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Isotopic variation of precipitation over eastern Mongolia and its implication for the atmospheric water cycle

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Received 29 August 2005; received in revised form 23 February 2006; accepted 24 July 2006

KEYWORDS

Atmospheric water cycle;
Stable isotope;
Rayleigh model;
Continental recycling;
Mongolia;
RAISE

Summary To investigate the atmospheric water cycle over eastern Mongolia and its surrounding regions, a monitoring network for stable hydrogen and oxygen isotopes in precipitation has been established. We obtained 66 monthly samples at six stations for a year from October 2002 to September 2003 and 204 daily samples at seven stations during the warm period from April to September of 2003. The observed isotopic composition showed considerable coherence in space and was characterized by a strong correlation with air temperatures (and a weak correlation with precipitation). A Rayleigh-type model involving the temperature and amount of precipitation successfully reproduced day-to-day variations in the observations with root mean square errors of 30‰ for δD and 2.7‰ for $\delta^{18}O$. This good reproducibility demonstrates that isotopic variation of precipitation can be explained by rainout history during transportation from a single vapor-source reservoir. The conditions of the reservoirs generally corresponded with those for a subtropical marine atmosphere, indicating that continental recycling generally has a minor effect at least on the temporal variability of isotopes in the precipitation. In July, however, observed δ values were very much lower than predicted. Considering the atmospheric circulation pattern and its relationship to isotopic data, it is inferred that the additive water vapor with low δ might be supplied from regions in the south (e.g., rice paddy fields in southeast China). Transient eddies can play an important role in transporting such water vapor to eastern Mongolia.

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Introduction

Objectives

The Mongolian landscape changes from desert in the south, through steppe in the middle, to forest in the north. Such an ecological transition zone (i.e., ecotone) is considered to be highly vulnerable to future climatic change including global warming (Roberts, 1994), and northeast Asia has indeed experienced one of the strongest warming signals on earth (Hansen et al., 1999; Chase et al., 2000). Global warming as an external force could potentially induce further changes in the local climate, hydrological cycle and ecosystem in Mongolia through interactions among them. According to numerical experiments, replacement of Mongolian grasslands with desert could reduce rainfall over and around the region through modification of water and energy exchanges between the land surfaces and the atmosphere (Xue, 1996). In fact, the summertime precipitation in Mongolia has shown a significant decreasing trend since the mid-1950s (Yatagai and Yasunari, 1995), although there are still uncertainties as to the mechanisms causing precipitation variability.

In order to understand precipitation variability and its relationship to land surface changes, it is essential to know the sources and transportation patterns of atmospheric moisture (e.g., Koster et al., 1986; Trenberth, 1999; Simmonds et al., 1999). Stable hydrogen and oxygen isotopes that constitute precipitating water molecules can be powerful tools in assessing these subjects (e.g., Yamanaka et al., 2002; Yoshimura et al., 2003). In the present paper, we address the atmospheric section of the hydrological cycle using isotopic tracers focusing on the Kherlen River basin, which occupies approximately one third of eastern Mongolia, under the framework of the The rangelands atmosphere–hydrosphere–biosphere interaction study experiment in northeastern Asia (RAISE) project (Sugita et al., this volume). Concrete objectives were twofold: (1) to clarify the characteristics of isotopic variation in precipitation over eastern Mongolia, and (2) to understand the mechanisms of the variation in terms of the continental-scale atmospheric water cycle. The following subsections summarize previous studies on the atmospheric water cycle and isotopes in precipitation over eastern Eurasia, and “Materials and methods” describes the methodology of the isotopic monitoring and meteorological data set utilized for the analysis. “Fundamental characteristics of isotopes in precipitation over eastern Mongolia” presents the fundamental features of the observed results, and in “Application of a Rayleigh-type model involving temperature and amount effects” a model to investigate isotopic variability and atmospheric hydrology in detail is proposed and tested. Finally, a general discussion and conclusions are given in “Summary and conclusions”.

Previous studies on atmospheric moisture transport and recycling in northeastern Eurasia

Trenberth (1999) has presented seasonal mean fields of vertically integrated water vapor flux over the globe. According to these, zonal moisture transport is dominant throughout

the year with strong westward components in the tropics and eastward components in the mid latitudes, whereas a southwesterly flux over the northern Indian Ocean and a northward flux over eastern China are strengthened in June, July and August (JJA) by the Asian summer monsoon. Simmonds et al. (1999) has decomposed the total summer moisture flux into mean and transient (less than a seasonal time scale) eddy components, and it was found that northward transport due to transient eddies is remarkable over northeastern Eurasia. In other words, atmospheric moisture transported by the mean flow from the northern Indian Ocean and western Pacific is further transported to higher latitudes by transient eddies. Eastern Mongolia (in contrast to western Mongolia) is clearly in the range of the transient moisture flux from the south, although the magnitude of the flux is less than that of the mean westerly flux across the Eurasian Continent.

Numaguti (1999) has assessed the origin of precipitating water over the Eurasian Continent using an atmospheric general circulation model (AGCM) with tagged waters. He showed that most waters precipitating over northeastern Eurasia ultimately originate from the northwestern Pacific, the northern Atlantic (both including the subtropics) and the northern Indian Ocean, while they experience a recycling process (i.e., having fallen onto the ground as precipitation they are then re-evaporated into the atmosphere) two or more times on average during JJA. It is noteworthy that the annual (JJA) mean ratio of precipitating water supplied most recently from the continent via evapotranspiration is calculated to exceed 0.5 (0.8) over northeastern Eurasia. Similar results for the continental recycling ratio have been obtained from another AGCM (Koster et al., 1986) and a simpler tagged-water transport model (Yoshimura et al., 2004b) driven by external meteorological forcing assuming a vertically well-mixed atmosphere. However, Bosilovich (2002) has shown that the vertical distributions of local (e.g., continental) and remote (e.g., oceanic) sources of water vapor can be different and that their relative contributions to precipitation and total precipitable water can also be different. These findings indicate not only the limitation of the “well-mixed” assumption but also the strong dependence of estimates against parameterization schemes in GCMs, such as cumulus convection.

Consequently, atmospheric moisture transport over northeastern Eurasia seems to be very complicated because the region is far from any ocean as the ultimate origin of the atmospheric moisture and has many potential sources for precipitation other than the ocean such as the semi-arid grasslands of central Asia, the boreal forest of Siberia, and the rice paddy fields of southeast China.

Previous studies on isotopes in precipitation over northeastern Eurasia

Stable hydrogen and oxygen isotopes in precipitation have been used for tracing the atmospheric water cycle. The major advantage of this approach is the capability of bringing out information about the real world, not a virtual world as in GCMs, even though there remain some limitations and uncertainties. To characterize the isotopic variability in precipitation, global network for isotopes in precipitation

(GNIP) has been established by the International Atomic Energy Agency (IAEA) in corporation with the World Meteorological Organization (WMO). The GNIP database (URL: <http://www.isohis.iaea.org>) comprises monthly data from more than 500 stations world-wide, clearly displaying the geographical distribution on a global scale and well-known isotopic effects (e.g., Clark and Fritz, 1997). In Mongolia, however, there is no station having at least one full-year record. As a basis for the present study, we will summarize several isotopic studies in adjacent regions.

Yamanaka et al. (2004) have classified the annual pattern variations in the isotopic composition of precipitation at GNIP stations in China into three types: summer depression, winter depression, and intermediate types. The summer depression type dominated by the amount effect is found in the southern parts, indicating strong influences due to the Asian summer monsoons. In contrast, the winter depression type, which occurs in the northern part, is dominated by the temperature effect, suggesting a weak contribution of monsoonal rains. Kurita et al. (2004) have investigated the spatial and temporal variation in isotopic composition of precipitation based on a monthly data set collected at 13 original stations across Russia, and showed the significance of the westerly vapor transport from the Atlantic Ocean in winter and the recycled water from the continent in summer.

