Flow Regimes from International Experimental and Network Data (FRIEND)

Volume III Inventory of streamflow generation studies

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3. The application of isotopic tracer techniques for hydrological process studies

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3.1 INTRODUCTION

In a rapidly changing world hydrologists are facing the development of scientific and public requirements with respect to increasing environmental problems, thus needing new ideas (technical-experimental; theoretical-modelling) in the diverse fields of scientific and applied ecohydrology. In this context, several recent comprehensive national and international state-of-the-art reports with priority research needs, for instance by Kundzewicz *et al.*, 1987 or US National Research Council (1991), are complaining about the deficient knowledge on the principal processes in the hydrologic cycle.

This deficiency above all concerns catchment flow processes where the Hortonian philosophy still persists, and retarding scientific progress after decades. Nevertheless, considerable refinement of process knowledge has been made concerning turnover mechanisms of water at a small drainage basin scale during the last two decades by the use of natural hydrological tracers and environmental isotopes in particular, and including processes controlling runoff generation.

The following brief outline of hydrological tracer techniques updates the earlier synopses by Stichler & Herrmann (1982, 1983) and Herrmann (1989, 1993) on hydrograph separations and mean transit time calculations of subsurface water. It focuses on the application of area-injected environmental isotopes by precipitation as natural tracers for small hydrological catchment systems. The benefit of deliberately point-injected artificial tracers, such as fluorescent dyes, is considered as far as additional information about physical processes on a small basin scale is possible. Other artificial tracers such as radioisotopes or activation-analytical substances and hydrogeogenic indicators which frequently reveal ambiguous tracing qualities, or helpful anthropogenic chemicals (pollutants, contaminants for ecosystems even if intentionally applied in low concentration as for instance bromide) are excluded from further consideration. But it should be recognised that the most comprehensive insight into natural hydrological system behaviour can be expected from the combined use of all modern tracer techniques.

Furthermore, this presentation concentrates on hydrological basin studies contributing to an improved knowledge about streamflow generating processes. Other specialised tracer hydrological investigations at single experimental sites, slope sections or aquifers are therefore left out unless being at least imbedded in a complete basin water balance assessment.

This paper describes, as example, some small European study basins with a high level of tracer data. Some of these basins are of multipurpose basic, comparative (regional) or applied research interest, but which also satisfy broader regional interests. This fact is supported by **Tables 3.1 and 3.2**. Some of these basins are also included in a recent comprehensive review of small Western European study basins for hydrochemical budgets by Hornung *et al.*, (1990) and in the FRIEND catalogue of small basin studies of streamflow generation (Robinson, 1993).

Table 3.1 *Synopsis of environmental isotope studies on direct runoff separation (after Stichler & Herrmann, 1982, 1983 and Herrmann, 1989, 1993; completed)*

SINGLE EVENTS

Author, Year/Reference	**Name, country**	**Surface area (km[2])**	**Altitude interval (m a.m.s.l.)**	**Indicator used[1])**	**Direct runoff % of storm total**	**melt events**	**Remarks**
Crouzet, Hubert, Olive, Siwertz, Marce 1970/J. Hydrol. 11	Pont des Blaves Charmoisy, France L'Eau Morte	15 5.7 91	?[2]) ?[2]) -460-2351	H-3 H-3 H-3	30[3]) 93[3]) 80	x x x	[2]) mountainous [3]) peak discharge
Behrens, Bergmann., Moser, Rauert, Stichler, Ambach, Eisner, Pessi 1971/Z. Gletscherkd. Glaziolgeol. 7, 1/2	Kesselwandbach Hintereisbach Austria Rofenache/Vent	5.7 20 96.2	2700-3500 2425-3739 1905-3772	H-3, H-2, cond H-3, H-2, cond H-3, H-2, cond	35-80 40-75 35-70	x x x	75% glaciers 56% glaciers 44% glaciers
Martinec 1972/Gas, Wasser, Abwasser	Modry Dùl, Czech Republ.	2.6	1010-1554	H-3	35-45	x	
Mook, Groeneveld, Brown, van Ganswijk 1974/Proc. IAEA Symp. Vienna	Hupselse Beek, Netherlands	6.5	several m	0-18	13	x	
Fritz, Cherry, Weyer, Sklash 1976/ Proc. IAEA Group Meet. Vienna 1975	Wilson Creek, Man. Kenora, Ont. Canada Big Creek, Ont. Big Otter Cr., Ont.	22 1.8	135 700	0-18, cond, ion 0-18, ion 0-18, cond 0-18, cond	10 55 45 60	x x x x	
Blavoux 1978/PhD Thesis Curie Univ. Paris	Le Maravant France Mélarchez	3.0 7.0	840-930 148-182	0-18, cond, ion 0-18, cond, ion	2-40 10-50	x x	
Stichler, Herrmann 1978/Dt. Gewässerkdl. Mitt. 22	Lainbach	18.8	670-1801	0-18	12	x	
Martinec, Oeschger, Schotterer, Siegenthaler, Nuti, Tongiorgi 1978/79/Rivista Ital. Geofis. Sci. Aff. 5	Dischma, Switzerland	43.3	1668-31465	H-3, 0-18	11	x	<3% glaciers
Herrmann, Martinec, Stichler 1979/Proc. CRREL Hanover NH Symp. 1978	Lainbach, Germany	18.8	670-1801	H-2	10-30	x	
Sklash, Farvolden 1979/J. Hydrol. 43	Ruisseau des subbasin 6 Eaux Volées subbasin 7A Hillman Cr., Ont., Canada	3.9 1.2 1	720-780 760-880 202-212	0-18 0-18 0-18, cond, ion	15-35[3]) 15-35[3]) <20[3])	x x x	

