



Contents lists available at ScienceDirect

Sustainable Environment Research

journal homepage: www.journals.elsevier.com/sustainable-environment-research/

Technical note

Experimental and modelling investigations of tracer transport in variably saturated agricultural soil of Thailand: Column study

Tulaya Masipan^a, Srilert Chotpantarat^{b, c, d, e, *}, Satika Boonkaewwan^{c, d, e}^a Inter-department of Environmental Science, Chulalongkorn University, Bangkok 10330, Thailand^b Department of Geology, Chulalongkorn University, Bangkok 10330, Thailand^c International Postgraduate Programs in Environmental Management, Chulalongkorn University, Bangkok 10330, Thailand^d Research Program of Toxic Substance Management in the Mining Industry, Center of Excellence on Hazardous Substance Management (HSM), Chulalongkorn University, Bangkok 10330, Thailand^e Research Unit of Site Remediation on Metals Management from Industry and Mining (Site Rem), Chulalongkorn University, Bangkok 10330, Thailand

ARTICLE INFO

Article history:

Received 27 July 2015

Received in revised form

2 October 2015

Accepted 3 December 2015

Available online 13 April 2016

Keywords:

Dispersivity

HYDRUS-1D

Unsaturated agricultural soil

ABSTRACT

Tracer (Bromide) movement through the unsaturated agricultural soil was investigated in soil columns. Two tracer column experiments, with a diameter of 7 cm and a depth of 25 cm, were vertically homogeneous packed with sandy loam and then carried out to investigate bromide (Br^-) transport under different water contents (at steady flow condition). One soil column (Column 1) represents the unsaturated agricultural soil in dry season (with water content ranging from 0.23 to 0.26) and the other (Column 2) represents the soil in wet season (water content from 0.24 to 0.35). Bromide samples were periodically collected by vacuum tubes inserted at 6.25 cm equally spaced intervals (e.g., 6.25, 12.5, 18.75 and 25 cm) along the length of the column and the effluent collected at the end of the column. The observed breakthrough curves (BTCs) of bromide in both columns represented a relative smooth and sigmoidal curves at different distances (sampling ports). Dispersivity (α , cm) for sandy loam at different locations was numerically estimated by curve fitting the experimental data with HYDRUS-1D. The α can be well described by the convection–dispersion equation and these values derived from Column 1 (ranging from 0.37 to 0.98 cm) are more than those from Column 2 (0.25–0.59). Moreover, the α in both columns increases with the travel distance due to the scale-dependent effect. Furthermore, the α values were plotted on a log–log scale against travel distances and they yield empirical power law relationships with an excellent correlation ($\alpha = 0.102 (L)^{0.697}$, $R^2 = 0.999$ and $\alpha = 0.086 (L)^{0.579}$, $R^2 = 0.963$ for Column 1 and 2, respectively).

© 2016 Chinese Institute of Environmental Engineering, Taiwan Production and hosting by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

The soil-column experiment was widely used to evaluate the transport model and determine the fate and migration of contaminants through soils [1,2]. The movement of water in unsaturated porous media and associated moisture contents are important in study of surface water and groundwater interaction with the average linear velocity for groundwater movement

estimation. However, effect of the variation of water velocity on solute migration is accounted for in the convection–dispersion equilibrium (CDE) with the hydrodynamic dispersion concept. Theoretically, the hydrodynamic dispersion coefficient (D) is the sum of mechanical dispersion and molecular dispersion. The dispersivity (α) reflects the degree of mechanical mixing, caused by variations in the local groundwater velocity. The value of α has traditionally been considered under saturated water condition and usually reported in the range of 0.1–2 cm for homogeneous saturated soils [3]. From previous literature, Dagan [4], Sudicky [5] reported that the α may increase with the travel time, distance, and/or experimental scales. However, in laboratory experiments, few studies have attempted to explore the transport parameters (e.g., α) through natural geologic media under various degree of water

* Corresponding author. Department of Geology, Chulalongkorn University, Bangkok 10330, Thailand.

E-mail address: csrilert@gmail.com (S. Chotpantarat).

