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HYDROLOGICAL APPLICATION OF ANALYSES OF STABLE ISOTOPE CONTENT

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1. Introduction

Of the stable environmental isotopes, deuterium (^{2}H) and oxygen-18 (^{18}O) are those primarily used for hydrological investigations because, being components of the water molecule, they fulfil to a large degree the requirements made of an ideal hydrological tracer.

Besides ^{2}H and ^{18}O, carbon-13 (^{13}C) has also come to achieve considerable significance in isotope hydrology. It serves primarily for studies of the carbon balance in the hydrosphere, generally in conjunction with the interpretation of a ^{14}C content determined in groundwater. This will be dealt with elsewhere during this conference.

The few hydrological applications of the sulphur-34 (^{34}S) content in water samples are based in part on the fact that the isotopic ratios in the sulphur from evaporite sulphates of different geological formation can be easily distinguisehd (NIELSEN and RAMBOW 1969, WEYER et al. 1979). Furthermore, the remaining sources of the sulphate content of groundwater can also in some circumstances be differentiated through the ^{34}S content.

Lastly, isotopic-ratio measurements, when measuring concentrations of inert gases in groundwater for determining palaeo-temperatures (MAZOR 1972), can also deliver additional hydrological data. The argon component, for example,

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that arises through radioactive decay of the potassium-4o contained in the aquifer, can be determined by measuring the isotopic ratio $^{4o}Ar/^{36}Ar$ and thus separated from the argon component that is introduced into the groundwater by precipitation and as a function of temperature (MAZOR et al. 1974).

The following paper is restricted to ^{2}H and ^{18}O since we have no experience of our own in the hydrological application of ^{34}S and inert-gas isotopic measurements and because the hydrological applications of ^{2}H and ^{18}O far exceed those of all other isotopes.

2. Some notes on the occurrence of ^{2}H and ^{18}O in the hydrosphere, on measuring ^{2}H and ^{18}O in water samples and on the ^{2}H and ^{18}O content in precipitation

Table 1 shows that in natural water there are approximately 2ooo molecules of $^{1}H_2{}^{18}O$ and 32o molecules of $^{2}H^{1}H^{16}O$ among $1o^6$ water molecules of $^{1}H_2{}^{16}O$. The spread of the fluctuations is caused by isotope-fractionation processes, which can occur in all phase changes through the different vapor pressures of waters of different isotopic species and during contact of the water with rock as a result of isotopic exchange. The isotope fractionations, which initially occur on contact surfaces, become volume effects through diffusion, dispersion and isotopic exchange within the water sample.

TAB.1: CONCENTRATION OF ENVIRONMENTAL ISOTOPES IN NATURAL WATER

ISOTOPE	WATER MOLECULE	ISOTOPE RATIO	PARTS PER MILLION	RANGE OF CONCENTRATION
^{2}H	$^{1}H\ ^{2}H\ ^{16}O$	$^{2}H/^{1}H$	90 - 170	-430 - +75 (‰)
^{18}O	$^{1}H_2\ ^{18}O$	$^{18}O/^{16}O$	1880 - 2010	-55 - +10 (‰)

Figure 1 is a schematic illustration of the causes in the natural hydrosphere leading to alterations of the ^{18}O and ^{2}H content in water.

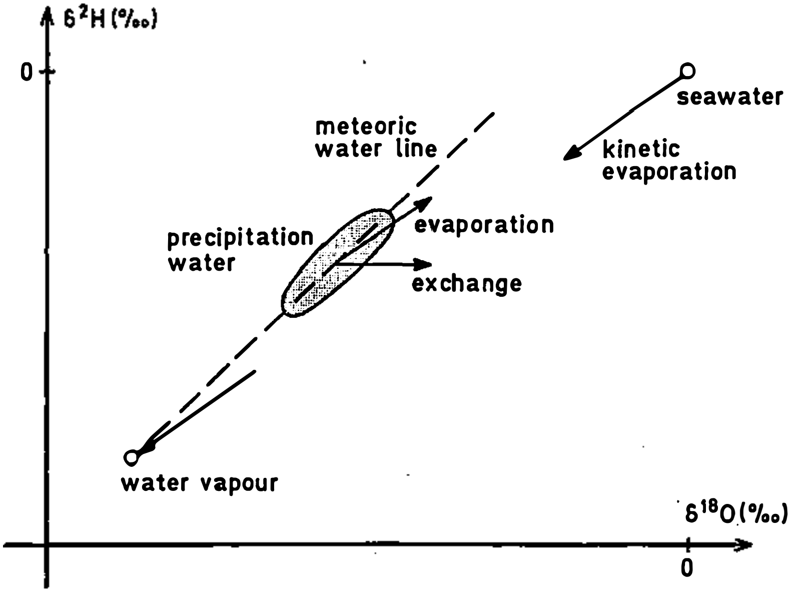

Fig. 1: Development of ^{2}H- and ^{18}O-content of liquid water and water vapor during evaporation, condensation and exchange processes within the natural water cycle

Starting with seawater there is first a heavy decrease of the content of ^{18}O and ^{2}H with the water vapour occurring in evaporation. This depletion and the associated $^{18}O/^{2}H$ relation depends on climatic conditions (primarily on temperature and the deficit of humidity in the atmosphere). During condensation there is then in reverse an increase of ^{18}O and ^{2}H content in the rain water, the magnitude of this depending on the condensation temperature. The remaining water vapour of the air mass continuously depletes itself of ^{18}O and ^{2}H, so with continued raining out of the air mass the ^{18}O and ^{2}H content of the precipitation also decreases. A linear relationship is maintained between the ^{18}O and the ^{2}H content, and this corresponds to the socalled "meteoric water line" (CRAIG 1961).

The ground and surface water formed of the precipitation can then be altered in some cases in its isotopic content through evaporation processes and through isotopic exchange with the aquifer. The enrichments that occur are generally relatively higher for ^{18}O than for ^{2}H and there is consequently a deviation from the precipitation line.

The measuring accuracy that is necessary for detecting the mentioned isotope fractionations has until now only been possible by using mass spectrometers specially equipped for isotopic-ratio measurements with a double-inlet system and a double or multiple collector.

The sample to be examined must be available in a gaseous state for such a measurement. The water sample is thus quantitatively reduced to hydrogen for the ^{2}H measurement and equilibrated with CO_2 for the ^{18}O measurement. Figure 2 shows a complete ^{18}O system, with which 3o samples can be fully automatically measured by mass spectrometer following the equilibration and the results computed and printed out together with the measuring error.

