Use of natural isotopes and groundwater quality for improved recharge and flow estimates in dolomitic aquifers

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Abstract

The application of a model to several dolomitic aquifers in the RSA is presented and has successfully simulated the reappearance of ^{14}C injected from nuclear tests in the discharge from springs. This is based on a new conceptual model, which accounts for the large variations of ^{14}C in the groundwater still representing recently recharged water. The input of ^{14}C is related to the recharge mechanism to yield low concentrations if the infiltration is direct; and higher concentrations if the recharge water interacts with biogenic CO_2 generated in the soil zone. The model has produced estimates of the recharge parameters and their controls but requires an independent estimate of the average recharge e.g. the chloride mass balance method. A close match has been obtained with ^{14}C measurements over the past three decades. A shallow component of the recharge mixes with a larger and older deep-water component. The model has provided the turn-over times of water in the system, which represent the storage capacity as multiples of the mean annual recharge. Quantitative estimates of the recharge of dolomitic aquifers could be derived from the bicarbonate concentrations of the spring waters.

The model has also been used to simulate the response of the limited temporal tritium measurements and single determinations of CFC for some springs. This has revealed significant differences that are related to additional dilution of the tritium tracer in the unsaturated zone, in comparison to ^{14}C and CFC.

Keywords: dolomite, karst, carbonate, aquifers, groundwater, model, carbon 14, ^{14}C, tritium, ^{3}H, CFC, recharge estimation, turnover time

Introduction

Background to the study

In view of their importance as significant groundwater resources the dolomitic aquifers of South Africa have been the focus of studies over many years (Enslin and Kriel, 1967; Fleisher, 1981; Taylor, 1983; Bredenkamp et al., (1985); Foster, 1988; Vegter and Foster, 1990; Simonic, 1993; Bredenkamp et al., 1994; Veltman, 2003; Stephens et al.,2004) A variety of methods have been applied to improve the assessment of the recharge and flow characteristics of these aquifers. The latest contribution involves the simulation of the ^{14}C pulse that had been injected into the atmosphere from thermonuclear tests and its reappearance in the spring discharge after a period of time. The ^{14}C content of several dolomitic springs has been measured at irregular intervals over more than 30 years. Previous attempts (Talma and Weaver, 2003) to simulate the breakthrough of the bomb ^{14}C have only been partially successful in view of:

- The difficulty to determine the initial ^{14}C concentration and account for the large variations in ^{14}C content of the recharge entering the aquifer
- Finding an acceptable mixing model for the water emanating from the spring.

According to the model the recharge comprises two components of different ^{14}C contents that are determined by a low- and a high-threshold rainfall value. Good simulations of ^{14}C have been achieved (Bredenkamp and Van Wyk, 2004).

The model has been further investigated to
- Determine the sensitivity of the model parameters
- Match the average recharge obtained from the model to the average recharge derived from the chloride mass balance method (CMB)
- Apply the model to simulate the response of the environmental tritium and CFC in groundwater
- Derive the age and turnover time of water in the aquifer, the latter representing the storage of groundwater as multiples of the average annual recharge. (Vogel, 1970; Bredenkamp and Vogel, 1970).

A WRC contract (K8/618) has been granted to further investigate the application of the method to dolomitic springs all over the country – see Fig. 1.

Rationale of the ^{14}C model

The new simulation model conforms to the lumped parameter approach (Maloszewski and Zuber , 1996) but incorporates a bi-modal recharge that could yield both low and high ^{14}C values, which still represent recent recharge (see Fig. 2). A two-box model has provided good results of the mixing occurring within these aquifers, without having to incorporate complex factors such as matrix flow, fracture flow, and dispersion of the tracer.

In the present ^{14}C simulation model the recharge of the dolomite comprises two components according to the conceptual model shown in Fig. 2 with differential uptake of ^{14}C. This yields both high and low ^{14}C content for recent recharge, which admixes with water flowing in the aquifer. Recharge in closer proximity to the spring mixes with the deeper flowing water, representing recharge that has occurred at greater distances from the spring outlet, which represents recharge from an earlier period. The ^{14}C content of the final mix therefore

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depends on the proportion of shallow water that mixes with a larger component of deeper water. However, the relative contributions of the shallow and deep components will always remain the same, as the recharge determines the groundwater levels and hydraulic head, which controls the fluctuating discharge of the springs.

