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### Recycling and the Hydrological Cycle

Banal as it may seem, the starting point of this paper is the hydrological cycle with its component processes of (i) interception of rainfall, followed by evaporation of what has been intercepted by the plant canopy; (ii) infiltration of the "net" rainfall (consisting of stemflow and throughfall) within the soil profile, whence it is eventually returned either to the atmosphere or to rivers as runoff; (iii) evapotranspiration of water either directly from forest leaf litter, or from the stomata of leaves following uptake by plant roots; (iv) runoff, regarded as the difference between net rainfall, evapotranspiration and change in soil moisture storage. By virtue of interception losses or evapotranspiration, water is returned to the atmosphere whence it came; if, within the extent of the tropical forest, it is reprecipitated once or more times, then recycling of water will have occurred. Studies of recycling mechanisms are particularly difficult since they require consideration of the movement of water not only in its liquid phase, but in its vapour phase also: the provenance, as well as the magnitude, of components of the hydrological cycle must be considered.

The purpose of this paper is to put forward some methods, as yet untried because the authors have had no access to data with which to test them, which take account of the probabilistic nature of the movement of water particles within a tropical forest. As the methods are here presented, many changes will no doubt be necessary before they can be considered as useful for the study of recycling processes, and our purpose in presenting them is to seek views regarding where the assumptions made in our essentially statistical approach to the problem are untenable, and regarding what alternative assumptions would be more appropriate.

### A Historical Perspective

One of the earliest studies of the recycling of rainfall from tropical rain forest is that by Bernard (1945) as described by Penman (1963). Bernard discussed the interaction of forest and rainfall in the Central Congo Basin, an area of about 1 million km<sup>2</sup>, mostly

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between 300 and 500 m altitude; the basin has a central region of swampy tropical jungle which gradually changes to savannah-type country away from the centre. On a rainfall map of the basin, the physical boundary of the swampy forest coincides with the isohyetal line of 1600 mm rainfall per annum. Beyond the savannah to the north, east and south there are dry lands, and on these sides distances to the nearest oceans are very large; to the west and southwest, however, the Atlantic Ocean is only 1100 km away, and Bernard regarded this as being the only external source of water vapour supplying the basin. The Atlantic was also regarded as a sink for runoff. Penman quotes average values of annual water transfer as: rainfall 1510 mm; runoff 337 mm; evaporation 1173 mm. This cycle Bernard divided into two components, "a device that may be valid for the tropics where the rainfall has an intensely local character, but which will be strongly challenged for extra-tropical areas" (quoted from Penman, 1963). Bernard suggested that the Congo has an interior cycle of 1173 mm of evaporation which supplies vapour for (on average) 1173 mm of rainfall, together with an external cycle in which 337 mm of rain are produced from water vapour from the Atlantic, returning to the Atlantic as runoff. According to Bernard's estimate, therefore, about 78% of rainfall on the Central Congo is recycled. Using Bernard's data shown in Table 1, Penman argued as follows: except where rainfall is obviously limiting, the mean annual evaporation is very nearly constant and independent of the type of vegetation in spite of a wide variation of rainfall, suggesting that the amount of rain determines the type of vegetation cover rather than the converse. Bernard and Penman arrived at the same conclusion by rather different arguments: namely, that the thick jungle inside the 1600 mm isohyet is a consequence of the high rainfall, and that if the trees were removed the evaporation would be maintained from swamps and low vegetation. The internal cycle, they argued, would be unaffected.

Bernard's early estimate of 78%, as the percentage of Central Congo rainfall that is recycled, must be compared with the value quoted by Villa Nova et al. (1976) for the Amazon basin, a vast plain of some 6 million km<sup>2</sup> with an altitude difference of only 120 m over a range of 3400 km (Salati, Dall'Olio, Matsui and Gat, 1979). Of the  $12 \times 10^{12}$  m<sup>3</sup> year<sup>-1</sup> of water falling on the Brazilian Amazon, runoff amounts to  $5.5 \times 10^{12}$  m<sup>3</sup> year<sup>-1</sup> and evapotranspiration to  $6.48 \times 10^{12}$  m<sup>3</sup> year<sup>-1</sup>; evapotranspiration therefore amounts to 54% of precipitation. Klinge et al. (1981) state that this figure has been recently confirmed by a study of the water budget of a tropical rainforest in Amazonian Venezuela (1979).