Daily (or event-based) isotope data are linked more directly with atmospheric moisture dynamics than the monthly data set. Kurita et al. (2003) have clarified that in eastern Siberia short-term isotopic variability can be explained by the Rayleigh distillation process (Dansgaard, 1964) for spring precipitation but not for summer precipitation. From a strong correlation between isotopic composition and the westerly component of the atmospheric moisture flux, they drew an inference that the summer precipitation is a composite of isotopically heavier waters from the west and lighter waters from other sources, for instance, evapotranspiration flux from thaw water. Yoshimura et al. (2003) have developed a global one-layer (i.e., with a “well-mixed” assumption) isotope circulation model including the Rayleigh process and succeeded in a more quantitative explanation of the short-term isotopic variability in precipitation in Southeast Asia, demonstrating that the isotopic compositions of precipitation reflect the transport and mixing processes of air masses. On the other hand, using deuterium excess, which is a parameter deduced from both hydrogen and oxygen isotopic compositions (see the next section for a definition), Yamanaka et al. (2002, 2004) have assessed source areas of precipitating water for each event over the Kanto Plain, Japan, and the northern North China Plain, suggesting that water vapors originating from their adjacent ocean areas are the main ingredients of precipitation.

Stable hydrogen and oxygen isotopes are not only promising tracers for investigating atmospheric hydrology but also good proxies for reconstructing climatic change. For example, Schotterer et al. (1997) have attempted to reconstruct past hydrometeorological conditions over several decades, based on the isotope record from the Tsast Ula ice cap in western Mongolia. Morley et al. (2005) investigated late glacial and Holocene environmental change using oxygen isotopes of biogenic diatom silica, which are sensitive to isotopic input (i.e., precipitation over the lake and its catchment area), in sediment cores from Lake Baikal in the north of Mongolia. Thus, an understanding of the factors determining the isotopic composition of precipitation over Mongolia is expected to contribute to an improvement in the reconstruction of climatic change in northeastern Eurasia, of which strong a continental setting is very rare in the world.

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Materials and methods

Isotopic monitoring

Precipitation samples were collected at seven sites covering the Kherlen River basin (Fig. 1). Monthly samples were collected throughout the year from October 2002 to September 2003, and daily samples were collected for the warm period from April to September 2003. The rain collector utilized in the present study was composed of a polyethylene tank and a funnel with a ping-pong ball to prevent evaporation. Snow samples were collected using a vat installed on the ground and then melted in an airtight container at room temperature. Rain and melted snow samples were transferred into 100 ml polyethylene bottles for monthly samples and 13 ml glass bottles for daily samples.

All the samples were analyzed for δD and $\delta^{18}O$ (δ (‰) = $(R_{\text{sample}}/R_{\text{standard}} - 1) \times 10^3$; R is D/H or $^{16}O/^{18}O$ ratio

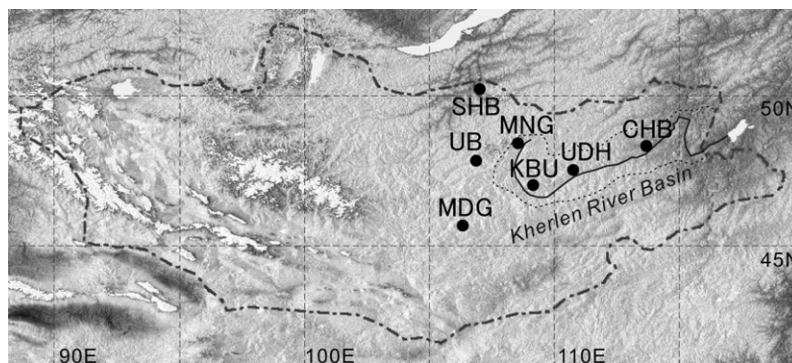


Figure 1 Location of isotopic monitoring stations (MNG, Mongonmorit; KBU, Kherlen-Bayan Ulaan; UDH, Underhaan; CHB, Choibalsan; UB, Ulaanbaatar; MDG, Mandalgobi; SHB, Sukhbaatar).

where the standard was the Vienna Standard Mean Ocean Water, V-SMOW) using a mass spectrometer (MAT252, Thermo Finnigan) at the University of Tsukuba. Hydrogen gas equilibration using a platinum catalyst for six hours for hydrogen and carbon dioxide gas equilibration for nine hours for oxygen were adopted for sample preparation. The measurement accuracy was consistently better than 1‰ for δD and 0.1‰ for $\delta^{18}O$. From the measured results, the deuterium excess d ($=\delta D - 8 \times \delta^{18}O$) was calculated for each sample. Since we lack many monthly samples at Ulaanbaatar, all the monthly data at the station were excluded from the following analysis.

Meteorological data set

At the seven monitoring stations, general meteorological components are routinely observed and compiled by the Mongolia Government via the Institute of Meteorology and Hydrology (IMH). We used surface air temperature and precipitation data for each station. Besides this data set, the National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis data set (Kalnay et al., 1996) was utilized to obtain the meteorological conditions in the upper air and wider region up to continental scale. The data are produced with a four-dimensional data assimilation technique and provided on a $2.5^\circ \times 2.5^\circ$ grid. In the present study, values at each grid point ($47.5^\circ N$, $110^\circ E$) were used as representative of the area covered by the isotopic monitoring network.

It has been reported that atmospheric moisture fields from NCEP/NCAR data contain some biases, especially in precipitation (e.g., Trenberth, 1999). The NCEP/NCAR daily precipitation was certainly underestimated as compared to the regional average of IMH observations at the seven sites. In contrast, the NCEP/NCAR daily surface air temperatures showed a high correlation with the IMH regional average (correlation coefficient $r = 0.993$, $p < 0.001$; data from April to September, 2003). Although the errors in specific humidity and wind fields at upper levels remain unclear, several studies have reported considerable validity of the data by comparing other data sets (e.g., Yoshimura et al., 2004a; Kurita et al., 2004).

Fundamental characteristics of isotopes in precipitation over eastern Mongolia

Temporal variation

Fig. 2 shows a plot of all observed data on the δD – $\delta^{18}O$ diagram. The δD ($\delta^{18}O$) ranges from -50‰ (-7‰) to -250‰ (-33‰) for monthly samples and from $+15\text{‰}$ ($+3\text{‰}$) to -230‰ (-30‰) for daily samples from April to September. For both monthly and daily data, the linear relationship between δD and $\delta^{18}O$ can be clearly identified. The regression coefficients (i.e., slope and intercept) for each site are not significantly different from those for the whole data set (Table 1). The variations of monthly δD and $\delta^{18}O$ clearly show the annual cycle with minor deviations among sites, though the deviation in deuterium excess seems to be relatively large (Fig. 3). The similarities in the δD – $\delta^{18}O$ relationship and the seasonal variation pattern among locations suggest

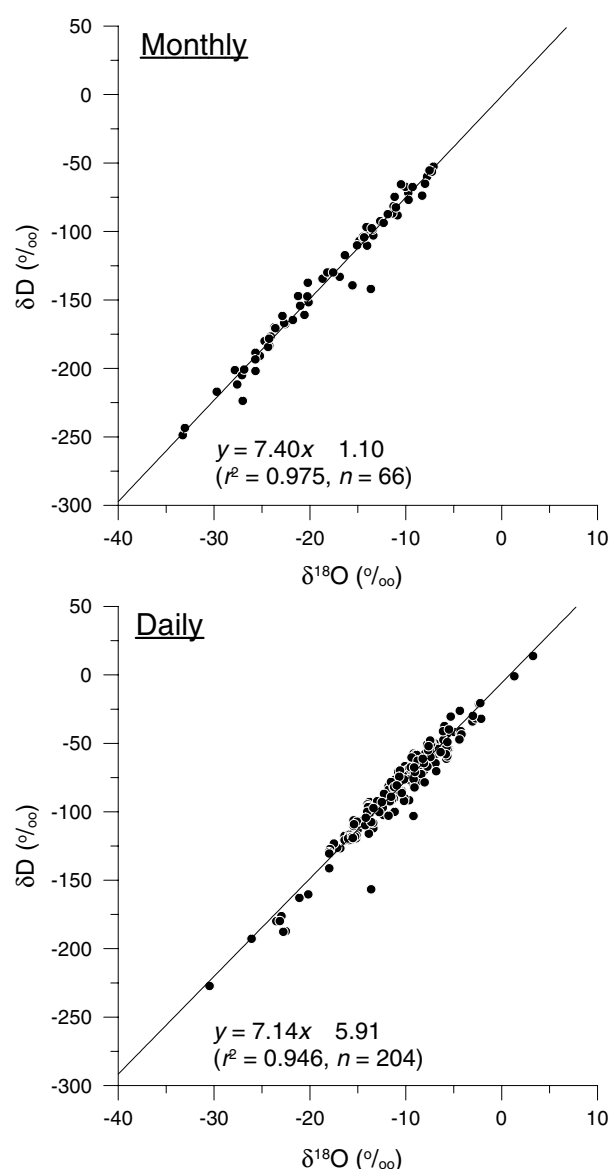


Figure 2 Relationship between δD and $\delta^{18}O$ of precipitation at all stations for (top) monthly samples and (bottom) daily samples.

