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ISOTOPIC COMPOSITION OF PRECIPITATION IN WESTERN TRANSYLVANIA (ROMANIA) REFLECTED BY TWO LOCAL METEORIC WATER LINES

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Abstract: The present study summarizes the isotopic composition of precipitation from two stations in western Romania, Rosia Montana and Cluj-Napoca. The isotopic data has been used for building the Local Meteoric Water Lines (LMWLs) for the two localities, the first situated in the Apuseni Mountains, at an elevation of about 1200 m.a.s.l, and the latter close to the western border of the Transylvanian Depression, at 360 m.a.s.l. Rainwater and snow samples from both stations have been collected on a monthly basis between August 2014 and August 2015, by following the IAEA standardized methodology. A Picarro water isotopic analyser was used for measuring the isotopic composition of water in terms of deuterium and oxygen-18 content. The equation of the LMWL for Rosia Montana is $\delta H=7.87^{18}O+11.72‰$ ($r^2=0.995$), while for Cluj-Napoca it is $\delta H=8.03^{18}O+11.29‰$ ($r^2=0.997$), with intercepts of 11.72, and 11.29 respectively. Both lines are close to the Global Meteoric Water Line, slightly above it. The features of the two lines were interpreted in terms of altitude effect, deuterium excess, and influence of temperature on $\delta^{18}O$. To date, there are few isotopic data from precipitation available for Romania, and the addition of two LMWLs would be useful as a reference for further studies that include an isotopic approach.

Keywords: stable isotopes, oxygen-18, deuterium, d-excess, Local Meteoric Water Line, rainfall, altitude.

1. INTRODUCTION

The stable isotopes of the water constituents, deuterium (^2H, D) and oxygen-18 (^{18}O) are widely used in hydrology and hydrogeology, but also in many other disciplines, due to their potential to track the transfer of water between different environmental compartments. The isotopes are useful to better understand the natural processes involving water in its different states, and in connection with the atmosphere, hydrosphere, biosphere, or lithosphere. These stable isotopes may provide help in improving the atmospheric circulation models, better managing the water resources, and elucidating aspects of the environmental change at a regional and global scale.

The first essential contributions in this field occurred after 1950, when several researchers have used stable isotopes as a tool to better understand the water movement in limited regions as a watershed, but rapidly extending to a global scale afterwards (Epstein & Mayeda, 1953; Craig, 1961; Dansgaard, 1964).

One of the most important steps in this new science was marked by Craig (1961), which identified a strong correlation between oxygen and deuterium in natural waters, thus developing an empiric equation that defines the Global Meteoric Water Line (GMWL), which will be the base for many other isotopic applications. In its original form, Craig's equation has the expression

$$\delta D = 8\delta^{18}O + 10 \text{ (‰) } (vs. \text{ SMOW}) \quad (1)$$

Based on a much larger amount of data, Rozanski et al. (1993) have refined the equation established by Craig in 1961. Using annual weighted precipitation data from the GNIP network, they proposed the expression:

$$\delta D = (8.17 \pm 0.06)\delta^{18}O + (10.35 \pm 0.65) \text{ (‰) } (vs. \text{ SMOW}) \quad (2)$$

Local sets of data may yield an equation slightly different from the GMWL. The slope of 8 in Craig's equation may generally vary in the range 8 ± 0.5, but values between 5 and 9 may also occur under particular circumstances. The value 8 of the slope corresponds to the equilibrium Rayleigh condensation of rain at 100% humidity (Kendall & Caldwell, 2006). Local Meteoric Water Lines have been developed in different countries in Europe, and at a worldwide scale (*e.g.*: Hungary – Deák, 1995; Slovenia and Croatia – Polona et al., 2006; Italy – Giustini et al., 2016; Sweden – Jonsson et al., 2009; Slovakia – Holko et al., 2012; USA – Lyn et al., 2004; China – Liu et al., 2010a; Australia – Liu et al., 2010b; India – Warrier & Babu, 2011; Antarctica – Fernandoy et al., 2012), etc. It is worth to mention the efforts of the International Atomic Energy Agency (IAEA) in Vienna, that initiated global networks of isotopes in precipitation and rivers (GNIP and GNIR, respectively). The Global Network of Isotopes in Precipitation (GNIP) was established by IAEA in collaboration with the World Meteorological Organization (WMO) in 1961, and takes advantage of the voluntary contribution of different institutions worldwide. Over 1000 stations from 125 countries and territories worldwide have provided samples of precipitation for deuterium, oxygen-18, and some of them also for tritium measurements. Due to this initiative, a huge amount of data is currently available to the scientific community.

