ISSN 0001-4370, Oceanology, 2017, Vol. 57, No. 1, pp. 31–40. © Pleiades Publishing, Inc., 2017.
Original Russian Text © E.O. Dubinina, S.A. Kossova, A.Yu. Miroshnikov, R.V. Fyaizullina, 2017, published in Okeanologiya, 2017, Vol. 57, No. 1, pp. 38–48.

MARINE CHEMISTRY

Isotope Parameters (δD, $\delta^{18}O$) and Sources of Freshwater Input to Kara Sea

E. O. Dubinina[a], *, S. A. Kossova[a], A. Yu. Miroshnikov[a], and R. V. Fyaizullina[b]

[a] *Institute of Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry, Russian Academy of Sciences, Moscow, Russia*

[b] *Moscow State University, Geological Faculty, Moscow, Russia*

**e-mail: elenadelta@gmail.com*

Received June 7, 2016

Abstract—The isotope characteristics (δD, $\delta^{18}O$) of Kara Sea water were studied for quantitative estimation of freshwater runoff at stations located along transect from Yamal Peninsula to Blagopoluchiya Bay (Novaya Zemlya). Freshwater samples were studied for glaciers (Rose, Serp i Molot) and for Yenisei and Ob estuaries. As a whole, δD and $\delta^{18}O$ are higher in glaciers than in river waters. isotope composition of estuarial water from Ob River is δD = −131.4 and $\delta^{18}O$ = −17.6‰. Estuarial waters of Yenisei River are characterized by compositions close to those of Ob River (−134.4 and −17.7‰), as well as by isotopically "heavier" compositions (−120.7 and −15.8‰). Waters from studied section of Kara Sea can be product of mixing of freshwater (δD = −119.4, $\delta^{18}O$ = −15.5) and seawater (S = 34.9, δD = +1.56, $\delta^{18}O$ = +0.25) with a composition close to that of Barents Sea water. isotope parameters of water vary significantly with salinity in surface layer, and Kara Sea waters are desalinated along entire studied transect due to river runoff. concentration of freshwater is 5–10% in main part of water column, and <5% at a depth of >100 m. maximum contribution of freshwater (>65%) was recorded in surface layer of central part of sea.

DOI: 10.1134/S0001437017010040

INTRODUCTION

The problem of identifying waters, their mixing processes, and formation of currents in the water column of Arctic Basin is among most important in studying the World Ocean. In this context, poorly studied isotope ($\delta^{18}O$ and δD) parameters of waters from the Russian Arctic shelf represent a huge gap in modern knowledge system. This is especially important for understanding the desalinization processes of Kara Sea water with an annual continental runoff of >1.5 × 10^3 km^3, which is more than a third of total continental runoff into Arctic water area [19]. More than 150 mln t of allochthonous material carrying a wide spectrum of pollutants (including radioactive) are discharged into the sea with runoff [6, 22]. Radionuclides entering the Kara Sea from catchment basins of the Ob and Yenisei rivers as both redistributed global precipitates and products of radiochemical factories are usually recorded in sea and river water mixing zones [4]. However, some pollutants pass frontal zones in estuaries both in dissolved form and in a sorbed state on suspension. Further migration of pollutants coming with river runoff to the shelf up to the coast of Novaya Zemlya is promoted by sharp stratification of surface waters of the Kara Sea, which was recorded in studies carried out in different years [3, 6, 7]. Thus, identification of sources and quantitative proportion of waters forming the surface desalinated layer in the Kara Sea is a very important task.

In this paper, we report the isotope composition of the desalinated surface layer on the basis of the isotope geochemistry of oxygen and hydrogen, which makes it possible to solve genetic tasks in hydrology of sea basins with a high level of evidence.

The natural isotope indicators of water (δD, $\delta^{18}O$) allow us to distinguish water flows and provide a quantitative estimate for content of waters of different origin in a water column, as well as to study phase transitions in evolution of the water column [5, 17]. Comparison of isotope data with data on salinity is highly informative in study of sea basins, since salinity and isotope composition of hydrogen and oxygen have a similar behavior during phase transitions and mixing of water of different origin [16]. Therefore, collection of samples accompanied hydrophysical data and accurate relationships to characteristics of water column is a key factor in isotope studies of seawater. fact that an isotope marker of water makes it possible to distinguish freshwater component of origin (glacial, river, and atmospheric) is an additional advantage to applying isotope methods to studying seawater desalination. In some cases, we can establish not only the type of freshwater component, but its source as well. For example, Russia's large northern rivers, which are the main contributors to freshwater runoff into the zone of Arctic seas, regularly differ in their isotope (δD and $\delta^{18}O$) parameters [8, 12, 15]. This makes accurate identification of water columns participating in desalination of Arctic seas promising.