that isotopic compositions of precipitation are primarily determined by large-scale (at least far greater than the eastern part of Mongolia) atmospheric moisture transport. Daily data are plotted generally along the seasonal variation curve but with slightly higher δ and lower d (Fig. 4). This indicates that some daily samples were isotopically altered by evaporation from falling raindrops under the cloud base, since light rain occurs more frequently than heavy rain and samples for light rain tend to be affected more strongly by evaporation.

Correlation with meteorological factors

Table 2 summarizes correlation coefficients between isotopic compositions and some selected meteorological factors based on monthly data. We find a strong temperature-effect

Table 1 Liner regression analysis statistics for local meteoric water lines

Site	Monthly data				Daily data			
	<i>s</i>	<i>i</i>	r^2	<i>n</i>	<i>s</i>	<i>i</i>	r^2	<i>n</i>
All sites	7.41 (± 0.15)	-1.10 (± 2.82)	0.975	66	7.14 (± 0.12)	-5.91 (± 1.43)	0.946	204
MNG	7.21 (± 0.11)	-1.72 (± 2.19)	0.997	12	7.04 (± 0.20)	-6.55 (± 2.44)	0.966	44
KBU	7.52 (± 0.19)	2.65 (± 3.75)	0.994	11	6.63 (± 0.26)	-8.92 (± 3.00)	0.952	36
UDH	7.71 (± 0.22)	3.59 (± 4.26)	0.992	12	6.90 (± 0.28)	-8.91 (± 2.93)	0.940	40
UB	N/A	N/A	N/A	N/A	6.59 (± 0.32)	-10.98 (± 3.92)	0.944	27
CHB	7.01 (± 0.49)	-9.68 (± 7.80)	0.953	12	7.46 (± 0.67)	-7.21 (± 6.45)	0.912	14
MDG	7.75 (± 0.77)	1.47 (± 14.19)	0.918	11	7.17 (± 0.27)	-3.25 (± 2.96)	0.965	28
SHB	7.46 (± 0.18)	3.19 (± 4.22)	0.996	8	7.50 (± 1.06)	-8.23 (± 18.74)	0.795	15

s, slope; *i*, intercept; r^2 , determination coefficient; *n*, number of samples.

The values in parentheses denote standard error of *s* or *i* estimates.

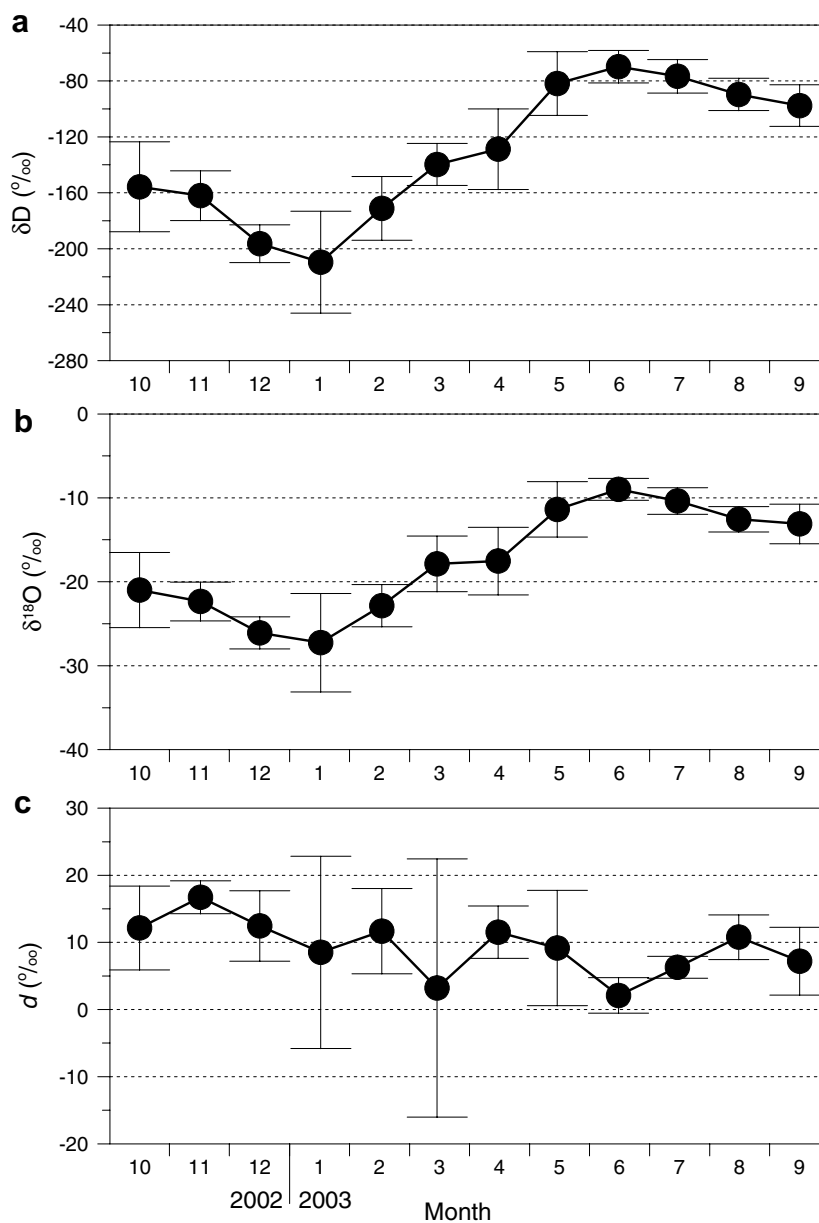


Figure 3 Annual variation of monthly (a) δD , (b) $\delta^{18}O$, and (c) deuterium excess d . Closed circles are spatial mean values, and vertical bars denote standard deviation among stations.

on δD and $\delta^{18}O$. The gradient of δ against surface air temperature is 2.9 ± 0.5 for δD and 0.37 ± 0.05 for $\delta^{18}O$ (mean $\pm$ SD for sites with $p < 0.01$, in $\text{‰}/^\circ\text{C}$). Although the other variables also show good correlations, most of these correlate with temperature and show slightly weaker correlations. On the other hand, the directions of atmospheric moisture transport (i.e., F_u/F and F_v/F ; F_u and F_v are the westerly and northerly components of the vertically integrated water vapor flux F , respectively), which are potentially linked to the air mass trajectory and/or vapor source, and show hardly any significant correlation. The features mentioned above can be also found for daily data (Table 3), while the correlation is generally worse for monthly

data, indicating that more complicated factors govern the day-to-day isotopic variability. The gradient of the δ -surface air temperature based on daily samples is 4.7 ± 0.9 for δD and 0.65 ± 0.07 for $\delta^{18}O$ (for sites with $p < 0.01$, in $\text{‰}/^\circ\text{C}$), being larger than that for monthly samples.