Peer review under responsibility of Chinese Institute of Environmental Engineering.

saturation, although many column studies of solute transport have been conducted in unsaturated porous media. As known, Thailand is the agricultural country and in order to yield high production, agrochemicals have been intensively applied for last decades. Consequently, such chemicals (pesticides and fertilizers) may, in turn, pass through unsaturated zone and eventually reach and contaminate groundwater system. Therefore, the purposes of this study were to investigate tracer transport in variably saturated agricultural soil and construct the relationship between α with travel distance under different volumetric water contents. The findings from this study derive the α of unsaturated agricultural soils for assessing groundwater contamination in agricultural areas of Thailand.

2. Materials and methods

2.1. Soil sampling

This study collected soil samples randomly in one of the intensively agricultural areas at the Huarua area, Muang district, Ubon-Ratchathani Province. This study soil is located in the northeastern part of Thailand. The soil texture of the present study is sandy loam, consisting of approximately 71% sand, 25% silt, and 4% clay. Soil samples were collected, packed in the zipper bag and then transported back to the laboratory. All soil aggregates were crushed, air-dried, and sieved through a sieve No. 10 (2 mm) before column studies.

2.2. Tracer experiment setup

The soil was uniformly wet-packed vertically into each column with 1-cm increment with an internal diameter of 7 cm and depth of 25 cm. The experimental setup using in this study are shown in Fig. 1, consisting of: 1) soil column (Column 1) with low volumetric water content (Fig. 1a), which represents soils in dry season and 2) soil column (Column 2) with high volumetric water content (Fig. 1b), which represents soils in rainy season. The groundwater table (saturated zone) is constant at the bottom end of the soil column by using overspill, connected with the bottom end of the soil column (Fig. 1b). The packed soil columns may be assumed to be homogeneous. A bulk density of agricultural soil from field measurement is $\sim 1.64 \text{ g cm}^{-3}$, for which packed soil inside the column should have the similar bulk density after the water saturation. Each column was reproduced by packing with the similar bulk density measured in the field. Firstly, deionized water from the bottom with at least 2–3 pore volumes (PVs) was used in saturated soil column to remove entrapped air and then allowed the water drain through the port at the bottom of the column. Then, a steady-state water flow was maintained by injecting the water from the

Table 1
Input parameters of the transport of bromide using in Hydrus-1D.

| Parameters | Value |
|--|-------|
| Saturated hydraulic conductivity, K_s (cm h^{-1}) | 0.41 |
| Residual soil water content, θ_r (-) | 0.16 |
| Saturated soil water content, θ_s (-) | 0.36 |
| Parameter α in the soil water retention function (-) | 0.007 |
| Parameter n in the soil water retention function (-) | 2.19 |
| Bulk density, B_d (g cm^{-3}) | 1.64 |
| Solution of bromide (Br^-), C (g L^{-1}) | 12.86 |

top of column and kept it constant at a rate of 0.41 cm h^{-1} at least 2–3 PV. After that, the solution of 12.86 g L^{-1} of bromide (Br^-) was injected at the top of the column by gravity at a rate of 0.41 cm h^{-1} (Table 1). The column effluent was periodically collected using vacuum tube water and then analysed using the electrical conductivity probe at a depth of 6.25, 18.75, 12.5 and 25 cm. The breakthrough curves (BTCs), expressed as the relative concentration (C/C_0) versus PV. Where C is Br^- concentration at time t and C_0 the influent Br^- concentration.

2.3. Water flow and solute transport equations

The one dimensional movement of water in unsaturated soils is a non-linear partial differential equation (Eq. (1)) as commonly known as Richards equation [6,7].

$$\frac{\partial \theta(h)}{\partial t} = \frac{\partial}{\partial z} \left[K(h) \frac{\partial h}{\partial z} \right] + K(h) \tag{1}$$

where $\theta(h)$ is the soil θ (volumetric water content) at the suction head ($\text{cm}^3 \text{ cm}^{-3}$); $K(h)$ is unsaturated hydraulic conductivity (cm h^{-1}); z is the vertical distance (cm) and t is time (h).

