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A Fifty-year chronicle of tritium data for characterising the functioning of the Evian and Thonon (France) glacial aquifers

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Abstract[1]

Using lumped models and a transfer function model, this paper deals with the interpretation of exceptionally long (up to 50 years) and precise tritium chronicles characterizing the rainfall, recharge (efficient rainfall) and outflow from various types of glacial aquifers from the French Alps (Evian-Thonon area). The efficient rainfall tritium chronicle was computed from tritium measurements performed for 11 years (1969-1979) in a lysimeter. The evapotranspiration induces a mean 15% drop of the annual tritium signal. The three superficial glacial aquifers (two fluvio-glacial kame terraces and a lateral till) provide similar results: a best fit with an exponential flow model (EM) (playing the major role) combined in parallel with a piston flow model (PFM), and a rather short mean transit time (T: 5 to 7 years). The deepest mineral aquifer (Evian) can only be fitted with the in a series combination of a highly dispersive model (DM; T 68 years, DP=0.5) and a piston flow model (T 2.5 years) or, better, by the in a series combination of an EM (T 8 years) modelling the subsurface aquifer and a DM (T 60 y, DP=0.75) and the same piston flow model (T 2.5 years) modelling the deep mineral aquifer, this latest combination of models providing the following parameters: T 70 years and median transit time 45.5 years. It is also to be noted that a very small part of the efficient rainfall, about 1.3%, avoids both the EM and the DM, and directly enters the PFM (at the Northern limit of the Gavot Plateau). These models are very sensitive regarding the T (±1 y, 0.25 y for the PFM), less so with DP. These results will prompt hydrologists to (re)work historical data

[1] **Abbreviations:**
amsl: above mean sea level (elevation)
CRG: Centre de Recherches Géodynamiques
DM: dispersion model
DP : dispersion parameter
EM: exponential flow model
BMMEP: in parallel combined piston flow and exponential models
IAEA: International Atomic Energy Agency
LM: linear model
LPM: combined linear flow and piston flow model
NMW: Natural Mineral Water
T: Mean Transit Time
PFM: piston flow model
TU: Tritium Unit
y BP: year Before Present

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to determine if important hydrologic information is available. The interest and limits of such a modelling, also for other constituents than tritium, along with the future for tritium as a tracer are discussed and it also provides new insights on the structure and functioning of alpine paleo glacial hydrosystems.

Highlights

- interpretation of an exceptionally long tritium chronicle (rainfall, natural mineral water and superficial springs)
- accurate characterization of a hydrological transfer function
- discussion of the interest and limits of lumped models
- new insights on the structure and functioning of glacial aquifers

Keywords

Evian; lumped model; natural mineral water; inverse modelling; tritium; glacial aquifers

1. Introduction - History of isotope measurements in the Evian-Thonon area

To evaluate the potential hydrogeological vulnerability of hydrosystems to anthropic pollution and predict any future changes, the pattern of the transit time of water in the aquifers should be defined. Isotopic measurements are presently used in field investigations and research projects to improve qualitative understanding of hydrosystems and identify the processes involved in water transfer (Turner and Barnes, 1998). Environmental tracers (^{18}O, ^{2}H and tritium) and specific flow models have been used by numerous authors to estimate the transit time of water in hydrological systems (Herrmann et al., 1990; Stewart and McDonnell, 1991; Maloszewski et al., 1983, 1992, 1995, 2002; Maloszewski et Zuber, 1982; Holko, 1995; Amin and Campana, 1996; Rodhe et al., 1996; Vitvar and Balderer, 1997; Etcheverry and Perrochet, 2000; Uhlenbrook et al, 2002; Viville et al., 2006; Hagedorn et al., 2011; Morgenstern and Daughney, 2012). One difficulty in estimating the transit time of water is the need of the input function, i.e. the concentration of tritium in the infiltrating water while only tritium in precipitation and precipitation amount are available. McGuire and McDonnell (2006) tried to construct better model to estimate the input function. Moreover, due to the insufficient length of input and output chronicles, hydrosystems with transit times of several decades sometimes cannot be studied.

Tritium (^{3}H) is a natural isotope of hydrogen (half-life 12.32 years; Lucas and Unterwerger, 2000), naturally present in the atmosphere at low concentrations (5 to 10 T.U.), and at higher concentrations (up to about 6000 T.U.) due to above ground nuclear weapons testing beginning in the mid 1940s, then dramatically increasing with the development of the hydrogen bomb (first test was in November 1952) until 1963, the year when the atmospheric atomic test was banned (the few small nuclear tests above ground since 1980 have just been too small to make any impact on the tritium burden). However the beginning of tritium recording started in August 1953 (Maloszewski and Zuber, 1994). It has been widely used in hydrology to estimate aquifers' mean transit time (or residence time) since it is a constituent of the water molecule and its concentration in rainfall has varied during the past sixty years, giving a well-known input function for hydrological models. Tritium better captures the longer term components of flow and recharge than other typical approaches (chloride or stable isotopes for instance) (Stewart et al., 2010).

The first tritium measurements of groundwater from the Quaternary deposits of the Geneva Lake basin were taken in the fifties, and have been densified since 1964, shortly after monthly analyses of tritium from rainfall began in June 1963 at the Thonon (France, Fig. 1) Centre de Recherches Géodynamiques (CRG, a branch of the Paris VI University), which was a member of the International Atomic Energy Agency (IAEA) isotopic monitoring network. As early as 1963 this new isotopic tool proved useful in characterizing the hydrogeological functioning of the complex glacial aquifers of the area (Blavoux et al., 1964; Fontes et al., 1967). A tritium

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content of several hundred TU (Tritium Units) was found in 1964 in the Thonon unconfined aquifer while in contrast the water from the Evian confined aquifer located about ten kilometres eastwards was still only a few TU. Sampling was subsequently performed quarterly in this area to monitor the evolution of the tritium signal until recently, providing probably the longest available detailed temporal record of tritium in groundwater.

In that framework, this paper deals with the interpretation of such an exceptionally long tritium chronicle characterising both the recharge and the outflow from various types of glacial aquifers, using lumped models and a transfer function model. It discusses the interest and limits of such modelling and also provides new insights on the structure and functioning of ancient alpine glacial hydrosystems. Such glacio-fluvial hydrosystems, either quite old, or "fossils" (see for instance Blavoux and Dray, 1971; Dray, 1993; Caballero et al., 2002; Nicoud et al., 1993; Henriot et al., 2013; Bayer et al, 2011) or subactual (see for instance McClymont et al. 2011, Rodhe and Seibert, 2011; Clow et al, 2003) are of high interest for their water resources in the many regions of the world that were glaciated, mostly during the Quaternary era, or are still at the margins of glaciated areas: the northern part of Eurasia, North and South America, alpine regions, etc. (see for example Roy and Hayashi, 2009; Beckers and Frind, 2000, 2001).