As compared to δ values, deuterium excess does not show clear correlations with specific factors for either monthly or daily data (Table 2 and 3). Monthly d at Mongonmorit (MNG) and Sukhbaatar (SHB), however, show relatively better correlations with temperature and other factors. This is due to simple seasonality in d at the two sites. In other words, d at the other sites includes a lot of noise that draws out its genuine variation. A cause of the

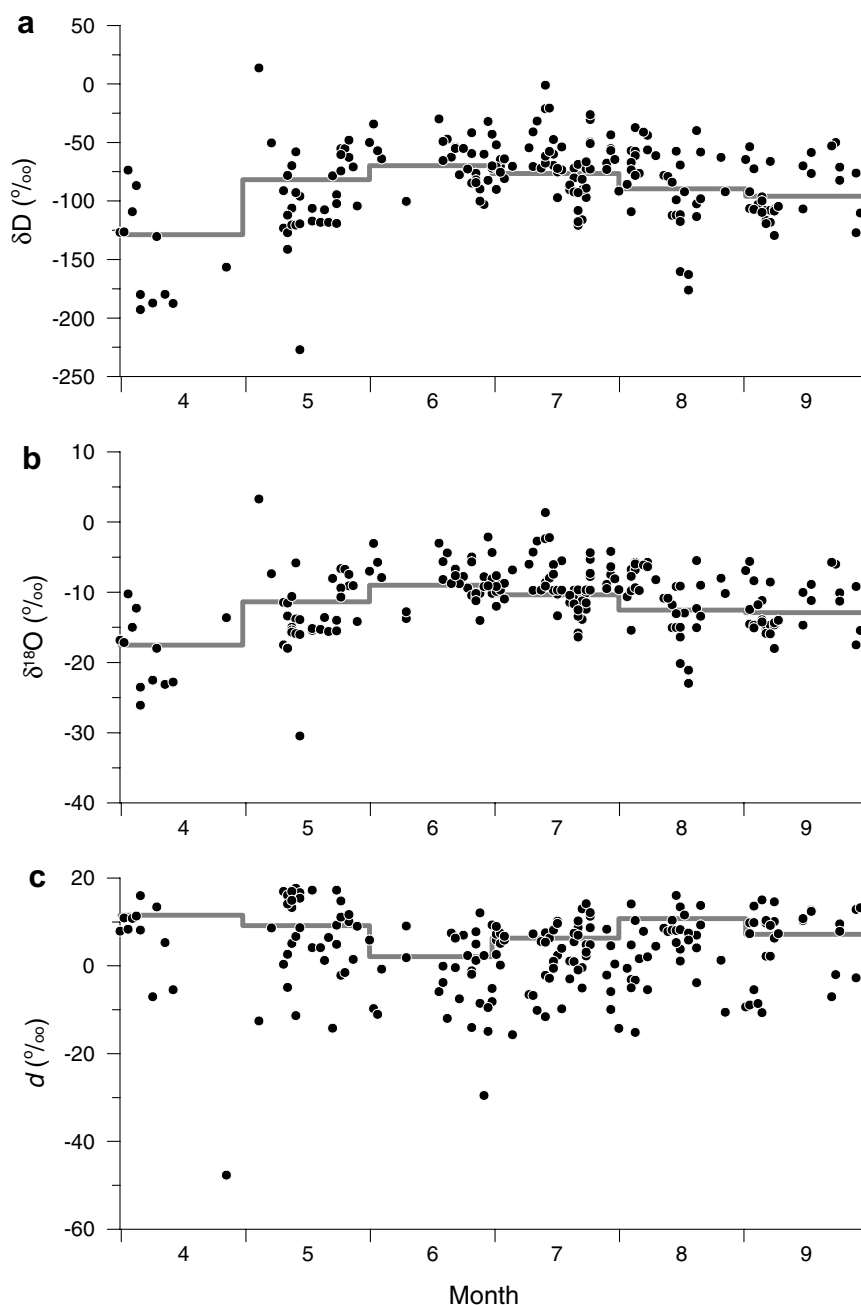


Figure 4 All daily data (closed circle) plotted with spatial mean monthly data (step-like line) for (a) δD , (b) $\delta^{18}O$, and (c) d .

noise is probably a postcondensation process, such as evaporation from falling raindrops. Fig. 5 clearly shows that negative d values, which are extraordinary and probably introduced by evaporation from raindrops, are generally

found in light rain (e.g., less than 5 or 10 mm/hr) though with some exceptions. On the other hand, even in light rain events δD and $\delta^{18}O$ show not only high values (i.e., may be affected by evaporative enrichment) but also extremely low

Table 2 Correlation coefficients between isotopic compositions observed at each site and meteorological variables based on monthly data

	MNG	KBU	UDH	CHB	MDG	SHB
<i>(a) Monthly δD</i>						
Pr ^a	0.55	N/A	0.69*	0.42	0.79**	0.85**
T _s ^a	0.94***	N/A	0.86***	0.89***	0.92***	0.89**
T ₇₀₀	0.94***	0.91***	0.86***	0.82***	0.88***	0.90**
T ₅₀₀	0.92***	0.90***	0.84***	0.83***	0.88***	0.91**
T ₃₀₀	0.92***	0.89***	0.85***	0.81**	0.86***	0.93***
q ₇₀₀	0.85***	0.80***	0.79**	0.70**	0.77**	0.84**
q ₅₀₀	0.86***	0.83**	0.79**	0.71**	0.78**	0.88**
q ₃₀₀	0.87***	0.86***	0.81**	0.73**	0.80**	0.91**
Pw	0.88***	0.83**	0.81**	0.74**	0.79**	0.89**
F	0.78**	0.68*	0.67*	0.71**	0.72*	0.82*
F _u /F ^b	0.05	0.06	0.23	0.14	0.11	0.66
F _v /F ^b	0.30	0.29	0.43	0.33	0.32	0.72*
<i>(b) Monthly $\delta^{18}O$</i>						
Pr ^a	0.53	N/A	0.65*	0.28	0.73**	0.85**
T _s ^a	0.93***	N/A	0.84***	0.83***	0.90***	0.89**
T ₇₀₀	0.93***	0.92***	0.85***	0.78**	0.85***	0.91**
T ₅₀₀	0.91***	0.90***	0.83***	0.78**	0.83**	0.92***
T ₃₀₀	0.91***	0.90***	0.84***	0.75**	0.83**	0.93***
q ₇₀₀	0.84***	0.80***	0.78**	0.64*	0.74**	0.85**
q ₅₀₀	0.85***	0.83**	0.78**	0.66*	0.76**	0.89**
q ₃₀₀	0.86***	0.87***	0.80**	0.70*	0.77**	0.91**
Pw	0.87***	0.84**	0.80**	0.69*	0.77**	0.89**
F ^b	0.77**	0.68*	0.65*	0.63*	0.68*	0.83**
F _u /F ^b	0.04	0.05	0.21	0.10	0.11	0.69
F _v /F ^b	0.29	0.29	0.42	0.29	0.29	0.73*
<i>(c) Monthly deuterium excess d</i>						
Pr ^a	-0.32	N/A	0.15	0.40	0.19	-0.61
T _s ^a	-0.73**	N/A	-0.10	-0.14	0.09	-0.74*
T ₇₀₀	-0.75**	-0.62*	-0.17	-0.17	0.16	-0.77*
T ₅₀₀	-0.72**	-0.61*	-0.15	-0.15	0.18	-0.77*
T ₃₀₀	-0.73**	-0.59	-0.20	-0.11	0.15	-0.75*
q ₇₀₀	-0.67*	-0.52	-0.15	-0.03	0.13	-0.74*
q ₅₀₀	-0.67*	-0.59	-0.20	-0.10	0.15	-0.76*
q ₃₀₀	-0.68*	-0.62*	-0.22	-0.16	0.15	-0.75*
Pw	-0.69*	-0.57	-0.19	-0.09	0.13	-0.76*
F ^b	-0.59*	-0.39	-0.01	0.04	0.18	-0.72*
F _u /F ^b	0.05	0.09	0.11	0.12	0.14	-0.77*
F _v /F ^b	-0.16	-0.13	-0.01	0.01	0.19	-0.76*

Pr, precipitation; T_s, surface air temperature; T and q, air temperature at three pressure levels (700, 500 and 300 hPa), q, specific humidity at three pressure levels (700, 500 and 300 hPa), Pw, precipitable water (i.e., total water vapor content in the atmospheric column); F, vertically integrated water vapor flux; F_u, westerly component of F; F_v, northerly component of F.