Some published studies including original LMWLs are available for the Romanian territory (Fig. 1). Țenu et al. (1981) have published a LMWL in Western Romania (Oradea – Satu Mare area) with the equation $\delta D = 6.7\delta^{18}O - 3.1‰$. Further on, Costinel et al. (2009) have developed a LMWL with the equation $\delta D = 8.079^{18}O+11.9‰$ in Raureni-Valcea area (Central-Southern Romania). Close to this area, in the Middle Olt River basin, the LMWL established by Popescu et al. (2014) has the equation $\delta D = 7.98\ \delta^{18}O + 9.82‰$, updated to $\delta D = 8.04\ \delta^{18}O + 9.79‰$ in Popescu et al. (2015). Ionete et al. (2015) published LMWLs for 6 localities with mineral water sources in the Romanian Carpathians. The approach used monthly cumulative samples collected in rain gauges for one year in 2001-2002.

The established equations are the following:

Borsec:	$\delta D = 7.68\delta^{18}O + 6.49$
Vatra Dornei:	$\delta D = 7.78\delta^{18}O + 6.80$
Bodoc:	$\delta D = 8.02\delta^{18}O + 4.98$
Domogled:	$\delta D = 8.72\delta^{18}O + 15.00$
Lipova:	$\delta D = 9.10\delta^{18}O + 16.41$
Stana de Vale:	$\delta D = 8.31\delta^{18}O + 5.72$.

As the available isotopic studies of precipitation and LMWLs are relatively few, we decided to build two new lines, based on a rigorous methodology, for Rosia Montana and Cluj Napoca localities. These may represent a useful reference for any further isotopic studies on atmospheric precipitation, surface-, and groundwater.

2. GEOGRAPHIC POSITION AND CLIMATIC SETTING

Two stations for collecting samples for the isotopic monitoring of precipitation were installed in Rosia Montana (46°18'00"N, 23°07'48"E) and Cluj-Napoca (46°46'48"N, 23°36'36"E) (Fig. 1).

2.1. Roșia Montană

Rosia Montana station (RM) is positioned at Rotunda weather forecast station, at an elevation of 1196 m.a.s.l., within the Southern Apuseni Mountains area. The average annual temperature, based on the 2015 recordings at the weather station,

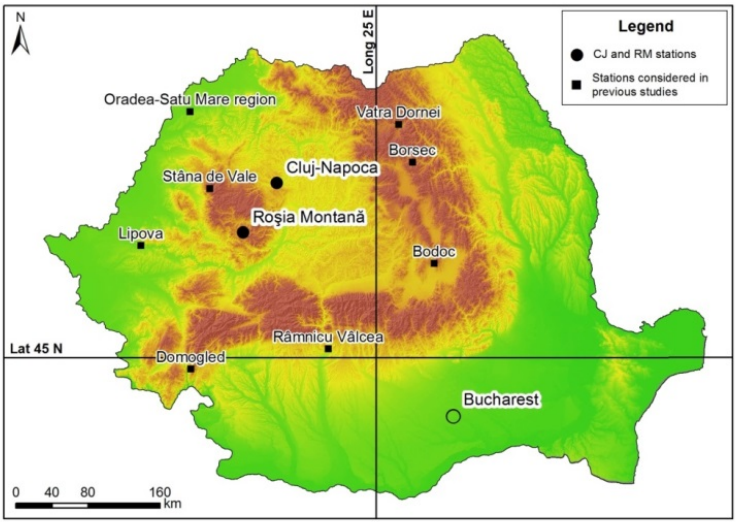

Figure 1. Map of Romania displaying the isotope collection stations

is 8.27°C, with a maximum monthly average of 18.6°C during the summer period, and minimum monthly average of –2.4°C in winter. The annual mean of the rainfall amount, calculated for the period of sampling of the performed research was 694.3 mm. The snow cap lasts for 3 to 4 months during the winter.