Figure 1 – Location and schematic geological map of the Evian-Thonon area. Location of springs and boreholes. Note the location of the rain gauge used since June 1963).

2. Geological and hydrogeological setting of the Evian-Thonon area

The studied area is about 20 km long by 10 km wide along the southern shore of Lake Geneva (Fig. 1 & 2). As in most of the Lemanic basin (Fiore et al., 2011), the land surface is almost exclusively covered by thick (up to 400 m or more in Evian; Triganon et al., 2005) Quaternary glacial sediments deposited over a major glacial erosion surface truncating the cemented, folded and thrusted Alpine rocks. The lake was dug during Quaternary glaciations by ice originating from the upstream Rhône valley catchment basin. The deep valley of the Dranse River (Fig. 1), the bottom of which exposes much older pre-Quaternary geological formations, divides the study area into two independent zones both from a geological and a hydrological viewpoint.

East of the Dranse River, the **Evian area** Quaternary geological formations are composed of glacial sediments deposited along the Rhône glacier's southern margin (Nicoud et al. 1993), well known from several kilometres of cored boreholes (Blavoux et al. 1971; Blavoux, 1988; Triganon et al. 2005) (Figs 1 & 2a):

- the shape of the pre-Quaternary substratum is the result of erosion from the pre-Wurmian pulsations of the Rhône glacier. This paleotopography is irregular, reaches an elevation less than 100 m amsl at Evian, and progressively rises towards the South, up to a few pre-Quaternary outcrops at the highest points of the Gavot Plateau;
- this depression is filled by about 100 m of thick sands and gravels (older than the last glacial cycle), corresponding to the northern tip of a paleodelta of the Dranse River, covered by a subglacial till up to 200 m thick from the Würmian maximum (65'000 to 35'000 y BP, Triganon et al., 2005), during which the glacier reached an elevation of about 1300 m amsl in the Evian area. This till surely prevents any groundwater flow between the pre-Quaternary substratum and the sediments described here below;
- glacio-lacustrine sediments (small local streams paleodeltas within lacustrine silts) from the "Inferior Complex", older than 30'000 y BP, sedimented during a major retreat phase of the Rhône glacier, overlie this till up to about 390 m amsl;
- the Gavot Plateau glacial margin formation, over 200 m thick, results from the progressive rise of the Rhône glacier during the Würmian "Lemanic stage" (30'000-27'000 y BP, Triganon et al., 2005, Fiore et al., 2011) when the glacier reached the

Geneva city area. It is composed of a subglacial till in the Evian area and up to 13 superimposed subglacial and lateral tills with peatbog sediments below the plateau;

- the partial glacier retreat down to an elevation of about 480 m amsl, and then to about 430 m amsl at Evian, was responsible for the deposition of the glacio-lacustrine sediments of the "Terminal Complex" (25'000-21'000 y BP, Triganon et al., 2005) with highly permeable (10^{-3} to 10^{-4} m/s) sands and gravels in the fans of the local paleostreams and low hydraulic conductivity silts laterally;
- finally, the last flood of the Rhône glacier, with the western tip of the glacier again reaching Western Lake Geneva - "le Petit-Lac" - (Copet and Nyon readvances, Fiore et al., 2011), covered all these sediments with a final subglacial till (the "terminal till") of 30 to more than 50 m thick up to an elevation of 650 to 750 m amsl in Evian. This till locally comprises kame terraces, mostly deposited at the mouth of local streams during the final deglaciation, and lateral tills.

The Natural Mineral Water (NMW) emerging from the confined aquifer of the "Terminal Complex", particularly at the Evian-Cachat spring, the highest-yield spring in Evian at 400 m amsl, has been famous since 1789. Since that time, it has been widely used for its medicinal properties (hydrotherapy). The Société Anonyme des Eaux Minérales d'Evian now bottles "évian®" NMW from these mineral springs. The terminal till prevents any infiltration of surface water into the aquifer and confines it in the downstream part of the aquifer. Recharge occurs on the Gavot Plateau, above the 650-750 m amsl maximum elevation of the last glacier advance (Blavoux, 1978; Henriot et al., 2013). Then the water slowly flows down and northward within the Gavot Plateau glacial margin formation that comprises several superimposed small confined and unconfined aquifers flowing mostly in the lateral tills, separated one from the other by the basal till layers that act as semi-confining units (Henriot et al., 2013). The mean elevation of the catchment area of the aquifer was estimated at 950-

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1000 m amsl by comparing the ^{18}O content of the Evian NMW aquifer with the ^{18}O content of precipitation at several altitudes and with the ^{18}O content of low discharge subsurface springs with a well-known catchment area altitude (Blavoux, 1978). Mean annual rainfall on the catchment area rises to 1’200 mm, and effective rainfall to about 750 mm. The specific hydrochemical characteristics of the Evian NMW, calcium (80 mg/l) and magnesium (26 mg/l) (that dominate the cations) on one hand, and silica (15 mg/l) on the other hand, are respectively the result of water-rock interaction with rocks of the aquifer (limestone and dolomitic sands) that were eroded locally, and with rocks (granites, schists, green rocks…) from buried lateral tills that were imported from the Rhône valley catchment by the glacier. The pH is neutral (7.2) and bicarbonates (360 mg/l) and sulphate (13 mg/l) dominate the anions. Due to the high inertia of the hydrogeological system, the mineral content and temperature (11.6°C) at the spring are perfectly stable (below the analytical thresholds) and its discharge shows less than 5% annual variations.

Small superficial aquifers are locally imbedded within the sands and gravels from the alluvial kame terraces interstratified in or deposited above the terminal till. These aquifers are recharged by the local precipitation. These waters can be easily distinguished from Evian Natural Mineral water as the mineral content is different from that of Evian NMW, for example in the Ca/Mg ratio and silica content.

Figure 2 - Schematic geological cross-sections of the Evian (2a - top) and Thonon (2b - bottom) areas (see the location of the cross-sections on Fig. 1).

