# Observation of isotopes in the water cycle—the Swiss National Network (NISOT)

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Abstract The Swiss National Network for the Observation of Isotopes in the Water Cycle (NISOT) includes eleven precipitation, seven surface water (river) and three groundwater stations, where tritium, deuterium and oxygen-18 are monthly measured in composite samples. The network provides a good overview of the characteristic isotope signatures in recharge waters in Switzerland and of the relations between isotopes and altitude, orography and the amount of precipitation. Mixing of air water vapour and surface waters can be observed along a NW/SE cross section through the Alps. With increasing length of the data series, the network provides a valuable contribution for national and international scientific and practical applications in surface and subsurface hydrology, climatology and biology. The Swiss Geological Survey at the Federal Office for Water and Geology operates the isotope network within the legal framework of the Federal Law on the Protection of Waters and guarantees quality, access and distribution of the isotope data.

Keywords National observation network  $\cdot$  Stable isotopes · Tritium · Water cycle · Switzerland

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# Introduction

The Swiss National Network for Isotopes in the Water Cycle (NISOT) has observed since 1992 monthly tritium, deuterium and oxygen-18 in precipitation, surface water (river) and groundwater. The network, operated by the Swiss Geological Survey at the Federal Office for Water and Geology (FOWG), provides specific long-term isotope data series for application and research for the following purposes: hydrological and climatological studies, groundwater management and protection, determination of groundwater origin and age, and calibration of flow and transport modelling (Etcheverry 2002; Etcheverry and Parriaux 1998). The objective of this paper is to present the organisational structure, the background and selected significant results of NISOT.

The stable isotopes oxygen-18 and deuterium, and the radioactive isotope tritium are constituents of the water molecule, and, as a consequence, are ideal natural tracers for describing phenomena of the water cycle. The  $\delta^{18}O$  and  $\delta^2$ H in waters are strongly dependent on meteoric processes, and so infiltration into a specific environment leads to a characteristic isotopic signature. This signature labels the origin of groundwater and surface water–groundwater interactions (e.g. Sonntag and others 1983). Compared to ocean water, the atmospheric moisture, precipitation, surface water, and groundwater are mostly depleted in the heavy isotopic species <sup>18</sup>O, <sup>17</sup>O and <sup>2</sup>H. The main reason for the depleted values of these waters is mass dependent fractionation during evaporation and condensation. A detailed description of these processes and the different measurement techniques of stable isotopes and of tritium are described in detail in Clark and Fritz (1997), Lucas and Unterweger (2000), Gat and others (2001), and Hoefs (2002).

The NISOT network for the observation of isotopes has to be considered in the context of other national observation networks in Switzerland such as the Environmental Radioactivity Survey Network (SFOPH 2002), the National Soil Monitoring Network NABO (Desaules and Dahinden 2000), the National Atmospheric Quality Network NABEL (SAEFL 2001), the National Long-Term Network of Swiss Rivers NADUF (Binderheim-Bankay and others 2000), the National Network for the Observation of Groundwater Quality NAQUA (SAEFL 1998; Greber and others 2002) and the National Network for Groundwater Quantity NABESS (Buttet and Eberhard 1995). The network for the

observation of isotopes provides a good overview of the main characteristic isotope signatures in Switzerland and, with the increasing length of the data series, also provides a valuable contribution to joint international isotope programmes. The Global Network for Isotopes in Precipitation (GNIP), initiated by the International Atomic Energy Agency (IAEA) in 1961, provides such a joint international isotope programme as a tool for climatological interpretation of palaeorecords, validation of global atmospheric circulation models and global scale water balances (IAEA/WMO 2001). Potential users of the NISOT data were initially expected to be hydrogeologists and engineers, but experience has shown that most data requests originate from climatology and biology. Isotope methods are probably still under-utilised in applied hydrogeology (Etcheverry 2002).

# Background and organisational structure

#### Background

In the 1960s, the Division of Climate and Environmental Physics of the University of Berne began to determine tritium and oxygen-18 in groundwater, surface water (river) and precipitation in Switzerland. Since 1970 tritium and oxygen-18 have been measured regularly in monthly precipitation composites at five observation stations: Berne, Meiringen, Guttannen, Grimsel and Locarno (Fig. 1).

