a second growing season. Root densities were estimated from two 2.5-cm core samples taken from each lysimeter at the time the barley was harvested and from the entire alfalfa root system, which was removed at the end of the lysimeter study. Treatment 6 (25% leaching fraction) received 165 cm of water and treatment 7 (10% leaching fraction) received 140 cm of water during the study period.

Soil solution suction samples taken on day 42, 140, 189, 227, and 248 were taken to the laboratory and pH, EC, Cl, and  $\text{HCO}_3^-$  were measured immediately (U. S. Salinity Staff, 1954). The samples were diluted with an equivalent volume of 0.1N HCl to prevent lime precipitation due to  $\text{CO}_2$  loss and to retard biological growth in the samples during storage. Sodium and potassium were determined by flame emission and Ca and Mg were determined by atomic absorption spectrophotometry. The  $\text{SO}_4$  was determined turbidimetrically.

To verify the chemistry and exchange subroutines used in this model, the soil physical and chemical properties, the irrigation water chemical composition, and the irrigation frequency and duration were used as computer program inputs. No attempt was made to reverify the water flow and salt transport portion of the model since that portion was assumed to have been validated (Childs and Hanks, 1975). The matrix potential-water content curves were calculated from 0, -0.2, and -5.0 bar potential data and then the hydraulic conductivity-water content relations were calculated from this data and the saturated hydraulic conductivity values from soil columns packed to the same bulk density as the lysimeter soils (Robbins, 1977). Since neither in situ pH nor  $P_{\text{CO}_2}$  data were obtained, soil profile pH values were used which best produced predicted  $\text{HCO}_3^-$  values that agreed with the measured soil solution values. Surface pH values of 7.3 and 7.6 were used for the Penoyer and Hunting soils. These were the respective saturation paste pH values. The input pH values increased linearly to 7.0 and 7.2 at 20 cm and then linearly to 6.2 and 6.3 at 90 cm. This pH profile agreed with the measured soil solution pH values at 0.25-, 0.50-, and 0.75-m depths and also gave satisfactory  $\text{HCO}_3^-$  concentration agreement between the measured and calculated values. The irrigation water rates were used that give either 10 or 25% leaching fractions. The lysimeter surfaces were 1.5 m above the soil surface and did not have a fetch around them. They were outside during the summer and in a greenhouse during the cold season. As a consequence, standard potential eva-

Table 1—Irrigation water compositions.

CaSO <sub>4</sub> content	Ca	Mg	Na	K	Cl	SO <sub>4</sub>	HCO <sub>3</sub>	Total	EC
High	12.0	1.0	1.5	0.5	1.5	12.0	1.5	15.0	1.10
Medium	7.0	2.0	5.5	0.5	6.5	7.0	1.5	15.0	1.33
Low	5.0	3.6	6.0	0.5	13.0	0.5	1.5	15.0	1.46

<sup>2</sup>C. W. Robbins, 1979. A salt transport and storage model for calcareous soils that may contain gypsum. Ph.D. Diss. Utah State Univ., Logan, Utah.

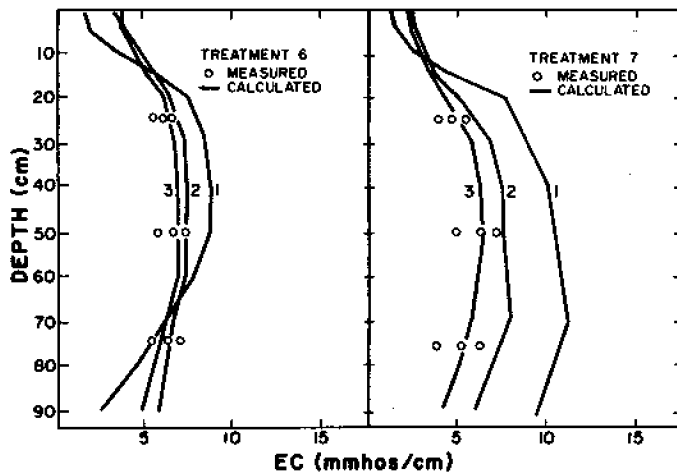


Fig. 1—Measured EC values and values calculated by (1) SALTFLOWI, (2) SALTFLOWII, and (3) SALTFLOWIII for treatments 6 and 7.

potranspiration (ET) input data for the treatments underestimated actual ET. Since the amount of water applied and the amount drained from the lysimeters was controlled, 90 and 75%, respectively, of the applied water was removed by evapotranspiration during the time between irrigations. Potential (ET) inputs to the model were therefore adjusted so that the computed water balance between ET, water storage, and drainage agreed with the measured values. Computed Ca, Mg, Na, Cl,  $\text{SO}_4$ ,  $\text{HCO}_3$ , EC, and SAR values were compared with the measured values for the three options SALTFLOWI, SALTFLOWII, and SALTFLOWIII.