^a Precipitation and surface air temperature are observed values at each site, and the other meteorological variables are from NCEP/NCAR reanalysis.

^b Vertically integrated water vapor flux was computed by approximating the continuous integral with finite differences based on the trapezoidal rule and by weighting the lowest layer by surface air pressure, same as in Simmonds et al. (1999).

* $p < 0.05$.

** $p < 0.01$.

*** $p < 0.001$.

values (i.e., probably not affected), indicating that these are insensitive to the secondary effect due to evaporation.

Negative correlations between local δ values and local precipitation can be found in the daily data (Table 3). While the correlations are weak, the upper limit of δ for a given precipitation tends to decrease with increase in precipitation (as illustrated by the dashed line in Figs. 5a and b). It is also noteworthy that if we take regionally representative values of δ (the average among different sites where precipitation was sampled) and precipitation (the average over all sites) in July–August, the value of the correlation coefficient reaches -0.62 ($p < 0.01$) for δD and -0.64 ($p < 0.001$) for $\delta^{18}O$. These facts suggest that the amount effect cannot be negligible particularly in summer, in which heavy rain often occurs. Calculations based on regional values give

the δ -precipitation gradient of $-3.7\text{‰}/\text{mm}$ for δD and $-0.53\text{‰}/\text{mm}$ for $\delta^{18}O$.

Spatial variation

Although the temporal variation patterns are not different among locations as shown above, we assessed the spatial variation of the annual mean isotopic compositions weighted by monthly precipitation, which is important for studying surface and subsurface hydrological processes. Because we lack all monthly precipitation data at Kherlen-Bayan Ulaan (KBU) and monthly isotope data at Ulaanbaatar (UB), data for the other five stations were available for this assessment. In calculating the weighted mean δ , some missing data were linearly interpolated. The weighted mean δ

Table 3 Correlation coefficients between isotopic compositions observed at each site and meteorological variables based on daily data

	MNG	KBU	UDH	UB	CHB	MDG	SHB
<i>(a) Daily δD</i>							
Pr	-0.14	-0.33*	-0.17	-0.24	-0.14	-0.07	0.30
T_s	0.69***	N/A	0.65***	0.59**	0.36	0.20	0.79***
T_{700}	0.74***	0.39*	0.60***	0.62***	0.33	0.12	0.86***
T_{500}	0.64***	0.41*	0.59***	0.56**	0.24	0.22	0.91***
T_{300}	0.52***	0.31	0.63***	0.55**	0.26	0.35	0.77***
q_{700}	0.13	0.06	0.26	0.20	-0.21	-0.17	0.48
q_{500}	0.39	0.27	0.38*	0.58**	-0.09	-0.09	0.42
q_{300}	0.23	0.32	0.41**	0.52**	-0.25	0.19	0.38
Pw	0.24	0.09	0.34*	0.36	-0.19	-0.17	0.54*
F	0.30*	-0.11	0.38*	0.45*	0.31	0.06	0.26
F_u/F	0.38*	0.02	0.27	0.08	0.34	0.33	-0.18
F_v/F	-0.06	-0.14	0.32*	0.11	0.02	-0.13	0.07
<i>(b) Daily $\delta^{18}O$</i>							
Pr	-0.20	-0.32	-0.26	-0.27	-0.20	-0.06	0.18
T_s	0.67***	N/A	0.71***	0.66***	0.29	0.20	0.75**
T_{700}	0.72***	0.38*	0.64***	0.68***	0.27	0.13	0.82***
T_{500}	0.60***	0.43**	0.62***	0.58**	0.14	0.22	0.81***
T_{300}	0.47**	0.32	0.63***	0.57**	0.13	0.35	0.57*
q_{700}	0.10	0.05	0.24	0.23	-0.31	-0.20	0.41
q_{500}	0.36*	0.26	0.35*	0.60***	-0.13	-0.11	0.42
q_{300}	0.18	0.27	0.38*	0.51**	-0.29	0.19	0.32
Pw	0.20	0.08	0.32*	0.39*	-0.28	-0.19	0.47
F	0.25	-0.04	0.35*	0.50**	0.22	0.06	0.31
F_u/F	0.41**	0.05	0.26	0.13	0.36	0.30	-0.08
F_v/F	-0.10	-0.18	0.27	0.10	0.00	-0.14	-0.01
<i>(c) Daily deuterium excess d</i>							
Pr	0.37**	0.15	0.42**	0.27	0.24	-0.03	0.29
T_s	-0.27	N/A	-0.49**	-0.60***	0.18	-0.08	0.18
T_{700}	-0.29	-0.20	-0.40**	-0.58**	0.19	-0.08	0.18
T_{500}	-0.17	-0.31	-0.35*	-0.40*	0.31	-0.10	0.31
T_{300}	-0.02	-0.23	-0.29	-0.37	0.43	-0.20	0.49
q_{700}	0.08	0.00	-0.04	-0.22	0.35	0.19	0.20
q_{500}	-0.06	-0.09	-0.06	-0.38*	0.14	0.14	0.05
q_{300}	0.12	-0.02	-0.07	-0.26	0.16	-0.07	0.16
Pw	0.04	-0.03	-0.06	-0.32	0.32	0.16	0.20
F	0.05	-0.20	-0.02	-0.46*	0.29	-0.05	-0.08
F_u/F	-0.35*	-0.12	-0.08	-0.22	-0.08	-0.02	-0.23
F_v/F	0.27	0.27	0.04	-0.02	0.09	0.13	0.16

for each station ranged from -84‰ to -118‰ for δD and from -11.7‰ to -16.4‰ for $\delta^{18}\text{O}$ (Table 4). Multiple linear-regression models based on geographic parameters were obtained as follows:

$$\delta\text{D} = 0.286X - 8.11Y - 7.37 \times 10^{-4}Z + 260$$

$$(r^2 = 0.952, n = 5) \quad (1)$$

$$\delta^{18}\text{O} = 0.0720X - 1.11Y - 9.32 \times 10^{-5}Z + 31.8$$

$$(r^2 = 0.966, n = 5) \quad (2)$$

where X is the longitude ($^{\circ}\text{E}$), Y the latitude ($^{\circ}\text{N}$), and Z the altitude (m). These models represent quantitatively the latitude and altitude effects. Partial correlation coefficients

are 0.29 for X , -0.95 for Y and -0.06 for Z in Eq. (1), and 0.55 for X , -0.96 for Y and -0.06 for Z in Eq. (2), emphasizing that the latitude effect is the most important. The coefficients of the latitude effect in the present models have the same order of magnitude as values for other mid- to high-latitude continental stations (-0.6‰ to -2‰ per degree latitude for $\delta^{18}\text{O}$; e.g., Clark and Fritz, 1997), while those of the altitude effect are one or two orders of magnitude smaller than common values for other mountainous regions over the world (-1.5×10^{-3} to $-5 \times 10^{-3}\text{‰}$ per 1 m rise in altitude for $\delta^{18}\text{O}$; e.g., Clark and Fritz, 1997; Poage and Chamberlain, 2001). Because of the smaller significance of the partial regression coefficient for altitude ($p = 0.96$ for