West of the Dranse River, the **Thonon slope** is mainly covered by the fifteen (Gagnebin 1933) Dranse River glacio-fluvial kame terraces (sands, gravels, and - limestone and less numerous crystalline - large pebbles) progressively deposited westwards against lateral tills (Figs. 1 & 2b), parallel to the Rhône glacier, during glacier retreat phases. The highest terraces deposited during the Gavot Plateau retreat (Geneva stage: 25’000 to 23’000 y BP)

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and the lowest during the final deglaciation of the "Petit Lac" stage (21'000 to 18'000 y BP). They all lie on a subglacial till contemporaneous with the formation of the Gavot Plateau ("Lemanic" stage), and the lowest lie on the till associated with the last flood of the Rhône glacier ("Petit Lac" stage), which there confines the oldest terraces. The terrace aquifer is generally unconfined and flowing northward. It exhibits a rather high hydraulic conductivity (about 5.10^{-4} $m.s^{-1}$; Blavoux, 1978). Numerous springs discharge from the terraces and some of them, "Les Blaves" for instance, are impounded for drinking water by Thonon town waterworks (Figs. 1 & 2b). The "Versoie spring" discharges from one of the lowest terraces at an elevation of 465 m amsl. Known since Roman times, it has been exploited as natural mineral water, known as "Thonon Water", for bottling and hydrotherapy since 1864. The "spring" is tapped with a 15 m deep radial well equipped with a submerged pump. The mean elevation of the Versoie spring catchment area has been estimated at about 600 m amsl from natural ^{18}O tracing (Blavoux, 1978; Dray, 1993). It corresponds to permeable kame terraces located between 650 and 465 m amsl, where mean annual precipitation reaches 1000 mm and effective rainfall 550 mm.

3. Material and methods - Sampling and analyses

Monthly samples of rain water have been collected since July 1963 at the CRG Thonon rain gauge station, at 385 m amsl on the shore of Lake Geneva and 10 km away from Evian (Fig. 1 and Fig. 3). Sampling consists of collecting and protecting from evaporation in a cumulative rain gauge all the rain falling on a 2000 cm^2 plastic rain gauge during each month.

Figure 3 CRG Thonon tritium monthly rainfall time series (since june 1963; previous annual data reconstructed from the Ottawa time series, see details in the text). Note the logarithmic scale of the Y axis and the 1960s very high peak.

The tritium levels of three springs, two NMW (Evian-Cachat and Versoie) and one superficial aquifer spring (Maxima), have been monitored since 1964; that is, during a crucial period

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after the 1963 major tritium peak in precipitation (Figs. 4A and 4B). Evian-Cachat and Versoie were sampled quarterly for 15 years (from 1964 to 1979); annual sampling continues today. Although more widely spaced, measurements on the Maxima spring are sufficiently numerous (22) that tritium restitution can be interpreted. Finally, the Larringes-Pugny well located on the Evian impluvium was sampled from 1966 to 1969, a time sufficiently long and close to the 1963 major tritium rain peak to give indications of the transformation of the tritium signal upon input to the system.

The water catchwork of the Evian-Cachat spring, built in 1911, reaches the sandy confined aquifer at the end of a 66 m long horizontal gallery crosscutting the till cover. As the aquifer is confined, the gallery is completely sealed and neither the aquifer nor the water is visible there. The artesian flow is limited by a valve to maintain the pressure. The samples were collected at a sampling tap located in the gallery; Natural Mineral Water is not treated, and of course not sterilised, consequently the samples were not subject to any treatment. Some sporadic measurements were made for comparison on the other boreholes of the same mineral hydrogeological system. The Versoie samples were collected on the pumping well from a tap located at the top of the pumping tube. The Maxima spring (Figs. 1 & 2) emerges from a superficial aquifer. It is located near the Lake's shore at 380 m amsl within a sandy kame terrace of the terminal till on the lowest part of the Evian area; its more than 40 years of tritium data are worth studying. The Larringes-Pugny well is located on the Gavot Plateau at 790 m amsl. This well exploited for drinking water taps a superficial aquifer of the Gavot Plateau glacial margin formations (4 m thick sand and gravel locally covered by 2 m of clay).

All the water samples were analysed for tritium at the Centre de Recherches Géodynamiques (CRG) in Thonon until it was closed in April 2005, and since then have been analysed at the Hydrogeology Laboratory of the University of Avignon. Tritium measurements were obtained by liquid scintillation counting after electrolytic enrichment. The associated error on the data varies with the tritium concentration in the samples; it is 1 TU for recent samples and was about 1 or 2 tens of TU for the samples which contained several hundred TU (see Figs 4A, 4B and 5). The limit of detection of the CRG equipment was 5 to 8 TU during the earliest years and about 1 TU since 1972 (see Fig. 5).

4. Theory/calculation - Tritium restitution modelling

Lumped parameter models are used in this study to estimate the transit time distribution of the groundwaters discharging from the Evian - Thonon area. In the lumped parameter approach, the output concentration is related to the input concentration using the following convolution integral (Equ. 1) (Maloszewski and Zuber, 1982; Zuber, 1986(a) and (b)):

$$C_{out}(t) = \int_0^{\infty} C_{in}(t - t')e^{-\lambda t'} g(t')dt' \qquad (1)$$

where t is the sample date, t' is the transit time of single tracer particle through the system, λ is the decay rate of the tracer, per year, $C_{in\_}$ and $C_{out\_}$ are the input and output tracer contents, and g(t') is the weighting function defining the transit time distribution.

Lumped parameter models assume a steady-state flow and that the selected tracers move with the water molecules, which is why tritium is an ideal tracer. Such a steady-state flow assumption is suitable for modelling the Evian and Thonon area aquifers; in fact, the water volume in these aquifers does not change significantly on a seasonal and yearly time scale as the variability in interannual discharge measurement is less than 5 % of the total annual discharge for each spring. In addition, as two of these springs (Evian-Cachat and Versoie) are bottled NMW, the spring discharges are monitored. In this paper, it is also considered that the aquifer system is mono-porous, thus that the portion of immobile water is negligibly small, which permits to use the mean transit time found from tritium as that of water.