Figure 1
Locality of dolomitic springs of the Central, North-Western and Eastern regions of the Republic of South Africa

Reliable recharge as reference

From a selection of available methods the chloride mass balance method representing the ratio of the rainfall chloride to that of the groundwater (CMB method - Beekman and Xu, 2003), is the best method to use, provided that no chloride is contributed by the geological formations or contamination. Refined measurement of the chloride in the rainfall has been initiated but needs to be continued over a longer period. The average recharge could be derived from the spring flows if the recharge area of the springs can be delineated reliably, and the chloride content of the rainfall could be inferred from the best match between the simulated chloride concentrations and the measured chloride of the spring water. These results have indicated that the relationship between chloride and rainfall (Bredenkamp et al., 1995), which has been based on reported measurements at the time, has overestimated the chloride concentration of the rainfall by about 40%, resulting in the same overestimation of the average recharge.
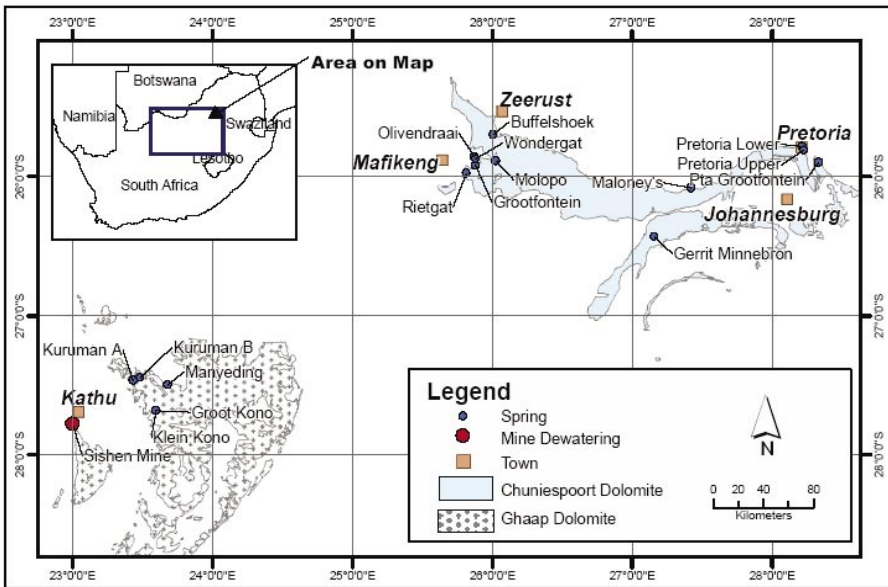
^{14}C simulation model (compiled in Excel)

Conceptual model

The rationale of the postulated bi-modal recharge is schematically presented in Fig. 2 and the recharge components of the model are set out below and have been adjusted so that the average recharge corresponds to independent estimates of the average recharge, e.g. from the CMB method:

- In addition to the 'normal' recharge (e.g. diffused flow through the soil zone), a recharge factor that operates during periods of high rainfall is introduced. The latter represents the more direct macro-pore recharge. This produces a gradual exponential increase in the recharge for increased precipitation.
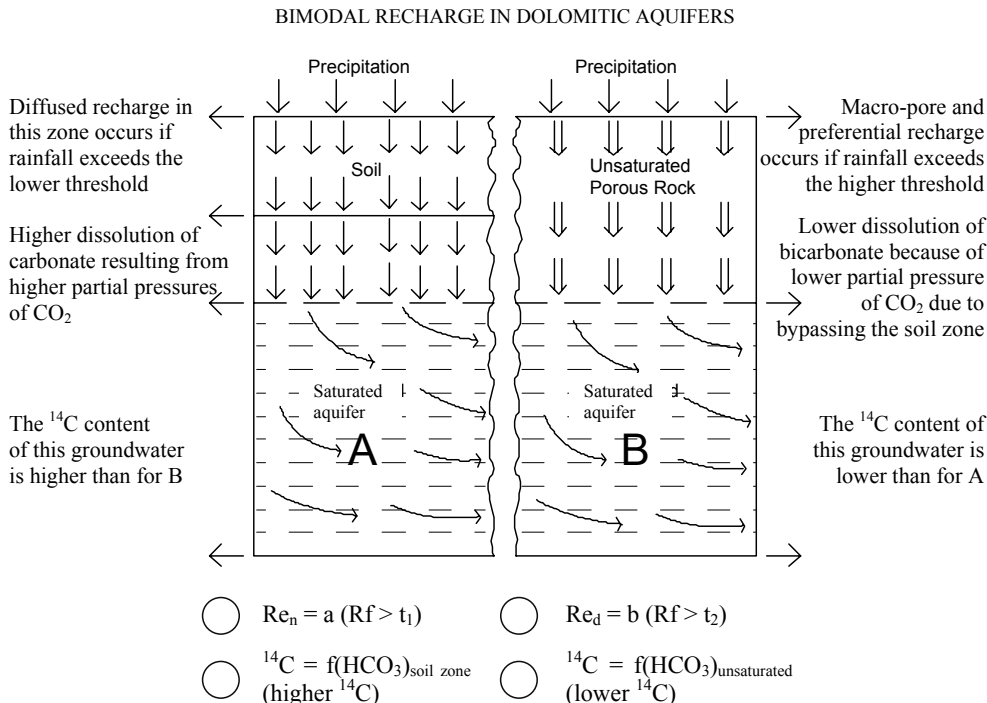
BIMODAL RECHARGE IN DOLOMITIC AQUIFERS