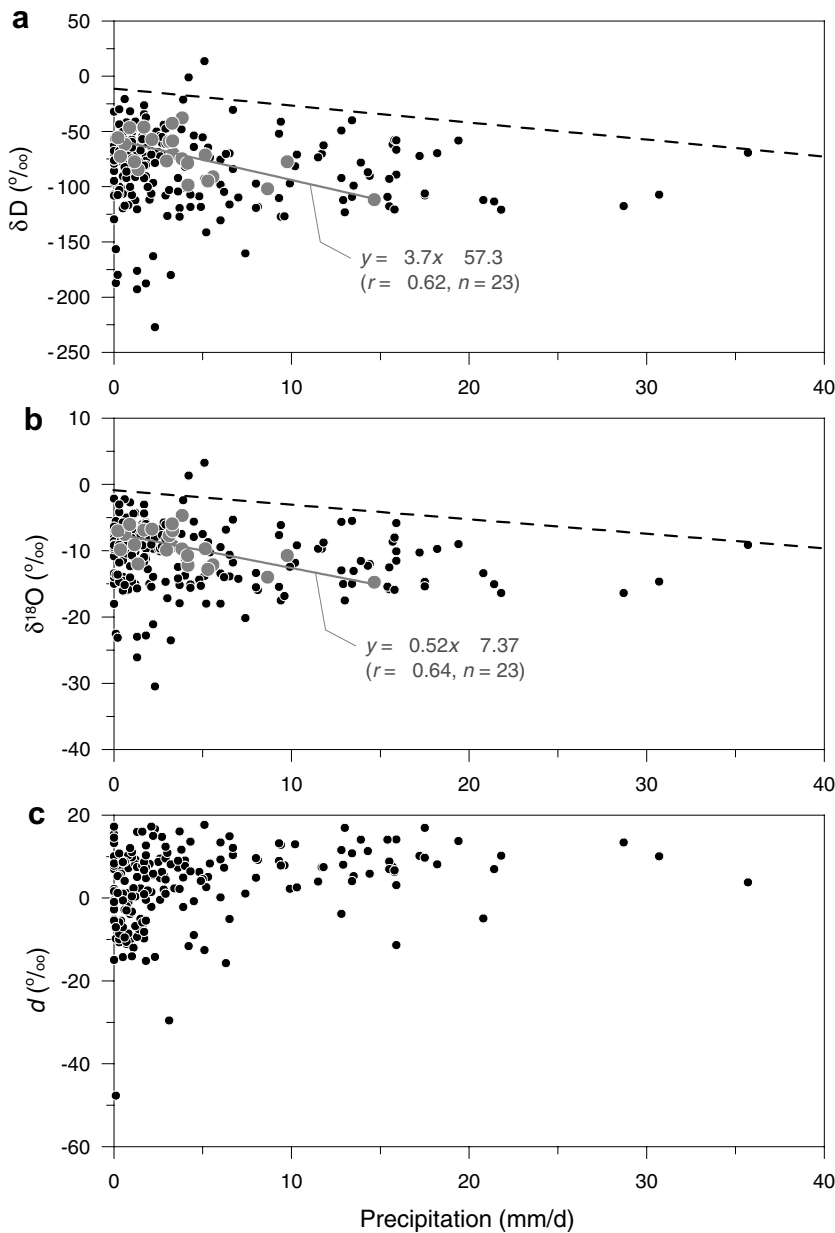


Figure 5 Relationships of precipitation to (a) δD , (b) $\delta^{18}\text{O}$, and (c) d , based on daily data for all stations. Gray plots and their regression lines represent the relationships between regional representatives (i.e., δ averaged over different stations where precipitation was sampled on same day and precipitation averaged over all sites for the corresponding day) in July–August.

Table 4 Geographic parameters and annual statistics for each sampling location

	MNG	KBU	UDH	UB	CHB	MDG	SHB
Latitude (°N)	48.20	47.28	47.32	47.92	48.07	45.75	50.25
Longitude (°E)	108.50	108.78	110.67	106.87	114.52	106.27	106.21
Altitude (m)	1439	1255	1033	1306	759	1393	626
Annual Pr (mm)	297	N/A	239	283	178	218	240
Annual mean T_s (°C)	−4.5	N/A	−1.6	−1.0	0.2	1.8	0.2
Weighted mean δD	−101.9	N/A	−88.5	N/A	−100.8	−84.1	−118.5
Weighted mean $\delta^{18}O$	−14.04	N/A	−12.26	N/A	−13.65	−11.65	−16.39

both equations), the altitude effect found in the present study may not be reliable.

Application of a Rayleigh-type model involving temperature and amount effects

Model description

As shown in the previous section, the temporal variation pattern of isotopes in precipitation over eastern Mongolia is highly uniform in space and is correlated primarily with temperature and partially with precipitation amount. In this section, we attempt to develop a simple model for investigating mechanisms that govern the observed isotopic variability.

The temperature effect is conventionally explained by the Rayleigh (distillation) process, which assumes that condensates are formed at isotopic equilibrium with the surrounding vapor in the cloud and are removed immediately as precipitation (e.g., Dansgaard, 1964; Jouzel and Merlivat, 1984). Under this process, the isotopic composition of the remaining water vapor, δ_v , can be predicted by the following Rayleigh equation:

$$\delta_v/1000 = (1 + \delta_{v0}/1000)f^{\alpha-1} - 1 \quad (3)$$

where α is the equilibrium fractionation factor, δ_{v0} is the initial value of δ_v , and f is the fraction of the remaining water vapor, defined by m_v/m_{v0} (m_v is the water vapor content and subscript “0” denotes the initial value). Assuming the condensation process to be caused by isobaric cooling without supersaturation, the m_v is in proportion to the saturation vapor pressure, e_{sat} , and the remaining vapor fraction is represented as follows:

$$f = e_{sat}(T_c)/e_{sat}(T_{c0}) \quad (4)$$

where T_c is the prevailing ‘in cloud’ temperature. Here, we assume that an air mass loses water vapor through rainout along a trajectory from its vapor source area. If the T_{c0} at the source area is nearly constant, T_c at a site where precipitation occurs is considered to be a proxy of f , in other words, of rainout history from the source area to the site.

While precipitation at a site is also accompanied by depression in T_c , it is difficult to evaluate the depression between the pre-event and the post-event using reanalysis data because of their coarse resolution in time (i.e., four times daily). Thus, in the present study, we predicted isotopic change due to on-site precipitation by another method. The instantaneous isotopic composition of precipitating water, δ_p , is equal to that of the condensate in isotopic

equilibrium with the surrounding vapor at that time and is related to δ_v by the following.

$$\delta_p/1000 = \alpha(1 + \delta_v/1000) - 1 \quad (5)$$

The isotopic composition of the total precipitation accumulated for an event, δ_p , is given by substituting Eq. (3) into Eq. (5) and then dividing its integral for f by the fraction of the removed water vapor (i.e., $1 - f$), as

$$\delta_p/1000 = (1 + \delta'_{v0}/1000)(1 - f')/(1 - f) - 1 \quad (6)$$

where f' is the remaining water vapor fraction for the event (not for the previous rainout history as defined in Eq. (1)), being expressed as

$$f' = (Pw - Pr)/Pw \quad (7)$$

where Pw is the precipitable water (i.e., total water vapor content in the atmospheric column), and Pr is the precipitation. The isotopic composition of the initial water vapor for the event, δ'_{v0} , can be given by δ_v computed from Eq. (3).

Finally, by assigning appropriate values for T_{c0} and δ_{v0} , we can predict event-based or daily isotopic compositions of precipitation using Eqs. (3) and (6) based on T_c , Pr and Pw . In the present study, we have assumed that T_c can be approximated by the temperature at the 700 hPa level, T_{700} . For initial values, we have further assumed that they correspond to those representing the subtropical ocean region, which is known to be the main source of global atmospheric water vapor (e.g., Craig and Gordon, 1965; Merlivat and Jouzel, 1979). This results in $T_{c0} = 10^\circ\text{C}$, a typical value of T_{700} over the region, and $\delta D_{v0} = -94\text{‰}$ and $\delta^{18}O_{v0} = -13\text{‰}$ (Craig and Gordon, 1965). Regionally averaged precipitation from surface observation and precipitable water data from NCEP/NCAR reanalysis were utilized in calculating Eq. (7). The equilibrium fractionation factor was computed with an empirical T – α relationship obtained by Majoube (1971) using the median between T_c and T_{c0} in Eq. (3) and T_c in Eq. (6). For simplification, the phase change from vapor to solid was not taken into consideration in this computation or in computing e_{sat} . All the assumptions included in the model will be validated in the following.

Application

Fig. 6 compares day-to-day variations of predicted and observed isotopic precipitation compositions. The observed values are regional representatives (see “Correlation with meteorological factors”), and two types of predictions are presented: one (hereafter, full model; FM) is computed using Eqs. (3) and (6), and the other (hereafter, temperature-effect model; TM) uses Eqs. (3) and (5) involving only

the temperature effect. Both the predictions reproduce the observed results very well. The two predictions are not very different, whereas the reproducibility of FM is better than that of TM for several events with minimal δ . The root mean square error (r.m.s.e.) for all periods is slightly improved from 37‰ in TM to 30‰ in FM for δD and 3.0‰ in TM to 2.7‰ in FM for $\delta^{18}O$. Thus, while the on-site precipitation process cannot be negligible, the isotopic composition of precipitation is primarily controlled by the rainout history during transportation from the vapor source area to eastern Mongolia.