^{18}O and ^{2}H contents are by international agreement referred to the SMOW standard, this corresponding approximately to the isotopic composition of seawater. The measure used is the socalled "δ-value" ($\delta^{18}O$, $\delta^{2}H$),

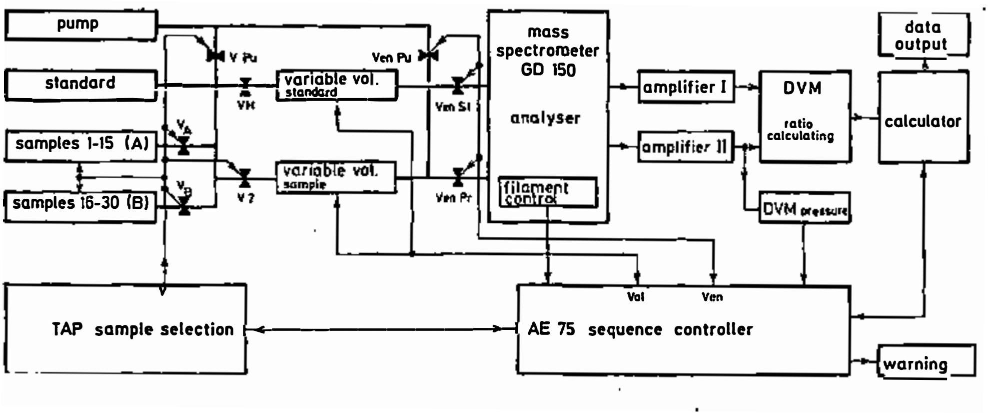

Fig. 2: Schematic diagram of measuring setup for the determination of the natural ^{18}O-contents of water samples (AUER et al. 1978)

which describes the relative deviation in per mil from the SMOW standard. The measuring accuracy generally achieved today (2σ criterion) is in the region of a $^{18}O/^{16}O$ and $^{2}H/^{1}H$ concentration of o.3 · $1o^{-6}$, corresponding to approximately o.1 per mil in the $\delta^{18}O$ value and 1 per mil in the $\delta^{2}H$ value.

Figure 3 shows the $\delta^{2}H$ values of all precipitation in a period of three years for the sampling station Lainbachtal (see fig. 8) in the Alps as a function of time. The spread of the fluctuation of these $\delta^{2}H$ values amounts to about 1oo per mil, caused by the different climatic conditions during the condensation of the different precipitations.

The frequency of the precipitations nevertheless produces monthly mean values of the ^{2}H content following the line of the monthly mean values for air temperature. In regions with rare precipitation (arid zones for instance) no such correlation with the monthly mean of the air temperature ("temperature effect") can be expected because of the heavy scatter of the δ-values.

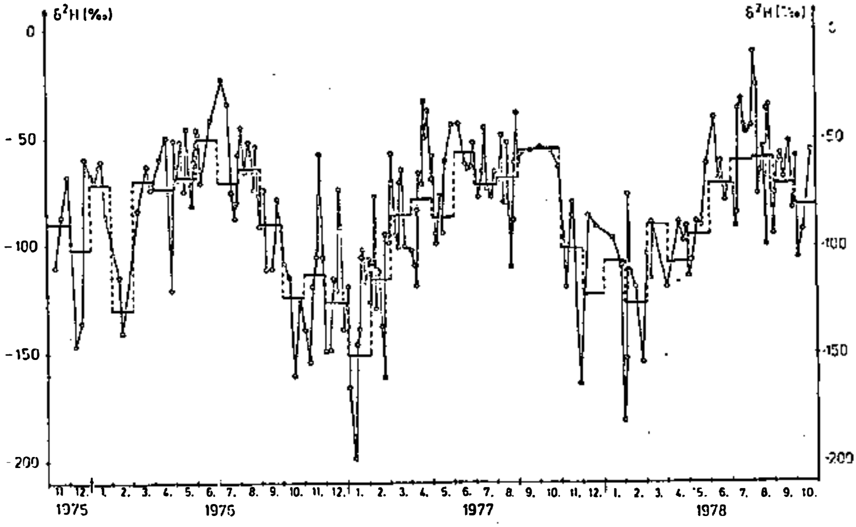

Fig. 3: ^{2}H-contents of every single precipitation events and of monthly precipitation at the Eibelsfleck, Lainbachvalley (see Fig. 8) from November 1975 till October 1978 (STICHLER & HERRMANN 1979)

Figure 4 shows the decrease of the ^{2}H content in precipitation with continuous raining out in the case of a sampling profile made in Chile between the Pacific Ocean and the Andes on the geographical latitude of Santiago. Besides the resulting "continental effect" there is an additional drop in the δ^{2}H values to be observed in the region of the orographic elevations of the coastal Cordillere and the main ridge of the Andes. This "altitude effect" can be explained by the intense raining out and the adiabatic cooling when the moist air masses slide up the mountain slopes and it amounts to $\delta^{2}H = 2$ to 4 per mil for 1oo meters of difference in altitude.

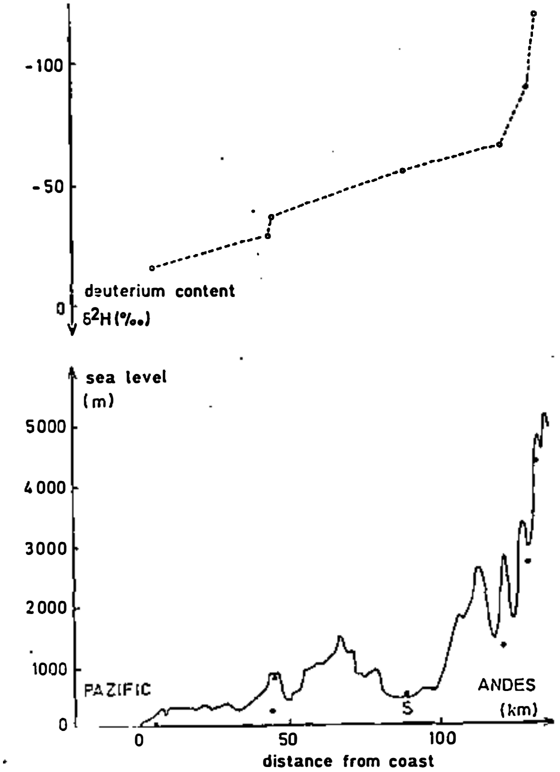

Fig. 4: ^{2}H-contents of mean monthly precipitation and West-East altitude profile across Chile at Santiago. The steady decrease in the δ^2H values from West to East corresponds to the continental effect, the sharp decreases at the coastal mountains and in the Andes are due to the altitude effect (MOSER et al. 1975)

3. Reflection of the isotopic content of precipitation in the snow cover

The vertical profile of the isotopic content in snow cover reflects the isotopic content of the individual snowfalls retained there. Figure 5 demonstrates this, taking as an example a snow profile from the Schneeferner glacier of the Zugspitze (Bavarian Alps), in which the layering of the isotopic content reflects the individual snowfalls of the preceding three months. Measurements of isotopic content also show how the snow cover reduces. In figure 6 it can be seen that the snow cover observed during a winter on the Weissfluhjoch near Davos reduces starting from the top, whilst the melting process illustrated in figure 7, of a snow cover in Chile, starts from the ground. In both cases the layering of the isotopic content is qualitatively retained during the period of ablation.