The following lumped parameter models were considered (Yurtsever, 1995; IAEA, 1996): the piston flow model (PFM), exponential flow model (EM), linear model (LM), and dispersive model (DM). According to the tritium restitution curves and the hydrogeological conceptual

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models (see chapter 2.), some of these models were also combined in parallel, thus constituting Binary Mixing Models (a terminology proposed for instance by Katz et al., 2001), which means a simple mixing of two - or more - endmembers occurring at the spring/outlet. Piston flow and exponential model (named after BMMEP) were combined. Some of these models were also combined in series. All these models take into account the natural radioactive decay of tritium. The choice of model/combination of models was made as a function of the appearance of the observed signal at each spring and the available knowledge on their hydrogeological functioning. Among the different computer codes available in the hydrological literature (for example FLOWPC, Maloszewski and Zuber 1994, 1996), we first used LUMPED (Ozyurt and Bayari 2003, available at http://www.sukimyasilab.hacettepe.edu.tr/english/software.shtml). In a second phase, the complex signal measured at the Evian-Cachat spring was interpreted using the TEMPO software developed at BRGM by Pinault (2001) (see for instance also Dörfliger et al., 2008), in which the equations of the models described by Maloszewski and Zuber (1994, 1996) have been implemented (the models developped by Maloszewski and Zuber have also been used to validate TEMPO). FLOWPC and TEMPO have the same input requirements and provide similar outputs. The subsurface aquifers of the Versoie, Larringes-Pugny, and Maxima springs were characterised using the exponential model (EM), and finally with a Binary Mixing Model combining exponential and piston flow models where the exponential part plays the major role. The Evian-Cachat aquifer was characterised with an in a series model combining dispersive and piston flow models (Fig. 10), and secondly with an in a series model combining exponential, dispersive and piston flow models (Fig. 13).

The piston flow model (PFM) assumes that all the flow lines have the same transit time and thus, the response function is the Dirac delta function $g(t)=\delta(t-T)$, with T: mean transit time. Hydrodynamic dispersion and diffusion are negligible (Maloszewski and Zuber, 1982, 1994).

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Tracer concentration is only affected by radioactive decay. It ideally corresponds to a confined aquifer with a narrow recharge area far from the sampling point.

In the EM, the distribution function of the transit time of water in the aquifer is exponential ($g(t)= T^{-1} exp(-t/T)$, with T = mean transit time, thus the average age of all parcels of water that entered the system and reached the discharge point). Water travelling along the shortest flow line has a transit time equal to zero while water travelling along the longest flow line has an infinite transit time. An exponential distribution of transit times in an aquifer can corresponds to multiple hydrogeological situations, including those where hydraulic conductivity decreases with depth or the aquifer has uniform permeability and porosity (Eriksson, 1958; Maloszewski and Zuber, 1982). Use of this model assumes that (1) there is no mixing of water among the various flow lines during transfer in the hydrosystem and (2) mixing of the water from the various flow lines occurs at the level of the sampling point.

The dispersive model (DM) allows the effects of transit time dispersion in the flow direction to be taken into account. The distribution function of transit times in this model uses the one-dimensional solution of the dispersion equation for a semi-infinite medium (Equ. 2) (Maloszewski and Zuber, 1982):

$$g(t) = \left(\frac{4\pi\, DP\, t}{T}\right)^{-\frac{1}{2}} exp\left[-\frac{\tau}{4\, DP\, T}\left(1-\frac{t}{\tau}\right)^{2}\right] t^{-1} \qquad (2)$$

with T = mean transit time and DP (without dimension) the dispersion parameter: the dimension of variance of the dispersive distribution of the transit time. This additional calibration parameter DP depends mainly on the distribution of transit times of the water (shape of the flow lines) in the aquifer. It thus includes apparent dispersion of the tracer in the system plus also apparent dispersion of the tracer in the recharge zone (recharge zone is in the lumped parameter approach reduced to one point, while space distribution of the input signal through the surface of the recharge zone, yields additional “dispersion”), and also the variable infiltration conditions over the recharge area. DP is generally greater (by one or more orders

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of magnitude) than the hydrodynamic dispersion parameter which usually represents only the dispersive properties of the material (Maloszewski and Zuber, 1994).

The distribution functions of transit times providing the best correspondence of the simulated concentrations to the experimental data were obtained by trial and error until a graphically good fit and a "good" Nash coefficient were obtained (at best, the Nash coefficient is 1).

5. Results

5.1. Observation results

The annual tritium levels of the precipitation at Thonon CRG (Table 1) have been computed from the monthly values weighted by the precipitation rate. For values prior to 1963, a regression (Equ. 3) was established with the annual levels measured in Ottawa from 1964 to 1985, a station at the same latitude as Thonon that has operated since 1953 (Brown, 1970).

$$[^3H]_{Thonon} = 0.892[^3H]_{Ottawa} + 9, \quad n=22, \quad R^2=0.9 \text{ (Molicova, 1991)} \qquad (3)$$

This chronicle of annual levels constitutes the input signal for the hydrogeological systems of the Evian-Thonon area (Figs. 4A and 4B). Since the start of thermonuclear weapons testing in the early 50's, continued more and more intensively until 1963, atmospheric tritium levels increased enormously. The levels in rainwater at Thonon reached several thousand TU in 1963, with a spectacular rise between 1961 and 1963 (Fig. 4B). From 1963 to 1966, nearly all atmospheric nuclear tests were stopped and levels dropped sharply with a pseudo-period close to one year, thus defining a very characteristic peak on the multiyear scale. From 1967 to 1980, atmospheric nuclear tests were of very low capacity, with less than 3% in total of all the tritium produced artificially, nevertheless leading to fallout that decreased slowly from 200 to 100 TU. Since 1980, there have been no atmospheric nuclear tests. With a pseudo-period of one year and according to tritium natural radioactive decay (half-life: 12.32 years), the level should have reached the natural level (5 to 10 TU) around 1985. However, it remained between 30 and 20 TU until 1996; this is due to the feedback from the oceans and surface

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waters that contain higher levels of tritium (Fine and Ostlund, 1977; Fine et al., 1987; Michel, 1992) and also the influence of the civilian nuclear industry. Since 2000, it seems that the rain signal at Thonon has entered a permanent regime around a value of 9 TU.