For this study, no attempt was made to predict crop yield because of the unusual growing conditions. The crops were grown primarily to extract soil moisture.

## RESULTS AND DISCUSSION

In all 12 treatments, SALTFLOWIII provided the best prediction of all measured parameters. Only SALTFLOWIII satisfactorily predicted EC (Fig. 1) and SAR (Robbins et al., 1980) after 140 days for any treatment. In a few cases one of the other calculation options also gave satisfactory predictions for a particular parameter, but these were limited. Only treatment 6 and 7 of the 12 treatments modeled will be discussed in detail. Treatment 6 was the irrigation of Penoyer loam with water containing 5.0 meq/liter Ca and 0.5 meq/liter  $\text{SO}_4$  (Table 2) at a 25% leaching fraction. The soil initially contained 0.7% gypsum by weight. This treatment produced the greatest gypsum dissolution rate of those used in this study. Treatment 7 consisted of irrigating Hunting silty clay loam with water containing 12 meq/liter Ca and 12 meq/liter  $\text{SO}_4$  at a 10% leaching fraction. This soil initially did not contain gypsum, but of the treatments

Table 2—Saturation extract data for the soils used.

Extractable ions	Penoyer soil	Hunting soil
	meq/liter	
Ca	33.4	18.0
Mg	9.7	9.0
Na	1.5	6.0
K	2.6	1.7
Cl	1.7	8.2
$\text{SO}_4$	39.5	17.3
$\text{HCO}_3$	5.8	8.9
	mmho/cm	
EC	3.0	2.7

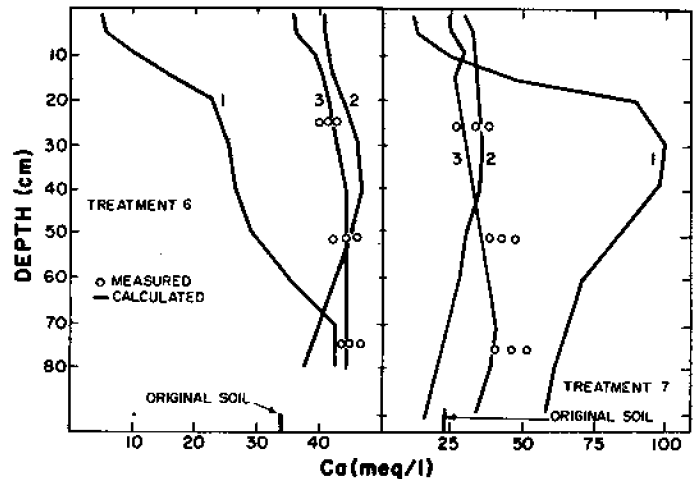


Fig. 2—Measured Ca concentrations and concentrations calculated by (1) SALTFLOWI, (2) SALTFLOWII, and (3) SALTFLOWIII for treatments 6 and 7.

applied to this soil, this treatment produced the greatest gypsum precipitation.

Electrical conductivity was satisfactorily predicted only by SALTFLOWIII for treatment 6 and 7. The other options overestimated EC for both treatments (Fig. 1). Calculated SAR values were also only satisfactorily calculated by SALTFLOWIII (Robbins et al., 1980).

To explain the differences in the ability of the three options to predict EC and SAR, it is necessary to look at the individual ions since each are handled slightly differently due to differences in chemical behavior.

The chloride ion was considered to move independently of chemical reactions and cation exchange and was satisfactorily predicted for all treatments when the calculated water movement into and out of the soil profiles agreed with the measured water movement. This would suggest that the water and salt transport model was working properly (Robbins, 1979).

