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

3  
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22

23 **Abstract<sup>1</sup>**

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

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**<sup>1</sup> 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

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

#### 46 **Highlights**

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

#### 52 **Keywords**

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

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

56 To evaluate the potential hydrogeological vulnerability of hydrosystems to anthropic pollution  
57 and predict any future changes, the pattern of the transit time of water in the aquifers should  
58 be defined. Isotopic measurements are presently used in field investigations and research  
59 projects to improve qualitative understanding of hydrosystems and identify the processes  
60 involved in water transfer (Turner and Barnes, 1998). Environmental tracers ( $^{18}\text{O}$ ,  $^2\text{H}$  and  
61 tritium) and specific flow models have been used by numerous authors to estimate the transit  
62 time of water in hydrological systems (Herrmann et al., 1990; Stewart and McDonnell, 1991;  
63 Maloszewski et al., 1983, 1992, 1995, 2002; Maloszewski et Zuber, 1982; Holko, 1995; Amin  
64 and Campana, 1996; Rodhe et al., 1996; Vitvar and Balderer, 1997; Etcheverry and Perrochet,  
65 2000; Uhlenbrook et al, 2002; Viville et al., 2006; Hagedorn et al., 2011; Morgenstern and  
66 Daughney, 2012). One difficulty in estimating the transit time of water is the need of the input

67 function, i.e. the concentration of tritium in the infiltrating water while only tritium in  
68 precipitation and precipitation amount are available. McGuire and McDonnell (2006) tried to  
69 construct better model to estimate the input function. Moreover, due to the insufficient length  
70 of input and output chronicles, hydrosystems with transit times of several decades sometimes  
71 cannot be studied.

72 Tritium ( $^3\text{H}$ ) is a natural isotope of hydrogen (half-life 12.32 years; Lucas and Unterwenger,  
73 2000), naturally present in the atmosphere at low concentrations (5 to 10 T.U.), and at higher  
74 concentrations (up to about 6000 T.U.) due to above ground nuclear weapons testing  
75 beginning in the mid 1940s, then dramatically increasing with the development of the  
76 hydrogen bomb (first test was in November 1952) until 1963, the year when the atmospheric  
77 atomic test was banned (the few small nuclear tests above ground since 1980 have just been  
78 too small to make any impact on the tritium burden). However the beginning of tritium  
79 recording started in August 1953 (Maloszewski and Zuber, 1994). It has been widely used in  
80 hydrology to estimate aquifers' mean transit time (or residence time) since it is a constituent  
81 of the water molecule and its concentration in rainfall has varied during the past sixty years,  
82 giving a well-known input function for hydrological models. Tritium better captures the  
83 longer term components of flow and recharge than other typical approaches (chloride or stable  
84 isotopes for instance) (Stewart et al., 2010).

85 The first tritium measurements of groundwater from the Quaternary deposits of the Geneva  
86 Lake basin were taken in the fifties, and have been densified since 1964, shortly after monthly  
87 analyses of tritium from rainfall began in June 1963 at the Thonon (France, Fig. 1) Centre de  
88 Recherches Géodynamiques (CRG, a branch of the Paris VI University), which was a member  
89 of the International Atomic Energy Agency (IAEA) isotopic monitoring network. As early as  
90 1963 this new isotopic tool proved useful in characterizing the hydrogeological functioning of  
91 the complex glacial aquifers of the area (Blavoux et al., 1964; Fontes et al., 1967). A tritium

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

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

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

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

112 The studied area is about 20 km long by 10 km wide along the southern shore of Lake Geneva  
113 (Fig. 1 & 2). As in most of the Lemanic basin (Fiore et al., 2011), the land surface is almost  
114 exclusively covered by thick (up to 400 m or more in Evian; Triganon et al., 2005)  
115 Quaternary glacial sediments deposited over a major glacial erosion surface truncating the  
116 cemented, folded and thrust Alpine rocks. The lake was dug during Quaternary glaciations

117 by ice originating from the upstream Rhône valley catchment basin. The deep valley of the  
118 Dranse River (Fig. 1), the bottom of which exposes much older pre-Quaternary geological  
119 formations, divides the study area into two independent zones both from a geological and a  
120 hydrological viewpoint.