The solute transport in homogeneous porous media under a steady flow of water at constant velocity equation (Eq. (2)) can be written as

$$\frac{\partial C}{\partial t} = D_L \frac{\partial^2 C}{\partial x^2} - V_x \frac{\partial C}{\partial x} - \frac{\rho}{\theta} \frac{\partial c^*}{\partial t} \pm \left[\frac{\partial C}{\partial t} \right]_{rxn} \tag{2}$$

where C is the concentration of solute in water (mg L^{-1}); V_x is the fluid velocity which passing through the pore of media (cm h^{-1}); D_L is the coefficient of dispersion length ($\text{cm}^2 \text{ h}^{-1}$); ρ is the soil bulk density (g cm^{-3}); C^* is the adsorption of solute per unit weight of the medium porous (mg g^{-1}); rxn is the subscript indicating a chemical or biological reaction of the solute (other than sorption) often defined as the sum of the effective molecular diffusion and

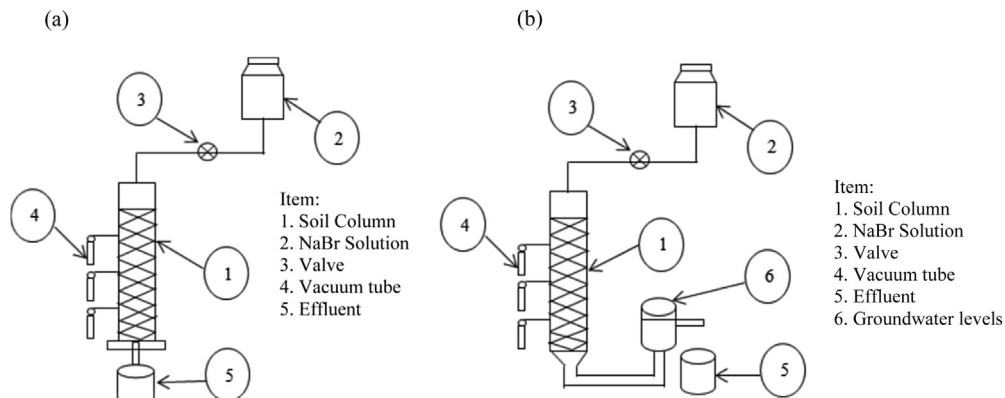


Fig. 1. Column experiment set up consisting of a) low water content (dry) and b) high water content (wet).

molecular diffusion (Eq. (3)). The ratio of the diffusion coefficient to the pore water velocity to derive α (Eq. (4))

$$D = D_w \tau_w + D_h \tag{3}$$

$$\alpha = \frac{D}{v} \tag{4}$$

where D_w is the molecular coefficient ($m^2 s^{-1}$); τ_w is a tortuosity (-); D_h is mechanical dispersion ($cm^2 h^{-1}$); v is velocity ($cm h^{-1}$), generally determined a characteristics of natural aquifer media under saturated condition. The tortuosity factor, dependent on water content, accounts for the pore geometry [8].

3. Results and discussion

3.1. Bromide BTCs: experimental results

Br^- used in the study was a non-reactive and applied to test hydrodynamic characteristics of agricultural sandy loam column. The BTC results of these column experiments are shown in Fig. 2. Under steady-state flow conditions, the BTCs of bromide show symmetrical shape, demonstrating equilibrium behaviour in sandy loam soil columns. The column was divided into 4 depth: (a) 6.25, (b) 12.5, (c) 18.75 and (d) 25 cm for inserting sampling ports. In Column 1, at 6.25 cm, bromide came out faster than those at depth of 12.5, 18.75 and 25 cm, indicating that there is no preferential flow in the column. Similar observation of BTCs in Column 2, at a depth of 6.25 cm, bromide came out faster than others. Interestingly, breakthrough of bromide in Column 2 came faster than those BTCs of the other column.

The various input parameters needed in Hydrus-1D, namely the saturated hydraulic conductivity (K_s), θ_s , θ_r , and van Genuchten parameters (α and n), bulk density (Bd) and concentration (C) were used to fit bromide transport in variably saturated soil columns as shown in Table 1.