Table 1 – (i) Rainfall tritium mean annual content at the Thonon CRG rain gauge, (ii) computed tritium efficient rainfall signal, and (iii) mean annual tritium content at the Evian-Thonon area springs

Figure 4 – Rainfall and springs tritium mean annual content at the Thonon CRG rain gauge and at the Evian-Thonon area springs (top: logarithmic scale; bottom: arithmetic scale)

The tritium content in rainfall shows a seasonal variation. Monthly levels in Thonon have a maximum in June-July and a minimum in November-December. For the temperate climate zone of the Northern hemisphere, this springtime increase results from direct discharge of tritium in the stratosphere toward the troposphere due to the discontinuities of the tropopause (Olive, 1970). The tritium-rich rain of the summer months is much more affected by evapotranspiration than that of autumn-winter, poor in tritium, therefore the transformation of the rain signal upon infiltration must be investigated (Foster and Smith-Carrington, 1980). A 1 m^2 lysimetric box covered with grass and filled with fluvio-glacial earth 1 m deep was installed in Thonon at the site of the rain gauge. The drainage of this box, equivalent to the effective rainfall, was monitored quantitatively and qualitatively on a monthly basis from 1969 to 1979. During these 11 years, the average annual rainfall was 944 mm and its weighted tritium content was 132 TU, while the drainage at the base of the box was 537 mm and showed a weighted annual tritium content of 114 TU. The average inter-annual evapotranspiration of 407 mm thus affects the tritium rain signal by a drop of 15%. This mechanism was repeated each year in similar proportions during the 11 years of monitoring. Consequently, the annual tritium levels in precipitation used as input data for modelling were systematically adjusted by a drop of 15% to estimate the infiltration signal toward the

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unconfined aquifers (Table 1). This correction is only really significant for the high tritium values close to the 1963 peak (Fig. 6). It seems illusory to specify the infiltration signal more precisely, in that the lumped models involve a steady-state regime that does not take into account the inter-annual variability of input volumes.

Figure 5 – Evian-Cachat spring tritium chronicle (note the increase in measurement precision with time; arrows indicate values below the detection limit (circle))

Figure 6 - Evian-Thonon area rainfall and effective rainfall tritium mean annual content.

Tritium restitution is very different according to the hydrogeological system (Fig. 4). The restitution at Versoie shows high values as of 1964 and a peak in 1965 in reaction to the 1963 peak in rainfall, but with concentrations reduced by almost 5 times compared to that in the rainfall (700 vs. about 3000 TU). The Maxima spring and the Larringes-Pugny well show behaviour close to that of Versoie. On the arithmetic scale figure (Fig. 4 bottom), the Evian-Cachat spring does not appear to be affected by the injection of atmospheric tritium. The use of a logarithmic scale (Fig. 4 top) and use of isolated data (Fig. 5) display the multimodal restitution of the tritium at the Evian-Cachat spring. An initial peak, transient and of very low amplitude, is observed from 1964 to 1966 (21 TU in 1965), followed by a dip and then a very broad peak, also of very low amplitude, that begins in 1969-1970 and displays a maximum around 1979 with levels of about 40 TU (41 TU in 1979), 70 times less than the levels in rainfall in 1963. The memory effect of this restitution is still perceptible in 2000 (16 TU).

From a methodological point of view, it appears (Figs. 4) that the period from the peak of 1963 to 1982 is decisive in distinguishing the different behaviour of the two types of hydrogeological systems, systems with low transit time on one hand (Versoie, Maxima, Larringes-Pugny) and that with a long transit time (Evian-Cachat) on the other hand.

5.2. Results of modeling

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Given the relatively large variation of the tritium signal at the **Versoie spring**, at least during the initial years of monitoring (1964 – 1974) (Figs. 4), the calculated annual tritium levels at the spring (Table 1) were averaged from the numerous individual levels measured on samples taken during the initial years of monitoring, every month until 1971, then every quarter until 1979. Finally, far from the 1963 thermonuclear peak in precipitation, the values used correspond to a single annual check. The shape of the observed tritium signal (Fig. 7) can be described using an exponential model (EM), taking a mean transit time (T) of 5 years (Fig. 7). This value is close to that of 4 years put forward by Molicova (1991) using a Gamma law on a shorter series, which was later confirmed by Hubert et Olive (1995) and Olive et al. (1996). A PFM only is definitely not appropriate (Fig. 7). Nevertheless, the signal measured at the Versoie spring shows a pronounced peak as of the beginning of monitoring in 1965, which probably indicates the existence of a component of water transfer by piston effect. Use of a Binary Mixing Model combining in parallel an EM with a PFM (BMMEP) thus allows the measured concentrations to be well reproduced (Fig. 7) with a 5.1 years mean transit time, and with the volume and flow parameters for the two reservoirs shown in Table 2**.** This transfer model is perfectly compatible with the hydrogeological context of the Versoie spring (§2. and Fig. 2) that involves two aquifers: (1) most of the flow (about 80%) comes from an unconfined aquifer that is attributed to the paleo-Dranse river lower fluvio-glacial kame terraces resting on the Petit Lac Stage impervious morainic substrate, and (2) a lower part of the flow (about 20%) is attributed to the earlier terraces aquifer, which is confined by the Petit Lac Stage impervious basal till and appears to be recharged upstream.

Figure 7 – Modelling the Versoie spring tritium time series (using LUMPED, Ozyurt and Bayari, 2003)

Table 2. – Summary of the parameters used to fit the Evian-Thonon area springs. Note that for the exponential-piston flow model, the two models are placed in parallel

The tritium signal measured at the **Maxima spring** of Amphion (380 m amsl), although including less data, has an appearance very similar to that of the Versoie spring (Figs 4). The same Binary Mixing Model (BMMEP) provides a very good fit (Fig. 8). Neither the PFM nor the EM are able to simulate the observed data. The simulation provides a mean transit time of 6.7 years (Table 2). This result is in good accordance with the hydrogeological structure of the Maxima spring, which outflows from a shallow semi-captive aquifer (local kame terrace) interstratified within the terminal basal till that appears to comprise both a confined and an unconfined component. The water molecule stable isotope content shows that this aquifer is very local (mean elevation of the recharge area of about 450 m amsl).

Figure 8 – Modelling the Maxima spring tritium time series (using LUMPED, Ozyurt and Bayari, 2003)

The tritium signal at the **Larringes-Pugny** well has been simulated with the same BMMEP with a good fit (Fig. 9) insofar as the scarce data available allow input and output data to be compared and a mean transit time of 6.3 years (Table 2). About 80% of the discharge of the spring is explained by the EM, while the last 20% is the result of PFM. The well also taps a shallow subhorizontal semi-confined aquifer flowing into a Gavot Plateau lateral till lying on a basal till which constitutes its impervious substratum. Hydrogeological data show that this aquifer is local, with a mean elevation of the recharge area of about 820 m amsl, similar to that of the well (790 m amsl). As the available data at this well are scarce, they can also be well fitted with an EM with T= 7 years (Fig. 9 and Table 2), which is not incompatible with the hydrogeological context (only locally confined aquifer).