SINGLE EVENTS

Author, Year/Reference	**Name, country**	**Surface area (km²)**	**Altitude interval (m a.m.s.l.)**	**Indicator used[1])**	**Direct runoff % of**	**storm total**	**melt events**	**Remarks**
Certer, Rauert, Stichler 1980/Proc. Int. Congr. Alp. Mét. Aix-les-Bains	Vernagtbach, Austria	11.4	2640-3628	H-3, H-2, cond	43)		x	82% glaciers [4])dir. snow-firn-icewater
Mérot, Bourguet, Le Leuch 1981/Catena 8	Nouvoitou, France	0.1	(8)	0-18, ions		x		
Rodhe 1981/Nordic Hydrol 12	Nåsten Stormayra, Sweden	6.8 4.0	18-55 10-77	0-18 0-18	9-16 11-14		x x	
Stichler, Herrmann 1982/Proc. Mississ. State Univ. Symp. 1981, WRP Littleton	Lainbach, Germany	18.8	670-1801	H-3, H-2, cond, ion	15-25	x		
Eden, Prösl. Stichler 1982/Proc. Berne Syp. 1982	Kreidenbach, Germany	1.8	1020-1360	0-18, cond, ion	25 50	x	 x	
Stichler, Herrmann 1982/Berne Symp. 1982	Lange Bramke, Germany	0.8	543-700	H-2	20	x	x[5)]	[5)]rain on snow
van de Griend, Arwert 1983/J/ Hydrol. 62	Rio delle Fonti, Italy	12.7	18.60-3479	0-18				[6)]rain metw. common. sep.
Siegenthaler, Schotterer, Müller 1984/Proc. IAEA Symp. Vienna 1983	Areuse, Switzerland[7)]	127	700[8)] 1115[9)]	H-3, 0-18 ion	9-25 16-45	x		[7)]karst spr. [8)]spring [9)]m. area altitude
Christophersen, Kjaernsröd, Rodhe 1984 In: Report 10 Nordic Hydrol. Progr. Oslo	Birkenes, Norway	0.4	-	0-18	10-40		x	
Körner, Agster, Einsele, Stichler 1986/	Goldersbach, Germany Kirnbach	1.1 9.1	Δ93 Δ168	H-2 H-2	35 40	x x		
Hooper, Shoemaker 1986/Wat. Resour. Res. 22	Hubbard Brook, NH, USA	0.4	-525-730	H-2, ion	25-40		x	
Kennedy, Kendall, Zellweger, Wyerman, Avanzine 1986/J. Hydrol. 84	Mattole R., CA, USA	620	-20-1200	H-3, H-2, 0-18 ion	20-40	x		
Pearce, Stewart, Sklash 1986/Wat. Resour. Res. 22	Tawhai, New Zealand	0.04	265-350	0-18, cond	10	x		

Author, Year/Reference	Name, country	Surface area (km^2)	Altitude interval (m a.m.s.l.)	Indicator used[1])	Direct runoff % of storm total melt events	Remarks
		SINGLE EVENTS				
Stichler, Herrmann, Rau 1986/IAHS Publ. no. 155 Herrmann, Koil, Schöniger, Stichler 1987/IAHS Publ. no. 167	Lange Bramke, Germany	0.8	543-700	0-18	20	x
Rodhe 1987/Uppsala Univ. Rep. Ser. A 41	Aspåsen	0.2	320-390	0-18	13 58 x	x
	Buskbäck	1.8	280-354	0-18	47	x
	Gårdsjön	0.04	115-150	0-18	15 44 x	x
	Nåsten Sweden	6.8	18-55	0-18	15 33 x	x
	Stormyra	4	10-77	0-18	30 29 x	x
	Svartberget	0.5	235-310	0-18	8 42 x	x
Leopoldo, Martines, Mortatti 1987/Proc. IAEA Symp. Vienna	Búfalos R., Sao Paulo,	1.6	-	0-18	18 x	
	Paraiso R. Brazil	3.3	-	0-18	37 x	
Yamagata, Okita, Kodair 1963/Proc. IAEA Symp. Tokyo 1963	River Tonegawa/Komatsu, Japan	4	--2000	Sr-89, Sr-90	21,72[10]	[10]sn.melt May 62; diff. Sr-90 ratios between surface and groundwater
Meiman, Friedman, Hardcastle; 1973/Proc. WMO/IAHS Symp. Banff 1972	Clear Creek Col.,	8.5	-3500[11]	H-2	?[12]	[11]elevation
	Little Beaver USA	31.8	-2800[11]	H-2	?[12]	[12]no exact notation
Ambach, Eisner, Elsässer, Löschhorn, Moser, Rauert, Stichler 1976/ J. Glaciol. 17	Hintereisbach, Austria	20	2425-3739	H-2	60[4]	56% glaciers; July-Sept. 69-74
Herrmann, Stichler 1980/Catena 7	Lainbach, Germany	18.8	670-1801	H-2	14-28 35-47 30-36	winter summer 1976-78 year
Martinec, Schotterer, Siegenthaler 1982/Beitr. Geol. Schweiz-Hydrol. 28	Dischma, Switzerland	43.3	1668-3146	H-3	38-48	snowmelt 1971, 1973-75
Eden, Prösl, Stichler 1982/Proc. Berne Symp.	Kreidenbach, Germany	1.8	1020-1360	0-18, cond	50	1980-81
Herrmann, Koll, Maloszewski, Rauert, Stichler 1986/IAHS Publ. no. 156	Lange Bramke, Germany	0.8	543-700	0-18/H-3	10	1980-86
Lepistö, Seuna 1990/In: Kauppi *et al.*, (eds) Acidificat. in Finland, Springer	Teeressuonoja, Finland	0.7	42-111	0-18, ion	15-30	melt seasons 1985, 1987

SINGLE EVENTS						
Author, Year/Reference	**Name, country**	**Surface area (km^2)**	**Altitude interval (m a.m.s.l.)**	**Indicator used[1])**	**Direct runoff % of storm total melt events**	**Remarks**
Cooper, Olsen, Solomon, Larsen, Cook, Grebmeier 1991. Water Resour. Res. 27	Imnavait Creek, Alaska USA	2.2	-875-955	0-18	>86[13)]	[13)]on frozen ground

[1)] [cond: electrical conductivity; ion = major ions; dye = dye tracers]

Table 3.2 *Synopsis of flow model applications for determination of mean transit times of water (t_o) in small drainage basins (after Stichler & Herrmann, 1982 and Herrmann, 1989; completed)*