The Swiss Geological Survey, formerly the Swiss National Hydrological and Geological Survey, observes in the scope

of its national isotope network (NISOT) monthly tritium, deuterium and oxygen-18 in precipitation, surface water and groundwater. The five long-term precipitation stations of the Division of Climate and Environmental Physics of the University of Berne, which have isotope data dating back to 1972, are part of this network; they also provide data for the network of the Swiss Federal Office of Public Health (SFOPH 2002), and until 1992 data for the Global Network for Isotopes in Precipitation GNIP (IAEA/WMO 2001). The 11 precipitation stations cover the different climatological areas of Switzerland such as the Jura Mountains, the Central Plateau, the Alps and the southern side of the Alps. They cover also the significant altitude sequence along the NW/SE profile through the Alps from Berne (541 m a.s.l.) over Grimsel (1,950 m a.s.l.) to Locarno (379 m a.s.l.). The seven surface water stations cover the major rivers of Switzerland and are mainly located at river inflows into major lakes: Rhine-Diepoldsau, Rhône-Porte du Scex, Ticino-Riazzino, and Aare-Brienzwiler (Fig. 1). The average altitudes of the river station catchments vary between 1,010–2,466 m a.s.l. (Table 1). The groundwater station of the Savigny Spring was added to the national network to observe climate-induced isotope changes in recharge within a small catchment (Table 1). Two groundwater pumping stations, which were added provisionally to the network in October 2002, observe fluctuations of isotopes in groundwaters in two characteristic gravel aquifers of Switzerland (Fig. 1). The Swiss Geological Survey operates the isotope network (NISOT) within the legal framework of Article 76 of the Federal Constitution of the Swiss Confederation of April 18, 1999 and of Article 57 of the Swiss Federal Law on the Protection of Waters of January 24, 1991. The Swiss



#### Fig. 1

Map of Switzerland showing the monitoring station localities of the Swiss National Network for the Observation of Isotopes in the Water Cycle (NISOT). Cross section A-A' is shown in Fig. 3

Table 1 Characteristics of the observation stations of the Swiss National Network for the Observation of Isotopes in the Water Cycle (NISOT)



Ordinance for extraneous substances and components in food products of June 26, 1995 defines the standards of water radioactivity for human consumption, for example the tritium concentration in drinking water should not pass 10 kBq/l (about 80,000 TU).

#### Organisational structure and methodology

The Swiss Geological Survey collaborates with the Universities of Berne (Division of Climate and Environmental Physics) and Lausanne (Institute for Mineralogy and Geochemistry) for tritium/oxygen-18 and deuterium measurements respectively. The contractors deliver the raw data twice a year to the Swiss Geological Survey for validation, and then both isotope laboratories check the data consistency of selected samples by realising additional analyses. A group of scientific and practical experts discusses once a year results, reports, organisational questions and the future development of the network. Figure 2 shows a flow chart of the organisation structure of the national network.

Oxygen-18 and deuterium are analysed by mass spectrometry at the Division of Climate and Environmental Physics of the University of Berne and at the Institute of Mineralogy and Geochemistry of the University of Lausanne with analytical errors of  $\pm 0.04$  and  $\pm 1\%$ , respectively. Tritium is measured at the Division of Climate and Environmental Physics of the University of Berne by counting  $\beta^-$  decay events in a liquid scintillation counter and by a gas counter as well. The detection limit on a  $2\sigma$ base ( $\sigma$ =standard deviation) is 1.6 TU for the liquid scintillation counter and 0.7 TU for the gas counter. Both laboratories participated at the IAEA interlaboratory comparison tests of stable isotopes and of low-level tritium





Organisational structure of the Swiss National Network for the Observation of Isotopes in the Water Cycle (NISOT)

measurements in water in 1999 and 2000, respectively (Lippmann and others 1999; Gröning and others 2001). All 11 stations for isotope measurements in precipitation are located next to the rain-gauging stations of ''Meteo-Swiss'' at Zurich, which provide specific data for precipitation, water vapour pressure and air temperature. The monthly composite sample of precipitation for isotope analysis corresponds to the sum of all precipitation events collected on a daily basis. Stations with discharge and