Authors of the papers so far cited had estimates of rainfall, runoff and evapotranspiration losses; they had no stable isotope data, and made no description of the movement of water, whether in vapour or liquid phases, above and within the basins considered. Deterministic models of these processes have been described by Eriksson (1965) and more recently by Merlivat & Jouzel (1979), whilst a simple model of isotopic fractionation of water in the Amazon basin has been given by Dall'Olio et al. (1979). These authors divided the Amazon region between 0° and 5°S into eight adjacent three-degree strips, the divisions being along lines of longitude 48°30', 51°30', ... 72°30'. Water

vapour from the Atlantic Ocean enters the most easterly strip; part of this input (determined from radiosonde measurements at Belém) falls as rain on the strip itself, whilst the remainder, augmented by evaporation from the first strip, passes to the next (second) strip to the west, where a similar budget is calculated. The budget is also calculated for the third, fourth ... strip successively. The model operates with a monthly time-step, and all water vapour appears to move westwards; because molecules of  $H_2^{18}O$  are more massive than those of  $H_2^{16}O$  they are precipitated from water vapour sooner and are evaporated from liquid water later, and the model by Dall'Olio et al. predicts the depletion of  $H_2^{18}O$  with distance from the Ocean.

### Stochastic Models of Water Recycling

To begin with, we restrict attention to but one species of water molecule, say  $H_2^{16}O$ . Once any one particular molecule has entered the Amazon basin, its subsequent fate shows probabilistic characteristics in that (a) the molecule is precipitated as rain after a certain interval of time  $t_0$ , after which, provided it remains within the basin, it will be evaporated following a further time interval  $t_1$ ; (b) its macroscale spatial movement within the basin may be described as erratic. With reference to (a) the quantities  $t_0$  and  $t_1$  (supplemented, if recycling occurs, by  $t_2, t_3, t_4 \dots$ ) may be considered as random variables, with the times  $t_0, t_2, t_4 \dots$  spent in the vapour phase having one probability distribution, whilst the times  $t_1, t_3, t_5 \dots$  spent in the liquid phase have another probability distribution. Moreover, since the intervals spent in liquid and vapour phases alternate, the history of any one particular molecule may be regarded as a realization of an **alternating renewal process** (Cox & Miller, 1970). Since molecules of different isotopic composition must be considered, two probability distributions (one of times spent in vapour phase, one of time spent in liquid phase) must be used for  $H_2^{16}O$ , two distributions for  $H_2^{18}O$ , two for  $HD^{16}O$ . With reference to (b) above, the spatial movement within the basin of a particular molecule in the vapour phase may be considered as a generalization of the familiar **random walk**; regarded as a stochastic process, the movement will be (i) two dimensional (since movement must be allowed for in both the north-south and east-west directions); (ii) with drift (since the probability of a westward (north-ward) step will not, in general, equal that for an eastward (south-ward) step. Moreover, a limiting process of the random walk model must be considered in which the magnitudes of steps become very small (tend to zero); in statistical terminology, we regard the spatial movement of any one particular molecule as being a **diffusion process with drift** (Cox & Miller, 1970). Just as for the alternating renewal process, described above, it is necessary to consider one diffusion process for each species of water molecule considered; parameters describing the drift may, as a first approximation, be related to the direction and velocity of prevailing wind. Other simplification will also be necessary; for example, the diffusion equation to be solved for the diffusion process model must be solved for certain prescribed boundary conditions, and the solution becomes more straight forward if the basin can be considered as having infinite extent.

The stochastic models outlined in this paper are based on the two component models described above: namely (a) the alternating renewal process for the times spent in vapour and liquid phases, and (b) the diffusion process for the movement of molecules in the vapour phase (possibly, another diffusion process will be necessary to define movement of water in its liquid phase, once it enters river channels). Both the alternating renewal process (a) and the diffusion process (b) will be required for each species of water molecule having different isotopic composition.

It is hoped that this paper demonstrates how the apparent foolishness of working on a molecular scale can be reconciled with the requirements of the problem at hand: namely to describe the recycling of water within the catchment area of the largest river basin on earth. To begin with, we consider how one particular model that has been widely used in the study of isotopic fractionation processes - namely the Rayleigh model - can be considered as a description of a stochastic process.