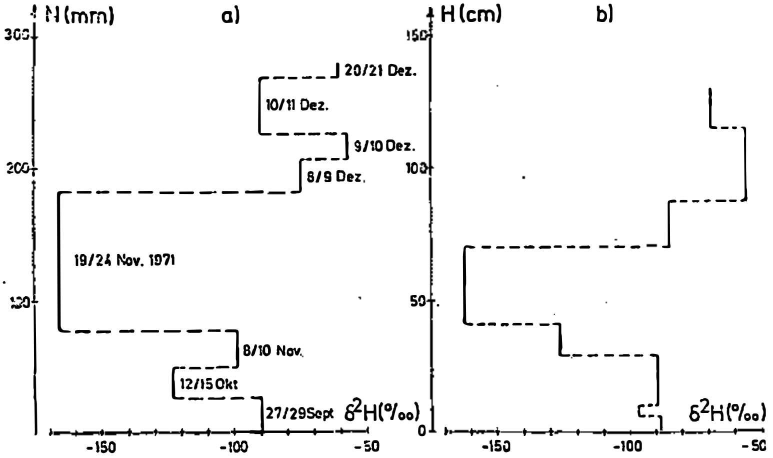

Fig. 5: a) δ^2H values of separate successive snowfalls on the Schneeferner glacier (Bavarian Alps) in relation to the water equivalents N (mm) of the single events at respective precipitation dates. b) δ^2H values of a snow profile sampled on 2 January 1972; H (cm) = snow depth (MOSER & STICHLER 1975)

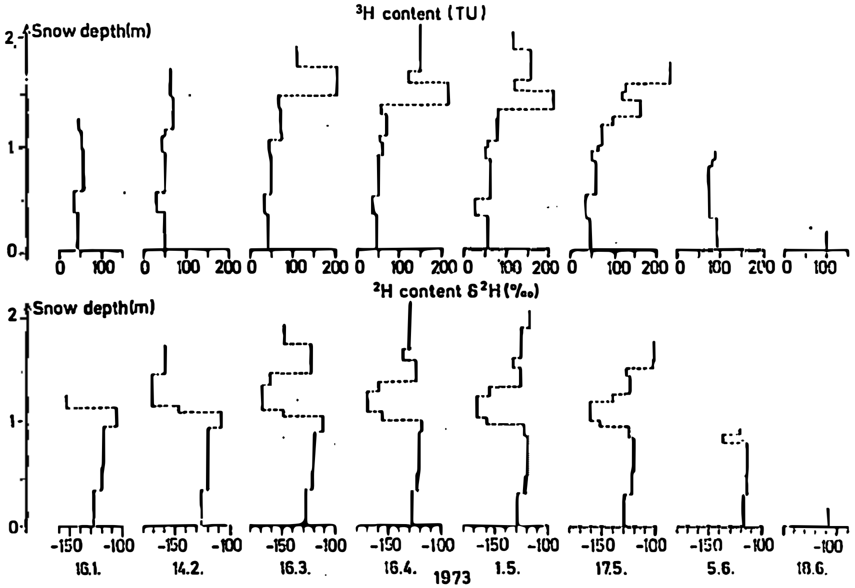

Fig. 6: Variations with time of the ^3H- and ^2H contents of snow profiles on the Weißfluhjoch, Davos (Switzerland) for the days indicated (STICHLER et al. 1979).

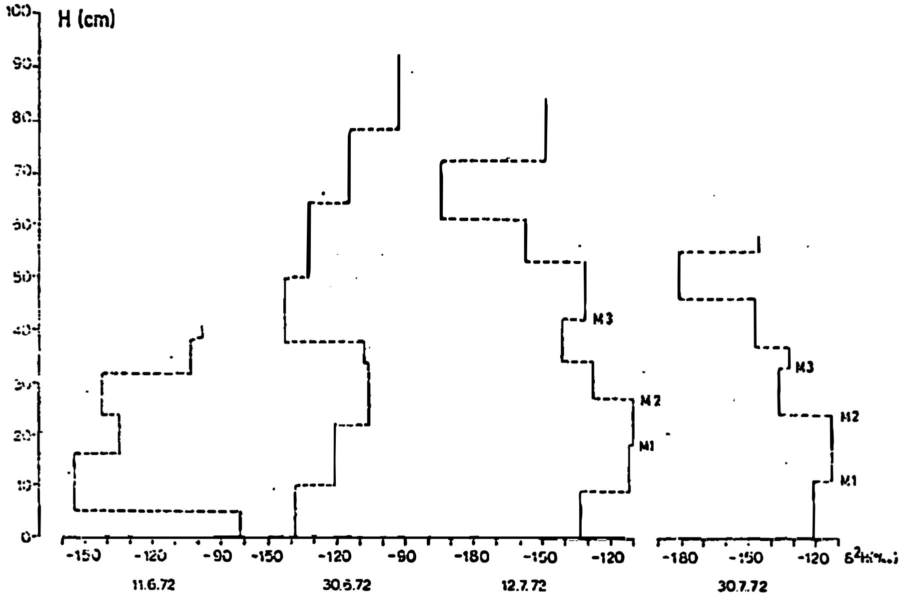

Fig. 7: Variation with time of the δ^2H values of snow profiles near La Parva (Chile). H = snow depth. The single snow layers have been identified by coloured threats (MOSER et al. 1975)

4. Precipitation/run-off relationship in regions with a temporary snow cover and with glaciers

For investigations of water balance and flood forecasts it is important to know what part of the rain water in a catchment area enters the receiving water course directly over the surface and what part infiltrates indirectly into the subsoil and thus possibly mobilizes subsoil water into a receiving water course. The same question poses itself in a modified form for the run-off of the snow water in a