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

- 125 - the shape of the pre-Quaternary substratum is the result of erosion from the pre-  
126 Würmian pulsations of the Rhône glacier. This paleotopography is irregular, reaches  
127 an elevation less than 100 m amsl at Evian, and progressively rises towards the South,  
128 up to a few pre-Quaternary outcrops at the highest points of the Gavot Plateau;
- 129 - this depression is filled by about 100 m of thick sands and gravels (older than the last  
130 glacial cycle), corresponding to the northern tip of a paleodelta of the Dranse River,  
131 covered by a subglacial till up to 200 m thick from the Würmian maximum (65'000 to  
132 35'000 y BP, Triganon et al., 2005), during which the glacier reached an elevation of  
133 about 1300 m amsl in the Evian area. This till surely prevents any groundwater flow  
134 between the pre-Quaternary substratum and the sediments described here below;
- 135 - glacio-lacustrine sediments (small local streams paleodeltas within lacustrine silts)  
136 from the "Inferior Complex", older than 30'000 y BP, sedimented during a major  
137 retreat phase of the Rhône glacier, overlie this till up to about 390 m amsl;
- 138 - the Gavot Plateau glacial margin formation, over 200 m thick, results from the  
139 progressive rise of the Rhône glacier during the Würmian "Lemanic stage" (30'000-  
140 27'000 y BP, Triganon et al., 2005, Fiore et al., 2011) when the glacier reached the

141 Geneva city area. It is composed of a subglacial till in the Evian area and up to 13  
142 superimposed subglacial and lateral tills with peatbog sediments below the plateau;  
143 - the partial glacier retreat down to an elevation of about 480 m amsl, and then to about  
144 430 m amsl at Evian, was responsible for the deposition of the glacio-lacustrine  
145 sediments of the “Terminal Complex” (25’000-21’000 y BP, Triganon et al., 2005)  
146 with highly permeable ( $10^{-3}$  to  $10^{-4}$  m/s) sands and gravels in the fans of the local  
147 paleostreams and low hydraulic conductivity silts laterally;  
148 - finally, the last flood of the Rhône glacier, with the western tip of the glacier again  
149 reaching Western Lake Geneva - “le Petit-Lac” - (Copet and Nyon readvances, Fiore  
150 et al., 2011), covered all these sediments with a final subglacial till (the “terminal till”)  
151 of 30 to more than 50 m thick up to an elevation of 650 to 750 m amsl in Evian. This  
152 till locally comprises kame terraces, mostly deposited at the mouth of local streams  
153 during the final deglaciation, and lateral tills.

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



166 1000 m amsl by comparing the  $^{18}\text{O}$  content of the Evian NMW aquifer with the  $^{18}\text{O}$  content of  
167 precipitation at several altitudes and with the  $^{18}\text{O}$  content of low discharge subsurface springs  
168 with a well-known catchment area altitude (Blavoux, 1978). Mean annual rainfall on the  
169 catchment area rises to 1'200 mm, and effective rainfall to about 750 mm. The specific  
170 hydrochemical characteristics of the Evian NMW, calcium (80 mg/l) and magnesium (26  
171 mg/l) (that dominate the cations) on one hand, and silica (15 mg/l) on the other hand, are  
172 respectively the result of water-rock interaction with rocks of the aquifer (limestone and  
173 dolomitic sands) that were eroded locally, and with rocks (granites, schists, green rocks...)  
174 from buried lateral tills that were imported from the Rhône valley catchment by the glacier.  
175 The pH is neutral (7.2) and bicarbonates (360 mg/l) and sulphate (13 mg/l) dominate the  
176 anions. Due to the high inertia of the hydrogeological system, the mineral content and  
177 temperature (11.6°C) at the spring are perfectly stable (below the analytical thresholds) and its  
178 discharge shows less than 5% annual variations.