Author, Year/Reference	**Name, country**	**Surface area (km^2)**	**Altitude interval (m a.m.s.l.)**	**Isotope**	**q(t)^2**	**t_o; t_t (yrs)**	**Remarks**
Dincer, Payne, Florkowski, Martinec, Tongiorgi 1970/Water resour. Res. 6	Modry Důl, Czech Repub.	2.6	1010-1554	H-3	BM	2.5	
Martinec, Siegenthaler, Oeschger, Tongiorgi 1974/Proc. IAEA Symp. Vienna	Dischma, Switzerland	43.3	1668-3146	H-3	EM DM BM	4.0 4.8 3.0	3% glaciers
Löschhorn, Ambach, Moser, Stichler 1977/Z. Gletscherkd. Glazialgeol. 12	Rofenache/ Vent, Austria	96.2	1905-3772	H-2	EM	100 days	meltwater comp.
Behrens, Moser, Oerter, Rauert, Stichler, Ambach, Kirchlechner 1979/Proc. IAEA Symp. Neuherberg 1978	Rofenache/ Vent, Austria	96.2	1905-3772	H-2	EM	$4^{2)}$	$^{2)}$from winter baseflow of subbasins; 44% glac.
Schotterer, Wildberger, Siegenthaler, Nabholz, Oeschger 1979/Proc. IAEA Symp. Neuherberg 1978	Rawil, Switzerland$^{3)}$	-100	1200-3250	H-3	EM	2-4	$^{3)}$karst region
Müller, Kiraly, Schotterer, Siegenthaler 1980/Steir. Beitr. Hydrol. 32	Areuse, Switzerland$^{3)}$	127	$700^{4)}$ $1115^{5)}$	H-3 0-18	EM	$0.75\text{-}2^{6)}$	$^{4)}$spring $^{5)}$mean area altitude $^{6)}$indir. springwater portion
Maloszewski, Rauert, Stichler, Herrmann 1983/J. Hydrol. 66	Lainbach, Germany	18.8	670-1801	H-3 H-2	EM DM DM I II EM DM I calc.II	2.2 2.4 $0.8^{7)}$ 7.5 2.1 0.6 6.6	$^{7)}$I, II represeparate sub-surface reservoirs
Eden, Pröskm /Stichler 1982/Proc. Berne Symp.	Kreidenbach, Germany	1.8	1020-1360	0-18 dyes	EM I II	1.4 $0.1^{7)}$ 5.3	
Stichler, Herrmann 1982/Proc. Berne Symp.	Lange Bramke, Germany	0.8	543-700	H-3 H-2	EM EM	$2^{8)}$ $1.9^{8)}$	$^{8)}$preliminary approximations
Pearce, Stewart, Sklash 1986/Water Resour. Res.	Tawhai, New Zealand	0.04	100-150	0-18	EM	0.35	

Author, Year/Reference	Name, country	Surface area (km²)	Altitude interval (m a.m.s.l.)	Isotope	q(t)[2]	t_o; t_t (yrs)	Remarks
Bergmann, Sackl, Maloszewski, Stichler 1986/Proc. 5th SUWT Athens	Pöllau, Austria	53.3	399-1280	0-18	EM	1.9	
Burgmann, Calles, Westmann 1987/Proc. IAEA Symp. Vienna	Sweden[9]	440-22000		0-18	EM	0.17- 2.25	[9]11 catchments
Maloszewski, Rauert, Stichler, Herrmann 1990/Freiberger Forschungshefte C442	Lange Brambe Saukappe[11], Germany	0.8 0.1	543-700 590-665	H-3 H-3	ODM ODM	2.7[10] 4.2[10]	[10]t_t [11]spring
Herrmann, Kill, Leibundgut, Maloszewski, Rau, Rauert, Schöniger, Stichler 1989/Landschaftsökol. u. Umweltforschung, Braunschweig	Lange Bramke	0.8	543-700	0-18 H-3	ODM DM	1.1[10][12] 1.5[13]	[12]unsat. soil zone [13]groundw. with retard. fact. 1.45 [14]groundw. with retard fact. 1.2
Maloszewski, Rauert, Trimborn, Herrmann 1993/J. Hydrol.	Wimbach, Germany	33.4	636-2713	H-3 0-18	EM ODM EM ODM	4.1 4.0[10] 4.0 4.0[10]	

[1)][BM=binomial model; EM=exponential model; ODM=ordinary dispersive model; DM=dispersive model]

The discussion uses the results of a recent and almost unique multilateral basin approach to demonstrate and encourage the diversification of applications of the environmental tracer technique in catchment hydrology and ecoysystem analysis. By this approach, from the hydrodynamic interpretation of (conservative) tracers, the behaviour for reactive substances transport is thought to become clear, provided that reaction dynamics are known.

3.2 THEORETICAL CONSIDERATIONS

3.2.1 Environmental isotopes as hydrological tracers

As constituents of the water molecule (H_20), the heavy isotopes of hydrogen (H^2, deuterium; H^3, tritium) and oxygen (^{18}O, oxygen-18) are assumed to be ideal tracers in the water cycle. Details about the physical fundamentals, measuring techniques, frequencies in natural waters and regional distributions can be found, together with many selected scientific and practical hydrological applications, in many publications including Moser & Rauert (1980), Fritz & Fontes (1980), and for stable isotopes (^2H; ^{18}O) in Gat & Gonfiantini (1981).

Stable isotopes are subject to temperature-dependent fractionation during phase changes of water (and tritium to radioactive decay with a half-life of 12.43 yrs.). A large variation in stable isotope concentrations occurs in natural waters. The most favourable tracing qualities of stable isotopes occur in the higher latitudes, largely due to sinusoidal seasonal variations, with the highest input concentrations from summer precipitation. The suitability is low in tropical and coastal regions.

Moreover, the use of these isotopes is quite attractive because sampling volumes of water of about 100 ml for the stable isotopes and 400 ml for the radionuclide (^3H) are very easy to carry out and analyse. According to **Tables 3.1 and 3.2** the use of tritium has been slightly replaced by the stable isotopes since the mid-1970s due to the greater ease of stable isotope measurements at a comparative accuracy level, and to worsening tracing suitability of tritium. The latter mainly originates from nuclear bomb tests in the atmosphere prior to the moratorium in 1963 (Fritz & Fontes, 1980, 1986; Moser & Rauert, 1980). Since then, depletion of ^3H concentrations in precipitation by two orders of magnitude has been observed, and has now reached a much less significant seasonal variation on an absolutely low activity level of 20-25 TU (1 TU=0.12 Bq in 1 l H_2O) in Central Europe (IAEA, 1992b). The hydrological application of tritium remains much more limited in the southern hemisphere where maximum bomb tritium content of precipitation was delayed by up to two years as compared to mid-latitudes of the northern hemisphere, and lower concentrations by about one order of magnitude (Taylor, 1966).

Unfortunately, isotopes of noble gases (e.g. ^3He, ^{85}Kr) cannot replace tritium at the moment which is still more convenient for sampling and analysis at a similar accuracy level (Weise *et al.*, 1992; Schlosser, 1992).

3.2.2 Tracer hydrological system analysis

Basic concepts and techniques of tracer hydrological system approaches are summarised in **Figure 3.1** representing a simple hydrological black-box model describing a natural system such as a catchment basin, aquifer or glacier. It is assumed that the time-dependent input and output water fluxes [$Q_{in}(t)$, $Q_{out}(t)$] and environmental tracer (isotope) concentrations [$C_{in}(t)$, $C_{out}(t)$] are known. The tracer technique makes use of the isotopic input signals (=functions) which can represent a Dirac impulse, for instance, and their transformation to specific output signals (breakthrough curves) to give information about the specific storage properties of the intercalated basin.