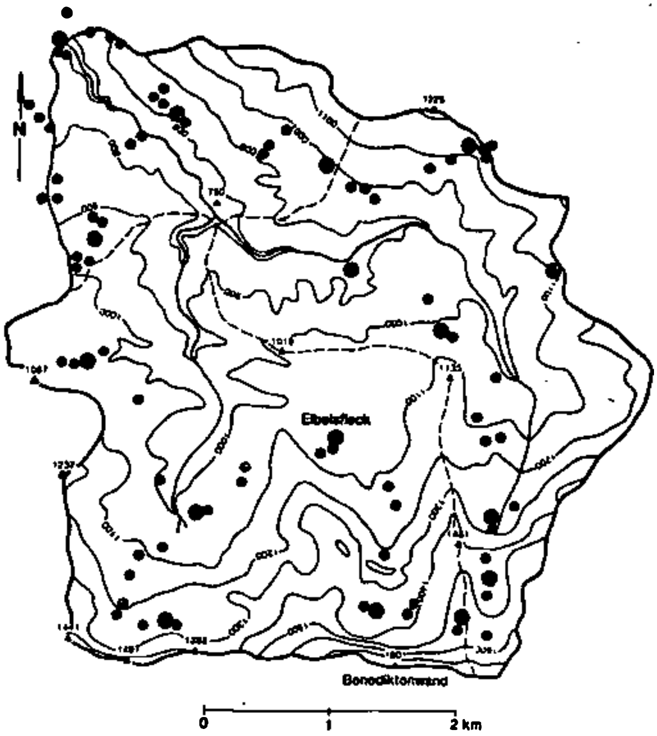

Fig. 8: Topographic map of Lainbach catchment area (Upper Bavaria) with sampling points for rainwater (●) and snow (•) (STICHLER & HERRMANN 1978a)

catchment area with a temporary snow cover and for the run-off processes in a glaciated catchment area during the period of ablation. The extent to which measurements of the ^{2}H and ^{18}O content can contribute towards answering these questions will be discussed in the following in the case of two catchment areas.

The Lainbach valley (Figure 8) is a very well installed, representative catchment area, 18.7 km^2 in size, in the boundary region of the northern limestone Lower Alps and Flysch Alps. Figure 9 shows the distribution in time for a period of three days of the precipitation events and of the Lainbach run-off in this catchment area together with the line of the $\delta^{18}O$ values in the Lainbach. The

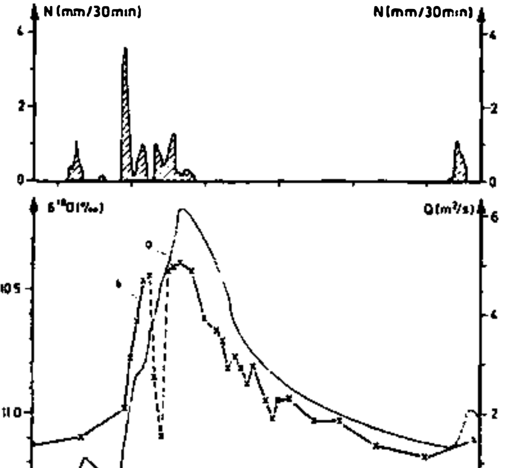

Fig. 9: Variation with time of ^{18}O-contents (δ) and run-off (Q) in the Lainbach creek (below) and precipitation distribution (N) at mean areal altitude (above) during the period of 14-16 July 1977 (STICHLER & HERRMANN 1978b)

regional precipitation during the period of observation was 23.5 mm and its mean $\delta^{18}O$ value δ_d = -7.45 ± o.5o per mil. Immediately preceding the run-off event a $\delta^{18}O$ value of δ_i = 11.14 ± o.o3 per mil was measured in the Lainbach.

For balancing the direct (above surface) and indirect (underground) run-off components (Q_d and Q_i) of the total run-off (Q_t) flowing into the Lainbach, it is possible to use the isotope balance equation

$$\delta_t Q_t = \delta_d Q_d + \delta_i Q_i \tag{1}$$

whereby δ_t stands for the $\delta^{18}O$ value in the total run-off. The evaluation of this balance showed that only 12 ± 3 % of the precipitated water run-off above ground, the remainder running off underground. The precipitation penetrating the soil produces an increased groundwater gradient, this leading to increased groundwater run-off into the receiving water course. Isotope balances such as this, however, can only be performed with sufficient accuracy if there are significant differences in the δ-values of the individual run-off components and if corresponding resolution in time of the δ-values is achieved.

For these reasons it is generally simpler to perform isotope run-off balances on melt water than on rain water; the difference between the δ-value of the snow precipitation during the cold season of the year and that of the groundwater is relatively high and the duration of the melting process relatively slow.

As an example of this, figure 1o shows the total run-off and the δ^2H line during a longish snowmelt period early in the year for the same catchment area of the Lainbach.

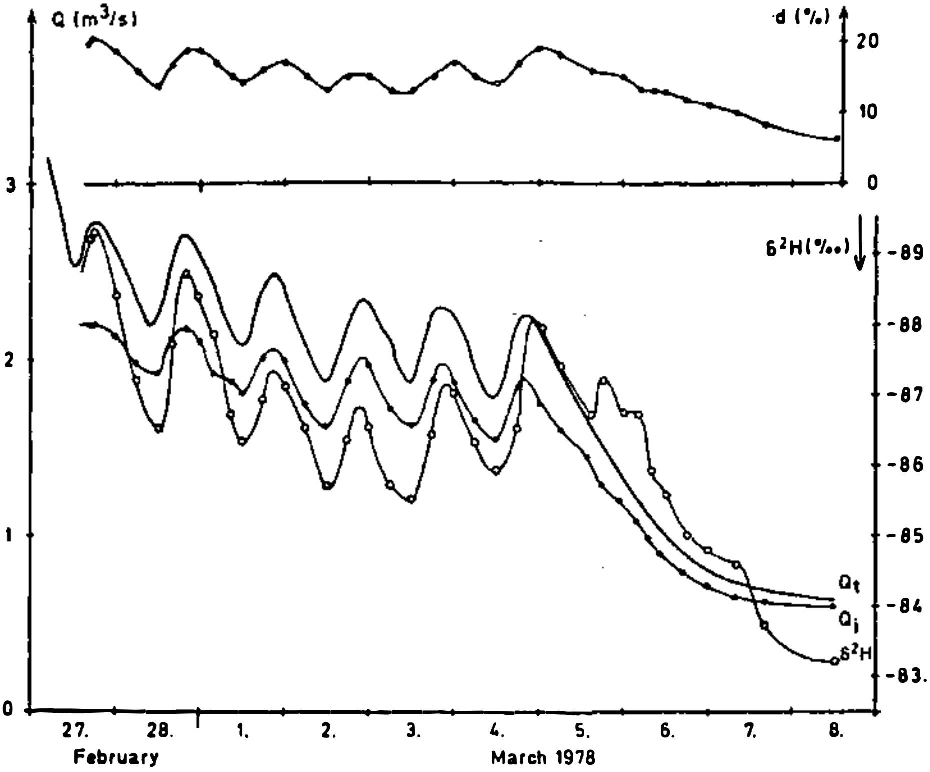

Fig. 1o: below: Variation with time of ^2H contents (δ^2H) and run-off (Q_t) in the Lainbach creek with calculated (by Eq.(1)) indirect run-off component (Q_i) during the period of 27.Febr.- 8 March 1978 above: relative amount of direct runoff ((Q_i/Q_t)· 1oo %) calculated from Q_t and Q_i for the same period (HERRMANN et al. 1978)