Figure 2
Schematic diagram of the bimodal ^{14}C input of the model that controls the input of ^{14}C derived from normal (Re_n) and direct (Re_d) recharge where t_1 and t_2 are the threshold rainfalls and a and b the recharge coefficients for the normal and direct recharge (see Eq.(1)

- A cut-off value to the monthly rainfall is applied to the 'normal' recharge component, while a significantly higher threshold applies to the high rainfall component. No water is contributed by the recharge component below these thresholds.
- Of the monthly excess rainfall only a fraction reaches the groundwater table - the rest is predominantly lost by evapotranspiration and runoff is negligible from these aquifers
- A further weighting factor is applied to the fractions. It increases/decreases as the average precipitation of preceding months is higher/lower than the long-term average and further contributes to the exponential response of recharge.
- The relative amount of recharge in a specific month is furthermore determined by the average of the excess rainfall values over a number of foregoing months.

The relative recharge of month i from the two components is thus given by:

$$R_i = \left(\frac{aw}{u} \sum_{i-u+1}^{i} (Rf_i > t_1) + \frac{bw}{v} \sum_{i-v+1}^{i} (Rf_i > t_2) \right) \qquad (1)$$

where:

$(Rf_i > t_1)$ and $(Rf_i > t_2)$ are the precipitation values for month i greater than t_1 and t_2, respectively

a and b are the respective fractions of the rainfall that reaches the saturated zone

$w = \frac{1}{x Rf_{ave}} \sum_{i=x+1}^{i} Rf_i$ is the weighting factor

Rf_{ave} is the long-term rainfall average

u and v and x are the number of months over which the summation is carried out. In this application they could vary between 12 and 48 months.

The model to simulate the ^{14}C in the spring discharge has the following features:

- The relative ^{14}C content of the two components of the recharge is calculated in a similar manner as for the actual amounts of the recharge. Thus the monthly value of the atmospheric ^{14}C times the excess rainfall amount is averaged over several previous months and multiplied by the fraction relating to the dilution of the ^{14}C during the dissolution of carbonate. The two components thus obtained are added and the sum is divided by the total recharge to give the ^{14}C content of the water reaching the water table for each month.
- The admixture of water in the aquifer is treated by two well-mixed boxes: a shallow box that contains the recent recharge and more rapidly reaching the spring outflow, and a deeper one that represents the average recharge over a preceding period of several decades.
- At the spring several multiples of the deep box mix with the water from the shallow box to determine the ^{14}C of the discharge
- Finally, a lag-time of up to a few years is introduced to the outcome of the simulated ^{14}C values. It mainly represents the time lag that is effected in the unsaturated zone, which has not been incorporated in the mixing model, as well as a possible delay in the transmission of the tracer through the aquifer; however, no provision has been made for dispersion.

The ^{14}C content of the water derived from the two-box model in month i is given by Eq. (2) below:

$$C_i = \frac{1}{p+1} \left(\left[\frac{gaw}{u} \sum_{i-u+1}^{i} A_i (Rf_i > t_1) + \frac{hbw}{v} \sum_{i-v+1}^{i} A_i (Rf_i > t_2) \right] \frac{1}{R_i} + \frac{p}{j-k} \sum_{i-k+1}^{i-1} C_i \right) \quad (2)$$

where:

A_i is the ^{14}C value of atmospheric CO_2 in month i

g and h are the fractions of A_i in the two components of the recharge determined from the model and the bicarbonate concentrations

p is the multiple of deep water that mixes with the shallow water fraction from the spring

(j-k) is the summation period, i.e. the number of months prior to j over which the deep water is averaged (usually several hundred months) and is summed up to month i-1 (i.e. values of C that are already calculated)

j is mostly 36 because of it being the rainfall period that on average corresponds best to the flow of the springs

The parameter p indicates that the deep flow component probably extends over a longer period than j-k, which is the reason why multiples of the deep flows have to be incorporated in view of limited historical ^{14}C input from too short a rainfall record.

The best simulation is obtained by interactive adjustment of the parameter values until the smallest least-square fit between the observed and simulated values has been obtained. At the same time, the simulated average recharge and average bicarbonate should correspond to the measured values.

Application of the model

Deriving the characteristics of dolomitic aquifer systems

The ^{14}C model has been applied to most of the monitored springs in the different areas and the best simulations of a selection of major springs are shown in Fig. 3 (next page), also indicating the ^{14}C input content in the atmosphere. The parameters that have been derived for this group are listed in Table 1.