Figure 9 – Modelling the Larringes-Pugny well tritium time series (using LUMPED, Ozyurt and Bayari, 2003)

The time variation of the tritium levels measured at the **Evian-Cachat spring** (Figs. 4 & 6) is complex and very different from the output signals observed at the springs previously studied.

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The signal is excessively attenuated; the maximum observed tritium levels are approximately 20 times lower. As it propagates over a greater time period, it appears to result from a transfer in a highly dispersive system. Moreover, the response of the hydrogeological system to infiltration of “tritiated” water is bimodal. Even if, due to their low activities, tritium measurements show a high uncertainty, an initial maximum, low in absolute value, is observed around 1966; the second, of significantly greater amplitude and very extended in time, appears much later, toward the 1980s. These two responses of the hydrogeological system to recharge by tritiated effective rainfall are the result of two different functional processes of the reservoir: (i) mainly a very highly dispersive aquifer system with very slow flow dynamics and (ii) a modest contribution of water with a less slow flow dynamics that doesn’t flow through the highly dispersive aquifer (Fig. 10). After several trials, a conceptual model with two models in a series (a DM and then a PFM) (Fig. 10) was adopted. In addition, a modest (about 1.3%) contribution of efficient rainfall directly enters the PFM and consequently avoids the DM.

Figure 10 - Conceptual model of the Evian-Cachat spring (in a series dispersive and piston flow model). “q” is the mean annual recharge and discharge of the spring

The less slow flow component of the system was studied together with the dispersive one, but was first calibrated. It was necessary first to identify the isotopic model best able to describe the time variation of the first tritium maximum observed at Evian-Cachat, and then to estimate the quantitative contribution of this less slow component to the overall functioning of the hydrogeological system. Only the PFM allowed the initial maximum observed at Evian-Cachat (Fig. 11A and B) to be satisfactorily reproduced. The mean transit time of these early waters lies around 2.5 years (± 0.25 y; Fig. 11A). The contribution of this first component appears to be very low, about 1.3 % (± 0.2 %; Fig 11B) of the total discharge at the spring.

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The other models tested are not acceptable; in particular, a DM significantly overestimates tritium levels during the decline that follows the initial maximum of 1966.

Figure 11 Modeling (TEMPO) of the Evian-Cachat spring. Fig. 11A: Sensitivity analysis of the PFM to the T. Fig. 11B: Sensitivity analysis of the PFM to the % of the less slow flow component.

Although not perceptible with the classical hydrochemical approaches, the computed parameters of this first component are consistent with the hydrogeological knowledge:

- the PFM and the perfect mixing before the outflow is consistent with the fact that the aquifer of the "terminal complex" is confined by the terminal till at and upstream from the spring. An EM (unconfined aquifer) would have been totally implausible;
- with the known parameters of this aquifer in the vicinity of the spring (hydraulic conductivity about 2.10^{-3} $m.s^{-1}$, hydraulic gradient about 1.10^{-3}, effective porosity about 5%), a 2.5 years mean transit time for the first component yields a recharge area about 3 km from the spring. This order of magnitude of a few kilometres (northernmost part of the Gavot Plateau) is in agreement with the maximum southern extension of the terminal till that, for a maximum elevation of about 650 to 750 m amsl (§ 2.), respectively, lies at a horizontal distance of about 1.5 to 1.9 km from the spring (Figs. 1 & 2a). Consequently, this first component is very probably a very small part of the total recharge (about 1.3 %) that infiltrates on the northernmost part of the Gavot Plateau and (this second condition is very important) is able to rapidly (in about one year) reach the "terminal complex" high hydraulic conductivity aquifer; in fact, as the Gavot Plateau is composed of several superimposed basal and lateral tills, its mean hydraulic conductivity is quite low and this explains, with the capping terminal till that avoids any infiltration downstream, the long transit time of Evian water (see the following part of this paper). Consequently, the infiltration of this first component

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occurs through few layers of superimposed basal and lateral tills, which is consistent with the geological structure of the northernmost tip of the Gavot plateau (Fig. 2a).

The effect of this less slow flow component is significant up to the mid-1980s; however, it is less than 5 TU starting from the mid-1970s.

Different types of isotopic transfer models (PFM, EM, BMM combining a PFM with an EM, DM) were tested to simulate the long term second part of the observed signal. Only the DM allows the restitution curve to be satisfactorily reproduced (Fig. 12) with T = 68 (±2; Fig. 12A) years, DP = 0.5 (±0.025; Fig. 12B), and a flow of 98.7% of the total discharge in the DM, the 1.3% remaining corresponding to the less slow flow component presented above. The sensitivity analysis shows that the DM is sensitive to small variations of both T and DP (Fig. 12A and B). This behaviour is in good agreement with the hydrogeological knowledge. The mineral aquifer recharge area, the 200 m-thick (Fig. 2a) Gavot Plateau, is composed of up to 13 superimposed Würmian basal and lateral till sequences. These formations respectively exhibit very low and quite low hydraulic conductivity. Hydrogeological and hydrochemical data from deep piezometers drilled on the plateau (Henriot et al., 2013) show the presence of several superimposed quite low hydraulic conductivity aquifers (flowing in the lateral tills) with hydraulic heads decreasing with depth, which is a strong clue for a vertical component of the groundwater flow. The groundwater is thus able to flow down through the basal till layers (either with leakage flow and/or in places where the basal till layer(s) is (are) missing). The hydrogeochemical data sampled in these deep piezometers (^{13}C and ^{14}C data, but also and mainly silica content, and Mg/Ca ratio, which are all residence time indicators) also show that most of the NMW mineralisation process occurs within this Gavot Plateau multilayer low hydraulic conductivity system, which is not capped by the thick low hydraulic conductivity superficial terminal till deposited below 650 to 750 m amsl. Consequently, such large dispersion behaviour is the result of the various pathways the future NMW can follow within

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the Gavot Plateau formation. The water then fast flows within the Terminal Complex aquifer. Consequently, with this set of parameters, the mean transit time in the Evian hydrosystem is about 70 years (T: 70.05 years): mostly (about 68 years) within the subsurface and deep aquifers of the Gavot plateau (DM) and about 2.5 years within the high hydraulic conductivity Terminal Complex (PFM). Statistical processing of the output data shows that the median transit time of the Evian NMW (a less biased estimator than the arithmetic mean for such non Gaussian distributions) is about 48 years (48.0 years).