For determining the quantity δ_d in equation (1), measurements of the ^2H content in the run-off were used from two snow lysimeters set up at a representative point of the catchment area (Figure 11). δ_i was derived as the mean value of the ^2H contents during low-water periods in the Lainbach. The result is shown in figure 1o as the hydrograph of the indirect run-off and the resulting proportion of the direct melt water run-off to the receiving stream.

Here too it is seen that only a small proportion of the melt water (15 to 3o %) reaches the Lainbach directly, the remaining melt water infiltrating and mobilizing groundwater.

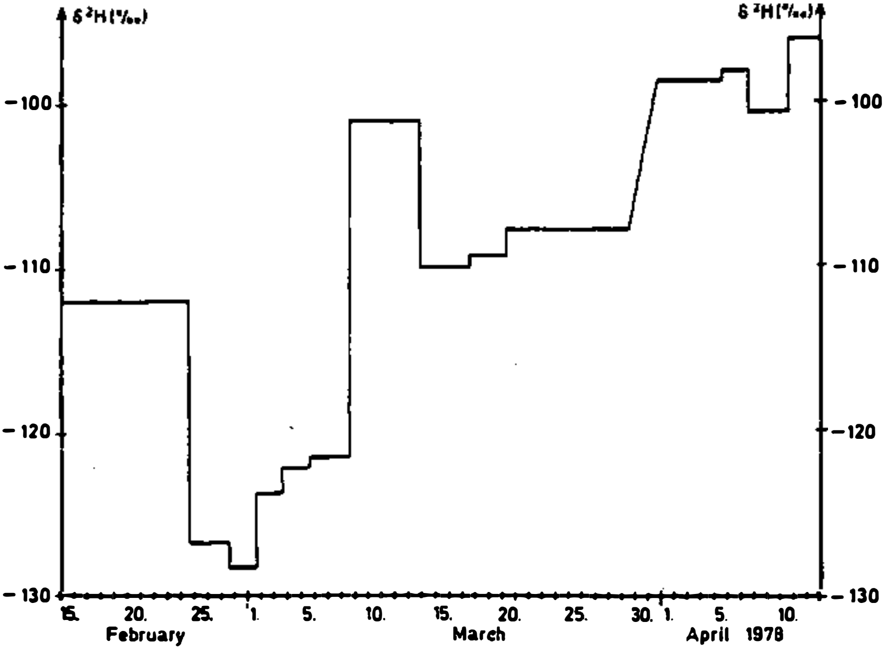

Fig. 11: ^{2}H contents in the outflow of lysimeters, situated in the Lainbach catchment area at 1o3o m a.s.l. (HERRMANN et al. 1978)

The second example is the glaciated catchment area of the Vernagt creek in the Oetztal Alps with a surface area of 11.44 km^2, covered 82 % by the Vernagt glacier (Figure 12). With the aid of isotopic contents and electrolytic conductivity it is possible to determine the run-off components of the glacier, of ice melt, of directly and indirectly (i. e. through the glacier) running off snow melt water and of water seeping through the ground moraine. Figure 13

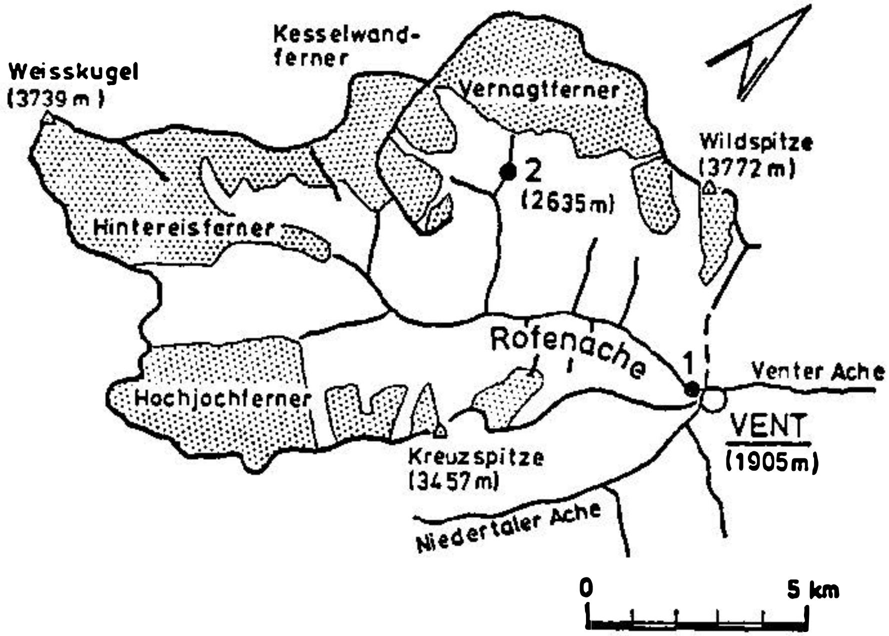

Fig. 12: Catchment area of Rofenache-creek (Oetztal Alps)