2.2. Cluj-Napoca

Cluj-Napoca station (CJ) was placed in the western part of the town of Cluj-Napoca at an altitude of 360 m.a.s.l. It is a hilly area, near the western border of the Transylvanian Depression. The climate is continental moderate, with average annual temperature of 11.6°C, with a maximum monthly average of 22.1°C during the summer season, and minimum monthly average of –0.3°C. The average annual precipitation for the period of the study was 682.8 mm, based on the recordings of Cluj-Napoca weather station.

3. METHODS

The precipitation samples for the isotopic study have been collected by following the IAEA standardized methodology. A Palmex rain sampler RS1 produced by Palmex Ltd. Zagreb, Croatia was used (Fig. 2). The rain water accumulated in the collector is recovered monthly, according to the protocol established by IAEA. The samples have been analysed in the Isotopes Laboratory of Babes-Bolyai University, and duplicates have been sent to IAEA for analysis and inclusion of the results in the GNIP database. The network was joined on a voluntary basis, given the insufficiency of currently available data on isotopes from precipitation in Romania. The two stations were labelled by GNIP as 1512001 Cluj-Napoca, and 1518401 Rosia Montana.

The meteorological data were provided by Rotunda Meteorological Station in Rosia Montana, and by the North-Transylvania Regional Meteorological Centre in Cluj-Napoca. Oxygen-18 and deuterium (^{2}H) isotopes in water were analysed by using a Picarro isotopic water analyser (Picarro CRDS analyser L2130-I), an instrument based on the cavity ring-down spectroscopy (CRDS) technology, able to measure simultaneously the deuterium and oxygen-18 ratios from liquid water samples. The isotopic ratios are reported using the δ notation relative to the Vienna Standard Mean Ocean Water (VSMOW) international standard. Two internal standards calibrated by using standards provided by IAEA Vienna, were used.

The data of ^{18}O and ^{2}H are expressed as:

$$\delta_{(‰)} = \left(\frac{R_x}{R_{std}} - 1\right) * 1000 \qquad (3)$$

The methodology recommended by Picarro for the laboratory analysis has been used. A quantity of 2 ml of re-filtered sample (0.22 μm pore size) is required.

Filters and syringes are individual for each sample (5 ml syringes). At every 5 samples, two different standards in terms of isotopic composition are injected in the analyser.

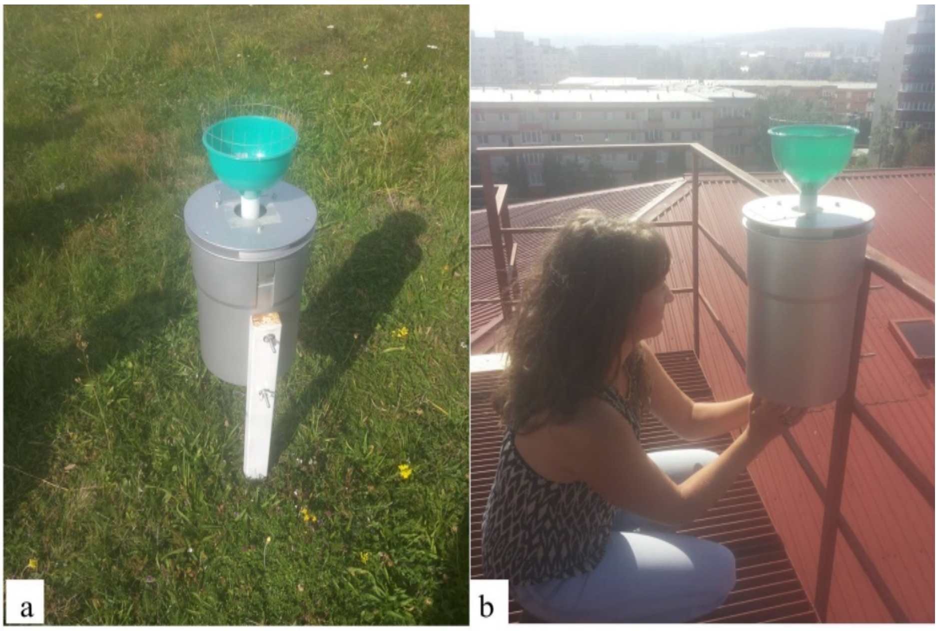

Figure 2. The rain samplers in operation: (a) Rosia Montana station, (b) Cluj-Napoca station

These two standard solutions are consistent with the international VSMOW standard. The analyser is set to perform 9 injections/sample or standard solution, each injection lasting for 15 minutes.