### The Rayleigh Model as a Stochastic Process

Suppose that at time zero there are  $N$  molecules of a particular isotopic species present in a parcel of water vapour, and that the probability that any one molecule is precipitated in a small time interval  $\Delta t$  is  $\alpha \Delta t$ , where  $\alpha$  is a constant. Let  $p(t)$  be the probability that a particular molecule has not been precipitated by time  $t$ ; then by the multiplication law for probabilities of independent events,  $p(t + \Delta t) = (1 - \alpha \Delta t) p(t)$  or  $[p(t + \Delta t) - p(t)] / \Delta t = \alpha p(t)$ .

Taking the limit as  $\Delta t \rightarrow 0$ ,  $dp(t)/dt = -\alpha p(t)$  whence  $p(t) = e^{-\alpha t}$  (assuming  $p(0) = 1$ ).

With  $N$  molecules initially present, and with the probability that any one of them has not been precipitated at time  $t$  being  $e^{-\alpha t}$ , then the distribution of  $K$ , the total number of molecules not precipitated at time  $t$  is

$$P(K) = \frac{N!}{(N-K)!K!} e^{-\alpha t K} (1 - e^{-\alpha t})^{N-K}$$

assuming that the probability of each molecule is independent of the others. This is a binomial probability law with mean value

$$N e^{-\alpha t} \quad (1)$$

Now consider the case where initially there are two isotopic species, one with  $N_1^0$  molecules, and the other with  $N_2^0$  molecules present (indices denote time, suffices the isotopic species). At time  $t > 0$ , the expected numbers of each species that are still present in vapour form are given by

$$N_1^t = N_1^0 \exp(-\alpha_1 t)$$

$$N_2^t = N_2^0 \exp(-\alpha_2 t)$$

where the  $\alpha_1$  and  $\alpha_2$  will be different for the two species. These two relations give, on eliminating  $t$ , the Rayleigh law in the form

$$\left( \frac{N_1^t}{N_1^0} \right) = \left( \frac{N_2^t}{N_2^0} \right)^{-\alpha_1/\alpha_2} \quad \text{or} \quad \left| \left( \frac{N_1^t}{N_2^t} \right) / \left( \frac{N_1^0}{N_2^0} \right) \right| = \left( \frac{N_2^t}{N_2^0} \right)^{\alpha_1/\alpha_2 - 1} \quad (2)$$

An alternative derivation is as follows. Let  $\Pi_i(t)$  be the probability that  $i$  molecules of particular isotopic species, out of an original  $N$ , still exist in vapour form at time  $t$ ; then

$$\begin{aligned}\Pi_i(t+\Delta t) &= \Pi_i(t) |1-\alpha\Delta t|^i + \Pi_{i+1}(t) (i+1) \alpha\Delta t |1-\alpha\Delta t|^{i-1} \\ &= \Pi_i(t) |1-\alpha\Delta t+0(\Delta t)^2|^i + \Pi_{i+1}(t) (i+1) \alpha\Delta t |1-0(\Delta t)|^{i-1}\end{aligned}$$

giving, as  $\Delta t \rightarrow 0$ ,

$$d\Pi_i(t)/dt = -\alpha i\Pi_i(t) + \alpha(i+1)\Pi_{i+1}(t). \quad (3)$$

Denoting the probability generating function of  $\Pi_i(t)$  by  $G(z,t)$ , so that

$$G(z,t) = \sum_{i=0}^{\infty} \Pi_i(t) z^i$$

we have from equation (3)

$$\frac{\partial G(z,t)}{\partial t} = (1-z)\alpha \frac{\partial G(z,t)}{\partial t}.$$

This has general solution

$$G(z,t) = f(e^{-\alpha t}(z-1))$$

where  $f(\cdot)$  is one arbitrary function determined by the boundary condition. For  $N$  molecules initially,

$$G(z,t) = \{1+(z-1)e^{-\alpha t}\}^N$$

which is the probability generating function of a binomial probability density function with mean  $Ne^{-\alpha t}$ ; this corresponds to the derivation leading to equation (1), and the rest of the argument, leading to the Rayleigh expression of equation (2), is identical.