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

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

186 West of the Dranse River, the **Thonon slope** is mainly covered by the fifteen (Gagnebin  
187 1933) Dranse River glacio-fluvial kame terraces (sands, gravels, and - limestone and less  
188 numerous crystalline - large pebbles) progressively deposited westwards against lateral tills  
189 (Figs. 1 & 2b), parallel to the Rhône glacier, during glacier retreat phases. The highest  
190 terraces deposited during the Gavot Plateau retreat (Geneva stage: 25'000 to 23'000 y BP)

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

### 206 **3. Material and methods - Sampling and analyses**

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

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

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

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

223 The water catchwork of the Evian-Cachat spring, built in 1911, reaches the sandy confined  
224 aquifer at the end of a 66 m long horizontal gallery crosscutting the till cover. As the aquifer  
225 is confined, the gallery is completely sealed and neither the aquifer nor the water is visible  
226 there. The artesian flow is limited by a valve to maintain the pressure. The samples were  
227 collected at a sampling tap located in the gallery; Natural Mineral Water is not treated, and of  
228 course not sterilised, consequently the samples were not subject to any treatment. Some  
229 sporadic measurements were made for comparison on the other boreholes of the same mineral  
230 hydrogeological system. The Versoie samples were collected on the pumping well from a tap  
231 located at the top of the pumping tube. The Maxima spring (Figs. 1 & 2) emerges from a  
232 superficial aquifer. It is located near the Lake's shore at 380 m amsl within a sandy kame  
233 terrace of the terminal till on the lowest part of the Evian area; its more than 40 years of  
234 tritium data are worth studying. The Larringes-Pugny well is located on the Gavot Plateau at  
235 790 m amsl. This well exploited for drinking water taps a superficial aquifer of the Gavot  
236 Plateau glacial margin formations (4 m thick sand and gravel locally covered by 2 m of clay).  
237 All the water samples were analysed for tritium at the Centre de Recherches Géodynamiques  
238 (CRG) in Thonon until it was closed in April 2005, and since then have been analysed at the  
239 Hydrogeology Laboratory of the University of Avignon. Tritium measurements were obtained  
240 by liquid scintillation counting after electrolytic enrichment. The associated error on the data

241 varies with the tritium concentration in the samples; it is 1 TU for recent samples and was  
242 about 1 or 2 tens of TU for the samples which contained several hundred TU (see Figs 4A, 4B  
243 and 5). The limit of detection of the CRG equipment was 5 to 8 TU during the earliest years  
244 and about 1 TU since 1972 (see Fig. 5).

#### 245 **4. Theory/calculation - Tritium restitution modelling**

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

$$250 \quad C_{out}(t) = \int_0^{\infty} C_{in}(t - t')e^{-\lambda t'} g(t') dt' \quad (1)$$