Since areal injection of environmental isotopes with rain and meltwater enables model treatments of the whole study basin, a direct flow portion (d) (=event water) which is defined as having the isotope concentration of the input, and a short mean transit time (t_o) of the order of some hours to days can be separated from total flow in the sense of a bypass component by applying a simple mixing formula. The other portion (i) (=pre-event water) is considered as an indirect flow with a longer t_o of months to years.

t_o (=V_m/Q; with V_m as the volume of mobile water in the system, and Q as the volumetric flow rate) is a hydraulic key and fitting parameter of specific transfer or basin response functions g(t) (for *a priori* g(t)-functions see **Table 3.3**), and the mobile water volume V_m (=Q t_o) a most important hydrologic parameter to be searched for.

Hydrological modelling of the system under consideration, according to relevant processes as they may occur in reality, is a main precondition to make the signalling effects of the environmental isotopes suitable for hydrological tracings. For this purpose, the hydrological system model shown in **Figure 3.1** may be extended by defining more boxes (storages; cf. **Fig. 3.2**). Another essential condition is the availability of reliable hydrological data. In this

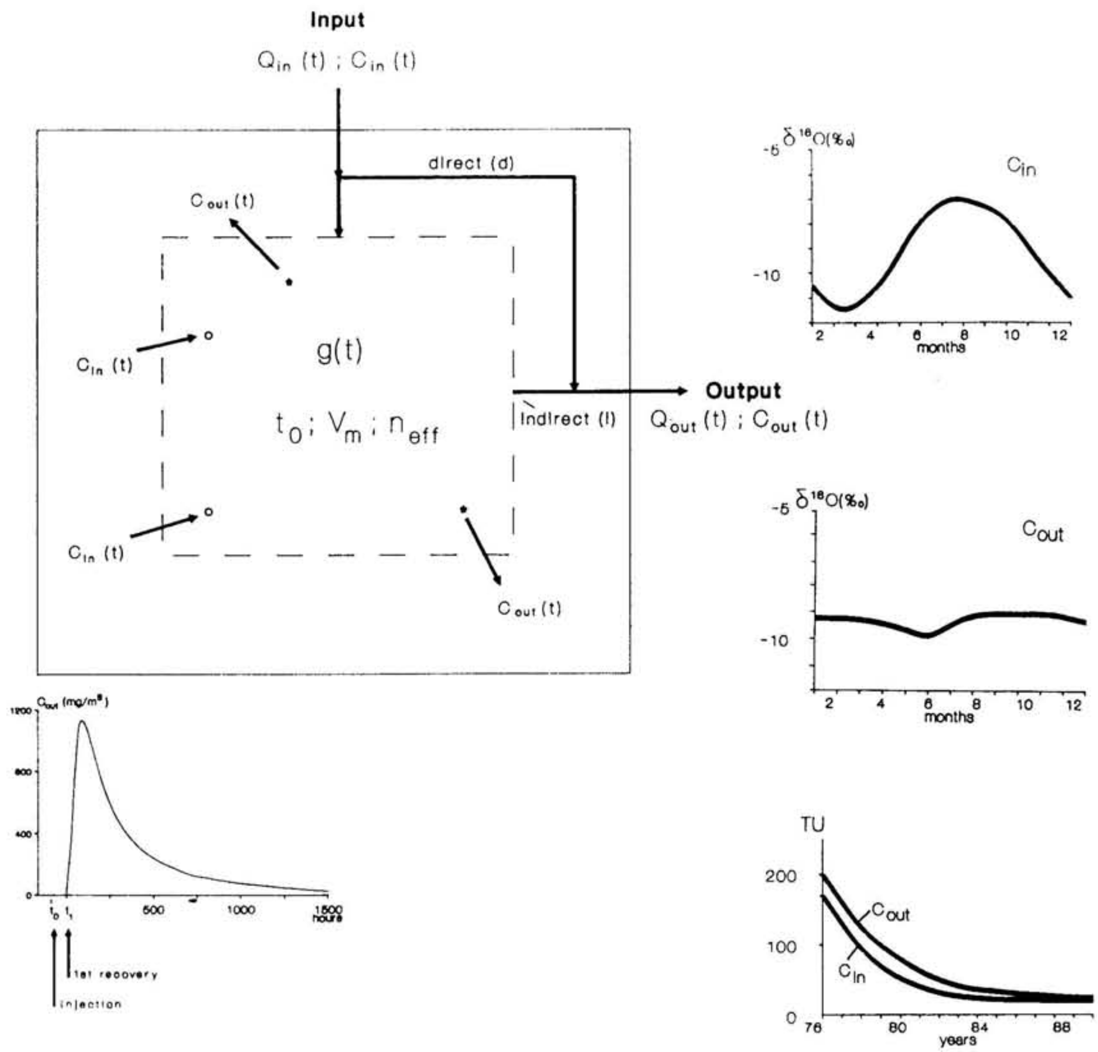

Figure 3.1 *Principle of tracer hydrological system analysis*

context it is also worth mentioning that the hydrological isotope techniques developed since the 1960s have primarily been for the interpretation of radioisotope data relating to springs and rivers. Comprehensive compilations of many hydrological applications of the environmental isotope technique by IAEA (1970, 1976, 1979, 1983a, 1983b, 1984, 1985, 1987, 1989a, 1989b, 1992a, 1992b) concern all components of the water cycle.

Additional tracer applications yield information about hydraulic connections and flow patterns, including dispersion within surface (rivers, lakes) and subsurface storage systems (soils, aquifers, glaciers) and flow velocities of tracer and water along distinct flow lines or pathways, using the input and output functions for artificial substances [$c_{in}(t)$, $c_{out}(t)$; cf. **Fig. 3.1**]. One should note that artificial tracings can only make selected flow lines of a system more apparent, depending on the site and time of tracer injection and tracer detection level and recovery rate. Field applications of the artificial tracer technique concentrate on subsurface hydrology with preference for karst systems and porous aquifers. Several compilations of appropriate contributions during the last decade are available as publications of the International Association of Tracer Hydrology (ATH), edited by Leibundgut & Weingartner (1982), Morfis & Paraskevopoulou (1986) and Hötzl & Werner (1992).