#### An "input-output" model describing $^{18}O/^{16}O$ ratios for the Brazilian Amazon

As a further example of a stochastic model, we consider one which uses data on  $^{18}O/^{16}O$  ratios and which, in theory, can give estimates of the proportion of water that is recycled within the Brazilian Amazon. This model is unsatisfactory for a number of reasons, amongst which is its inability to make use of the data on spatial variability of  $^{18}O/^{16}O$  ratios within the basin itself; it uses only

a) The radiosonde measurements of wind and humidity which are available for Belém, Manaus and Vilhenna. Salati et al. (1979) stated that these are insufficient for quantifying the moisture flux; however, following other workers (Dall'Olio et al. 1979), who used only the Belém radiosonde data, we recognize this as a deficiency which need not represent too great a shortcoming for an initial exploratory model. "The isotopic composition of the moisture flux is calculated by taking the mean value for precipitation at the ocean side stations of Belém, Amapá, and three stations on the Marajó Island (São Sebastião, Santa Cruz do Arari and Cachoeira do Arari" (Salati et al. 1979).

b) The  $^{18}O/^{16}O$  ratio in discharge from the Amazon. "The discharge was sampled near Water recycling in ...

its mouth in the 'Canal Sul' north of the Island of Marajó (Salati et al. 1979) for the determination of  $^{18}\text{O}/^{16}\text{O}$ . Estimates of discharge are also available at Óbidos (Salati et al. 1979).

Thus, monthly estimates of  $^{18}\text{O}/^{16}\text{O}$  (and, incidentally, of D/H, the data for which may be treated in the same manner as that described below) are available for (a) the vapour input from the Atlantic Ocean, together with its isotopic composition, and (b) the discharge from the Amazon, together with its isotopic composition.

For the **structure of the model**, we begin by assuming that any water molecule (whether it be  $\text{HD}^{16}\text{O}$ ,  $\text{H}_2^{16}\text{O}$ , or  $\text{H}_2^{18}\text{O}$ ), can exist within the basin in any one of the following three states:

1. the vapour phase within the atmosphere;
2. in the liquid phase as precipitated water available for evaporation back to the atmosphere;
3. as liquid water removed from the basin as runoff.

Transitions between these three states can occur; if a molecule is in state 1 (as water vapour), then we assume that the probability that it changes to state 2 (as precipitated water) within a short interval of time  $\Delta t$ , is  $\alpha_i \Delta t$ . If a molecule is in state 2, it may transfer back to state 1 with probability  $\beta_i \Delta t$ , or to state 3 (runoff) with probability  $\gamma_i \Delta t$ . We assume that no direct transition from state 1 to state 3 is possible, and that no transition from state 3 to either of states 1 or 2 is possible. For any molecule of isotopic composition  $i$ , therefore, the matrix of transition probabilities may be written as follows:-

		1	2	3
State at time	1	$1 - \alpha_i \Delta t$	$\alpha_i \Delta t$	0
$t - \Delta t$ :	2	$\beta_i \Delta t$	$1 - \beta_i \Delta t - \gamma_i \Delta t$	$\gamma_i \Delta t$
	3	0	0	1

where the parameters  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$  will differ for molecules of the different isotopic compositions  $\text{HD}^{16}\text{O}$ ,  $\text{H}_2^{16}\text{O}$ ,  $\text{H}_2^{18}\text{O}$ . For these three isotopic compositions, therefore, 9 parameters (3  $\alpha$ 's, 3  $\beta$ 's, 3  $\gamma$ 's) are required to define the model.

**Formulation of the model equations** then proceeds as follows. Since we consider separately the isotopic species defined by parameters  $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$ , we drop the suffices  $i$ ; the probability that a particular molecule of a given isotopic species is in state 1 at time  $t + \Delta t$ , denoted by  $P(1, t + \Delta t)$ , is related to  $P(1, t)$  and  $P(2, t)$  by the equation

$$P(1, t + \Delta t) = (1 - \alpha \Delta t)P(1, t) + \beta \Delta t P(2, t)$$

which, following a Taylor expansion, gives

$$dP(1, t)/dt = -\alpha P(1, t) + \beta P(2, t).$$

Similarly

$$dP(2, t) / dt = \alpha P(1, t) - (\beta + \gamma) P(2, t)$$

$$dP(3, t) / dt = \gamma P(2, t)$$

and, in matrix notation

$$\frac{d\underline{P}}{dt} = \begin{vmatrix} -\alpha & \beta & 0 \\ \alpha & -(\beta + \gamma) & 0 \\ 0 & \gamma & 0 \end{vmatrix} \underline{P}$$

where  $\underline{P}$  is the vector  $[P(1, t), P(2, t), P(3, t)]^T$ . The solution of this equation is