Deriving the turnover time and age of water in the aquifer

The turnover time T of water discharging from the different springs and the *average age* of the water emerging from the springs are derived according to Eqs. (3) and (4). The turnover time represents the ratio of the total groundwater in storage to the average annual recharge (Bredenkamp and Vogel, 1970), and it provides an indication of the time it would take for a contamination pulse to be flushed from these aquifers.

$$T = \frac{n_1 + n_2 * p}{(p+1) * 12} \qquad (3)$$

where:

n_1 = the period representing the shallow water
n_2 = the period for the deep water contribution
p = the multiples of deep flow relative to the shallow component

which yields:

total age of water (yr) = turnover time (yr) + final lag L (months)/12 (4)
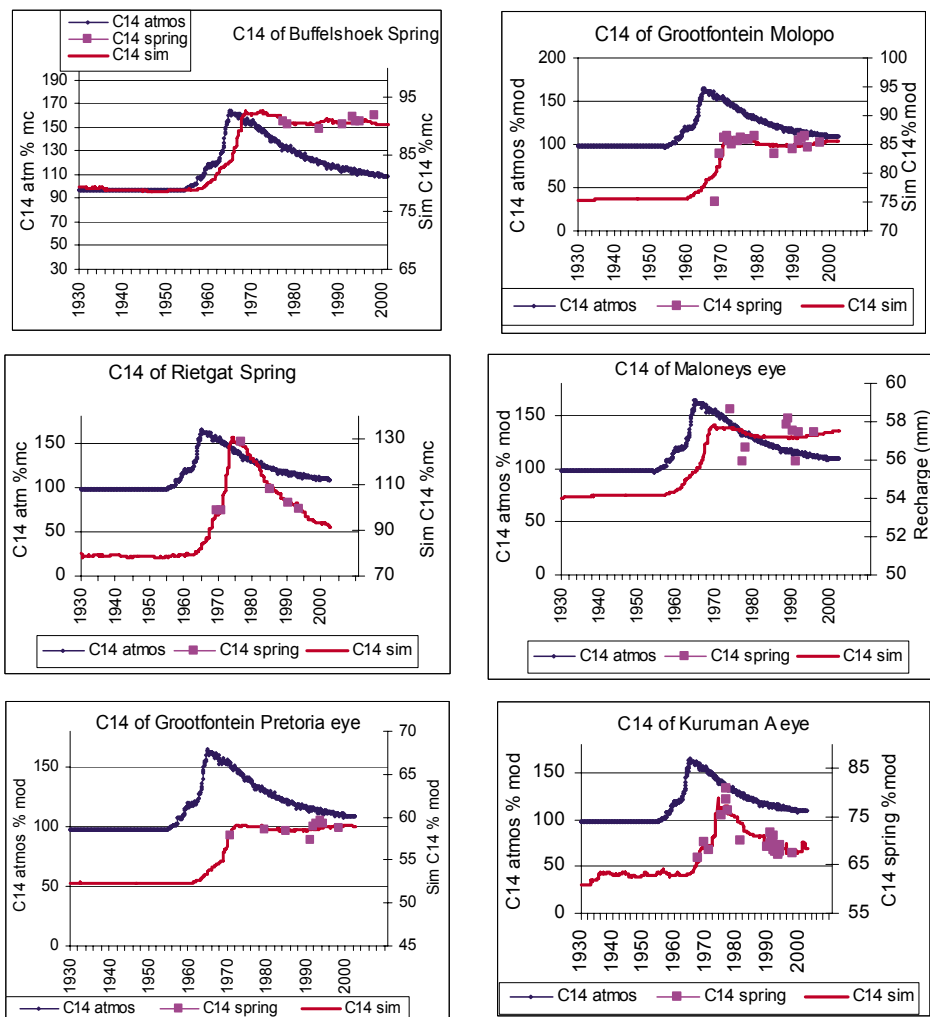
Figure 3
Graphs of the ^{14}C observations in comparison to the simulated values for a selection of dolomitic springs in different areas

The final lag L (Table 1) in fact means that the measured ^{14}C content of a specific month has been the response to rainfall effectively starting L months earlier than month i.

The model has provided reliable estimates of the average recharge that corresponded to the values derived from the CMB method and conforming to the time-varying response of the spring discharge. The same applies to the average simulated and measured bicarbonate content of the spring.

Table 1 shows good comparison of parameters for the various aquifers derived from the ^{14}C modelling, e.g.:

- The high threshold rainfalls are very similar and variations of the lower threshold can be related to the thickness and extent of the superficial cover overlying the aquifer. In the case of Rietgat Spring and Kuruman A Spring the surfaces are largely covered by calcrete, which seems to be linked to a large final lag that has to be further investigated.
- For most of the aquifers, the normal recharge coefficient is the dominant component.
- The ^{14}C coefficient of the normal recharge component is always higher than that of the direct recharge.
- The multiples of deep flow (p) that have yielded the best simulation for the smaller compartments and larger for the big compartments, with Buffelshoek Spring being an exception, apparently because of overflow occurring from compartments higher up. For Rietgat the value of p= 0 indicates that there is no deep flow component but its long lag still needs further clarification.