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

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

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

266 models (see chapter 2.), some of these models were also combined in parallel, thus  
267 constituting Binary Mixing Models (a terminology proposed for instance by Katz et al.,  
268 2001), which means a simple mixing of two - or more - endmembers occurring at the  
269 spring/outlet. Piston flow and exponential model (named after BMMEP) were combined.  
270 Some of these models were also combined in series. All these models take into account the  
271 natural radioactive decay of tritium. The choice of model/combination of models was made as  
272 a function of the appearance of the observed signal at each spring and the available  
273 knowledge on their hydrogeological functioning. Among the different computer codes  
274 available in the hydrological literature (for example FLOWPC, Maloszewski and Zuber 1994,  
275 1996), we first used LUMPED (Ozyurt and Bayari 2003, available at  
276 <http://www.sukimyasilab.hacettepe.edu.tr/english/software.shtml>). In a second phase, the  
277 complex signal measured at the Evian-Cachat spring was interpreted using the TEMPO  
278 software developed at BRGM by Pinault (2001) (see for instance also Dörfliger et al., 2008),  
279 in which the equations of the models described by Maloszewski and Zuber (1994, 1996) have  
280 been implemented (the models developed by Maloszewski and Zuber have also been used to  
281 validate TEMPO). FLOWPC and TEMPO have the same input requirements and provide  
282 similar outputs. The subsurface aquifers of the Versoie, Larringes-Pugny, and Maxima  
283 springs were characterised using the exponential model (EM), and finally with a Binary  
284 Mixing Model combining exponential and piston flow models where the exponential part  
285 plays the major role. The Evian-Cachat aquifer was characterised with an in a series model  
286 combining dispersive and piston flow models (Fig. 10), and secondly with an in a series  
287 model combining exponential, dispersive and piston flow models (Fig. 13).  
288 The piston flow model (PFM) assumes that all the flow lines have the same transit time and  
289 thus, the response function is the Dirac delta function  $g(t)=\delta(t-T)$ , with T: mean transit time.  
290 Hydrodynamic dispersion and diffusion are negligible (Maloszewski and Zuber, 1982, 1994).

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

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

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

$$307 \quad g(t) = \left( \frac{4\pi DP t}{T} \right)^{-\frac{1}{2}} \exp \left[ -\frac{\tau}{4 DP T} \left( 1 - \frac{t}{\tau} \right)^2 \right] t^{-1} \quad (2)$$

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

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

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

## 321 **5. Results**

### 322 **5.1. Observation results**

323 The annual tritium levels of the precipitation at Thonon CRG (Table 1) have been computed  
324 from the monthly values weighted by the precipitation rate. For values prior to 1963, a  
325 regression (Equ. 3) was established with the annual levels measured in Ottawa from 1964 to  
326 1985, a station at the same latitude as Thonon that has operated since 1953 (Brown, 1970).

$$327 \text{[}^3\text{H]}_{\text{Thonon}} = 0.892\text{[}^3\text{H]}_{\text{Ottawa}} + 9, n=22, R^2=0.9 \text{ (Molicova, 1991)} \quad (3)$$

328 This chronicle of annual levels constitutes the input signal for the hydrogeological systems of  
329 the Evian-Thonon area (Figs. 4A and 4B). Since the start of thermonuclear weapons testing in  
330 the early 50's, continued more and more intensively until 1963, atmospheric tritium levels  
331 increased enormously. The levels in rainwater at Thonon reached several thousand TU in  
332 1963, with a spectacular rise between 1961 and 1963 (Fig. 4B). From 1963 to 1966, nearly all  
333 atmospheric nuclear tests were stopped and levels dropped sharply with a pseudo-period close  
334 to one year, thus defining a very characteristic peak on the multiyear scale. From 1967 to  
335 1980, atmospheric nuclear tests were of very low capacity, with less than 3% in total of all the  
336 tritium produced artificially, nevertheless leading to fallout that decreased slowly from 200 to  
337 100 TU. Since 1980, there have been no atmospheric nuclear tests. With a pseudo-period of  
338 one year and according to tritium natural radioactive decay (half-life: 12.32 years), the level  
339 should have reached the natural level (5 to 10 TU) around 1985. However, it remained  
340 between 30 and 20 TU until 1996; this is due to the feedback from the oceans and surface

341 waters that contain higher levels of tritium (Fine and Ostlund, 1977; Fine et al., 1987; Michel,  
342 1992) and also the influence of the civilian nuclear industry. Since 2000, it seems that the rain  
343 signal at Thonon has entered a permanent regime around a value of 9 TU.