3.2.3 Hydrograph (direct flow) separation

For total basin runoff R_t the flow proportions d and i in **Fig. 3.1** are obtained from the mixing formula:

Table 3.3 *Relevant mathematical flow models with weighting functions g(t), fitting (flow) parameters and main fields of application (after Zuber, 1986; completed)*

Model/Author	g(t) [1]	Parameters [1]	Application
exponential (EM) (Eriksson 1958)	$t_o^{-1}\exp(-t/t_o)$	$t_o\ [=\omega^{-1}(f^{-2}-1)^{1/2}]$	porous aquifers
dispersive (DM) (Maloszewski & Zuber 1982)	$(4\pi t D/vxt_o)^{-1/2}\exp[-(1-t/t_o)^2\ vxt_o/4Dt]$	$t_o\ [=\omega^{-1}[-\ln f/(D/vx)]^{1/2}$ D/vx	porous aquifers
ordinary dispersive (ODM) (Maloszewski & Zuber 1985)	$(4\pi t D^*/vxt_o)^{-1/2}\exp[-(1-t/t_o)^2 vxt_o/4D^*t]$	t_t; D^*/vx	fissured rock aquif. ($t_o \geq 1$ yr.) unsat. soil zone ($t_o \approx$ months)
parallel fissure dispersive (PFDM) (Maloszewski & Zuber 1985)	2)	t_o; D/vx; a; l	fissured rock aquifers
single fissure dispersive (SFDM) (Maloszewski & Zuber 1985)	3)	t_o; D/vx; a;	fissured rock aquifers ($t_o \leq 1$ month)

1) Definitions:

a	$(h^{-1/2})$	$(n_p/2b)\sqrt{D_p}$	R_p		retardation factor:
2b	(mm)	fissure aperture			V/V_m; t_t/t_o
D_p	(m^2/s)	molecular diffusion			$\approx 1+(n_p/n_f)$
D/vx		dispersion parameter	t_o	$(yr.^{-1})$	mean transit time of water:
f		amplitude ratio			V_m/Q; t_t/R_p
l		$(L-2b)/2\sqrt{D_p}$	t_t	$(yr.^{-1})$	mean transit time of tracer:
L	(cm)	fissure width			V/Q; $t_o\ R_p$
n_f	(1)	fissure porosity	V	(m^3)	total volume of water
n_p	(1)	matrix porosity	V_m	(m^3)	volume of mobile water
Q	$(m^3/yr.)$	volumetric flow rate	ω		$2\pi/1$ yr.

2) not quoted because practically not applicable

3)
$$2a/\pi \int_{\theta}^{\infty} u(r)[t-u(r)]^{3/2}\exp\left\{-r^2\{1-[u(r)/t_o]\}^2\right\}\exp\left\{-[a^2u^2(r)/t-u(r)]\right\}dr$$

with $u(\tau) = t_o/[4D\tau^2/vx]$
$\Theta = \frac{1}{2}(vxt_o/Dt)^{1/2}$
a (see annotation 1))

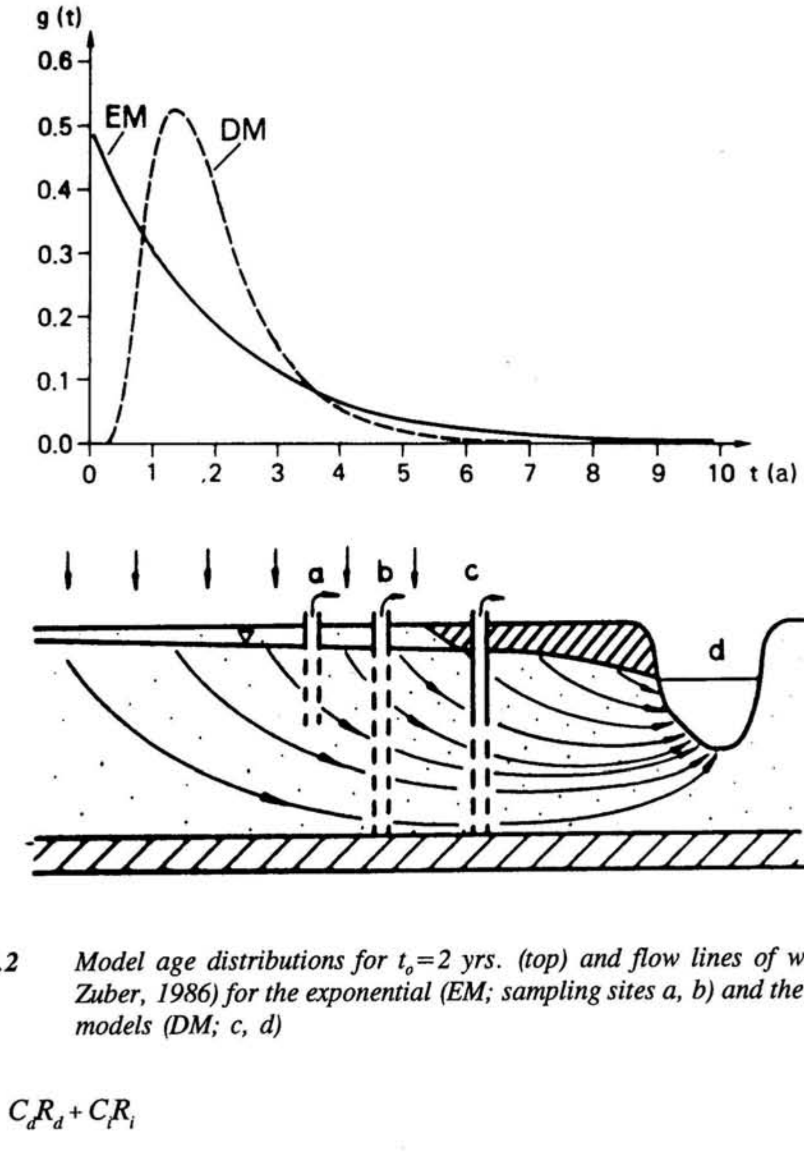

Figure 3.2 *Model age distributions for $t_o = 2$ yrs. (top) and flow lines of water (after Zuber, 1986) for the exponential (EM; sampling sites a, b) and the dispersive models (DM; c, d)*

$$C_tR_t = C_dR_d + C_tR_i$$

with $t + d + i = 1$ where d stands for event water, and i for older, pre-event water.

Today, isotopic flood hydrograph separations are best performed using the stable isotopes (cf. **Table 3.1**) because of the reduced atmospheric input of tritium. The applicability and accuracy of the isotope technique depends on the difference between isotope contents of the components to be separated from each other. For instance, for a difference of 5‰ for δ^2, accuracy of the resulting mixing ratio is 30%; for 30‰ it amounts to 5%.

Indeed, n independent tracers enable the separation of n+1 flow components. Since ^2H and ^{18}O as tracers have the same source (Fritz & Fontes, 1980; Moser & Rauert, 1980), the two-component case prevails in **Table 3.1** which causes complications where young soil water (as compared to older groundwater) isotopically influences the separation procedure and, therefore, the clearness of results. A good example of this case is given by Kennedy *et al.*, (1986) where delayed interflow was important in the runoff process, and even dominated the groundwater component. Additional observations of water chemistry might be helpful, but one must know that flow components are not directly comparable when using isotopically or chemically based definitions, and much more experience is needed in this field. In this context, refinement is also necessary in continuous isotopic hydrograph separation, instead of using isotopical bulk inputs where information about input concentration change is available during a single storm (Herrmann *et al.*, 1987) or snowmelt event (Stichler *et al.*, 1986a).