- The final lag shows a large variation ranging from a few months to 86 months for Rietgat and 133 months in the case of Kuruman. The monthly variability of the recharge of springs with flow records corresponds well with the value simulated from the ^{14}C model.

The successful simulation of the ^{14}C of springs in dolomitic aquifers of the Northwest, Gauteng, Far West Rand and Kuruman regions indicates that the parameters conform to the conceptual bi-modal recharge and mixing model. The results reveal that the smaller the contribution of the deep flow, the more the outflow ^{14}C pulse conforms to the shape of the input tracer pulse. The larger a compartment is, the higher the deep flow component generally is, whilst a greater final lag apparently applies when the unsaturated zone comprises calcrete deposits. Typical examples are those of the Rietgat Spring, Molopo Eye and Kuruman A Spring as is indicated in Table 1. The controlling parameters of the recharge, e.g. the threshold rainfalls and coefficients of recharge of the two components showed a great deal of similarity. The low threshold rainfall is in the range of 20 mm and the high threshold is about 60 mm per month for direct recharge to occur.

The low concentrations of ^{14}C of Maloneys Eye and Pretoria Grootfontein Springs initially appeared non-typical of dolomitic springs. However, a reassessment of the Grootfontein Spring (Pretoria) has revealed that the recharge coefficient is about 16% and is entirely derived from the dolomite and not from the adjacent quartzite aquifer.

TABLE 1 **A summary of results of the ^{14}C simulations of selected springs from the different dolomitic areas**							
SPRINGS	**Buffels-hoek**	**Grootftn Mol**	**Rietgat**	**Grootftn Pretoria**	**Maloneys**	**Gerrit Minnebron**	**Kuruman A**
Low threshold (t_1)	12.5	29.7	26.8	23.5	15.5	10	20
High threshold (t_2)	63.9	38.2	51.4	61.4	55.9	74.5	65
Normal recharge factor (a)	0.112	0.084	0.065	0.15	0.109	0.09	0.106
Direct recharge factor (b)	0.065	0.114	0.034	0.084	0.056	0.06	0.07
^{14}C factor (N) (g)	0.83	0.853	0.881	0.545	0.57	0.75	0.658
^{14}C factor (D) (h)	0.592	0.551	0.682	0.504	0.558	0.50	0.551
Deep flow mix factor (p)	3.19	4.12	0	4.29	9.55	8.83	2.31
Mix period 1 (n1)	36	36	36	36	36	36	240
Mix period 2 (n2)	348	338	No deep	329	460	548	597
Turnover time (years)	11.6	12.8	1.5	12.7	16.1	22	22.4
Rf weighting w (months)	24	24	24	36	36	36	60
Aver. period (>N-thresh) (mths)	12	36	36	36	36	36	36
Aver. period (>H-thresh)(mths)	12	36	36	24	36	36	36
Final lag L Sim ^{14}C (months)	74	57	86	61	29	16	133
Sim HCO$_3$ of spring (mg/ℓ)	228.6	222.1	238.8	79.8	97.0	181	149
Measured HCO$_3$ (mg/ℓ)	228.8	228	285*	117	130	201*	162
Recharge area (km^2)	32	92	38	28.7	234	796	286
Cl of spring (mg/ℓ)	7.4	6.4		4.8	3.3	7.5	5.3
Recharge % of average Rf	7.5	8.9	7	13	12	8.4	8.5
^{14}C Recharge % of av. Rf	7.5	8.9	9.6	13	12	8	8

Slight pollution of aquifer

Comparison of the ^{14}C simulations with tritium and CFC measurements

Concurrently with the ^{14}C input from thermonuclear tests, a similar tritium pulse has been released into the atmosphere, although its maximum concentrations have been 30 times that of the pre-bomb levels compared with 1.6 times in the case of ^{14}C. The pre-bomb tritium levels have increased from about 2 TU (tritium units) to about 60 TU in 1962 and have thus provided a distinct signal to the groundwater recharge to trace its propagation through the aquifer. However, in view of the radioactive decay of tritium (half-life of 12.3 years), the tritium input into the aquifer had to be corrected for each of the simulated months with incorporation of the final lag as well.

Measurements of tritium have been carried out only for some of the springs and were modelled according to the same set of parameters that have been derived from the best ^{14}C simu-
lations, except that there is no differentiation in the bi-modal input. Some of the results for tritium are shown in Figs. 4a and 4b. Table 2 indicates that larger contributions of deep flow have to be incorporated to obtain a good fit in the case of tritium than for the ^{14}C simulation, while for CFC they were smaller than for ^{14}C.