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water temperature devices were chosen for the measurement of isotopes in surface waters (rivers), and the selected stations are, if possible, included in the National Long-Term Network of Swiss Rivers (NADUF). The isotopes at the NADUF-stations (the River Rhine at Diepoldsau, the River Aare at Brugg, the River Rhône at Porte du Scex, the River Ticino at Riazzino, and the River Inn at S-chanf) are determined from a 28-day, flow rateproportional composite sample. Surface water is pumped proportionally to the flow rate of the river during 28 days and is collected in a bottle, which is then sent to the laboratories for isotope analysis. On the other hand, a water sample is taken in the first and third week of each month at Aare-Brienzwiler and Aare-Thun (Fig. 1), and then both samples of each month are mixed and analysed for isotopes. The monitored Savigny Spring belongs to the AQUITYP-network of the Laboratory of Engineering and Environmental Geology at the Federal Institute of Technology of Lausanne, which has installed a flow gauging station and which determines monthly different physical and chemical parameters at the spring. Table 2 summarises the different parameters which are measured monthly in precipitation, surface water and groundwater at the different observation stations.

#### Data access/periodic publications for users

The oxygen-18, deuterium and tritium data are available for users six months after the end of the year by contacting the Swiss Geological Survey, Federal Office for Water and Geology. The isotope data and the associated hydrometeorological data are published as graphs in the Hydrological Yearbook of Switzerland (e.g. FOWG 2002), which can be downloaded as a PDF-file from the Internet (www.bwg.admin.ch). Brief scientific results based on the measured isotope data are published at five-year intervals to provide standard citation (Schotterer and others 1995, 2000). Data from the network for the observation of isotopes have also been published as a contribution to the Hydrological Atlas of Switzerland (Parriaux and others 2001).

## Results

#### Deuterium and oxygen-18 in precipitation

The climate in Switzerland is characterised by precipitation coming from the west from the Atlantic Ocean and by cold, dry winds coming from the east from the European continent. The influence of the Mediterranean Sea is generally restricted to the southern side of the Alps (Pfister

1999; Wanner and others 1998). The continental effect, which is the progressive  $^{18}$ O-depletion in precipitation with increasing distance from the ocean (Rozanski and others 1982; Sonntag and others 1983), is small within Switzerland. The Alps influence the transport of the air mass as a climate divide and thus provide characteristic isotopic signatures in precipitation (Schotterer and others 1993). The influence of the orography can be seen in the increase of precipitation with altitude on the northern side and due to a barrier effect on the southern side of the Alps; inner-alpine regions experience often dryness (Frei and Schär 1998; Kirchhofer and Sevruk 1992).

Figure 3 shows the schematic NW/SE cross section from La Brévine, situated within the Jura Mountains, to Locarno on the southern side of the Alps. It also shows the annual average precipitation and the average  $\delta^{18}$ O and  $\delta^2$ H at the different observation stations of the NISOT network along this cross-section for the observation period between January 1994–December 2001. The  $\delta^{18}$ O and  $\delta^2$ H in precipitation at La Brévine  $(1,042 \text{ m } a.s.$ ].), where the annual average precipitation reaches 1,643 mm, is comparable to that of Berne (541 m) despite the higher altitude of the La Brévine station. This is a result of the rainout effect. Clouds coming from the West from France are forced to rain out when they reach the Jura Mountains and therefore the air mass arriving at Berne is depleted in  $\delta^{18}$ O and  $\delta^2$ H respectively. The annual average precipitation at Grimsel in the Alps is about twice the annual average precipitation in Berne and attains 2,058 mm. Precipitation at Locarno on the southern side of the Alps, which attains an annual average precipitation of 1,874 mm, is less depleted in the heavy isotope species than precipitation on the northern side of the Alps (Fig. 3), which is probably due to the higher average air temperature in Locarno and the vicinity to the Mediterranean Sea.

#### Altitude effect

The isotopic composition of precipitation changes with the altitude of the terrain because of the uplift and cooling of air masses and thus becomes more and more depleted in oxygen-18 and deuterium. Therefore, the altitude effect is also related to the air temperature (Gat and others 2001). Different relations between altitude and stable isotopes are discussed in the literature for Switzerland: Jura Mountains (Kullin and Schmassmann 1991; Muralt 1999), Central Plateau (Kullin and Schmassmann 1991; Vuataz 1982), Alps (Siegenthaler and Matter 1982; Kullin and Schmassmann 1991).