$$P(1, t) = (\lambda_2 - \lambda_1)^{-1} \{ (\lambda_2 + \alpha) e^{\lambda_1 t} - (\lambda_1 + \alpha) e^{\lambda_2 t} \} \quad (5a)$$

$$P(2, t) = \alpha (\lambda_2 - \lambda_1)^{-1} \{ e^{\lambda_2 t} - e^{\lambda_1 t} \} \quad (5b)$$

$$P(3, t) = \gamma (\lambda_2 - \lambda_1)^{-1} \{ \lambda_2 e^{\lambda_1 t} - \lambda_1 e^{\lambda_2 t} \} \quad (5c)$$

where  $\lambda_1, \lambda_2$  are the roots of the equation  $\lambda^2 + \lambda(\alpha + \beta + \gamma) + \alpha\gamma = 0$ . If the initial time  $t_0$ , is not zero, then  $t - t_0$  must be written instead of  $t$  in this solution.

A solution of this form can be calculated for each of the isotopic combinations  $HD^{16}O, H_2^{16}O, H_2^{18}O$ . The solution gives the probabilities that a molecule of (for example)  $HD^{16}O$ , entering the basin as vapour at time  $t_0$  is either (i) vapour within the basin, (ii) precipitated water within the basin, or (iii) emerging as runoff at a later time  $t$ .

We are now faced with the problem of estimating the parameters  $\alpha, \beta, \gamma$  for each isotopic species.

To begin with, we assume that the input (radiosonde) data can be used to give estimates of the "instantaneous influxes" (or, more precisely, the fluxes at discrete timesteps) of  $HD^{16}O, H_2^{16}O, H_2^{18}O$ ; call these inputs  $I_1(t), I_2(t), I_3(t)$ . Similarly, suppose that the output (estimated discharge) data give fluxes  $O_1(t), O_2(t), O_3(t)$ . Then the solution of equations (4), represented by (5), gives the probabilities that a molecule of  $HD^{16}O$  is in states 1, 2 or 3 at time  $t$ , given that it entered the basin at time  $t_0$ . To relate the output  $O_i(t)$  to the input  $I_i(t)$ , however, we need the probability that the molecules enters state 3 (from which, of course, it cannot return) in the interval  $(t - \Delta t, t)$ ; this must therefore be equal to the probability that the molecule is in state 2 at time  $t - \Delta t$ , multiplied by the probability that there is a transition from state 2 to state 3 in the interval  $(t - \Delta t, t)$ . The probability that the molecule enters state 3 in  $(t - \Delta t, t)$  is therefore

$$\gamma \Delta t \cdot P_i(2, t) \quad (6)$$

where the suffix  $i$  applies to isotopic species  $i$ . The relation between  $O_i(t)$  and  $I_i(t)$  must therefore be

$$O_i(t) = \gamma \int_{t_0=0}^{t_0=\infty} P_i(2, t_0) I_i(t-t_0) dt_0. \quad (7)$$

Equation (7) is the convolution of  $P_i(\cdot)$  with  $I_i(\cdot)$ , allowing for the fact that the output at time  $t$  can be made up of components which entered the basin at all earlier times  $t-t_0$ , and which only effect the final transition to runoff (state 3) at time  $t$ . In equation (7), the quantity  $P_i(2, t_0)$  is a function of the parameters  $\alpha_i, \beta_i, \gamma_i$ ; since we are assuming that the inputs  $I_i(d)$  and outputs  $O_i(t)$  are known from data, the parameters  $\alpha_i, \beta_i, \gamma_i$  can, in theory, be estimated by minimization of

$$\int_{\tau} \{O_i(\tau) - \gamma \int_{t_0=0}^{t_0=\infty} P_i(2, t_0) I_i(\tau-t_0) dt_0\}^2 d\tau \quad (i = 1, 2, 3)$$

with respect to  $\alpha_i, \beta_i, \gamma_i$ . This estimation procedure can be readily adapted to allow parameter estimation from the data on  $^{18}O/^{16}O$  ratios directly.