The higher contribution of deep flow in the case of tritium is probably caused by the fact that additional dilution of the input of tritium occurs in the unsaturated zone overlying the aquifer. This is not the case with the ^{14}C input, which is effected at the depth of the root zone. In the model, the mixing only occurs as though the recharge of a specific month enters the saturated zone without any dilution. A larger multiple of deep flow is therefore required in the case of tritium to best simulate the measured levels. Hence, the larger deep-flow component for the tritium simulation than that of the ^{14}C simulation is an indication of the impact of the unsaturated zone.
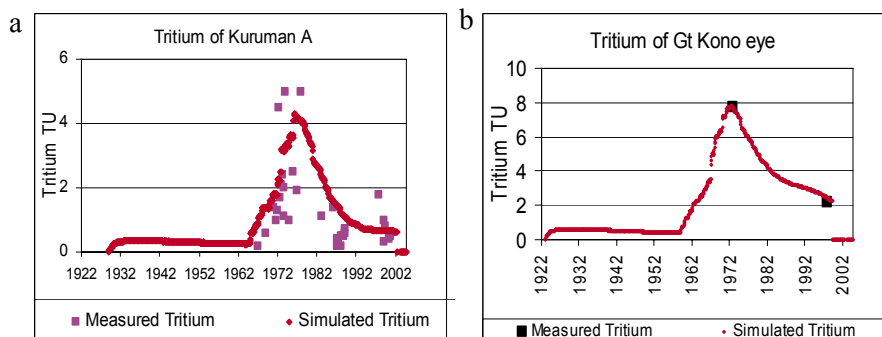

Figure 4a and b
Typical simulations of the tritium content of springs based on the parameter values for the best ^{14}C model, which in this case required a higher contribution of deep flow than for ^{14}C (see Table 2).
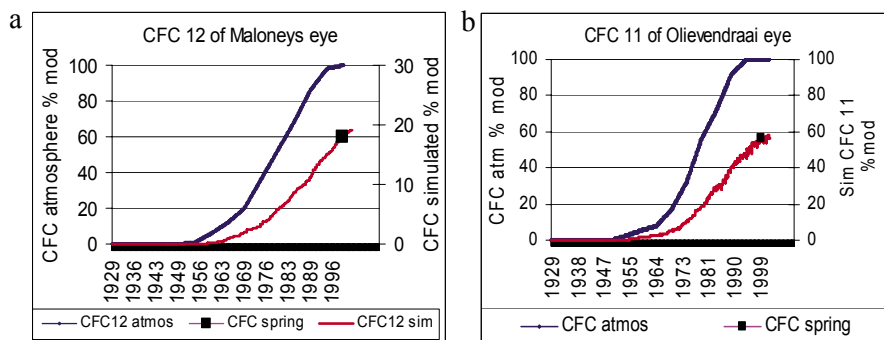
Figure 5a and b
Simulations of CFC concentrations of two springs using the parameter values that have produced the best fit for the ^{14}C model (see Table 2)

It appears that in the case of CFC a smaller contribution of deep flow is required than for the ^{14}C simulations, which conforms, as is generally assumed, to the input of CFC occurring at the interface between air and the saturated water level of the aquifer. Therefore, it would seem that the different responses of the tracers could be explained by their input mechanisms. The ^{14}C simulations are more reliable in view of fewer analysis of tritium and only single CFC measurements have been carried out.

Assessment of the pollution of an aquifer based on the ^{14}C model

Simulation of the transmission of pollution through a dolomitic aquifer

Springs in the West Rand dolomitic aquifers (Fig. 1) have been polluted by the gold mines, e.g. Turffontein and Gerrit Minnebron. In cases of accidental spillage a reliable evaluation of the environmental impact and the effectiveness of clean-up measures is often required. The parameters obtained from the ^{14}C model and the mixing models have been used to derive the pollution input and its temporal variation for different recharge scenarios (see Fig. 6). From inverse modelling it appears that the pollution had commenced at about 1959 and had remained constant up to about 1977, whence it had increased and remained effectively constant from 1988 until 2001 (Fig. 6). The decontamination response can be simulated assuming that the influx of the pollution stopped in 2001 and is being flushed out, by extending the rainfall sequence by the series experienced over the past 60 years. This indicates that the pollution pulse would only be reduced to 100 mg/ℓ after 64 years.

Bicarbonate concentrations

The bicarbonate of the bi-modal recharge components of the ^{14}C model is based on a linear interpolation between the two extremes in the bicarbonate dissolution-equilibrium shown in Fig. 7. A maximum concentration of 280 mg/ℓ of bicarbonate is attained during slow movement of recharge through the unsaturated zone where biogenic CO_2 is released by the plant roots. This corresponds to a ^{14}C factor of 0.9 used in the model. A minimum concentration of 51 mg/ℓ represents a ^{14}C factor of 0.5 that applies if recharge occurs directly to the saturated zone to dissolve carbonate having by-passed the root zone of the vegetation. Depending on the relative proportions of direct and normal recharge, the bicarbonate concentrations of the water emanating from a spring will vary between these extreme values.