It is common to study one of the isotope systems of water (usually oxygen) and interpret the data within correlation of salinity and $\delta^{18}O$ values [8, 9, 13–15, etc.]. Meanwhile, analysis of two isotope systems (hydrogen and oxygen) yields better identification of components in δD–$\delta^{18}O$ coordinates that have both physical and genetic meaning [23]. Moreover, these coordinates make it possible to verify the models of complex with two or more mixing components, as well as study evaporation and freezing in evolution of water columns.

The study of seawaters requires highly accurate isotope analysis, because variations in δD and $\delta^{18}O$ in a seawater column usually do not exceed a few ppm, whereas in actively mixed segments. These values can be at level of analytical error. For example, δD variations in the central part of the Black Sea do not exceed a few ppm. These variations could be indistinguishable in an analysis of hydrogen isotope composition using old (before 1990s) methods with an accuracy of ±5‰. Recent methods for precisely analyzing the hydrogen isotope composition have an accuracy higher by 1.5 orders of magnitude (about ±0.3‰), which has shown, for example, slight isotope variations and made it possible to distinguish different waters in Black Sea column [1].

With allowance for the abovementioned factors, we can summarize certain requirements on procedure for isotope–hydrological studies of seawater: correct sample collection accompanied by detailed hydrophysical data, study of two isotope systems (δD and $\delta^{18}O$), and application of highly accurate isotope methods. All these factors were observed in studying the Kara Sea water column in order to establish sources and configuration of the desalinated layer and to obtain quantitative estimates for the contribution of freshwater runoff to the Kara Sea.

MATERIALS AND METHODS

The oxygen and hydrogen isotope compositions were studied in water samples from Kara Sea collected during cruise 128 of R/V *Professor Shtokman* in 2014 [6]. The location scheme of the studied stations is shown in Fig. 1. Seawater samples were collected at stations along a transect from Yamal Peninsula to Blagopoluchiya Bay of Novaya Zemlya archipelago (stations 4–7 and 9–12). Freshwater samples were collected in Rose and Serp i Molot glaciers and in estuaries of Ob (station 18) and Yenisei (stations 25–28) rivers. Samples were collected with a Rosette hydrophysical complex equipped with necessary detectors and a set of Niskin bathometers. The vertical distribution profile of hydrophysical parameters and δD, $\delta^{18}O$ was studied at each station.

The oxygen isotope analysis was carried out by isotope balancing of water with CO_2 using a GasBench II instrumental complex and a PAL autosampler. The size of water sample, temperature, and duration of isotope exchange reaction were 0.5 cm^3, 32°C, and 18 h, respectively. The oxygen isotope composition in CO_2 was analyzed on a DELTA V+ mass spectrometer in the mode of a constant helium flow (the CF IRMS method). The hydrogen isotope composition was carried out by decomposition of microsamples (0.001 cm^3) of water on hot (800°C) chromium using an H/Device instrumental complex and a DELTAplus mass spectrometer operating in dual inlet mode (the DI IRMS method). δD and $\delta^{18}O$ values in water samples were calibrated against V-SMOW–V-SLAP scale using internal standards and samples for comparison MAGATE (OH-1–OH-4 and OH-13–OH-16), which are regularly calibrated in laboratory in with respect to V-SMOW and V-SLAP standards. The accuracy of $\delta^{18}O$ and δD measurements was ±0.2 and ±0.3‰, respectively. The isotope data for waters of stations along the Yamal transect are given in Table 1 together with data on sampling depth and salinity. The isotope data for glacial and river waters are given in Table 2.