Figure 3 shows the decrease in oxygen-18 in precipitation in Switzerland from  $-9.69\%$  in Berne at 541 m a.s.l. to  $-13.25\%$  at Grimsel at 1,950 m a.s.l.. It continues to

#### Table 2

Parameter determination in precipitation, surface water and groundwater





Fig. 3

Schematic NW-SE cross section through Switzerland with annual average precipitation and average  $\delta^{18}$ O and  $\delta^2$ H in precipitation and surface water. Location of the cross section is shown in Fig. 1

decrease to the summits of the Bernese Alps where a longterm average of  $-17\%$  in oxygen-18 has been observed in an ice core at an altitude of 3,900 m (Schotterer and others

2001). Figure 4 shows the relation between the altitude of the observation stations and the average, extreme minimum and extreme maximum of  $\delta^{18}$ O and  $\delta^2$ H in precipitation for the period between January 1994–December 2001. The average decrease of oxygen-18 in precipitation in the Swiss Alps of about  $0.2\%$  per 100 m corresponds well with earlier findings (Siegenthaler and Oeschger 1980;



Fig. 4 Altitude effect of  $\delta^{18}O$  and  $\delta^2H$  in precipitation in the Swiss Alps

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station at Berne

Siegenthaler and Matter 1982). However, Fig. 4 indicates that Locarno on the southern side of the Alps and probably also Berne on the Central Plateau belong to another precipitation regime with different rainout mechanisms.

#### Seasonality of stable isotopes in precipitation

Figure 5 shows, as an example, the dataset obtained at the Berne precipitation station between January 1994– December 2001, including precipitation, air temperature, water vapour pressure, oxygen-18, deuterium and tritium values. The seasonal evolution of oxygen-18 and deuterium in precipitation is correlated to air temperature and water vapour pressure (e.g. Rozanski and others 1982;

Schoch-Fischer and others 1984). Oxygen-18 and deuterium attain generally maximum values in summer and autumn, when the air temperature and vapour pressure are high, and minimum values in winter, when the air temperature and vapour pressure are low. The three precipitation stations (Berne, Grimsel and Locarno) reflect the seasonality of the isotope distribution along the NW-SE cross section through the Alps (Fig. 3). Figure 6 shows the seasonal evolution of oxygen-18 and tritium for these three stations from January 1994– December 2001. The seasonality of oxygen-18 at these three stations is similar. The average amplitude of the oxygen-18 fluctuations from 1994–2001 attains about  $9.3\%$ 



Fig. 6 Monthly oxygen-18 and tritium in precipitation, January 1994–December 2001, network stations at Berne, Grimsel and Locarno

at Berne on the Central Plateau, about 9.5 $\%$  at Locarno on the southern side of the Alps and about  $10.6\%$  at Grimsel within the Alps. The difference between the extremes of  $\delta^{18}$ O is generally higher at Locarno (Fig. 4). This might be ascribed to a significant wet anomaly in the annual average precipitation in the Lake Maggiore region, in particular at the observation station in Locarno (Frei and Schär 1998). Moreover, the Lake Maggiore region is often affected by severe precipitation events (Geiger and others 1992). However, the observed relation between amount of precipitation and  $\delta^{18}O$  (Dansgaard 1964) cannot be verified by our data (Fig. 7a). A reason for the more pronounced extremes of  $\delta^{18}$ O at Locarno may also be evaporation during warm summer seasons with less precipitation during which the stable isotopes shift to more positive values.