We must not lose sight of the fact that the purpose of the model is to assess **recirculation of water vapour within the Amazon Basin**. When  $\alpha_i, \beta_i, \gamma_i$  have been estimated, we are now in a position to examine the recirculation of molecules within the basin. For a molecule of isotopic composition  $i$ , the probability that it passes from state 1 (water vapour) to state 2 (precipitated water) and thence to state 3 (runoff leaving the basin) **without** recirculation is

$$\gamma_i / (\beta_i + \alpha_i). \quad (8)$$

The probability that the molecule passes from state 1 to state 2 with one or more transitions back to state 1 before emerging as runoff (state 3) is therefore

$$1 - \gamma_i / (\beta_i + \alpha_i). \quad (9)$$

As a by-product of the fitting procedure that leads to the estimation  $\alpha_i, \beta_i, \gamma_i$ , standard errors of the estimates  $\hat{\alpha}_i, \hat{\beta}_i, \hat{\gamma}_i$ , can be calculated.

#### A Model For Isotopic Movement Within the Brazilian Amazon

The model of the last section uses only 'input' and 'output' data, and made no use of records of isotopic ratios in rainfall at the 19 sites within the basin; we now proceed to outline a model which includes not only the 'alternating renewal process' component, but also the 'diffusion with drift' component described earlier.

As before, the external source of water vapour is taken to be the Atlantic Ocean, and we assume a point source  $X_0 \equiv (x_0, y_0)$  for all vapour, which may be taken as Belém; this assumption simplifies the development of the model, but can be dispensed with. Whilst a particular water molecule is in the atmosphere (state 1), it can move in a



manner which will be extremely complex in that it includes vertical motion within particular storms; we take no account of this, but assume that molecules move two-dimensionally in a 'random' manner with the velocity and direction of drift being determined by prevailing wind. The probability distribution of the position  $\underline{X} \equiv (x, y)$  of a molecule starting at  $\underline{X}_0$  and having been in the atmosphere for a length of time  $t$  is taken as Gaussian with mean  $\underline{X}_0 + \underline{\mu}t$  and variance  $\underline{\Sigma}t$ ; here the parameter  $\underline{\mu}$  is a two-dimensional vector which determines the direction and velocity of drift, and  $\underline{\Sigma}$  is a  $2 \times 2$  matrix associated with the variability of molecular movement.

As in the last section, whilst a molecule is in the atmosphere (state 1) there is the probability of a transition to state 2 (as precipitated water) which, for a time interval  $\Delta t$ , is  $\alpha \Delta t$ . Similarly, while in state 2, as precipitated water available for re-evaporation, there is a probability  $\beta \Delta t$  that a molecule effects the transition back to vapour by evapotranspiration, and probability  $\gamma \Delta t$  that it is removed as runoff. The parameters  $\alpha, \beta, \gamma$  will differ for the different isotopic species  $\text{HD}^{16}\text{O}, \text{H}_2^{16}\text{O}, \text{H}_2^{18}\text{O}$ . The model of the last section can also be developed to show that the time that a molecule spends in state 1 before its transition to state 2 (precipitated water) has a simple exponential distribution  $p_1(t)$ , where

$$p_1(t) = \alpha \exp(-\alpha t). \quad (10)$$

Similarly, the distribution  $p_2(t)$  of the time that a molecule spends in state 2, having entered it, is

$$p_2(t) = (\beta + \alpha) \exp(-(\beta + \gamma)t). \quad (11)$$

From these results, we can find the probability distribution of the time spent in the atmosphere before the  $i$ th occasion on which a water molecule precipitates; this is the Gamma distribution  $G_v(\alpha, i)$ , where

$$G_v(\alpha, i) = \alpha^i \exp(-v\alpha) v^{i-1} / \Gamma(i). \quad (12)$$

Using this result, and the assumptions made about the movement of water molecules within the atmosphere, it can be shown that the distribution of the position at which the molecule is precipitated for the  $i$ th time is

$$p(\underline{X}, i) = \int_{v=0}^{\infty} N_X(\underline{X}_0 + \underline{\mu}v, \underline{\Sigma}v) G_v(\alpha, i) dv \quad (13)$$

where  $\underline{X} \equiv (x, y)$  are the coordinates of position, and  $N_X(\cdot)$  is the bivariate Gaussian distribution given by

$$N_X(\underline{X}_0 + \underline{\mu}t, \underline{\Sigma}t) = \{2\pi t \sigma_{xx} \sigma_{yy} \sqrt{1 - \rho_{xy}^2}\}^{-1} \exp\{-1/(2(1 - \rho_{xy}^2)) [\theta^2 / \sigma_{xx}^2 t - 2\rho_{xy} \theta \phi / \sigma_{xx} \sigma_{yy} t + \phi^2 / \sigma_{yy}^2 t]\}$$