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

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

349 The tritium content in rainfall shows a seasonal variation. Monthly levels in Thonon have a  
350 maximum in June-July and a minimum in November-December. For the temperate climate  
351 zone of the Northern hemisphere, this springtime increase results from direct discharge of  
352 tritium in the stratosphere toward the troposphere due to the discontinuities of the tropopause  
353 (Olive, 1970). The tritium-rich rain of the summer months is much more affected by  
354 evapotranspiration than that of autumn-winter, poor in tritium, therefore the transformation of  
355 the rain signal upon infiltration must be investigated (Foster and Smith-Carrington, 1980). A 1  
356 m<sup>2</sup> lysimetric box covered with grass and filled with fluvio-glacial earth 1 m deep was  
357 installed in Thonon at the site of the rain gauge. The drainage of this box, equivalent to the  
358 effective rainfall, was monitored quantitatively and qualitatively on a monthly basis from  
359 1969 to 1979. During these 11 years, the average annual rainfall was 944 mm and its  
360 weighted tritium content was 132 TU, while the drainage at the base of the box was 537 mm  
361 and showed a weighted annual tritium content of 114 TU. The average inter-annual  
362 evapotranspiration of 407 mm thus affects the tritium rain signal by a drop of 15%. This  
363 mechanism was repeated each year in similar proportions during the 11 years of monitoring.  
364 Consequently, the annual tritium levels in precipitation used as input data for modelling were  
365 systematically adjusted by a drop of 15% to estimate the infiltration signal toward the



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

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

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

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

385 From a methodological point of view, it appears (Figs. 4) that the period from the peak of  
386 1963 to 1982 is decisive in distinguishing the different behaviour of the two types of  
387 hydrogeological systems, systems with low transit time on one hand (Versoie, Maxima,  
388 Larringes-Pugny) and that with a long transit time (Evian-Cachat) on the other hand.

## 389 **5.2. Results of modeling**

390 Given the relatively large variation of the tritium signal at the **Versoie spring**, at least during  
391 the initial years of monitoring (1964 – 1974) (Figs. 4), the calculated annual tritium levels at  
392 the spring (Table 1) were averaged from the numerous individual levels measured on samples  
393 taken during the initial years of monitoring, every month until 1971, then every quarter until  
394 1979. Finally, far from the 1963 thermonuclear peak in precipitation, the values used  
395 correspond to a single annual check. The shape of the observed tritium signal (Fig. 7) can be  
396 described using an exponential model (EM), taking a mean transit time (T) of 5 years (Fig. 7).  
397 This value is close to that of 4 years put forward by Molicova (1991) using a Gamma law on a  
398 shorter series, which was later confirmed by Hubert et Olive (1995) and Olive et al. (1996). A  
399 PFM only is definitely not appropriate (Fig. 7). Nevertheless, the signal measured at the  
400 Versoie spring shows a pronounced peak as of the beginning of monitoring in 1965, which  
401 probably indicates the existence of a component of water transfer by piston effect. Use of a  
402 Binary Mixing Model combining in parallel an EM with a PFM (BMMEP) thus allows the  
403 measured concentrations to be well reproduced (Fig. 7) with a 5.1 years mean transit time,  
404 and with the volume and flow parameters for the two reservoirs shown in Table 2. This  
405 transfer model is perfectly compatible with the hydrogeological context of the Versoie spring  
406 (§2. and Fig. 2) that involves two aquifers: (1) most of the flow (about 80%) comes from an  
407 unconfined aquifer that is attributed to the paleo-Dranse river lower fluvio-glacial kame  
408 terraces resting on the Petit Lac Stage impervious morainic substrate, and (2) a lower part of  
409 the flow (about 20%) is attributed to the earlier terraces aquifer, which is confined by the Petit  
410 Lac Stage impervious basal till and appears to be recharged upstream.