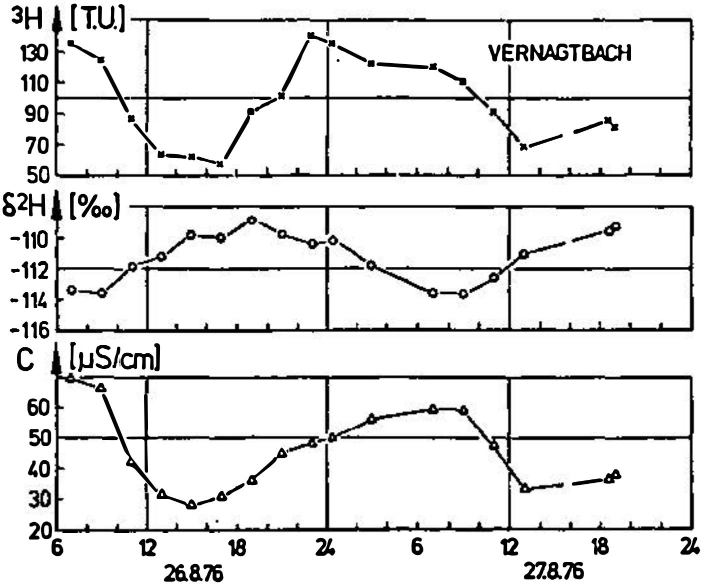

Fig. 13: Variations with time of ^{3}H- and ^{2}H-contents and of electrolytic conductivity in the discharge of Vernagt-glacier (Oetztal Alps) during 26 and 27 August 1976 (BEHRENS et al 1979)

shows this in the case of daily variations of ^3H and ^2H content and electrolytic conductivity in the glacier run-off during the summertime ablation period. It can be seen that during the time of highest melting in the early afternoon the ^3H content and the conductivity reach their minimum and the ^2H content reaches its maximum. This is conditioned by the fact that at this time the majority of the melt water consists of old glacial ice, which includes no bomb tritium, and of the snow of high ^2H content that has fallen in the summer; the proportion of mineralized " groundwater " from the glacier bed is low compared to the melt water. The lines can be quantitatively evaluated with the aid of a balance equation corresponding to the equation (1).

$$c_t Q_t = \sum_n c_n Q_n \qquad (2)$$

whereby c stands for the isotopic contents or the electrolytic conductivity and Q for the run-off. t represents the total run-off values and n the values of the individual run-off components. The result of this model calculation is shown in figure 14 for the period of time represented in figure 13. One can see that the main run-off component comes from the ice melt, followed by directly running off snow water, this arriving later, however, because of the longer distance that it has to flow. The components that run off within and under the glacier have substantially different residence times (LÖSCHHORN et al. 1976, OERTER et al. 1978). Measurements of ^3H content lasting several years, for instance, have shown that the water flowing from the glacier in winter has a mean residence time of approximately five years in the glacier (BEHRENS et al. 1979).

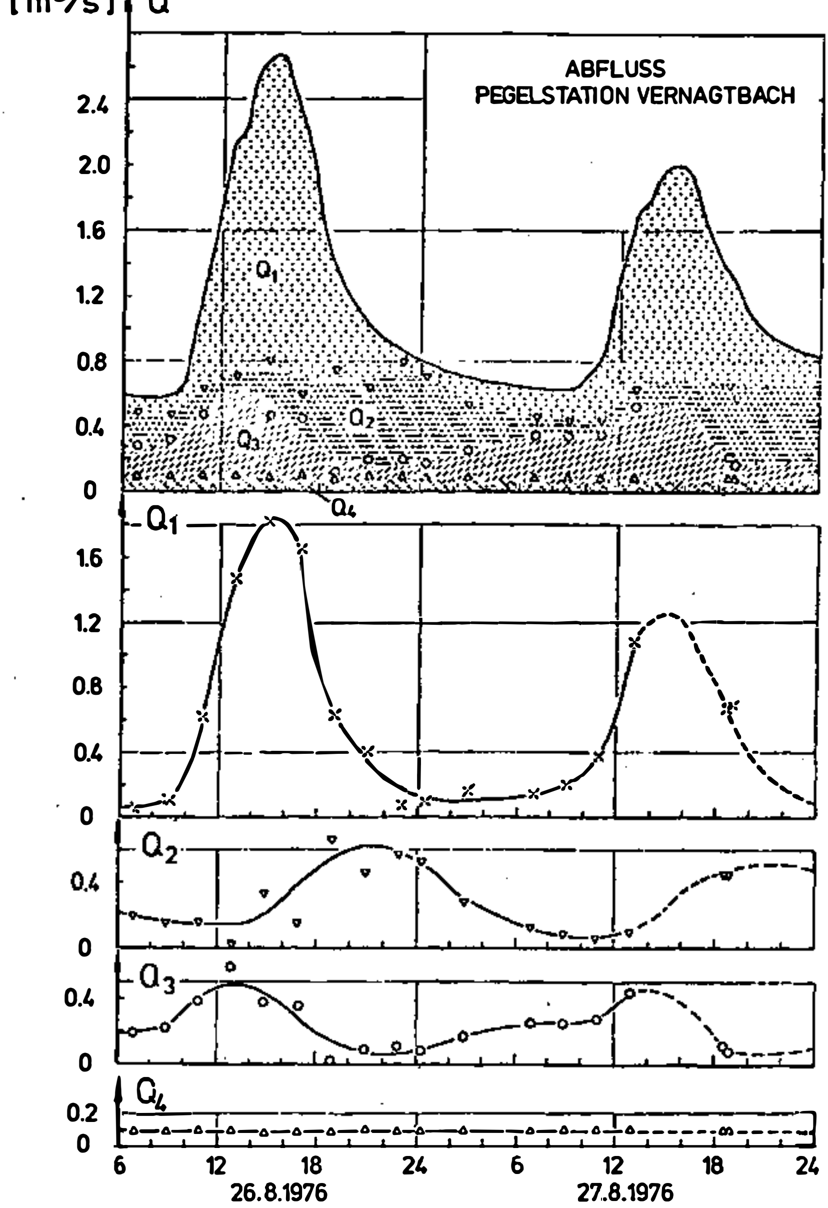

Fig. 14: Total run-off Q and run-off components from ice melt (Q_1), directly running off snow melt water (Q_i), indirectly running off snow melt water (Q_3) and groundwater (Q_4) in the discharge of Vernagt glacier during 26 and 27 August 1976 (BEHRENS et al. 1979)

5. Communication of groundwater and surface water

Similarly to precipitation/run-off relationship, the communication of groundwater and surface water can be investigated by measurements of the ^{2}H or ^{18}O content if significant differences exist between these contents or in their relation. This is the case, for instance, if the groundwater and the surface water have catchment areas lying at different altitudes or if the surface water is enriched in ^{2}H and ^{18}O through evaporation. As an example figure 15 shows the results of a series of measurements in which the effect of an excavated lake on groundwater was to be investigated. The positions of the groundwater measuring points around this lake, which