where  $\theta = x - x_0 - \mu_x t$ ,  $\phi = y - y_0 - \mu_y t$ ,

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$$p(\underline{X}, i) = \int_{v=0}^{\infty} N_X(\underline{X}_0 + \underline{\mu}v, \underline{\Sigma}v) G_V(\alpha, i) dv \quad (13)$$

where  $\underline{X} \equiv (x, y)$  are the coordinates of position, and  $N_X(\cdot)$  is the bivariate Gaussian distribution given by

$$N_X(\underline{X}_0 + \underline{\mu}t, \underline{\Sigma}t) = \{2\pi t \sigma_{xx} \sigma_{yy} \sqrt{1 - \rho_{xy}^2}\}^{-1} \times \exp\{-1/(2(1 - \rho_{xy}^2)) [\theta^2/\sigma_{xx}^2 t - 2\rho_{xy} \theta \phi / \sigma_{xx} \sigma_{yy} t + \phi^2/\sigma_{yy}^2 t]\}$$

where  $\theta = x - x_0 - \mu_x t$ ,  $\phi = y - y_0 - \mu_y t$ ,

$$\underline{\Sigma} = \begin{Bmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yx} & \sigma_{yy} \end{Bmatrix}$$

and  $\rho_{xy}^2 = \sigma_{xy}^2 / \sigma_{xx} \sigma_{yy}$ .

Allowing for the probability that, for  $i > 1$ , the molecule may not be precipitated on more than  $(i-1)$  occasions, and using the assumption that the mean number of molecules entering the basin from the Atlantic Ocean in a short time interval  $\Delta t = N^0 \Delta t$ , then the mean number of molecules precipitated in an area  $|x_1, x_2; y_1, y_2|$  in a period of time  $T_2 - T_1$  can be shown to be

$$(T_2 - T_1) N^0 (\beta + \gamma) / \gamma \int_{x_1}^{x_2} \int_{y_1}^{y_2} p(\underline{X}) \, dx dy \quad (14)$$

where

$$\begin{aligned} p(\underline{X}) &= \sum_{i=1}^{\infty} p(\underline{X}, i). \text{ prob (molecule precipitates on exactly } i \text{ occasions)} \\ &= \sum_{i=1}^{\infty} p(\underline{X}, i) \{ \beta / (\beta + \gamma) \}^{i-1} \{ \gamma / (\beta + \gamma) \}. \end{aligned}$$

This can be shown to be

$$p(\underline{X}) = \int_{v=0}^{\infty} N_X(\underline{X}_0 + \underline{v}, \underline{\Sigma v}) E_v(\alpha \gamma / (\beta + \gamma)) \, dv \quad (15)$$

In which

$$E_v(\alpha \gamma / (\beta + \gamma)) = \alpha \gamma / (\beta + \gamma) \exp\{-\alpha \gamma v / (\beta + \gamma)\}.$$

Subject to the assumptions made, equation (14) gives the distribution of rainfall over the basin. It is possible to write the expression for  $p(\underline{X})$  in (15) in terms of standard mathematical functions, and the integration required for the evaluation of (14) may be performed numerically.

An expression for the distribution of evapotranspiration of molecules of a particular isotopic species over the basin can be found using similar arguments to those put forward when deriving the distribution of precipitation. A water molecule evaporating for an  $i$ th time must have been precipitated for an  $i$ th time, after which it is potentially available for evapotranspirations at the site where it fell; the distribution of the time spent in the atmosphere is therefore the same as that derived for the  $i$ th occasion on which the molecule precipitated, namely

$$G_v(\alpha, i) = \alpha^i \exp(-\alpha v) v^{i-1} / \Gamma(i).$$

The distribution of the position at which a water molecule evaporates for the  $i$ th

time is likewise the same as that computed for the  $i$ th precipitation, that is

$$p(\underline{X}, i) = \int_{v=0}^{\infty} N_X(\underline{X}_0 + \underline{\mu}v, \underline{\Sigma}v) G_V(\alpha, i) dv.$$

Since the probability that a molecule evaporates exactly  $i$  times, given that  $i$  is at least one, is

$$\{\beta/(\beta+\gamma)\}^i \cdot \{\gamma/\beta\},$$

the distribution of the position of a water molecule on the last occasion on which it evaporates is

$$\begin{aligned} p(\underline{X}) &= \sum_{i=1}^{\infty} p(\underline{X}, i) \cdot \{\beta/(\beta+\gamma)\}^i \{\gamma/\beta\} \\ &= \int_{v=0}^{\infty} N_X(\underline{X}_0 + \underline{\mu}v, \underline{\Sigma}v) E_V(\alpha\gamma/(\beta+\gamma)) dv. \end{aligned}$$