3.2. Bromide BTCs: Hydrus-1D modelling results

The HYDRUS-1D program is a model for simulating one-dimensional water flow and solute transport in variably saturated

media through different soil types, concerning the impacts of physical and chemical non equilibrium conditions [9]. Figs. 3 and 4 show the observed data and fitted BTCs for bromide using HYDRUS-1D with the CDE model. The dispersivity values were fitted as shown in Table 2. According to fitted results with HYDRUS-1D, Column 1 (dry season) in Fig. 3, the dispersivity at depth of 6.25, 12.5, 18.25 and 25 cm are 0.37, 0.59, 0.78 and 0.98 cm, respectively with the order of R^2 from 0.9922 to 0.9952. For Column 2 (rainy season) in Fig. 4, the dispersivity at depth of 6.25, 12.5, 18.25 and 25 cm are 0.25, 0.39, 0.43 and 0.59 cm, respectively, with the order of R^2 from 0.9930 to 0.9951 (Table 2). Porro et al. [10] found that the dispersivity in homogeneous unsaturated column increased from 2.2 cm to 7.8 m at the travel distances of 82 and 400 cm, respectively. Similarly, Huang et al. [11] presented the dispersivity values increased from 0.1 to 5.0 cm as increasing distance in the homogeneous saturated sandy column of 12.5 m long. A log–log plot of dispersivity versus column length Fig. 5 shows that the dispersivity is higher under lower water content (Column 1) and tends to linearly increase as travel distance increases. They yield empirical power law relationships with an excellent correlation as follows: $\alpha = 0.102 (L)^{0.697}$ ($R^2 = 0.999$) and $\alpha = 0.086 (L)^{0.579}$ ($R^2 = 0.963$) for Column 1 and 2, respectively. These results are in agreement with the study of Neuman [12] who found that dispersivity for the flow distance less than 3500 m, can be calculated from the power law relationship as follows: $\alpha = 0.0175 (L)^{1.46}$.

The comparison of the dispersivity between two columns at each location indicates that the values of dispersivity in Column 1 were higher than those in Column 2, suggesting that most soil pores are partially filled resulting in high tortuous factor; consequently, the local velocity varies and dispersivity values become higher. Chou and Wyseure [13] found that dispersivity decreased as the volumetric water content increased.

4. Conclusions

In this present study, the BTCs of bromide transport in homogeneous sandy loam columns under different water contents and travel distances are demonstrated. Both columns showed that, at a depth of 6.25 cm, bromide came out faster than those at deeper locations. Values of the dispersivity derived from Column 1 (ranging from 0.37 to 0.98 cm) is higher than those from Column 2

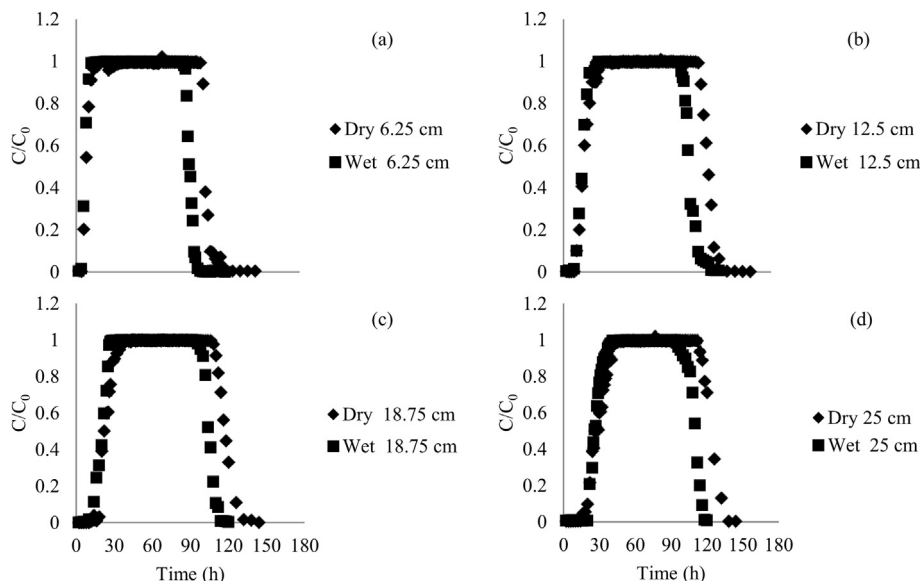


Fig. 2. Experimental BTCs for NaBr of dry and wet column at (a) 6.25 (b) 12.5 (c) 18.75 (d) 25 cm.