TABLE 2
A comparison between the multiples of deep flow for ^{14}C tritium and CFC for typical springs that have been monitored

| Spring	Multiple of deep flow	
Buffelshoek Eye	^{14}C	3.18
Doornplaat	^{14}C	3.96
	CFC	1.67
Gerrit Minnebron*	^{14}C	15.2
	CFC	contam
Gftn Mol	^{14}C	4.12
	Tritium	6
	CFC*	0.33
Kuruman A Eye	^{14}C	2.68
	Tritium	12
Kuruman B	^{14}C	4.94
	Tritium	11
Manyeding	^{14}C	0.82
	Tritium	7
Molopo Eye	^{14}C	7.62
	Tritium	11.5
Olievendraai	^{14}C	1.08
	CFC	1.0
Paardevlei	^{14}C	2.03
Pta Erasmus fount	^{14}C	17.18
Pta Grootfontein	^{14}C	11.98
Renosterfontein	^{14}C	2.59
	CFC	0.95
Schoonspruit Eye	^{14}C	1.5
	Tritium	10.5
Turffontein	^{14}C	2.01
Upper Mooirivier	^{14}C	10.86
	CFC	3.45
Vergenoegd Eye	^{14}C	2.53
Groot Kono Eye	^{14}C	1.5
	Tritium	6.5
Klein Kono Eye	^{14}C	1.9
	Tritium	4.71
Pta Fount Upper	^{14}C	2.62
Pta Sterkfontein	^{14}C	2.23
Maloneys	^{14}C	9.55
	CFC	5.95

Sample contaminated
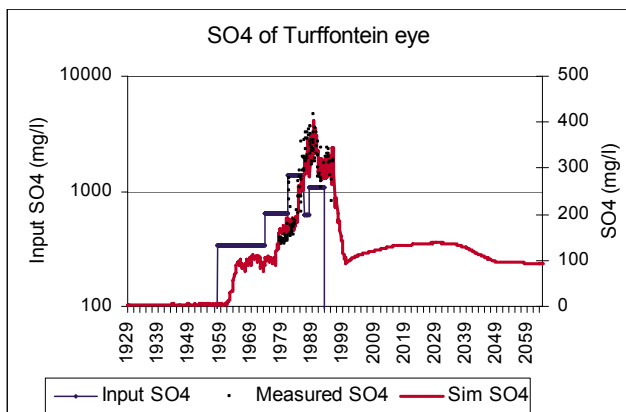
Figure 6
The simulated pollution of Turffontein Spring vs. the measured concentrations
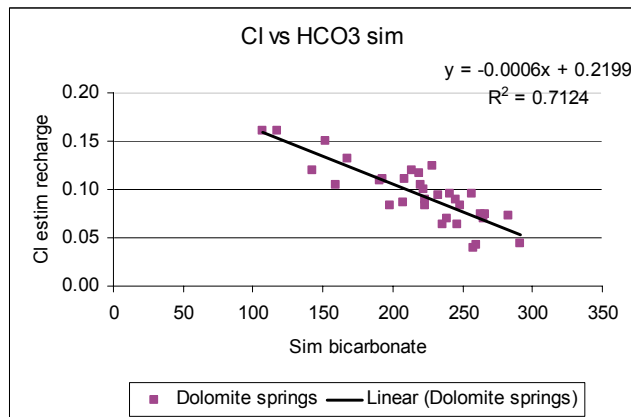

Figure 8
Relationship between the bicarbonate concentrations of the different dolomitic springs and the average recharge coefficients derived from the chloride method
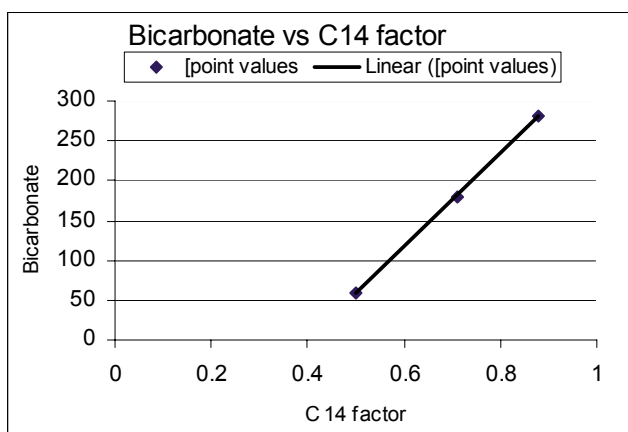