Similarity between the TM outputs and the observed results is to be expected from the high correlation between temperature and δ values (Table 3), whereas the remarkably good reproducibility of not only the correlation but also the variation range and slope value supports the idea that isotopic variation of precipitation over Mongolia can be explained by global- or continental-scale rainout history from a single vapor-source reservoir rather than by a local precipitation system. It should be noticed that the initial conditions of temperature and isotopic composition correspond with those for a subtropical marine atmosphere. The other sets of temperatures and δ values could also produce the same result. However, such conditions themselves should be of an air mass that originated in a subtropical ocean re-

gion as they are predicted by our model. Thus, the present result suggests that the ultimate origin of Mongolian precipitation is a subtropical marine atmosphere. However, we cannot reject the possible contribution from re-evaporation in other regions if the T – δ relationship is hardly affected by the re-evaporation process. Clearly more studies are needed to draw a concrete conclusion.

Only in July, do the models exceptionally overestimate precipitation δ values (Fig. 6). Potential causes of this overestimation are threefold: (1) underestimation in T_{c0} , (2) overestimation in δ_{v0} , and (3) contribution of continental recycling water with a strong isotopic signal. It is difficult, using the first two causes to explain the reason for the fact that the exceptional overestimation occurs only in July.

On the other hand, it can be explained by a continental recycling process. Fig. 7 is a rose diagram for the 700 hPa wind for each month, showing that the steady northwesterly wind weakened only in July. In contrast, a southerly wind occurred more frequently in July than in other months. Fig. 8 demonstrates that the observed δ tends to have much smaller values than predicted when water vapor comes from the south. In particular, although there are not many southeasterly events, they are linked to small δ values with a strong probability. We can infer from these facts that continental vapors originating in the south of Mongolia

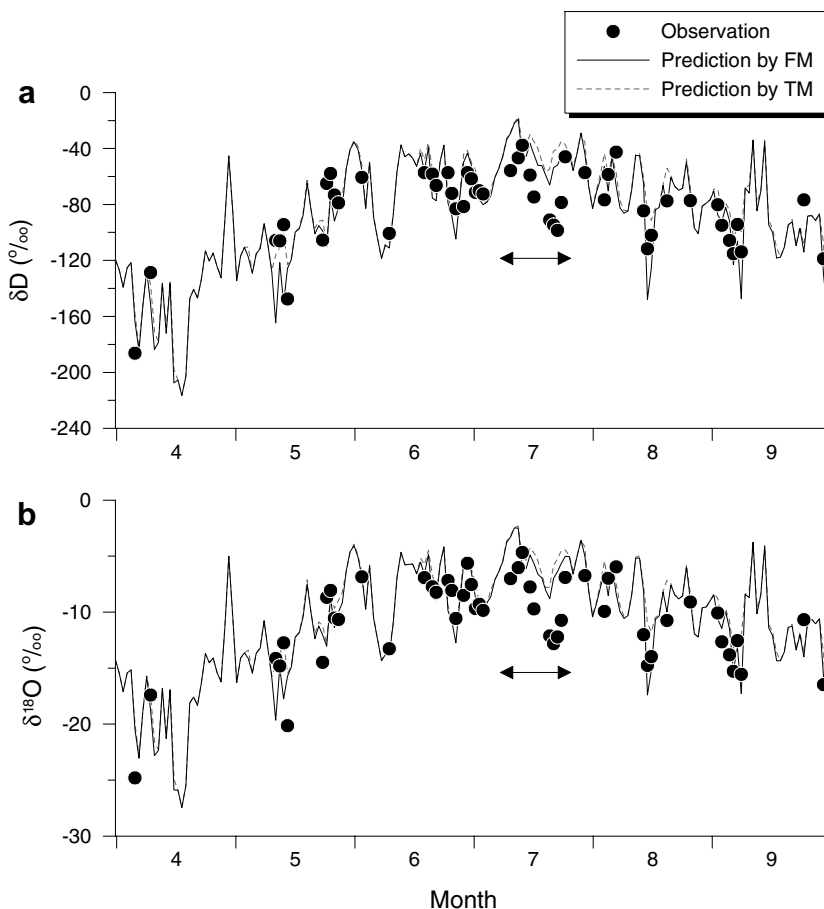


Figure 6 Comparison of day-to-day variations between predicted and observed isotopic compositions of precipitation. The observed values are regional representatives. Two types of predictions are presented: one is from the full model (FM; Eqs. (3) and (6)) involving both the temperature and amount effects, and another is from the temperature-effect model (TM; Eqs. (3) and (5)) involving only the temperature effect. Arrow indicates a period in which the models clearly overestimate.

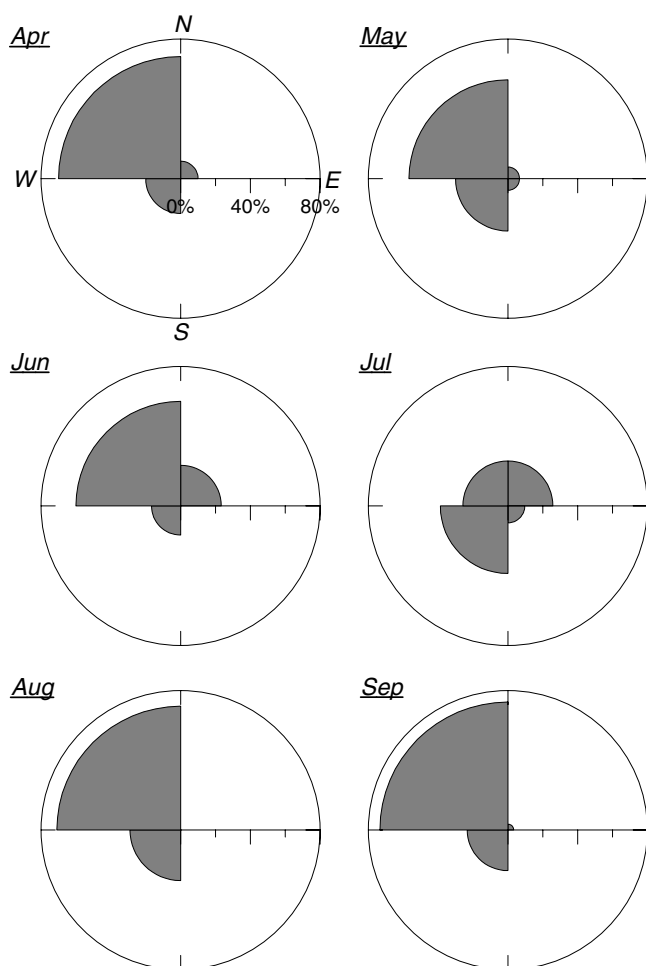


Figure 7 Relative frequency distribution of wind directions at 700 hPa level for each month.

contribute to a considerable portion of the atmospheric moisture in July and provide smaller δ values than those expected from the Rayleigh model for eastern Mongolian precipitation. One of the most likely source-areas may be southeast China, where evaporation occurs most intensively in eastern Eurasia and its adjacent seas (Fig. 9a). In southeast China, precipitation δ in summer is lowered by the amount effect due to the strong monsoon activity (Yamanaka et al., 2004). The precipitating water is stored in flooded, rice paddy fields, which is the most dominant land cover in the region, and then re-evaporates into the atmosphere. The evaporation from the water surface in paddy fields is subject to isotopic fractionation and produces water vapor further depleted in heavy isotopes. Figs. 9b and c suggest that such water vapor can be transported from southeast China into eastern Mongolia by transient eddies not by mean flow. Although the monthly mean value of the transient eddy flux is not always greater than the mean flow component, rainfall events with low δ are strongly associated with southerly or southeasterly winds, which are caused by transient eddies (Fig. 8), indicating that the transient eddy is important for transporting water vapor as an ingredient in precipitation over eastern Mongolia. All of the above facts are consistent with the idea that deviation of observations from the model predictions is caused