For the period between January 1994–December 2001, the increase of  $\delta^{18}$ O with air temperature in monthly composites from Berne, Locarno and Grimsel attains 0.56, 0.70 and 0.71  $\frac{9}{20}$ /1 °C, respectively (Fig. 7b) and correspond to the range of observed values in the Alps (e.g. Rozanski and others 1993; Schotterer and others 2000). In Fig. 8 the relation of  $\delta^2 H - \delta^{18} O$  is shown for the monthly composites of precipitation between January 1994 and December 2001 at Berne, Grimsel and Locarno respectively. The slope of the individual regression lines is about rno station (representing also the other network stations) 8. The term "deuterium excess" d, defined as  $d = \delta^2 H - 8 \times \delta^{18} O$  shows seasonal fluctuations around the Global Meteoric Waterline defined as  $\delta^2$ H=8× $\delta^{18}$ O+10 (Yurtsever and Gat 1981). These fluctuations are due to evaporation conditions in the oceanic source area and to evaporation effects on falling raindrops and re-evaporated soil moisture mainly during summer (Schotterer and others 2000).

#### Tritium in precipitation and surface waters

The distribution of the NISOT-stations provides a good spatial resolution of the tritium signature in precipitation in Switzerland (Schotterer and others 2000; FOWG 2002). Increased tritium concentrations are frequently found in precipitation samples taken close to some industrial sites (SFOPH 2002; see also Fig. 1). Tritium concentrations (given here in the still commonly used Tritium Units) in precipitation above 80 TU (1 TU corresponds to 0.12 Bq/l) are only observed at the NISOT-stations in Berne and in Buchs/Suhr (Figs. 1, 5). Important sources are industrial plants close to Berne, La Chaux de Fonds, and Teufen producing tritium filled vials serving as light sources and incineration plants handling slightly contaminated waste, mainly from the watch industry (SFOPH 2002). Nevertheless, these tritium emissions are well below Swiss limits according to the Swiss regulations. Occasional contamination of the atmosphere by industrial tritium release is restricted to the lower atmospheric air mass, from which it is precipitated. Because of exchange and dilution with uncontaminated water vapour in the atmosphere and because of the orography of Switzerland, elevated tritium values are mainly observed close to the few and wellknown tritium emitting plants.

The seasonal fluctuations of tritium at Grimsel and Locaare characterised by maximum values in late spring and summer and minimum values in winter. The high values are due to the tritium input from the stratospheric reservoir (still containing higher tritium levels from the nuclear weapon test period) in spring and the specific meteorological conditions (precipitation distribution). In summer, evapo-transpiration from land surface may also add higher tritium concentrations to air moisture. During late autumn





a Relation between precipitation and oxygen-18. b Relation between air temperature and oxygen-18. Monthly composites from January 1994–December 2001, network stations at Berne, Grimsel and Locarno



and winter time the stratospheric tritium input is already diluted from oceanic water vapour with low tritium concentrations (Schotterer and others 1995). Swiss nuclear power plants (NPPs) hardly contribute to increased atmospheric tritium compositions. However they are the main source for prominent tritium peaks in samples from the River Aare at Brugg (Fig. 9). This river receives tritium from a NPP, which is situated 25 km upstream of the observation station and which releases controlled amounts of wastewater once a year. Part of the tritium in the river may be also related to the elevated

tritium concentrations in the precipitation over the Central Plateau, due to non-nuclear industrial activities (SFOPH 2002). Therefore mean tritium compositions are higher in the River Aare than in the other two large Swiss rivers Rhône and Rhine, but they generally do not exceed 25 TU (Fig. 9). Of course, mixing relations of the main rivers and their tributaries by means of their individual tritium concentrations, and the extension of tritium contamination plumes in rivers and in the atmospheric water vapour, cannot be determined with the low station density of the NISOT-network.





Monthly discharge, oxygen-18 and tritium in river waters, January 1994–December 2001, network stations at Rhine-Diepoldsau, Aare-Brugg and Rhône-Porte du Scex

#### Deuterium and oxygen-18 in surface water

Figure 9 illustrates the seasonal evolution of discharge, of oxygen-18 and tritium in the River Rhine, River Aare and River Rhône from January 1994–December 2001. The mean  $\delta^{18}$ O of the Rhine is -12.60% for the period from January 1994–December 2001. The variation patterns in the  $\delta^{18}$ O composition of the Rhône and the Aare correspond to those observed in the Rhine (Fig. 9). The different  $\delta^{18}$ O of the Rhine, Rhône and Aare reflect the different  $\delta^{18}$ O in precipitation governed by the different altitudes in the catchments (Table 1). Seasonal changes in isotopic composition in rivers are more pronounced in alpine basins due to enhanced snowmelt mixing and modification of groundwater-surface water exchange during varying winter periods. Dams for energy production and lakes may change short-term variability in river discharge which is due to mixing and discharge controls. As a result, the annual cycle of the  $\delta^{18}$ O composition of the rivers differs significantly from the isotopic cycle of precipitation with minimum  $\delta^{18}$ O values in winter and maximum values in summer.