Figure 12 - Modelling (TEMPO) and sensitivity analysis of the slow flow component of the Evian-Cachat spring. Fig. 12A: Sensitivity analysis of the DM to the T. Fig. 12B: Sensitivity analysis of the DM to DP

Another approach to modelling the transit time of the Evian-Cachat spring NMW in its reservoir was attempted on the assumption that across the recharge area of the Gavot Plateau, the infiltration component first crosses through a subsurface aquifer. In fact, the catchment area of the Evian NMW system, about 35 km^2, has a mean elevation of about 950-1000 m (see § 2.) and corresponds to the Gavot Plateau and Mont Bénant area (Fig. 1). Several local subsurface aquifers of limited extent are known in this area. The rather low discharge springs emerging from these aquifers are locally tapped to supply the few villages of the plateau. These subsurface aquifers constitute the first step in the functioning of the mineral hydrosystem. In this case, the tritium input signal of the deep mineral reservoir is transformed by its stay in the subsurface aquifer and so can in fact be different from the tritium chronicle resulting from effective rainfall. To simulate this new input signal, we thus assumed that the functioning of the various superficial aquifers of the Evian watershed area (Gavot Plateau) can be described using a mixing model (EM) with a transit time on the order of several years (Fig. 13). Various simulations were performed, taking among others mean transit times in the subsurface aquifer increasing from 6 to 10 years (Fig. 14). The monitoring of tritium levels in

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one of these subsurface aquifers at Larringes-Pugny from 1966 to 1969 is actually compatible with this choice (Fig. 9).The use of these data from the EM as new input data for the Evian-Cachat dispersive model (the two models are in a series) requires the parameters of this latest model to be revised. The tritium levels in the Evian-Cachat spring can only be well simulated if it is taken into account that the mean transit time in the EM is about 8 years (Fig. 14A), with a T of 60 (±1) years (Fig. 14B) in the deep aquifer system, DP = 0.75 (±0.025; Fig. 14C), and still a flow of 98.7% of the total discharge in the EM and DM, the 1.3% remaining corresponding to the less slow flow component presented above that only flows in the PFM (Figure 13). The first result (8 years in the EM) is in good accordance with the fit obtained at the Larringes-Pugny well (7 years) tapping one of the Gavot Plateau superficial aquifers (see section 5. and Fig. 9). Consequently, the mean transit time in the whole Evian hydrosystem computed from the 3 models in a series is about 69.5 years: 8 years within the subsurface aquifers of the Gavot plateau (EM), 60 years within the deep aquifers of the Gavot plateau (DM) and 2.5 years within the Terminal Complex (PFM). The median transit time of the Evian NMW is then 45.5 years. More in detail, the comparison of the results obtained with both in a series modelling approaches ((Model 1 - M1) DM+PFM and (Model 2 - M2) EM+DM+PFM) for a perfect tracer (without the tritium radioactive decay) shows interesting results (Fig. 15):

- the 4 first years are governed by the PFM and show no difference between M1 & M2;
- during the following 14 years the difference between the two models is rather low. The effect of the EM is nevertheless visible, with, during the first 8 years (up to a total duration of about 12 years), a restitution higher for M2 (EM8 + DM 60) than for M1, mostly showing the influence of the EM and of the lower T of the DM;
- after that, the differences are governed by the DM with logically a higher peak for M2.

Due to the difference of the two curves shapes, even if the T of both M1 and M2 are rather similar (about 70 years), their median are significantly different with a higher one for M1 (48 y) than for M2 (45.5 y). Consequently, the two different models provide significantly different results particularly during some important phases of the output, and particularly during the second part of the restitution (beyond 18 years) with significant absolute differences reaching about -0.12% (M1-M2), and consequently relative differences of about 8% ((M1-M2)/(Average of M1,M2)).

Figure 13 - Conceptual model of the Evian-Cachat spring (in a series exponential, dispersive and piston flow model). "q" is the mean annual recharge and discharge of the spring

Figure 14 – Modelling (Tempo) of the Evian-Cachat spring for various mean transit time (T) within the superficial aquifer (EM). Fig. 14A: Sensitivity analysis of the EM to the T. Fig. 14B: Sensitivity analysis of the DM to the T. Fig 14C: Sensitivity analysis of the DM to DP

Figure 15 – Modelling of a perfect tracer one year duration Dirac (100%, arbitrary unit; year 0) input for the Fig. 10 D- (T 68 y, DP = 0.5) + PF- (2.5 y) model (M1), and the Fig. 13 E- (T 8 y) + D- (T 60 y, DP = 0.75) + PF- (2.5 y) model (M2). The figure also provides the difference between the two outputs (M1 - M2)

6. Discussion

6.1. On the need of anthropogenic tracers long term database in hydrological studies

It appears that, even in quite heterogeneous and complex aquifers such as those of the Evian-Thonon area, the use of quite simple lumped models leads to very accurate results. Such tools allow transfer model types to be obtained, their parameters inferred, and aquifer transit time distributions computed.

Use of the exceptionally long and precise Evian-Thonon rainfall, effective rainfall and spring tritium content time series allows existing knowledge about the structure and functioning of such complex aquifers to be much better characterised and/or assessed. Among other results,

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the computed median transit time in the Evian-Cachat aquifer, about 45 years, appears to be much longer than previously expected from simpler approaches. Knowledge of this distribution function of transit times, allowing mass transfer of chemical elements within the hydrosystem to be imaged, has been especially useful in establishing a policy to protect the mineral resource (see for instance Buric et al., 2011, Devier et al., 2013; Lachassagne et al., 2011).

The long tritium chronicle available at the Evian-Cachat spring even allows a 1.3% contribution to the spring discharge that, due to its low value, was not identifiable by other hydrodynamical or hydrogeochemical means. However, this identification results from a very opportune and exceptional combination of circumstances: the fact that tritium monitoring at the Evian-Cachat spring began only a few years (2 years) after the main period of tritium injection into the atmosphere, and was carried out continuously for several decades. The scientists at the Centre de Recherches Géodynamiques were able to convince stakeholders to carry out such monitoring.

These results should encourage the owners of the old tritium database, even if sparse or incomplete, to make use of them, at least with such simple computational models. It should also encourage young scientists to be on the lookout for emerging tracers, and consequently to acquire the corresponding data on such high-stakes aquifers. A few years ago, noble gases and chlorofluoromethanes were such emerging tracers. These methods are presently being tested on the Evian aquifer and the results will be compared to those of this study.