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

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

415 The tritium signal measured at the **Maxima spring** of Amphion (380 m amsl), although  
416 including less data, has an appearance very similar to that of the Versoie spring (Figs 4). The  
417 same Binary Mixing Model (BMMEP) provides a very good fit (Fig. 8). Neither the PFM nor  
418 the EM are able to simulate the observed data. The simulation provides a mean transit time of  
419 6.7 years (Table 2). This result is in good accordance with the hydrogeological structure of the  
420 Maxima spring, which outflows from a shallow semi-captive aquifer (local kame terrace)  
421 interstratified within the terminal basal till that appears to comprise both a confined and an  
422 unconfined component. The water molecule stable isotope content shows that this aquifer is  
423 very local (mean elevation of the recharge area of about 450 m amsl).

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

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

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

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

440 The signal is excessively attenuated; the maximum observed tritium levels are approximately  
441 20 times lower. As it propagates over a greater time period, it appears to result from a transfer  
442 in a highly dispersive system. Moreover, the response of the hydrogeological system to  
443 infiltration of “tritiated” water is bimodal. Even if, due to their low activities, tritium  
444 measurements show a high uncertainty, an initial maximum, low in absolute value, is  
445 observed around 1966; the second, of significantly greater amplitude and very extended in  
446 time, appears much later, toward the 1980s. These two responses of the hydrogeological  
447 system to recharge by tritiated effective rainfall are the result of two different functional  
448 processes of the reservoir: (i) mainly a very highly dispersive aquifer system with very slow  
449 flow dynamics and (ii) a modest contribution of water with a less slow flow dynamics that  
450 doesn't flow through the highly dispersive aquifer (Fig. 10). After several trials, a conceptual  
451 model with two models in a series (a DM and then a PFM) (Fig. 10) was adopted. In addition,  
452 a modest (about 1.3%) contribution of efficient rainfall directly enters the PFM and  
453 consequently avoids the DM.

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

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

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

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

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

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

489 occurs through few layers of superimposed basal and lateral tills, which is consistent  
490 with the geological structure of the northernmost tip of the Gavot plateau (Fig. 2a).  
491 The effect of this less slow flow component is significant up to the mid-1980s; however, it is  
492 less than 5 TU starting from the mid-1970s.  
493 Different types of isotopic transfer models (PFM, EM, BMM combining a PFM with an EM,  
494 DM) were tested to simulate the long term second part of the observed signal. Only the DM  
495 allows the restitution curve to be satisfactorily reproduced (Fig. 12) with  $T = 68 (\pm 2)$ ; Fig.  
496 12A) years,  $DP = 0.5 (\pm 0.025)$ ; Fig. 12B), and a flow of 98.7% of the total discharge in the  
497 DM, the 1.3% remaining corresponding to the less slow flow component presented above.  
498 The sensitivity analysis shows that the DM is sensitive to small variations of both  $T$  and  $DP$   
499 (Fig. 12A and B). This behaviour is in good agreement with the hydrogeological knowledge.  
500 The mineral aquifer recharge area, the 200 m-thick (Fig. 2a) Gavot Plateau, is composed of up  
501 to 13 superimposed Würmian basal and lateral till sequences. These formations respectively  
502 exhibit very low and quite low hydraulic conductivity. Hydrogeological and hydrochemical  
503 data from deep piezometers drilled on the plateau (Henriot et al., 2013) show the presence of  
504 several superimposed quite low hydraulic conductivity aquifers (flowing in the lateral tills)  
505 with hydraulic heads decreasing with depth, which is a strong clue for a vertical component of  
506 the groundwater flow. The groundwater is thus able to flow down through the basal till layers  
507 (either with leakage flow and/or in places where the basal till layer(s) is (are) missing). The  
508 hydrogeochemical data sampled in these deep piezometers ( $^{13}\text{C}$  and  $^{14}\text{C}$  data, but also and  
509 mainly silica content, and Mg/Ca ratio, which are all residence time indicators) also show that  
510 most of the NMW mineralisation process occurs within this Gavot Plateau multilayer low  
511 hydraulic conductivity system, which is not capped by the thick low hydraulic conductivity  
512 superficial terminal till deposited below 650 to 750 m amsl. Consequently, such large  
513 dispersion behaviour is the result of the various pathways the future NMW can follow within