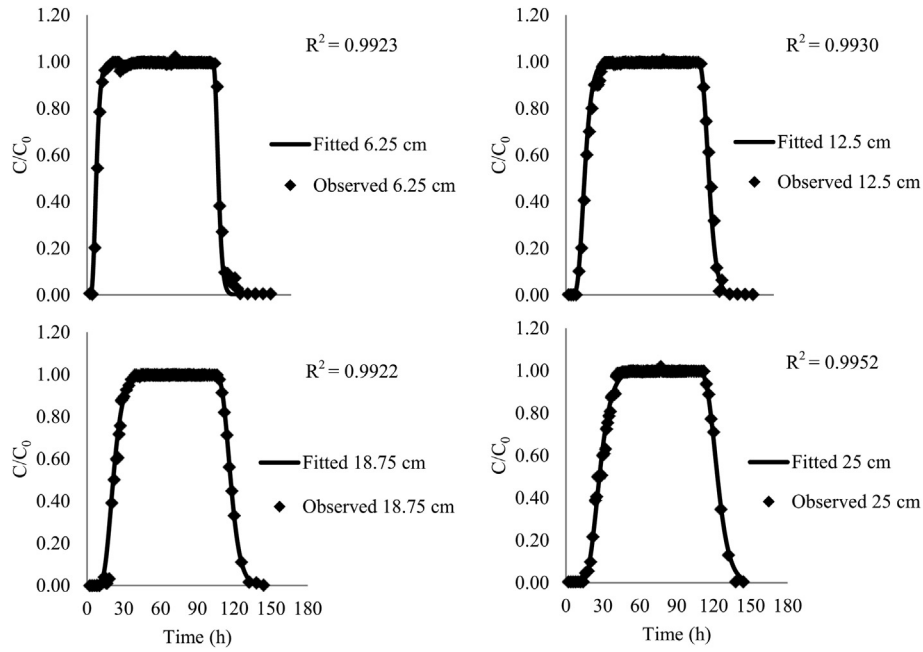


Fig. 3. Fitted curves of the observed data in Column 1 (dry).

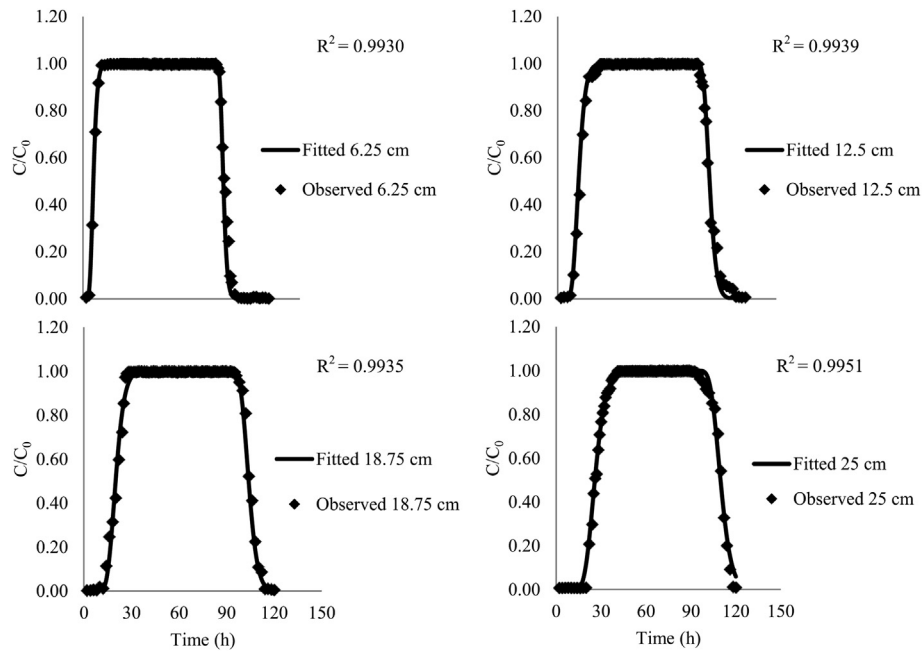


Fig. 4. Fitted curves of the observed data in Column 2 (wet).