The first 5 injections are performed for washing the previous sample, and only the last 4 readings for oxygen and the last 3 readings for hydrogen are taken into account. The latest readings are averaged, and this will be the value attributed to the sample. The precision of the $\delta^{18}O$ and δ^2H measurements is ±0.12‰ and ±0.8‰, respectively.

4. RESULTS

$\delta^{18}O$ and δ^2H variations in precipitation

The isotopic ratios (δ^2H and $\delta^{18}O$) of precipitation measured in the two chosen locations were used for the construction of the local meteoric water lines (CJ LMWL and RM LMWL). The rain and snow samples were collected on a monthly basis from August 2014 to August 2015. Table 1 and 2 show characteristics for both stations: stable isotopes (oxygen-18 and deuterium) ratios, standard deviation, deuterium excess, air temperature, and rainfall amount.

The two regression lines for the isotopic ratios in rainwater were plotted. The δD of the precipitation in Rosia Montana ranged between –115.58‰ (February 2015) and –25.88‰ (August 2015), with an average of –65.00‰ ± 0.09‰. The $\delta^{18}O$ ranged between –15.17‰ (December 2014) and –3.78‰ (August 2015), with an average of −9.74‰ ± 0.01‰. Based on our data, the equation of the LMWL in Rosia Montana area is:

$$\delta D = 7.87^{18}O + 11.72‰ \quad (4)$$

The δD of precipitation at Cluj-Napoca station ranged between –107.49‰ (December 2014) and –23.79‰ (July 2015), with an average of –63.33‰ ± 0.09‰. The $\delta^{18}O$ ranged between –14.89‰ (December 2014), and –4.27‰ (July 2015), with an average of –9.29‰ ± 0.03‰. The equation of the LMWL in Cluj-Napoca area is:

$$\delta D = 8.03^{18}O + 11.29‰ \quad (5)$$

The position of the local meteoric water lines fall close to the GMWL defined by Craig (1961), both above it (Fig. 3).

The CJ LMWL is located much closer to the global line. The intercept values of the two lines drawn for Rosia Montana and Cluj-Napoca stations are 11.72, and 11.29 respectively, which are slightly higher than that of the GMWL. The correlation between oxygen and deuterium is 0.995 in the case of Rosia Montana, and 0.997 in the case of Cluj-Napoca station. The isotopic ratios show a large variation between the cold season and the warm one, due to the range of temperatures. The most negative values in the chart represent the values recorded in the cold season, both for RM and CJ. As it was expected the minimum value in the whole study was recorded in Rosia Montana, a mountain region with low temperature and important amount of snow. Some of the Local Meteoric Water Lines created in Romania (Costinel et al., 2009; Ionete et al., 2015), and also some Local Meteoric Water Lines from Europe (Vystavna et al., 2016; Holko et al., 2012; Giustini et al., 2016) are plotted in figure 4.

5. DISCUSSION

The slope and intercept of any Local Meteoric Water Line can be significantly different from the Global Meteoric Water Line. Distinct deviations of the two established LMWL from the GMWL are due to several factors that will be discussed below. Compared with other local meteoric water lines from Europe, RM and CJ are plotted almost in the same range with lines from Slovakia and Ukraine. The lines from Italy may show a different position and slope, due to the proximity of the Mediterranean basin.