RESULTS

Kara Sea water. Waters of stations 4–7 and 9–12 located along the transect from Yamal Peninsula to Blagopoluchiya Bay (Yamal transect) show dilution by the freshwater component, which is heterogeneous in space. A sharp variation in salinity and isotope parameters of water is observed for the thin surface layer (≤25 m; on average, ~10 m) (Fig. 2). This makes it possible to conclude that dilution by freshwater has a sharp depth gradient (Fig. 2), i.e., a layer of strongly desalinated waters with low salinity and isotope parameters close to composition of atmospheric waters (e.g., δD reaches –80‰) in the surface zone of the central part of the sea. A linear trend formed by samples of the Yamal trend ($y = 7.60x–1.46$, $R^2 = 0.997$) is clearly observed in δD–$\delta^{18}O$ isotope diagram (Fig. 3).

Waters of the studied section are characterized by a strong correlation of isotope parameters with salinity (Fig. 4), which indicates two-component mixing of freshwater and seawater components. Extrapolation of the linear trend (Fig. 4) to zero salinity (and a similar dependence of oxygen isotope composition on salinity) makes it possible to calculate characteristics of freshwater component for waters of Kara Sea along Yamal transect (Table 2, Fig. 4).

River waters. Ob River water is characterized by δD and $\delta^{18}O$ values of –131.4 ± 0.2 and –17.6 ± 0.03‰, respectively. Yenisei River water shows a bimodal behavior of isotope parameters. δD and $\delta^{18}O$ analyzed