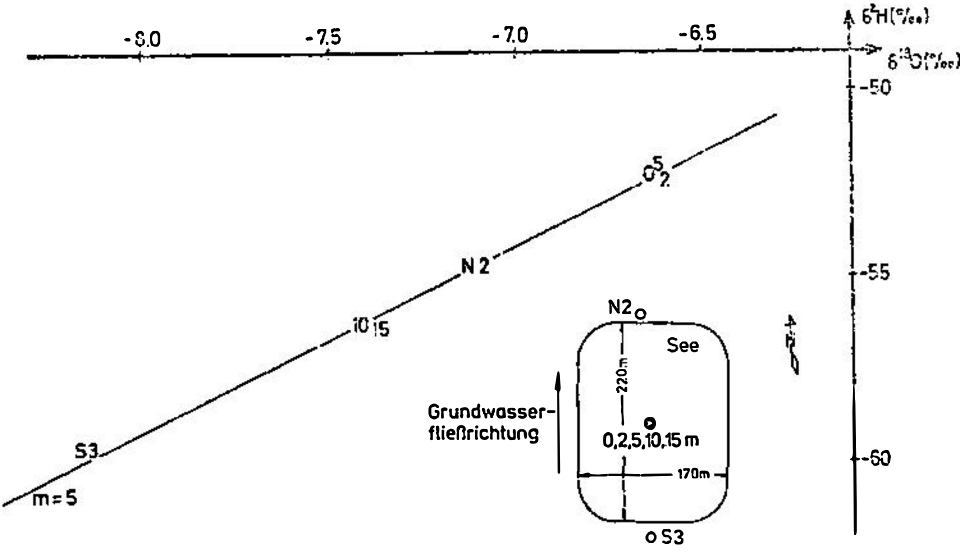

Fig. 15: ^{2}H- and ^{18}O-contents of lake water and groundwater in the area of an excavated lake in the Rhine valley together with the location of sampling points (o groundwater, • lake water). Depths of seawater sampling are given in meters; m = slope of $\delta^{2}H$-$\delta^{18}O$-line (STICHLER & MOSER 1977)

is located in the Rhine valley near Ludwigshafen and is about 15 meters deep, can be seen from the plan in figure 15. From the position of the measuring points in the δ^2H-$\delta^{18}O$ diagram it can be seen that the groundwater that flows approximately from S to N is enriched in the lake in ^2H and ^{18}O through evaporation and thus distinguished from the groundwater that does not flow through the lake. This lake water can be verified in the groundwater measuring points situated in the north of the lake. The quantitative evaluation of the time series recorded for isotopic content at different depths of the lake and in the groundwater measuring points provides information about the mixing processes in the lake and in the surrounding groundwater, about the groundwater flow field (i.e. velocity and direction of flow) in the vicinity of the lake and thus about possibilities of the quality of the groundwater being affected by such gravel pits to the depth of the groundwater.

6. Origin and residence time of groundwater

The isotopic effects in the formation of precipitation enable conclusions about the origin of the groundwater or its catchment area and the climatic conditions prevailing at the time of the groundwater recharge.This is possible because the ^2H and ^{18}O content of the precipitation seeping into the ground remains largely unaltered, disregarding isotopic-exchange effects in thermal waters. Investigations of isotopic content thus aid chemical investigations in providing information about the aquifer, i.e. the path of the groundwater.

An example for the application of the isotopic altitude effect is shown in figure 16. To arrive at information about the altitude of the catchment area of brine springs in the region of Reichenhall (Upper Bavaria), the dependence

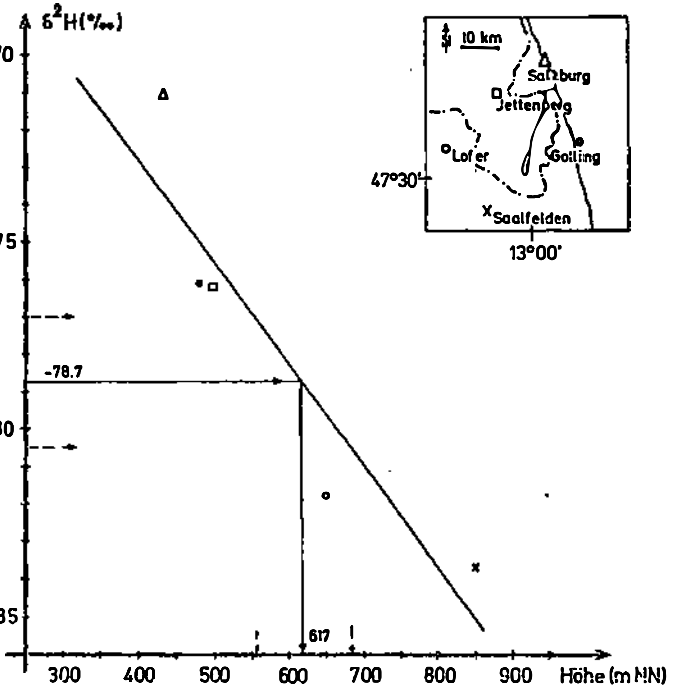

Fig. 16: Estimation of mean altitude of the catchment area of brine sources near Reichenhall (Upper Bavaria) from isotope altitude effect of precipitation in the environment of Reichenhall (for sampling points, see the map in the right corner) (RAUERT & STICHLER 1979)

on altitude of the δ^{2}H values of the precipitation was first measured, using five stations in the surroundings of the spring region. The measuring points entered in figure 16 are the weighted annual averages of these δ^{2}H values for the period 1975/1976 as a function of the altitude of the precipitation collecting stations. If one puts an averaging straight line through these measuring points, the result is that the ^{2}H content of the precipitation decreases by approximately 2.7 % for every 1oo meters of altitude, and this corresponds to experience gained in other areas that have been investigated. If one enters the mean value of the δ^2H value of the brine springs in this relationship, the brines according to its ^3H content and its δ^2H-$\delta^{18}O$ relationship having been formed of recent precipitation, the result is a mean altitude of the catchment area of somewhat more than 600 meters above sea level with an uncertainty of 130 meters. Such an altitude for the catchment area is completely in accord with hydrogeological facts.

For determining the residence time of groundwater, with relatively fast run-off (e.g. in karst), the seasonal ^2H or ^{18}O content, often depending on the individual event, can be used. Figure 17 shows the variation in time of the ^2H content in karst springs in the catchment area of the Ljubljanica (Slovenia), these reflecting the ^2H content of the precipitation. The high δ^2H value observed in September 1973 as the result of a single precipitation, originating most likely from tropic air masses, falls at different rates in the individual sources. It can thus be concluded that the springs 1.32 and 1.34 are fed by a run-off system without a largish reservoir, whilst the water of the springs 1.36 and 1.37 is retained longer in the body of karst water. Assuming an exponential model, it is possible to estimate a mean residence time of approximately one year.