Figure 7
Relationship between the bicarbonate concentrations and the ^{14}C factor used in the simulation model to derive the bicarbonate concentrations
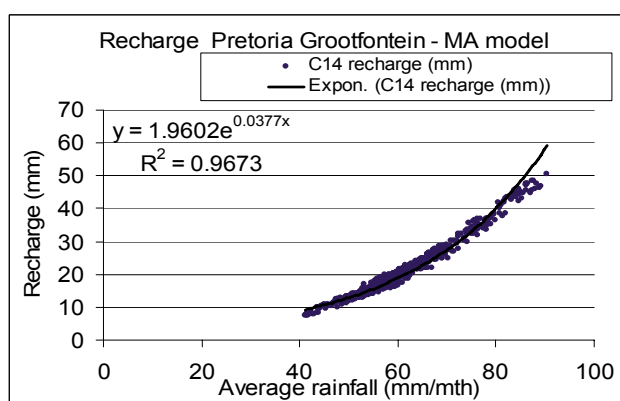

Figure 9
Exponential relationship between the monthly recharge derived from the ^{14}C simulation model and the average rainfall over the preceding 36 months without the incorporation of the threshold rainfalls

The bicarbonate content of the spring water that has been derived from the relationship shown in Fig. 7 with revised chloride values of Pretoria dolomite, has shown an inverse linear relationship if plotted against reliable estimates of recharge (see Fig. 8). The recharge derived from the chloride mass-balance ratio has been used as the reference recharge values. The bicarbonate concentrations of the spring waters could thus be used to obtain a provisional independent estimate of the average recharge of dolomitic aquifers for all dolomitic aquifers of South Africa. Similarly, the bicarbonate of individual boreholes would reflect the spatial variability of recharge experienced over the recharge area of the various springs.

Simplified rainfall-recharge relationship

An important outcome of the ^{14}C model is that the monthly recharge values reduce to an exponential relationship in relation to the average rainfall over a characteristic period ignoring the threshold rainfall values and weighting factors that have been used in the ^{14}C model.

The exponential relationships shown in Figs. 9 and 10 are similar to the one derived by Van Rensburg (2004) to simulate the monthly recharge from rainfall, but which still incorporates a cut-off threshold rainfall. The newly derived relation-
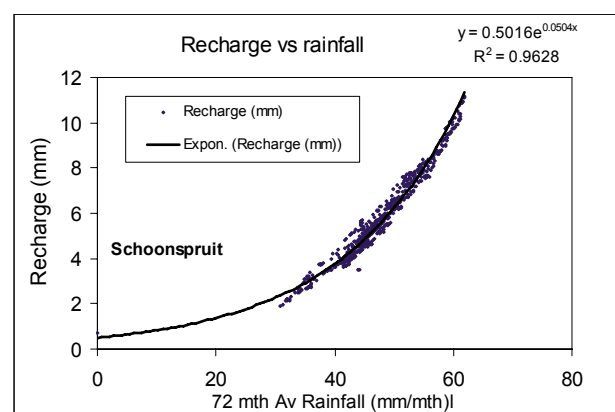

Figure 10
The monthly natural flows of Schoonspruit eye (mm/month) in relation to the average rainfall over 72 months for a recharge area of 842 km² as derived by Veltman (2003), without the incorporation of threshold rainfalls

ship provides a method to simulate the temporal response of the groundwater levels from which the reserve and the allocations of groundwater in the periods of surplus could be derived.

It is evident that the ^{14}C model could be simplified by deriving the monthly recharge using the new exponential relationship to determine the ^{14}C input. Likewise the bi-modal recharge components could be substituted by a single recharge coefficient with a corresponding initial ^{14}C coefficient that is determined by the bicarbonate relationship shown in Fig. 7.

Conclusions

The successful simulation of the reappearance of the bomb ^{14}C in dolomitic springs, based on the new tracer model programmed in Excel, has:

- Provided evidence in support of the bi-modal recharge model controlling the input of ^{14}C, although it appears that reasonably good simulations could also be obtained by using a simpler model
- Clearly indicated that a two-box model, mixing recharge from a recent period with water from a longer preceding period, is an essential component of the model
- Confirmed that reliable estimation of groundwater recharge is critical to obtain reliable estimates of the aquifer parameters
- Provided successful simulations of the ^{14}C of several springs in spite of the availability of only intermittent measurements over a period of more than 30 years
- Enabled derivation of the turnover time and 'age' of water in the dolomite aquifers and from it the ratio of the aquifer storage to the average annual recharge, which would provide an estimate of the storage water in relation to the annual recharge
- Indicated a linear relationship between the bicarbonate concentrations of the springs and the recharge coefficients as derived from the chloride mass balance method, assuming the latter to be reliable.

Furthermore:
- The model could be used to assess the reliability of tritium and CFC as tracers and to interpret differences in their results in terms of the conceptual model
- It could be shown that an exponential regression relationship exist between the rainfall and recharge that has been obtained from the ^{14}C model to be used as a general equation to obtain the variability of monthly recharge from rainfall
- It has demonstrated the significance of ^{14}C, tritium and CFC to provide a better understanding of the flow of water in dolomitic aquifers, more effective utilisation and better assessment of pollution in these valuable groundwater resources.

Acknowledgements

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