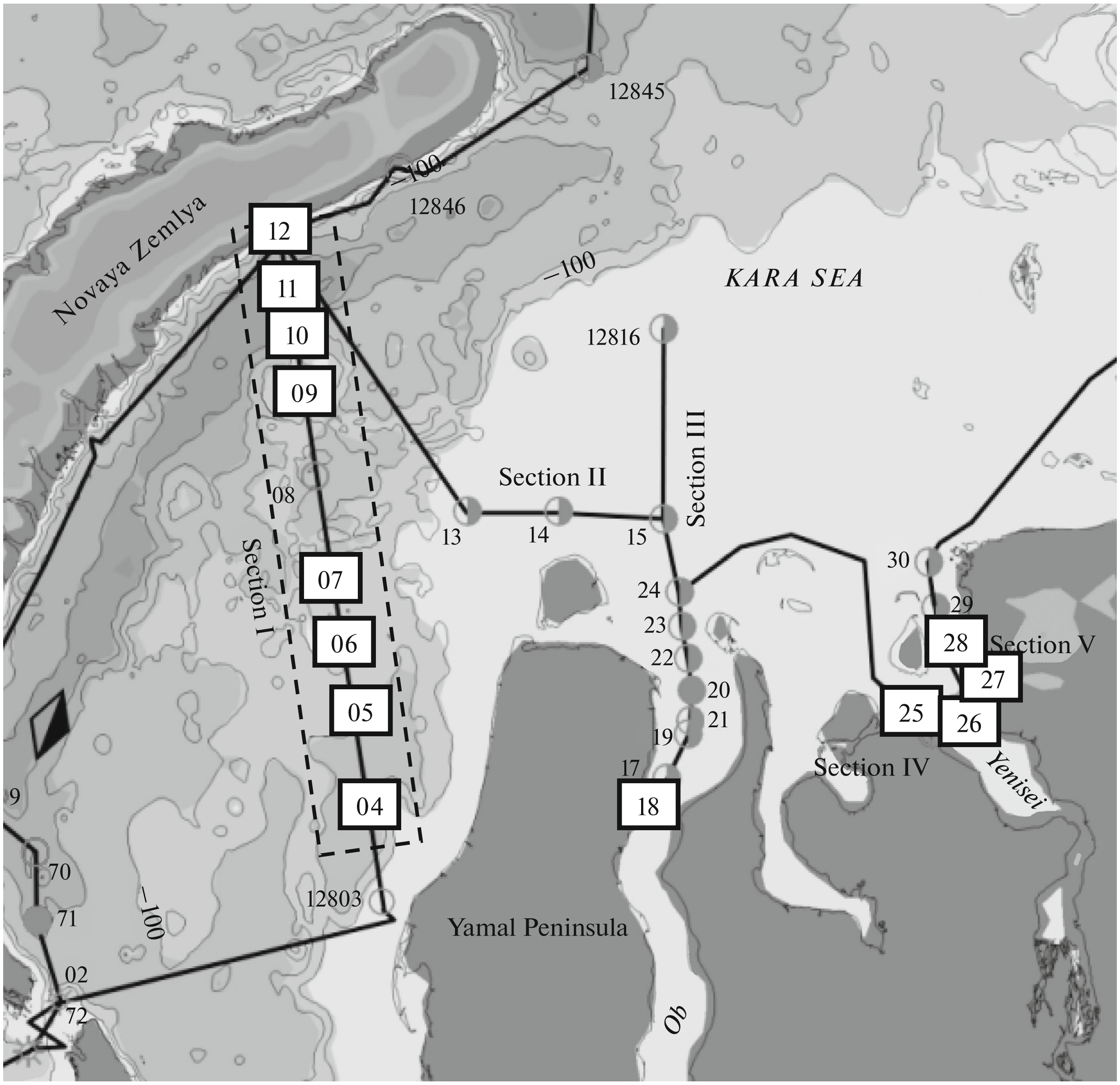

Fig. 1. Scheme of sampling on cruise 128 of R/V *Professor Shtokman* (2014) in Kara Sea. Rectangles with numbers are stations at which material for isotope studies was collected: stations 04–07 and 09–12, Yamal transect along desalination zone; 18, Ob River; 25–28, Yenisei River.

for stations 25 and 26 are close to those obtained for the Ob River (–134.4 and –17.7‰), whereas at stations 27 and 28, these values are significantly higher (–120.7 and –15.8‰). Such behavior of isotope parameters within a small area of the estuary of the same river (Fig. 1) is evidence for transformation of waters, most likely due to mixing or freezing. According to available data, Ob River waters are characterized by higher δ^{18}O values (by ~2‰) than those obtained for Yenisei River waters [8, 15, and references therein]. Our data for stations in estuaries do not agree with these general relationships, but such deviations in isotope parameters were previously observed for these rivers as well [7].

Glaciers of Novaya Zemlya. As a whole, oxygen and hydrogen isotope compositions of glaciers of Novaya Zemlya (δD = –94.1…–122.8, δ^{18}O = –13.4…–17.0) are “heavier” than the composition of waters of main river (Ob and Yenisei) runoff (Fig. 5). Compositional points on δD–δ^{18}O isotope diagram plot along Craig line $\delta D = 8.06 \times \delta^{18}O + 14.2$ ($R^2 = 0.997$), which is evidence for their atmospheric origin with a high excess of deuterium typical of sediments from this region [11]. Serp i Molot glacier in Tsivol’ka Bay is characterized by higher δD and δ^{18}O values than Rose glacier. Most likely, this is explained by different isotope compositions of atmospheric precipitates within

Table 1. Results of isotope studies of waters from Kara Sea

Station no., (depth), (coordinates; N, E)	Depth, m	*S*, PSU	$\delta^{18}O$, ‰	δD, ‰	*X* (portion of freshwater component)
1	2	3	4	5	6
128-04 (154) (71°45.2′; 65°45.6′)	3	25.88	–4.6	–36.3	0.31
	8	26.24	–3.2	–26.2	0.23
	15	32.34	–1.1	–10.8	0.10
	20	33.17	–0.7	–7.7	0.08
	30	33.58	–0.7	–7.6	0.08
	40	33.72	–0.5	–6.4	0.07
	80	34.39	–0.2	–2.8	0.04
	110	34.57	–0.5	–4.7	0.05
	125	34.64	–0.6	–5.7	0.06
	151	34.77	–0.7	–6.7	0.07
128-05 (105) (72°25′; 65°28′)	2	23.66	–5.7	–46.4	0.40
	5	23.79	–4.6	–37.0	0.32
	10	30.41	–2.0	–17.3	0.16
	20	33.35	–0.5	–6.8	0.07
	36	33.63	–0.7	–6.7	0.07
	50	33.71	–0.6	–6.7	0.07
	75	34.34	–0.5	–4.7	0.05
	101	34.46	–0.6	–4.9	0.05
128-06 (76) (72°53′; 65°30′)	2	22.28	–6.0	–48.4	0.41
	7	22.84	–5.1	–41.5	0.36
	16	32.61	–0.8	–9.7	0.09
	25	33.44	–0.5	–6.6	0.07
	30	33.56	–0.3	–6.2	0.06
	40	33.66	–0.4	–5.7	0.06
	60	33.98	–0.5	–5.8	0.06
	74	34.10	–0.5	–4.9	0.05
128-07 (64) (73°20′; 65°40′)	1	11.69	–10.5	–80.6	0.68
	4	11.88	–10.2	–79.1	0.67
	8	16.98	–7.8	–60.6	0.51
	18	32.84	–1.5	–13.0	0.12
	25	33.53	–0.6	–6.1	0.06
	35	33.64	–0.7	–7.2	0.07
	50	33.81	–0.5	–5.7	0.06
	62	34.09	–0.6	–4.7	0.05
128-09 (61) (74°42.3′; 64°54′)	2	25.91	–4.9	–36.4	0.31
	4	27.77	–2.5	–20.0	0.18
	8	29.24	–1.7	–13.7	0.13
	17	33.25	–0.4	–4.8	0.05
	25	33.45	–0.3	–2.7	0.03
	35	33.72	–0.2	–1.9	0.03
	45	33.86	–0.1	–1.7	0.03
	58	33.92	–0.1	–1.6	0.03