Table 2
Dispersivity fitted from HYDRUS-1D at different locations in both columns.

| Column | Depth (cm) | Average linear velocity (cm h^{-1}) | Dispersivity (cm) | R^2 |
|----------------|------------|--|-------------------|--------|
| Column 1 (dry) | 6.25 | 0.41 | 0.37 | 0.9923 |
| | 12.5 | 0.41 | 0.59 | 0.9930 |
| | 18.75 | 0.41 | 0.78 | 0.9922 |
| | 25 | 0.41 | 0.98 | 0.9952 |
| Column 2 (wet) | 6.25 | 0.41 | 0.25 | 0.9930 |
| | 12.5 | 0.41 | 0.39 | 0.9939 |
| | 18.75 | 0.41 | 0.43 | 0.9935 |
| | 25 | 0.41 | 0.59 | 0.9951 |

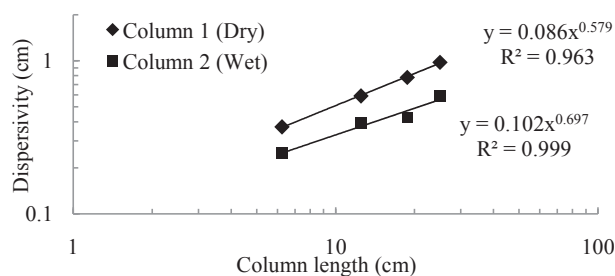


Fig. 5. The power law relationship between dispersivity versus travel distance of Column 1 (dry) and Column 2 (wet).

(0.25–0.59). Moreover, the dispersivity values in both columns increase with the travel distance due to the scale-dependent effect. Moreover, dispersivity of sandy loam is the important factor on the movement of the BTCs and depends on the travel distance and water content.

Acknowledgements

The researchers would like to thank the Inter – Department of Environmental Science, the 90th Anniversary of the Chulalongkorn University Fund, the Office of Higher Education Commission (OHEC) and the S&T Postgraduate Education and Research Development Office (PERDO) for the financial support of the Research Program and thank the Ratchadaphiseksomphot Endowment Fund, Chulalongkorn University for the Research Unit and the

Ratchadaphiseksomphot Endowment Fund 2013 of Chulalongkorn University (CU-56-901-FW).

References

- [1] Dontsova KM, Yost SL, Simunek J, Pennington JC, Williford CW. Dissolution and transport of TNT, RDX, and Composition B in saturated soil columns. *J Environ Qual* 2006;35:2043–54.
- [2] Hrapovic L, Sleep BE, Major DJ, Hood ED. Laboratory study of treatment of trichloroethene by chemical oxidation followed by bioremediation. *Environ Sci Technol* 2005;39:2888–97.
- [3] Bear J. *Dynamics of Fluids in Porous Media*. New York: Elsevier; 1972.
- [4] Dagan G. Solute transport in heterogeneous porous formations. *J Fluid Mech* 1984;145:151–77.
- [5] Sudicky EA. A natural gradient experiment on solute transport in a sand aquifer: spatial variability of hydraulic conductivity and its role in the dispersion process. *Water Resour Res* 1986;22:2069–82.
- [6] Chotpantarat S, Limpakanwech C, Siriwong W, Siripattanakul S, Sutthirat C. Effects of soil water characteristic curves on simulation of nitrate vertical transport in a Thai agricultural soil. *Sustain Environ Res* 2011;21:187–93.
- [7] Chotpantarat S, Sutthirat C. Different sorption approaches and leachate fluxes affecting on Mn^{2+} transport through lateritic aquifer. *Am J Environ Sci* 2011;7: 57–64.
- [8] Nielsen DR, Vangenuchten MT, Biggar JW. Water flow and solute transport processes in the unsaturated zone. *Water Resour Res* 1986;22:89–108.
- [9] Radcliffe DE, Simunek J. *Soil Physics with HYDRUS: Modeling and Applications*. Boca Raton, FL: CRC Press; 2010.
- [10] Porro I, Wierenga PJ, Hills RG. Solute transport through large uniform and layered soil columns. *Water Resour Res* 1993;29:1321–30.
- [11] Huang K, Toride N, Vangenuchten MT. Experimental investigation of solute transport in large, homogeneous and heterogeneous, saturated soil columns. *Transp Porous Med* 1995;18:283–302.
- [12] Neuman SP. Universal scaling of hydraulic conductivities and dispersivities in geologic media. *Water Resour Res* 1990;26:1749–58.
- [13] Chou PY, Wyseure G. Hydrodynamic dispersion characteristics of lateral inflow into a river tested by a laboratory model. *Hydrol Earth Syst S. C* 2009;13:217–28.