This is identical to the equivalent distribution for precipitation. Using this result, it is easy to show that the expected number of water molecules evaporating in an area  $\{x_1, x_2, y_1, y_2\}$  in a period of time  $T_2 - T_1$  is

$$(T_2 - T_1) N^0 (\beta/\gamma) \int_{x_1}^{x_2} \int_{y_1}^{y_2} p(\underline{X}) dx dy.$$

It should be noted that the difference between the expected rainfall and the expected evapotranspiration is

$$(T_2 - T_1) N^0 \int_{x_1}^{x_2} \int_{y_1}^{y_2} p(\underline{X}) dx dy$$

which is the expected number of molecules entering the river in the area  $\{x_1, x_2; y_1, y_2\}$  and time interval  $T_2 - T_1$ .

The above development, leading to equation (14), has assumed that all molecules are identical; it is possible, however, to treat each isotopic combination  $HD^{16}O, H_2^{18}O, H_2^{16}O$  separately, thus deriving expressions such as (14) in which the parameters  $\alpha, \beta, \gamma, \underline{\Sigma}, \underline{\mu}$  depend on the isotopic combination considered. It is then easy to show, for example that the ratio of the numbers of molecules of two isotopic species at any time instant  $T$  is

$$\frac{N_1^T}{N_2^T} = \frac{N_1^0}{N_2^0} \cdot \frac{(\beta_1 + \gamma_1)}{\gamma_1} \cdot \frac{\gamma_2}{(\beta_2 + \gamma_2)} \cdot \frac{p_1(\underline{X})}{p_2(\underline{X})} \quad (16)$$

Here  $\underline{X}$  is the point with coordinates  $(x, y)$  as before, and the subscripts 1, 2 denote quantities associated with molecules of isotopic species 1 and 2 respectively. There is clearly an analogy between the expression (16) and the expression (2) above for the water recycling in ...

Rayleigh model.

### Fitting the Model for Isotopic Movement to the Brazilian Amazon Data

The model of the last section, capable as it is of utilizing the available data on the isotopic composition of rainfall at the 19 stations described by Villa Nova *et al.* (1976), gives predictions of observable quantities; mainly, the  $^{18}\text{O}/^{16}\text{O}$  ratio in rainfall month by month. These predictions depend on the values of certain parameters, some of which ( $\alpha_i$ ,  $\beta_i$ ,  $\gamma_i$ , for example) are totally unknown, whilst others (such as  $\mu_x$ ,  $\mu_y$ ) have some physical interpretation and can perhaps be estimated independently. The model parameters may be estimated - that is to say, the model may be fitted - by choosing some criterion by which its predictions are similar to the observable quantities, subject to any necessary constraints on the parameter values. Least squares is an example of one such fitting procedure.

At the present stage, more work must be done on the model before it could be considered a useful tool for the study of recycling processes within the Amazon. An example of its inadequacy is given by the fact that unless parameter values are allowed to vary seasonally, the mean rainfall at any given point is constant from month to month, whereas the rainfall in some parts of the basin is strongly seasonal; thus at Alto Tapajos ( $7^{\circ} 20' \text{S}$ ,  $57^{\circ} 30' \text{W}$ ) the mean July rainfall (1931-1960) is 11 mm, compared with 434 mm in March. Further changes to the model may be necessary to allow for changes in the origins of water vapour inputs and their magnitudes; similarly, allowance for water movement in river channels may be required, possibly by the superposition of another diffusion process.

Table 1. Water balance for natural regions of the Congo basin.

Dominant vegetation	River	From source to:	Rain	Water (mm) Runoff	Evap	E/R%
Wet tropical forest	<b>Fimi</b>	Mouth	1700	661	1039	61
Forest and savannah	Congo	Banana	1510	337	1173	78
Savannah with forest glades	Kasai	Kutu-Moke	1371	289	1082	79
Dry savannah	{ Lualaba	Nzilo	1100	186	914	83
	{ Lufira	Chute Cornet	1172	135	1037	88
	{ Lukuga	Deversoir	850	29	821	97

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