Table 1. (Contd.)

Station no., (depth), (coordinates; N, E)	Depth, m	*S*, PSU	$\delta^{18}O$, ‰	δD, ‰	*X* (portion of freshwater component)
1	2	3	4	5	6
128-10	1	29.90	–0.8	–6.9	0.07
(260)	7	30.04	–0.7	–6.2	0.06
(75°03.7′; 64°34.2′)	14	32.97	–0.7	–5.9	0.06
	20	33.36	–0.6	–5.3	0.06
	32	33.58	–0.5	–5.6	0.06
	50	33.97	–0.2	–2.7	0.04
	65	34.31	–0.1	–1.7	0.03
	100	34.37	0.0	–1.5	0.02
	140	34.40	0.1	–1.0	0.02
	200	34.46	0.2	–1.0	0.02
	225	34.50	0.1	–0.9	0.02
	256	34.61	0.3	–0.4	0.02
128-11	1	29.53	–0.7	–6.0	0.06
(350)	8	32.05	–0.6	–5.7	0.06
(75°23.1′; 64°18.1′)	15	32.36	–0.6	–5.0	0.05
	20	32.51	–0.5	–5.4	0.06
	30	33.13	–0.5	–4.9	0.05
	45	33.66	–0.3	–3.6	0.04
	75	34.06	–0.2	–2.9	0.04
	100	34.36	0.0	–1.5	0.03
	150	34.48	0.1	–1.2	0.02
	200	34.56	0.1	–0.7	0.02
	250	34.62	0.2	–0.4	0.02
	300	34.69	0.2	–0.7	0.02
	348	34.73	0.2	–0.1	0.01
128-12	2	31.03	–1.5	–11.2	0.11
(30)	5	31.41	–0.7	–5.3	0.06
(75°35.6′; 63°41.9′)	10	32.56	–0.5	–4.4	0.05
	15	33.22	–0.4	–3.6	0.04
	20	33.46	–0.4	–3.3	0.04
	27	33.77	–0.2	–2.8	0.04

the Novaya Zemlya archipelago. Waters of a small river with a source under Serp i Molot glacier have δD and $\delta^{18}O$ ranges typical of samples from the glacier (Fig. 5). The composition of these waters can be taken as the average composition of Serp i Molot glacial deposits.

DISCUSSION

In contrast to situations described in other water areas of the Arctic region [8–10, etc.], simple mixing of isotopically heavy saline and isotopically light freshwater components without evidence of participation of other water sources (e.g., sea ice) is observed in the center of the Kara Sea. In this case, the task of searching for the source of and estimating the contribution of freshwater runoff for the center of the Kara Sea is significantly simplified. It is necessary to establish isotope parameters for saline (seawater) and freshwater end-members to calculate the contribution of the freshwater component. The seawater component can be represented as composition of nondesalinated water of Barents Sea with a salinity of 34.90 ± 0.05 [2], which plots in the upper part of the linear trend of Yamal transect waters in the δD–$\delta^{18}O$ diagram. The