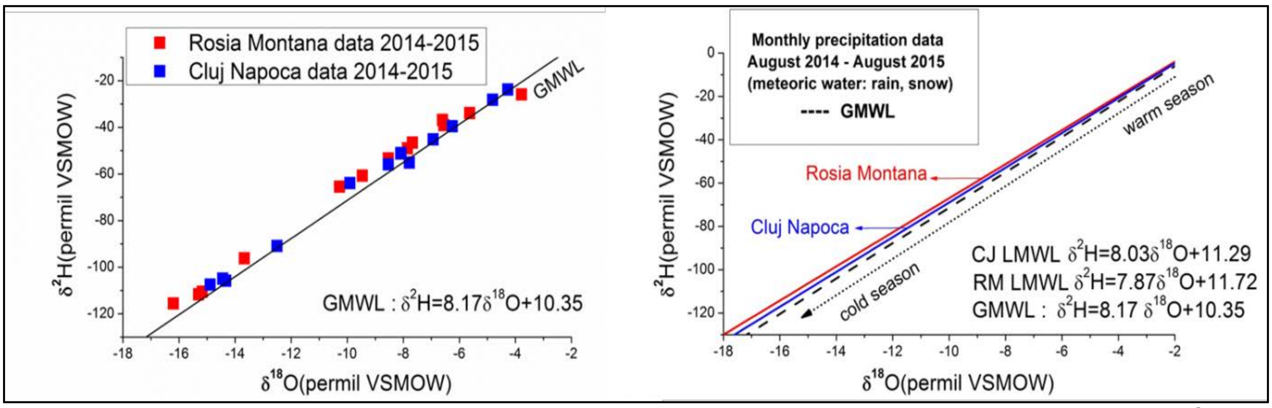

Figure 3. The data used to set up the two Local Meteoric Water Lines (LMWL) (left), and the relation between δ^2H and $\delta^{18}O$ of precipitation for both stations against the global meteoric water line (GMWL) (right).

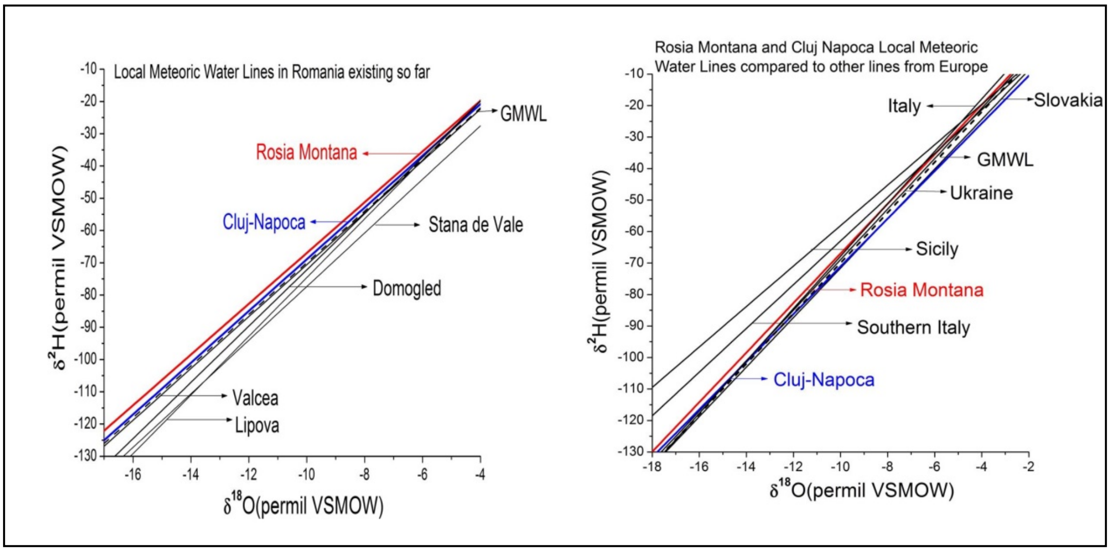

Figure 4. The RM LMWL and CJ LMWL position compared to other localities from Romania (left), and from Europe (right) (after Costinel et al., 2009; Ionete et al., 2015; Vystavna et al., 2016; Holko et al., 2012; Giustini et al., 2016).

The equation for the Mediterranean Meteoric Water Line established by Gat & Carmi (1970), shows an intercept quite higher than the rest (MMWL $\delta D = 8\delta^{18}O + 22$). The range of the intercept of all local meteoric water lines drawn so far for different regions in Romania is between –3.1‰ (Oradea-Satu Mare region) calculated by Țenu et al. (1981) and +16.41‰ (Lipova) obtained by Ionete et al. (2015). The differences between local meteoric water lines are induced by climate variations, seasonality, and geographic position. These differences are reflected in the deviations of the slope, and the d-excess value (Michener & Lajtha, 2007).