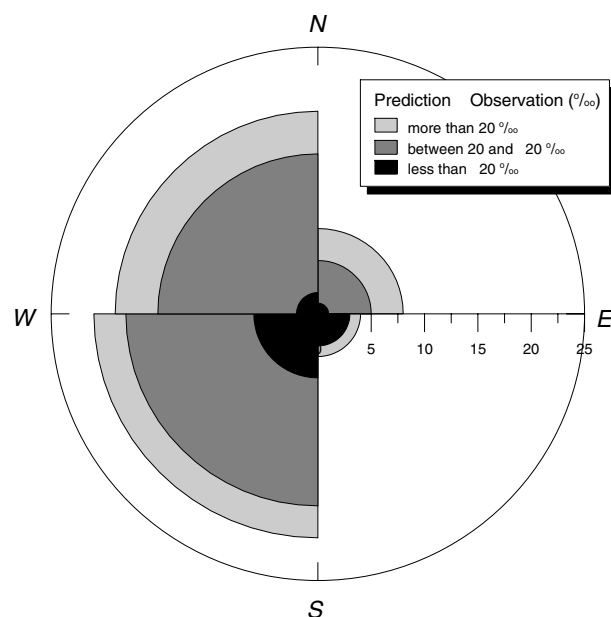


Figure 8 Histogram of wind directions at the 700 hPa level with classification by deviation of observations from predictions for days in which precipitation was sampled more than once from April to September 2003. Each bin represents the number of times the wind blew from a given direction. The bin is divided up into three sub-sections by the range of differences between observations and predictions by FM.

by the contribution of water vapor originating in southeast China. Unfortunately, we lack direct evidence for identifying vapor source areas and thus cannot reject other possibilities. For specifying them, deuterium excess is usually more useful than δ (Yamanaka et al., 2002). However, in the present study it has been altered for many events by the evaporation of falling rain drops under the cloud base. This is due to the arid climate in the study area. In addition, d of water vapor evaporating from rice paddy fields or any other land cover is still unknown, so that it is difficult in the present state to specify source areas of precipitation using d . Further investigations will be necessary to solve the above problems and examine our findings.

Throughout the present analysis, the new and most interesting finding is the fact that the simple Rayleigh-type model can simulate isotopic variability of precipitation in Mongolia at not only monthly but also daily time-scales. Although there are some uncertainties, the present result may be related to the unique geographical conditions of Mongolia. Since Mongolia is situated far from any ocean, the effects of the source region's meteorology and isotopic variations would be averaged during long-range transport by mixing of vapors having different sources and histories, so that various isotopic signals may cancel each other out (except for the strong signal in July) and the original signal from the main source reservoir can stand out. In addition to this, because of the high altitude of the Mongolian Plateau, temperature and isotopic variations are likely to be insensitive to water and energy exchanges between land surfaces and the atmosphere during the transportation process over surrounding regions. Therefore, the initial condition of an air parcel can be treated as an average, and the

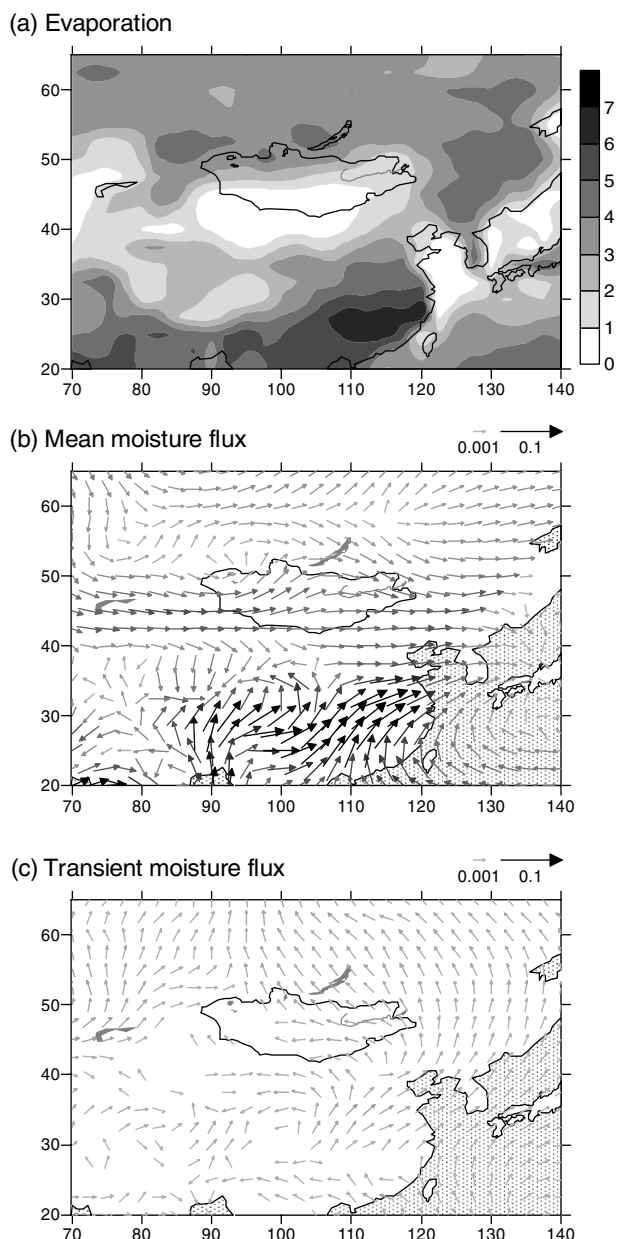


Figure 9 Spatial distribution of (a) evaporation (mm/d) and water vapor fluxes at 700 hPa ($\text{kg kg}^{-1} \text{m s}^{-1}$) due to (b) mean flows and (c) transient eddies, in July 2003. Water vapor flux was partitioned into the two components according to conventional breakdown (e.g., Peixoto and Oort, 1992).

isotopic variation of precipitation is most dependent on local temperature that reflects the mean rainout-history of the parcel coming day-by-day. To verify this speculation, the correlative variation in global temperature and isotope fields should be reexamined at a shorter (e.g., daily) time scale.

Summary and conclusions

The present paper describes the isotopic characteristics of precipitation over eastern Mongolia and assesses their

causes in terms of a continental-scale atmospheric water cycle. The main results and conclusions are summarized as follows:

- (1) Seasonal and day-to-day variations of isotopic composition in precipitation are characterized by a strong correlation with air temperature and a weak correlation with precipitation amount. Temporal variation patterns are almost uniform among sites within the study area.
- (2) A Rayleigh-type model successfully reproduces not only the seasonal variation but also the day-to-day variation of precipitation δ values, indicating that isotopic composition of precipitation is primarily controlled by rainout history during transportation from a vapor-source-reservoir. The assumption of a single source is effective, and its conditions can be treated in the same way as for a subtropical marine atmosphere. This result also suggests that the continental recycling process generally has a minor effect on isotopic variations in precipitation over, at least, eastern Mongolia.
- (3) Exceptionally, the model overestimated the δ values only in July, suggesting a contribution of isotopically light, additive water vapors. Because the overestimation tended to occur in southerly wind conditions and the most dominant wind in July was southerly, one of the remarkable vapor sources is likely to be rice paddy fields in southeast China, where evaporation occurs most intensively in eastern Eurasia and evaporated water vapor is further depleted in heavy isotopes by the isotopic fractionation effect. However, further research will be necessary to specify the source area more definitely.

Acknowledgements

This research was partly supported by the Japan Science and Technology Agency under the Core Research for Evolutional Science and Technology program (RAISE: the rangelands atmosphere–hydrosphere–biosphere interaction study experiment in northeastern Asia) and by a Grant-in-Aid for Scientific Research (No. 15740289, T.Y.). We are grateful to staff members of the Mongolian meteorological observatory and the Institute of Meteorology and Hydrology for their assistance in collecting precipitation samples for isotopic analysis. This manuscript has been improved by comments from M. Sugita, X. Song and two anonymous reviewers.

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