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

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

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

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

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



563 Due to the difference of the two curves shapes, even if the T of both M1 and M2 are rather  
564 similar (about 70 years), their median are significantly different with a higher one for M1 (48  
565 y) than for M2 (45.5 y). Consequently, the two different models provide significantly  
566 different results particularly during some important phases of the output, and particularly  
567 during the second part of the restitution (beyond 18 years) with significant absolute  
568 differences reaching about -0.12% (M1-M2), and consequently relative differences of about  
569 8%  $((M1-M2)/(\text{Average of } M1, M2))$ .

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

572 *Figure 14 – Modelling (Tempo) of the Evian-Cachat spring for various mean transit time (T)*  
573 *within the superficial aquifer (EM). Fig. 14A: Sensitivity analysis of the EM to the T. Fig.*  
574 *14B: Sensitivity analysis of the DM to the T. Fig 14C: Sensitivity analysis of the DM to DP*

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

## 579 **6. Discussion**

### 580 *6.1. On the need of anthropogenic tracers long term database in hydrological studies*

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

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

588 the computed median transit time in the Evian-Cachat aquifer, about 45 years, appears to be  
589 much longer than previously expected from simpler approaches. Knowledge of this  
590 distribution function of transit times, allowing mass transfer of chemical elements within the  
591 hydrosystem to be imaged, has been especially useful in establishing a policy to protect the  
592 mineral resource (see for instance Buric et al., 2011, Devier et al., 2013; Lachassagne et al.,  
593 2011).

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

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

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

613 6.2. *What is the future for Tritium as a tracer?*

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

633 **7. Conclusion**

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

638 The evapotranspiration induces a mean 15% drop of the tritium signal. The three superficial  
639 glacial aquifers (two fluvio-glacial kame terraces and a lateral till) provide similar results: a  
640 best fit with an exponential flow model (EM) (playing the major role) combined in parallel  
641 with a piston flow model (PFM), and rather short mean transit time (T: 5 to 7 years). The  
642 deepest mineral aquifer (Evian) can only be fitted with the in a series combination of a highly  
643 dispersive model (DM; T 68 years, DP=0.5) and a piston flow model (2.5 years) or, better, by  
644 the in a series combination of an EM (T 8 years) modelling the subsurface aquifer and a DM  
645 (T 60 y, DP=0.75) and the same piston flow model (2.5 years) modelling the deep mineral  
646 aquifer (note that a very small part of the efficient rainfall, about 1.3%, avoids the DM (and  
647 the EM), and directly enters the PFM). This latest in a series combination of models is rarely  
648 used in hydrology even if it is conceptually highly credible in such a hydrogeological context.  
649 It provides the following parameters: T 69.5 years and median transit time 45.5 years. These  
650 models are very sensitive regarding the T ( $\pm 1$  y, 0.25 y for the PFM), less so with DP.  
651 With this study the interest and limits of such a modelling and the future for tritium as a tracer  
652 can be discussed and it also provides new insights on the structure and functioning of alpine  
653 paleo glacial hydrosystems.

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661

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- 828

829 **Figure captions**

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

832

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

835

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

839

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

842

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

845

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

847

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

850

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

853

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

856

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

859

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

863

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

867

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

870

871 Figure 14 – Modelling (Tempo) of the Evian-Cachat spring for various mean transit time (T)  
872 within the superficial aquifer (EM). Fig. 14A: Sensitivity analysis of the EM to the T. Fig.  
873 14B: Sensitivity analysis of the DM to the T. Fig 15C: Sensitivity analysis of the DM to DP

874

875 Figure 15 – Modelling of a perfect tracer Dirac (100%, arbitrary unit; year 0) input for the  
876 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  
877 y, DP = 0.75) + PF- (2.5 y) model (M1). The figure also provides the difference between the  
878 two outputs (M1 - M2)

879

880 **Table captions**

881

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

885

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