With longer residence times of groundwater in the aquifer the seasonal fluctuations of the ^2H and ^{18}O content are compensated by hydrodynamic dispersion. The longterm fluctuations of temperature and humidity associated with climatic changes, however, are reflected in the δ^2H and $\delta^{18}O$ values and their relationship of formerly recharged groundwater. The comparison illustrated in figure 18 between isotopic contents of old groundwaters infiltrated during the period of the Spätwürm - according to ^{14}C measurements -

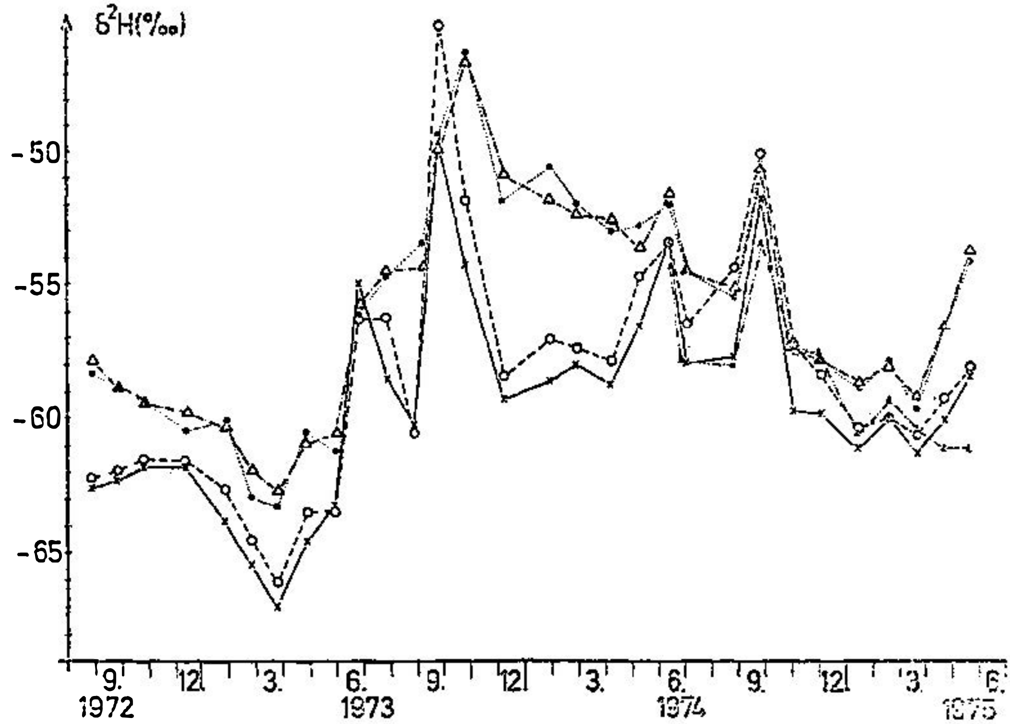

Fig. 17: Variations in time of ^{2}H-contents in various Karst springs in the catchment area of Ljubljanica (Slowenia) during the period of August 1972 to Mai 1975 (MOSER et al. 1976)

x spring No. 1.32
o " " 1.34
• " " 1.36
△ " " 1.37
▲ " " 1.38

from eastern Saudi Arabia and of more recent water from wadis of the same region shows that the Pleistocene water originates from a cooler and more moist climatic period than that of today. This is a result of the lower δ^2H and δ^{18}O values, which are also below the line presently valid for Central Europe $\delta^2H = 8\ \delta^{18}O + 10$. The low deuterium excess points to the fact that the precipitations that renewed the groundwater in former times originated from water vapour evaporated from the ocean at a lower moisture deficit than is the case today (see figure 1). We arrived at similar results with old groundwaters from Libya that was investigated in cooperation with the Ruđer Bošković Institute (SRDOČ et al. 1978).

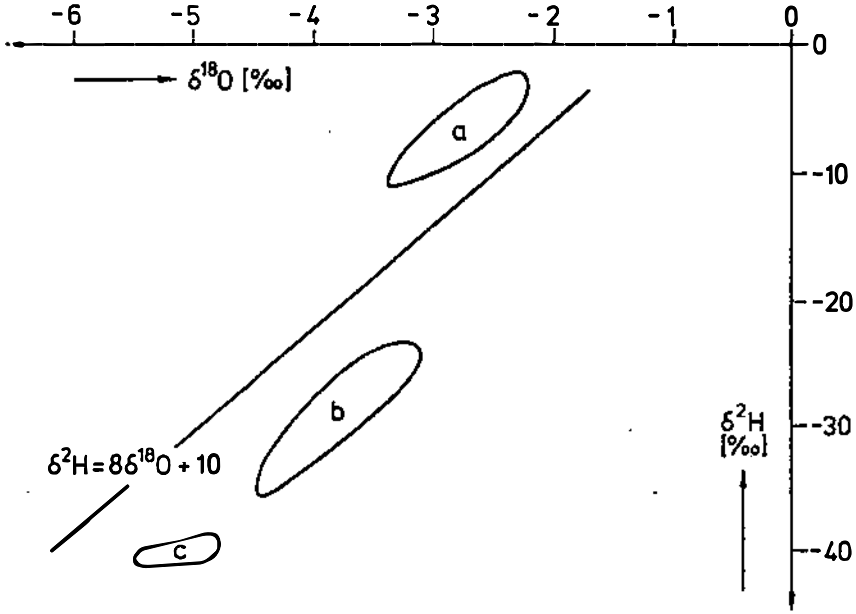

Fig. 18: ^{2}H- and ^{18}O contents of groundwater and Karst water from Saudi Arabia (sampling period March/April 1973)
(a) recent groundwater from wadi aquifers (gravel, sands), Eastern Saudi Arabia near Ryadh.
(b) Karst springs near Al Qatif, East coast, 2o samples (^{14}C model age of 2 samples > 33 ooo y and > 22 ooo y b.p.)
(c) Karst water from oasis area Al Hasa, ca. 55 km from East coast 2 samples (^{14}C model age of 2 samples > 33 ooo y b.p.) (MOSER et al. 1978)

7. Summary

The examples briefly given here are intended to show the kind of contributions towards solving hydrological problems that can be expected from analyses of ^{2}H and ^{18}O content. This review was, of course, because of the restricted amount of time available, incomplete as regards the hydrological problems encountered and also subjective, because only the work of the Institute for Radiohydrometry of the GSF has been reported on.

The narrowing down of the discussion to the applications of ^{2}H and ^{18}O analyses should not create the impression that this method alone is sufficient for handling the hydrological problems in the examples given. In many cases this would most certainly lead to misinterpretations. The generally very complex situation in nature calls rather for as many complementary methods as possible, and among these ^{2}H and ^{18}O analyses should most definitely be included.

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