The most favourable conditions for hydrograph separation with stable isotopes occur during snowmelt seasons which are characterised by isotopically very light input signals as compared to the heavy pre-event components. This is valid for all investigated subpolar and medium latitude, alpine, highland and lowland environments (cf. **Table 3.1**), where successful multi-component separations are restricted to glaciated basins such as that of the Vernagtferner Glacier in the Ötztal Alps where the combined use of ^3H and ^2H enabled the simultaneous identification of ice, firn and snow meltwater components in streamflow (Oerter *et al.*, 1980).

For long-term separations of direct flows (over months, seasons, years; cf. **Table 3.1**) the theoretical stable isotope content of the indirect (pre-event) component is needed in the common case of the actual input isotopically oscillating around the background values of this component. It can be found indirectly by flow model g(t) application to tritium system functions as demonstrated by Herrmann *et al.*, (1989) for the Lange Bramke highland research basin using monthly values. For greater time steps and more distinct differences in isotope contents of the respective flow components, the application of the mixing formula to the weighted average isotope contents leads to realistic solutions for flow proportions, as demonstrated for half-years and years for the alpine Lainbach basin by Herrmann & Stichler (1980).

Finally, the benefit from major ions, specific compounds or even electrical conductivity measurements at relevant basin water fluxes should not be underestimated for discriminating natural hydrological catchment systems. However, the behaviour of reactive substances in the system is rather ambiguous when compared to conservative environmental isotopes.

3.2.4 Application of mathematical flow models g(t)

Mathematical flow models are basin response or exit age distribution functions g(t), thus corresponding to weighting functions that define the transit time of tracer transport through a system in the convolution integral, which describes the relationships between tracer input and output concentrations for steady-state conditions:

$$C_{out}(t) = C_{in}(t)\ (t-t')\ g(t')\ \exp\ (-\lambda t')dt'$$

With the decay constant $\lambda=0$ for stable isotopes, g(t) are ascertained by the response of the system to a pulse injection, and theoretically found by solving one (dispersion) or two transport equations (dispersion; diffusion) according to the model used. Relevant flow models in **Table 3.3** have recently been discussed by Zuber (1986). Their hydrological application demands solution of the mathematical inverse problem which consists of determining flow parameters such as the mean transit times (t_o; t_t) by solving the convolution integral. Respective flow parameter values are found by fitting theoretical $C_{out}(t)$-values as closely as possible to measured values. Problems might result from independent evaluation of findings by adequate field surveys, whereas the reliability of answers to the direct (migration) problem, which consists of assessing $C_{out}(t)$ as a result of specific $C_{in}(t)$ and g(t), is rather easy to verify. The development of migration (transport) models is, at least for conservative substances, considered the next urgent step in flow model application for ecosystem analysis, after the solution of the hydraulic inverse problem as discussed here. Some isotope hydrological bank filtration studies of Stichler *et al.*, (1986) and Maloszewski *et al.*, (1990a) have made progress in that direction.

Table 3.3 compiles those g(t) functions which are relevant to catchment hydrology, or are frequently applied for practical reasons. g(t) apparently differ in the number and meaning of fitting (flow) parameters but with all containing t_o as the hydrologically most important hydraulic parameter except in the ordinary dispersive model (ODM). The early single-parameter (t_o) exponential model (EM), which is mathematically equivalent to the well-mixing model, represents only a very rough approximation of reality, whereas the two-parameter (t_o; D/vx) dispersive model (DM) yields the most reliable solutions for t_o and porous media. Since the application of the convolution integral does not include any integration over the recharge area, values of the dispersion constant (D/v) are several orders greater than those found from artificial tracer experiments on macro-dispersion on the field scale. It may be expected that the apparent values of D/v in environmental isotope studies will be roughly equal to the length (x) of recharge zones measured along the streamlines.

For the case of double-porosity media (soils with macropores, or solid rock aquifers with fissures and fractures in a porous matrix) tracer diffusion in the matrix is an additional transport mechanism to be considered besides dispersion. According to Maloszewski & Zuber (1985) environmental tracer output concentrations can be interpreted using the ordinary dispersive model (ODM) provided that $t_o \geq 1$ yr. The two fitting parameters of ODM are: the mean transit time of tracer (t_t) and the purely mathematical parameter (D^*/vx) describing the variance of transit time distributions of the tracer in the system as a result of dispersion in macropores or fissures, and diffusion in the porous matrix.

Hydrologists should, therefore, be aware of the fact that they determine t_t instead of t_o and the whole (mobile plus stagnant) water volume (V) and not V_m which they are usually looking for; the related problems are discussed in Maloszewski *et al.* (1990b). Because of $R_p = t_t/t_o = V/V_m \approx 1 + (n_p/n_f)$ where R_p is the retardation factor as a result of tracer diffusion in the matrix, at least both porosities (n_p; n_f) should be known in order to calculate t_o from t_t ($t_o = t_t/R_p$) which is R_p times greater than t_o.

Two flow models in **Table 3.3** apply to the interpretation of isotope data from fissured rock aquifers: the three-parameter (t_o; D/vx; a) single fissure dispersive model (SFDM), and the four-parameter (t_o; D/vx; a; l) parallel fissure dispersive model (PFDM). With a great number of solutions possible due to the four unknowns, a reasonable application of PFDM to experimental tracer data is practically impossible. Therefore, Maloszewski & Zuber (1985) suggest using ODM, and at least SFDM for the case of $t_o \leq 1$ month, i.e. for short distance tracer experiments. This would allow indirect local solutions for n_f ($=2b/L$ with $2b = (n_p/a)\sqrt{D_p}$; for definitions see **Table 3.3**) which is, unlike n_p, difficult to determine but needed for assessing the retardation factor R_p ($\approx 1 + [n_p/n_f]$).

What makes the application of environmental isotopes in connection with g(t) functions so attractive and indispensable in some cases are the following basin parameters which hydrologists frequently require: the volume of mobile water (V_m) and effective porosity n_{eff} ($=V_m/V_t$; with V_t as the total volume of the reservoir) of the system under consideration and, last but not least, mean aquifer thickness H_{aq} ($=H_w/n_{eff}$; with H_w as mobile water volume V_m expressed as a water column). Another often neglected aspect with respect to catchment hydrological benefit is to supply check data as, for instance, the storage volume of mobile water (V_m) which should agree with the natural groundwater recharge rate for the system considering mean groundwater transit time (t_o). This is of course also valid for the output from any other quantitative approach to groundwater recharge. Information about true flowpaths of water in the basin should come from complementary artificial tracer experiments.