5.1. Altitude effect

As a general trend, precipitation becomes more depleted in heavy isotopes of oxygen and hydrogen, with the increase of elevation. This is obvious on the windward side of the mountains, with typical gradients between –0.15‰ and –0.5‰ per 100 m for ^{18}O, and –1.5‰ and –4‰ per 100 m for D (Clark & Fritz, 1997). This trend is less visible on the leeward side of the mountains, or in the case of snow. The average of the isotopic composition for RM station is –9.74‰ for ^{18}O, and –65.00‰ for D. In the case of CJ station, the average values are –9.29‰ and –63.33‰ for ^{18}O and D, respectively. Taking into account the elevation difference of 836 m between the two stations, the gradient is –0.05‰ for ^{18}O, and –0.20‰ for D. These values are lower than the typical gradients, reflecting the inner position of RM station by respect to the mountain range. Therefore, RM sampling site does not suffer the direct influence of the westward winds, which are dominant in the region.

5.2. Seasonal variation

The δ^2H and $\delta^{18}O$ follow very similar trends for the two stations (Fig. 5), with some minor differences. In case of RM station, the isotopic compositions are more depleted compared to CJ station, with two exceptions in May and August 2015, with slightly higher values compared to CJ station. The contrast between the depleted values during the cold season (months 5 – 8; December – March, isotope values from snow or mixing snow and rain), and the enriched isotopic composition during the warm season, is clearly shown by the graph. There is a relatively large gap between the isotopic compositions recorded during the cold and warm season. The amplitude of the variation is larger during the warm season, presumably due to the variation of temperature and evaporation.

5.3. Pattern of deuterium excess

Deuterium excess (d-excess) has been defined as the intercept of the Global Meteoric Water Line when the slope is 8 (Dansgaard, 1964):

$$d - excess = \delta D - 8 \cdot \delta^{18}O \ (‰) \quad (6)$$

This parameter offers information on the physical conditions in the vapour source area, moisture recycling, and may contribute to the calibration of the general atmospheric circulation models (Merlivat & Jouzel, 1979; Froehlich et al., 2002).
In our case, the d-excess of precipitation is 11.72 for RM station, and 11.28 for CJ station. The monthly values for d-excess varied between 4.36‰ in August 2015 and 16.69‰ in November 2014 for RM station, and in the range 7.09‰ in May 2015 to 15.26‰ in November 2014 for CJ station (Table 1).

Table 1. Rosia Montana station - isotopic data for the period August 2014- August 2015

MONTH	$\delta^{18}O$	SD	$\delta^{2}H$	SD	d-excess (‰)	T°C	Rainfall (mm)
Aug-14	-6.6	0.03	-36.86	0.04	15.94	15.6	121
Sep-14	-9.46	0.02	-60.73	0.1	14.95	12.2	37.6
Oct-14	-7.67	0	-46.61	0.03	14.75	12.5	41.6
Nov-14	-10.27	0.01	-65.47	0.06	16.69	4.7	39.5
Dec-14	-15.17	0.03	-110.57	0.07	10.79	-1.2	66.2
Jan-15	-13.67	0.02	-96.22	0.05	13.14	-2.4	61.6
Feb-15	-16.2	0.01	-115.58	0.09	14.02	-2.1	29.3
Mar-15	-15.3	0.05	-111.72	0.3	10.68	1	24.5
Apr-15	-8.53	0	-53.34	0.15	14.90	4.4	45.2
May-15	-6.55	0.02	-39.07	0.1	13.33	11.3	58.8
Jun-15	-7.85	0.02	-49.04	0.08	13.76	14.7	80.8
Jul-15	-5.63	0.02	-33.93	0.08	11.11	18.3	21.2
Aug-15	-3.78	0.01	-25.88	0.06	4.36	18.6	67
Average	-9.74		-65.00		12.96	Total	694.3

Table 2. Cluj-Napoca station - isotopic data for the period August 2014- August 2015