This research also shows that the standard simple models can easily be improved with some kind of plug-in, for instance, to compute the behaviour of superficial aquifers. The next steps at Evian will be to more precisely compute the role of the high hydraulic conductivity aquifer of the "Terminal Complex" in the framework of a complete process-based modelling of the aquifer.

6.2. What is the future for Tritium as a tracer?

The tritium tool was especially effective in the vicinity of the 1963 rainfall peak and up to approximately 1980. But new prospects are emerging. Since 2000, the mean annual tritium signal in rainfall at Thonon has oscillated around a level of 9± 1TU. Comparison with the data available at the Orléans-BRGM station (Parisian Basin) shows the same trend around a level of 6 TU, and good correlation. Thus there seems to be a return to the case of a constant influx of tracer. In this case, the transit time of the water would be accessible starting from one or more tritium measurements using simple empirical models that require only this parameter; here, the exponential model (EM) and the Piston Flow model (PFM). For unconfined aquifers likely to function according to an exponential model (of course if the vadose zone is not too thick, in order to allow the arrival of water with a theoretically zero residence time), the tritium level could thus be translated into the apparent age of the water, on the express condition of having good knowledge of the local input rainfall signal. Given the low levels and the necessary precision, correlations with a too-distant rain gauge station are now inadequate, and a denser network would be necessary. This projection gives 6.7± 0.7 TU for Versoie in the immediate future. In 2009, its content was measured at 7.1 ± 0.6 TU. Measurements on such systems already studied near the 1963 peak would be worth repeating today to establish these new prospects. It would be interesting to verify whether slight differences still exist between families previously defined by their tritium level during former regional prospecting, so as to offer new standards for interpretation.

7. Conclusion

This paper deals with the interpretation, with lumped models and a transfer function model, of exceptionally long (up to 50 years) and precise tritium chronicles characterizing the rainfall, the recharge (efficient rainfall) and the outflow from various types of glacial aquifers from the French Alps (Evian-Thonon area).

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The evapotranspiration induces a mean 15% drop of the tritium signal. The three superficial glacial aquifers (two fluvio-glacial kame terraces and a lateral till) provide similar results: a best fit with an exponential flow model (EM) (playing the major role) combined in parallel with a piston flow model (PFM), and rather short mean transit time (T: 5 to 7 years). The deepest mineral aquifer (Evian) can only be fitted with the in a series combination of a highly dispersive model (DM; T 68 years, DP=0.5) and a piston flow model (2.5 years) or, better, by the in a series combination of an EM (T 8 years) modelling the subsurface aquifer and a DM (T 60 y, DP=0.75) and the same piston flow model (2.5 years) modelling the deep mineral aquifer (note that a very small part of the efficient rainfall, about 1.3%, avoids the DM (and the EM), and directly enters the PFM). This latest in a series combination of models is rarely used in hydrology even if it is conceptually highly credible in such a hydrogeological context. It provides the following parameters: T 69.5 years and median transit time 45.5 years. These models are very sensitive regarding the T (±1 y, 0.25 y for the PFM), less so with DP.

With this study the interest and limits of such a modelling and the future for tritium as a tracer can be discussed and it also provides new insights on the structure and functioning of alpine paleo glacial hydrosystems.

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Figure captions

Figure 1 – Location and schematic geological map of the Evian-Thonon area. Location of springs and boreholes. Note the location of the rain gauge used since June 1963).

Figure 2 - Schematic geological cross-sections of the Evian (2a - top) and Thonon (2b - bottom) areas (see the location of the cross-sections on Fig. 1).

Figure 3 - CRG Thonon tritium monthly rainfall time series (since june 1963; previous annual data reconstructed from the Ottawa time series, see details in the text). Note the logarithmic scale of the Y axis and the 1960s very high peak.

Figure 4 – Rainfall and springs tritium mean annual content at the Thonon CRG rain gauge and at the Evian-Thonon area springs *(top: logarithmic scale; bottom: arithmetic scale)*

Figure 5 – Evian-Cachat spring tritium chronicle (note the increase in measurement precision with time; arrows indicate values below the detection limit (circle))

Figure 6 - Evian-Thonon area rainfall and effective rainfall tritium mean annual content.

Figure 7 – Modelling the Versoie spring tritium time series (using LUMPED, Ozyurt and Bayari, 2003)

Figure 8 – Modelling the Maxima spring tritium time series (using LUMPED Ozyurt and Bayari, 2003)

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Figure 9 – Modelling the Larringes-Pugny well tritium time series (using LUMPED, Ozyurt and Bayari, 2003)

Figure 10 - Conceptual model of the Evian-Cachat spring (in a series dispersive and piston flow model). "q" is the mean annual recharge and discharge of the spring

Figure 11 – Modelling (TEMPO) of the Evian-Cachat spring. Fig. 11A: Sensitivity analysis of the PFM to the T. Fig. 11B: Sensitivity analysis of the PFM to the % of the "less slow flow component".

Figure 12 - Modelling (TEMPO) and sensitivity analysis of the slow flow component of the Evian-Cachat spring. *Fig. 12A: Sensitivity analysis of the DM to the T. Fig. 12B: Sensitivity analysis of the DM to* DP

Fig. 13 - Conceptual model of the Evian-Cachat spring (in a series exponential, dispersive and piston flow model). "q" is the mean annual recharge and discharge of the spring

Figure 14 – Modelling (Tempo) of the Evian-Cachat spring for various mean transit time (T) within the superficial aquifer (EM). Fig. 14A: Sensitivity analysis of the EM to the T. Fig. 14B: Sensitivity analysis of the DM to the T. Fig 15C: Sensitivity analysis of the DM to DP

Figure 15 – Modelling of a perfect tracer Dirac (100%, arbitrary unit; year 0) input for the Fig. 10 D- (T 68 y, DP = 0.5) + PF- (2.5 y) model (M1), and the Fig. 13 E- (T 8 y) + D- (T 60 y, DP = 0.75) + PF- (2.5 y) model (M1). The figure also provides the difference between the two outputs (M1 - M2)

Table captions

Table 1 – (i) Rainfall tritium mean annual content at the Thonon CRG rain gauge, (ii) computed tritium efficient rainfall signal, and (iii) mean annual tritium content at the Evian-Thonon area springs

Table 2 – Summary of the parameters used to fit the Evian-Thonon area springs. Note that for the exponential-piston flow model, the two models are placed in parallel