In treatment 6, Ca was dissolved from the surface of the gypsiferous soil when irrigated with a low  $\text{SO}_4$  water. When gypsum dissolution was not considered, Ca concentration predictions were lower and SAR pre-

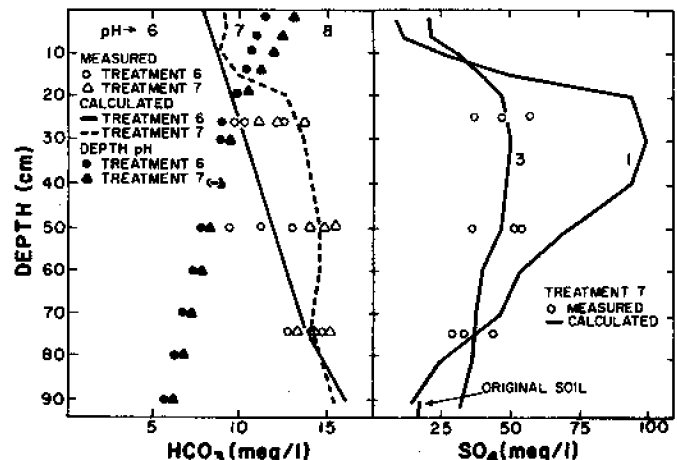


Fig. 3—Measured and calculated (SALTFLOWIII)  $\text{HCO}_3$  concentrations shown with the simulated pH profiles and the measured  $\text{SO}_4$  concentration and the concentrations calculated by (1) SALTFLOWI and (3) SALTFLOWIII.

dictions were higher than measured. When chemical precipitation was not considered in treatment 7, Ca concentration predictions were extremely high (Fig. 2). This explains in part the overestimation of EC and the underestimation of SAR for treatments where gypsum was precipitating when only salt transport (SALTFLOWI) was simulated, and also explains the results of Melamed, (1977). Cation exchange calculation was also necessary for satisfactory Ca prediction.

Estimations of Na, Mg, and K concentrations were satisfactory only when SALTFLOWIII was used (Robbins et al., 1980).

The pH profiles used are shown in Fig. 3 along with the measured and calculated  $\text{HCO}_3^-$  concentrations for the two treatments. Of the ions measured  $\text{HCO}_3^-$  is the most sensitive to the input pH values used. Carbonate and Ca are less sensitive. Carbonate concentration data were calculated and printed but are not shown here since in these pH ranges the carbonate concentrations were too low to be measured. The  $P_{\text{CO}_2}$  values calculated by this method were between 0.006 at the surface and 0.08 at the 90-cm depth and varied slightly with (Ca) changes.

When gypsum solubility was calculated but cation exchange was not considered (SALTFLOWII),  $\text{SO}_4$  predictions were in error in the opposite direction to that of Ca, since the ion activity product of (Ca) and ( $\text{SO}_4$ ) is a constant at a given temperature. When  $\text{SO}_4$  was calculated with SALTFLOWI,  $\text{SO}_4$  was considered an inert salt, and thus was not affected by precipitation or dissolution of gypsum and  $\text{SO}_4$  concentration predictions by this option were extremely low for treatment 6 and extremely high for treatment 7 (Fig. 3). SALTFLOWIII satisfactorily predicted solution  $\text{SO}_4$  concentrations in all treatments.

## CONCLUSIONS

Interfacing a chemical precipitation-dissolution subroutine and a cation exchange subroutine with an existing water movement-salt transport model provided a computer program that satisfactorily predicted EC, SAR, and Ca, Mg, Na, K, Cl,  $\text{SO}_4$ , and  $\text{HCO}_3^-$  concentrations in the soil solution for the twelve treatments studied. The chemistry subroutine was necessary for Ca,  $\text{SO}_4$ ,  $\text{HCO}_3^-$ , and  $\text{CO}_3$  predictions when precipitation or dissolution reactions were involved. The cation exchange subroutine was required for satisfactory Ca, Mg, Na, K, and  $\text{SO}_4$  predictions when the cation ratios in the irrigation water differed from those on the soil solution. Both subroutines were required for reasonable EC and SAR calculation. Likewise Jury and Pratt (1980) concluded that cation exchange and lime and gypsum chemistry was necessary for satisfactory EC prediction. However, they did not compare predicted and calculated SAR values.

The  $P_{\text{CO}_2}$ - $\text{HCO}_3^-$ - $\text{CO}_3$ -pH interaction calculations are considered the most serious weakness of the chemistry model. pH was assumed constant with time for each depth increment and decreasing with depth, and the  $P_{\text{CO}_2}$  was calculated from pH and (Ca) and then

( $\text{HCO}_3^-$ ) and ( $\text{CO}_3$ ) were calculated from (Ca),  $P_{\text{CO}_2}$  and pH. Under field conditions  $P_{\text{CO}_2}$  and pH are also related to other factors including the ratios between various cations and Cl and  $\text{SO}_4$  content. Even with this weakness SALTFLOWIII was quite satisfactory in predicting salt movement and storage under the conditions tested.

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