MONTH	$\delta^{18}O$	SD	$\delta^{2}H$	SD	d-excess (‰)	T°C	Rainfall (mm)
Aug-14	-4.81	0.02	-28.19	0.08	10.29	20.4	45.4
Sep-14	-6.25	0.02	-39.59	0.07	10.41	16.9	26.2
Oct-14	-8.09	0.03	-51.26	0.13	13.46	11.5	72.0
Nov-14	-9.90	0.01	-63.94	0.05	15.26	5.8	48.5
Dec-14	-14.89	0.03	-107.49	0.08	11.63	1.5	77.3
Jan-15	-12.5	0.03	-90.94	0.10	9.06	-0.3	28.6
Feb-15	-14.44	0.09	-104.94	0.15	10.58	1	24.4
Mar-15	-14.33	0.01	-105.86	0.13	8.78	5.8	36.8
Apr-15	-8.08	0.03	-51.03	0.08	13.61	9.6	35.6
May-15	-7.78	0.01	-55.15	0.04	7.09	15.7	77.4
Jun-15	-6.93	0.05	-45.18	0.19	10.26	19.1	119.2
Jul-15	-4.27	0.02	-23.79	0.09	10.37	21.8	32.4
Aug-15	-8.53	0.04	-55.92	0.07	12.32	22.1	59.0
Average	-9.29		-63.33		11.01	Total	682.8

The d-excess average for CJ station (11.01‰) is lower than that of RM station (12.96‰). Both values are higher than +10‰, thus suggesting the recycling of continental vapour (Dansgaard, 1964). Figure 6 does not show a clear correlation between the monthly amount of precipitation and the calculated d-excess.

5.4 Relation between δ^{18}O and temperature

The isotopic ratios in precipitation are dependent on the air temperature, the concentration of oxygen-18 increasing with the temperature. Based on data collected from North Atlantic coast stations and Greenland ice cap stations, Dansgaard (1964) has established a relation between annual averages of δ^{18}O in precipitation and temperature:

$$\delta^{18}O = 0.69t_a - 13.6‰ \quad (7)$$

where t_a is the air temperature. Based on a complete set of data collected by the IAEA/WMO network, Rozanski et al. (1993) have calculated a general slope of the best fit line of 0.58‰/1°C. This empirical equation could get changes due to local temperature and other factors. Figure 7 displays the correlation between oxygen-18 in precipitation and temperature for the two stations. The equations describing the relationship between δ^{18}O and temperature for the two stations were deducted by the least-squares fit method, as follows:

For RM station:

$$\delta^{18}O = 0.48t_a - 13.7‰ \text{ (n = 13; r = 0.86) (8)}$$

For CJ station:

$$\delta^{18}O = 0.39t_a - 13.8‰ \text{ (n = 13; r = 0.77) (9)}$$

As shown by Rozanski et al. (1993), the slope is higher in cold climate (0.67 to 0.90‰/1°C in Arctic and polar regions), and lower (generally below 0.5‰/1°C) in temperate and warmer regions.

The slopes calculated for RM and CJ stations are moderate by respect to the values recorded at a worldwide scale, and also reflect the climate difference between the two locations.

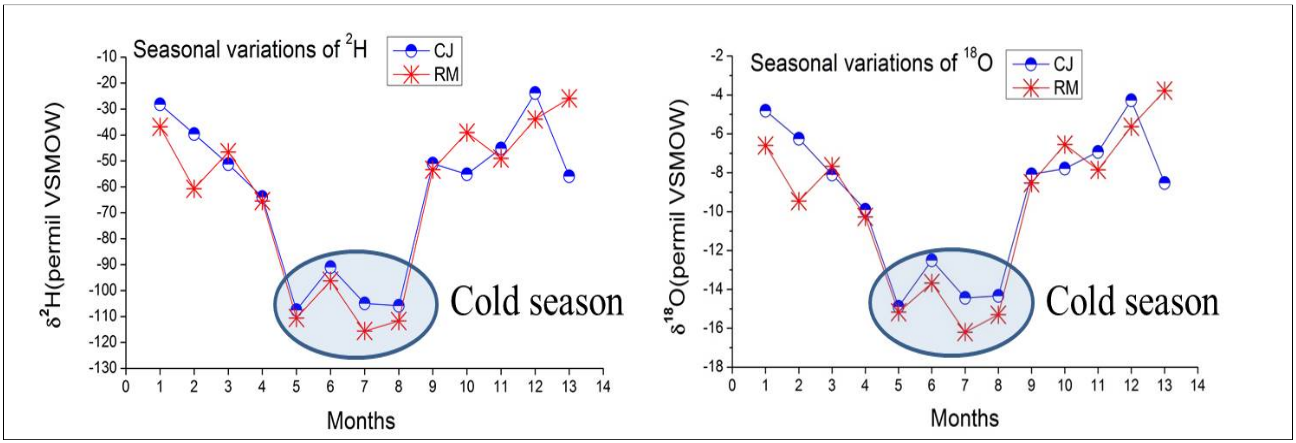

Figure 5. Seasonal variation of δD and $\delta^{18}O$ for RM and CJ stations. Month 1 corresponds to August 2014, and month 13 to August 2015