In this context, the time scale of one month and less which was suggested for SFDM applications allows the interpretation from their breakthrough curves of vertical tracer transport through topsoils or weathered and fractured bedrock, or lateral transport through vein fissures and major faults. If the same experimental data are ascertained for mono-porous media, they would be interpreted with the DM model. As with many other simple applications of 1-D or 2-D analytical numerical solutions for the mass (tracer) transport equation and determination of transport parameters - such as flow velocities of water and dispersivities - note that this should be always done by calibration of chosen transport models on experimental data (Maloszewski & Zuber, 1990). If, for example, the approximative momentum method is used for interpretation of artificial tracer data, one should always check that the calculated concentration curve fits the experimental data, otherwise the transport model may produce wrong results. Another problem is the highly deficient mass recovery in many tracer experiments which lead to false parameter predictions if not adequately considered.

To conclude, g(t) transfer functions apply to a great variety of time scales and both environmental and artificial tracers, thus contributing to various dimensions of hydrological process studies on a small catchment scale.

3.3 EXPERIMENTAL RESULTS

The following discussion of experimental results from the application of the environmental tracer technique focuses on the benefits for the recognition and confirmation of dominant catchment hydrological processes such as runoff formation and groundwater recharge, with a short comment on artificial tracer experiments.

For this purpose, the present state of knowledge on hydrograph separation and mean transit time determination will be briefly described with reference to the literature reviews presented in **Tables 3.1 and 3.2**. By taking the extensive multidisciplinary research activities in a complex Central European research basin as an example, a realistic evaluation of present progress and future needs is developed with respect to specific hydrodynamic processes.

3.3.1 Global synopses of d and t_o resp. t_t

The updated listings of experimental work on direct runoff (R_d) separation and mean transit time (t_o, t_t) calculation in **Tables 3.1 and 3.2** in principle confirm the comments by Stichler & Herrmann (1982, 1983) and Herrmann (1989, 1993) thus also preserving earlier research needs as well. These concern, above all, the need for more widespread use of the stable isotopes (^2H, ^{18}O) instead of tritium because of the rapidly decreasing concentrations of ^3H inputs from precipitation. Furthermore, extended and more systematic regional applications of the isotope technique should contribute to developing a modern regionalisation concept for hydrodynamic catchment systems as discussed below. Accordingly, further refinement of combined experimental and modelling techniques seems necessary for solving difficult hydrodynamic interactions on the small basin scale.

According to **Table 3.1**, the finding that groundwater (pre-event) rather than surface or near-surface (event) water dominates in rain and snowmelt flood hydrograph generation is the most outstanding hydrologic result. In most cases, only minor fractions (less than 10% of actual input volumes) would rapidly leave basins within days. Therefore, classical runoff formation concepts should be revised for both quantitative and qualitative approaches to natural basin systems. The proposed hydraulic turnover mechanism will be discussed later.

Unfortunately, as already mentioned, the methodical problem of continuous hydrograph separation with changing input concentration remains unsolved. Methodical progress is limited to long-term d-separations for months, half-years and years in connection with theoretical stable isotope contents of the indirect runoff component found from joint use of ^3H and ^2H or $^{18}0$ and the appropriate mathematical flow models (Herrmann *et al.*, 1986, 1989).

As to the findings for mean transit times in **Table 3.2**, certain scepticism might arise when considering the warning remarks of Maloszewski *et al.* (1990b) about the meaning of t_o from DM for the case of double porosity storage media which are quite common. In fact, there are many examples of this type, with the results thus representing t_t rather than t_o and, therefore, an overestimate for the mean transit times of water. One such fallacy concerns the Lainbachtal basin (Maloszewski *et al.*, 1983). Hence, users of mathematical flow models g(t) should be conscious of that problem.

The dominant use of ^3H for determining t_o of very young groundwater may diminish with the increased use of the stable isotope technique (but which has to be improved for this purpose) or even of ^{85}Kr where large water samples of 0.2 m^3 and budgets for costly analyses are available. Moreover, ^3He as a decay product of bomb tritium has proved to be a reliable but not very practicable tracer (Weise & Moser, 1987). In the case of ambiguous ^3H results (recent or >100 yrs) the combined use of helium-3 and tritium ($^3He/^3H$ method) may be advantageous. From these considerations it follows that ^3H is difficult to replace as a hydrological tracer in the water cycle.

3.3.2 Lange Bramke research basin

First instrumented in 1949, Lange Bramke (0.76 km^2, 543-700 m a.m.s.l., 90% forested) in the Harz Mountains, Germany belongs to the best studied drainage basins of the world, with the hydrologic tracer investigations starting in 1980. **Fig. 3.3** represents the hydrological model based on major experimental results where water fluxes and storage volumes have been assessed from isotopic data as described in a comprehensive report by Herrmann *et al.*, 1989.

Basic findings such as $d \approx 10\%$ ($=0.06 \pm 0.02$ $10^6 m^3$), i.e. direct flow most frequently amounts to $\leq 1\%$ of actual input volumes, and $t_o \approx 1.5$ yrs. ± 1 month for the groundwater system, are fully compatible with the literature findings in **Tables 3.1 and 3.2** considering that in reality t_t instead of t_o was published in many cases. Moreover, Zuber *et al.*, (1986) confirm that it is valid to assume steady-state conditions for flow modelling for at least Lange Bramke, and Herrmann *et al.* (1987, 1989) found that interflow is negligible (cf. **Fig. 3.3**).

One main consequence from the surprisingly high groundwater supply to total runoff is a rather short t_o, in the order of 1.5 yrs, corresponding to a considerable annual recharge rate of 650 mm for the active groundwater fraction (in the hydrodynamic sense), as compared to 210-290 mm/yr found from traditional computation techniques. This is apparently due to many inadequate runoff generation concepts which are, therefore, to be reexamined.

The turnover mechanism of water during single flood events in the frequent case of permeable soils and subsoil-rock interfaces due to efficiently draining macropore and fissure systems has been discussed in detail by Herrmann *et al.* (1987). Accordingly, during the infiltration process groundwater potential spontaneously increases preferentially at lower slopes, mainly because of compression of the capillary fringe and a natural hydraulic barrier at the fissured rock/porous aquifer interface which accounts for hydraulic conductivities in the order of 10^{-5}-10^{-6}m/s (Schöniger & Herrmann, 1990). Quick and quantitatively important lateral water transport occurs through some major cross faults dividing the fractured rock groundwater system.

Discharge and groundwater tables show significantly close relationships which are hysteretic. One of the main conclusions from such combined isotope hydrological and hydrogeological investigations is that exfiltrating groundwater can contribute considerably to flood hydrograph generation, and may be subsequently recharged through directed macropore/fracture systems of the unsaturated zone, the quantitative balance of the aquifer thus being maintained. In other respects, such short-circuited subsurface storage systems may cause water quality problems where there is limited buffering capacity and where air or surface pollution plays an important role.