The seasonality of the isotope concentration is dampened as river water mixes with lake water, but the seasonality still remains (Schotterer and others 2002). The damping influence of a great lake (Lake Constance) and the changing influence of snowmelt floods on the isotope distribution of river run-off can be observed along the River Rhine (Fig. 10). The seasonality of the isotope concentration is significant in the Rhine at Diepoldsau before entering Lake Constance. For example, the  $\delta^{18}O$  in the Rhine at Diepoldsau fluctuated in 1999 between  $-11.77$ and  $-13.58\frac{\%}{\%}$ . Downstream of Lake Constance at Rekingen, water mixing with tributaries, in whose catchments snow cover plays a minor role, eliminates almost all this seasonality (Fig. 10). The  $\delta^{18}O$  in the Rhine increases along its flow direction as the average altitude of the river catchment decreases. The snowmelt induced seasonality of the isotope concentration in the Rhine recovers downstream the confluence with the River Aare (Rhine at Weil, Figure 10). Discharge of the Aare is generally higher at the confluence than discharge of the Rhine (FOWG 2002).

## Conclusions

The Swiss National Network for the Observation of Isotopes in the Water Cycle (NISOT), operational since 1992, provides a significant resource for various scientific and practical applications in surface water and groundwater hydrology, and in climatology. Since isotopic fractionation during phase transitions is temperature dependent, the seasonally changing stable isotope composition of precipitation produces a specific input signal for recharge areas. The average  $\delta^{18}$ O and  $\delta^2$ H in precipitation decrease in the Alps with the altitude. Seasonal fluctuations of  $\delta^{18}O$ and  $\delta^2$ H in precipitation are significant and their amplitude depends not only on the meteorological parameters



#### Fig. 10

Seasonal evolution of  $\delta^{18}O$ , January 1996–August 2000, network stations at Rhine-Diepoldsau and Aare-Brugg (both NISOT-stations), Rhine-Reckingen and Rhine-Weil (both stations of University of Berne) (adapted from Schotterer and others 2002)

and the altitude, but also on the local topography. No relation is found along the NW/SE cross-section through the Alps between  $\delta^{18}$ O and the amount of precipitation in monthly composite samples but the correlation of  $\delta^{18}$ O with ambient temperature is significant (0.5–0.7  $\frac{\omega}{10}(1 \text{ °C})$ . Although the long-term trend of stable isotopes in the NISOT is generally similar, it does not override characteristic differences between the northern and southern side of the Alps originating from the different precipitation characteristics. The lower isotope ratios of river water, reflecting the higher mean elevation of its catchment area, are distinctly different from the local groundwater and rain water of lower elevation that yield higher isotope ratios. Therefore, long-term observation of the isotopic ratios in precipitation and surface waters is necessary and helpful to detect regional climate induced changes. The intense variability of isotopes due to the particular topography of Switzerland can thus provide a valuable contribution to the Global Network for Isotopes in Precipitation (GNIP) for climatological interpretation of palaeorecords and present changes in Europe. The significant maximum values of tritium monitored between 1994–2001 in precipitation at Berne (120–270 TU) and in the River Aare at Brgg (80–250 TU) show that the network for isotopes also provides important information about industrial releases to the atmosphere, to surface waters and to groundwater. Of course, the density of NISOT-stations is too low to establish mixing ratios between precipitation, surface, and groundwater using individual short-term contamination plumes, but the observed long-term local and regional differences of tritium input concentrations are a base for estimating recent components to old tritiumfree groundwater.

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# Legal bases

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Swiss Ordinance for extraneous substances and components in food products of June 26, 1995. Swiss Federal Council (817.021.23)

Switzerland's new Federal Constitution of April 18, 1999. Swiss Federal Council (101), www.admin.ch