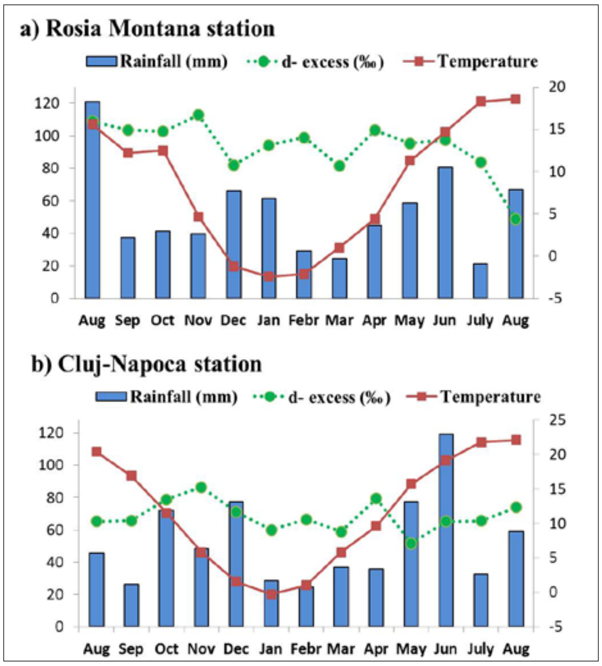

Figure 6. Monthly variation of precipitation amount, d-excess and temperature for RM and CJ station in the time period August 2014-August 2015.

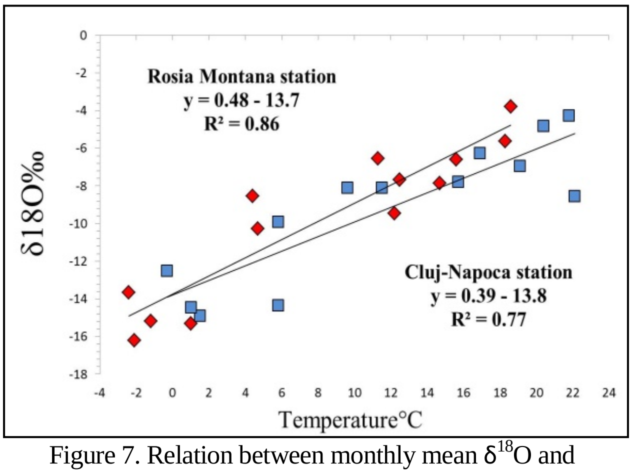

Figure 7. Relation between monthly mean $\delta^{18}O$ and temperature for RM station (diamonds) and CJ station (squares).

6. CONCLUSIONS

Two Local Meteoric Water Lines have been drawn for Rosia Montana and Cluj-Napoca stations in Western Romania. The isotopic data were collected monthly, over one year, by following a rigorous procedure. Both lines are located approximately parallel to the GMWL, and slightly above it. Their equations do not differ significantly among them, and compared to the GMWL. By comparison to other lines established for different locations in Romania, the new LMWLs are plotted in a higher position.

A first interpretation of the features of the two lines was performed, outlining the importance of different factors in the variation of the isotopic composition of precipitation in the study area. The isotopic ratios vary within a wide range, thus demonstrating the importance of the seasonality, with an obvious gap between the cold and warm seasons.

The altitude effect is not very sharp, due to the geographical position of the two stations, on the leeward side of the Apuseni Mts. (CJ station), and in a middle position by respect to the mountain range (RM station). The deuterium excess (11.28 for CJ, and 11.72 for RM station) suggest recycling of continental vapour. The correlation between $\delta^{18}O$ and temperature gives typical slopes for the temperate region.

The two new lines will represent a baseline for different types of studies (hydrological, ecological, atmospheric precipitation, climate, etc.), to be conducted in the region, that may include an isotopic approach.

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