A computer-aided analysis of hydrograph recession limbs over almost 40 years of flow record at Lange Bramke yielded runoff components and residence times based on the linear storage concept (Schwarze *et al.*, 1989). This led to comparable but even more detailed results for direct flow proportions and mean residence times of the quick, delayed and slow subsurface flow components (Schwarze *et al.*, 1991). This promising result encourages the continued development of a feasible method to regionalise hydraulic system features such as residence times and direct runoff proportions by only using official basic hydrological and climatological network data.

Additional information from artificial tracer experiments, apart from the single well technique (Drost, 1983), commonly concentrate on determining hydraulic connections, flow velocities and dispersivities, but some also deal with hydraulic conductivities and the difficult determination of fissure porosities for fractured rock aquifers by combined pumping and dye tracing as proposed by Maloszewski & Zuber (1990). Preliminary results for the Lange Bramke basin are given in Herrmann *et al.* (1989), but interpretation of multiple well test results in fractured media remains a crucial problem as compared to the positive experiences with porous aquifers.

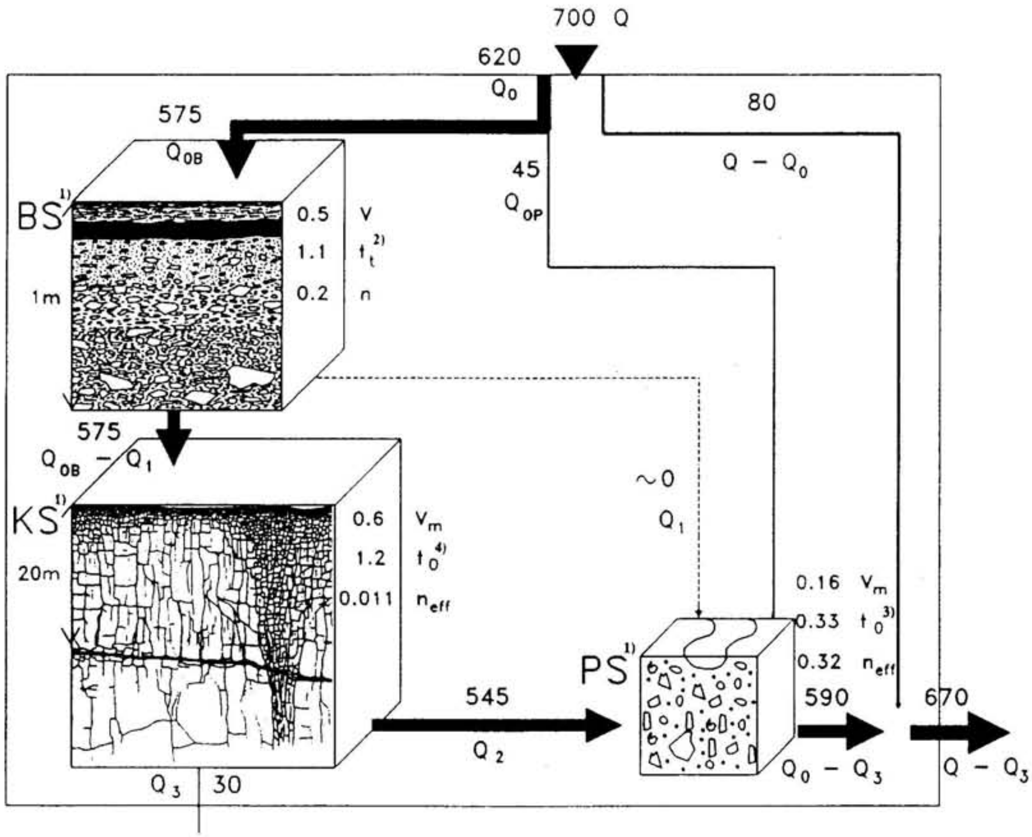

Figure 3.3 *Hydrological system model of Lange Bramke basin with mean annual flow rates 1980-89 (in mm water column) and reservoir features (V and V_m in $10^6 m^3$; t_o and t_t in yrs.)*

[1] BS: unsaturated soil zone (residual weathering, allochthonic pleistocene solifluidal materials on fissured and faulted rock)
PS: porous aquifer (valley filling)
KS: fissured rock aquifer (folded and fractured lower Devonian sandstones, quartzites, slates)
[2] from application of flow model ODM
[3] from application of flow model DM
[4] from $t_o = t_t/R_p$ with $R_p = 1.45$ [determined from weighted mean areal porosities (n_p; n_f)]

Furthermore, such point tracer applications in the saturated zone are of only limited, i.e. local value. The same is true, of course, for most artificial tracer experiments in the unsaturated zone (IAEA, 1985) leading to point information about seepage velocities. Local importance might be also attributed to most artificial injections in hydrologic karst systems. However, a recent experiment of basinwide relevance can be reported from Lange Bramke where artificial pulse injections of a major cross fault with heavy water, bromide, uranine, eosine and naphthionate led to some encouraging hydraulic results supporting the quick seepage and quantitatively significant groundwater exfiltration hypotheses during single events. Hydraulic parameters (t_o, D/vx, a; cf. **Table 3.3**) for this major subsurface drainway of the basin have been identified by applying SFDM (Maloszewski & Herrmann, 1993).

3.4 CONCLUSIONS

Existing runoff formation and groundwater recharge concepts still ignore the basin turnover mechanisms of water. In the future, adequate hydraulic algorithms should be developed and prepared for inclusion in appropriate modular hydrodynamic basin models for the simulation of relevant runoff components (e.g. direct, delayed direct and groundwater flows) as, for instance, described in Herrmann *et al.*, (1990). By starting with the updated modular Brook90 version of the BROOK model (Federer & Lash, 1978) for forested basins and the ACRU model (Schulze *et al.*, 1989) for agricultural basins, or with similar conceptual system models which are suitable for both kinds of ecosystem and a daily data interval, a priority task consists of developing adequate transfer functions for the subsurface storage systems. Finally, model calibration and verification of water quantities are assumed to consider the hydraulic results from environmental tracer studies which obviously represent good approximations of reality.

Accordingly, the synopses of **Tables 3.1 and 3.2** should contribute to such a model development by considering closely connected regional varieties of catchment systems and hydrogeological conditions. Realistic simulation of discharge quantities for single runoff components from small hydrologic systems would then constitute a big step forward towards areal dissemination, i.e. regionalisation of hydrodynamic knowledge, as for instance proposed by Schwarze & Herrmann (1993). Moreover, such a hydraulically-based basin model should allow more reliable interpretations of basin output concentrations of conservative and reactive solutes (provided that information about reaction kinetics is available) thus providing benefit from the conjuctive use of both conventional and expensive isotopic investigation methods.

Flow Regimes from International Experimental and Network Data (FRIEND)

Volume III Inventory of streamflow generation studies

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Mark Robinson

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