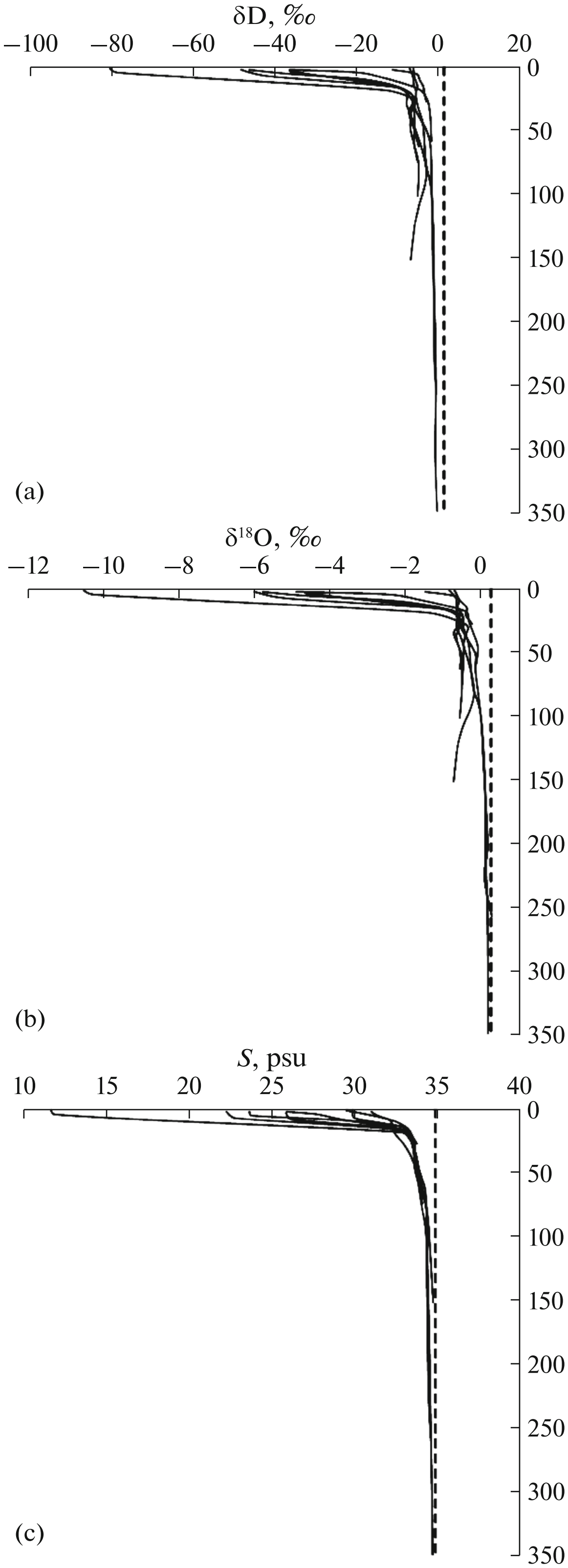

Fig. 2. Vertical distribution of δD (a), δ^{18}O (b) and salinity (c) at stations of Yamal transect. Depth (m) is shown on *Y* axis. Dashed lines indicate parameters of nondesalinated waters of Barents Sea (S = 34.9, δD = 1.56 ± 0.4, δ^{18}O = 0.25 ± 0.1‰).

analyzed composition of Barents Sea waters plots in the range of δD (0 to +2) and δ^{18}O (+0.3 to –0.2) reported by different authors as a characteristic value for Arctic waters [18, 21].

The isotope parameters of the freshwater component can be determined by analyzing the data in isotope composition–salinity coordinates by extrapolating the linear trend to zero salinity. As is evident from the δD–*S* diagram (Fig. 4), waters of the studied profile of the Kara Sea contain a freshwater component with a hydrogen isotope composition of δD = –119.4. A similar trend is observed in δ^{18}O–*S* for the oxygen isotope composition of freshwater component δ^{18}O = –15.5‰ (Table 2). These compositions are very similar to compositions of waters collected at stations 27 and 28 in the Yenisei River estuary. The position of the lower part of the Yamal transect trend and calculated composition of the freshwater component in the δD–δ^{18}O diagram (Fig. 5) show that river waters, but not Novaya Zemlya glaciers, yield freshwater component for the central part of the Kara Sea.

The mostly river origin of freshwater component in the Kara Sea is not in doubt, but now it is impossible to obtain more precise information, for example, a quantitative estimate of individual contribution of each of large rivers. This is explained by the absence of unambiguous estimates of isotope characteristics for the Ob and Yenisei rivers. For example, data on behavior of δ^{18}O values with seasonal variations and on a long-term scale [8, 12, 15] are fragmentary, and the hydrogen isotope composition in waters of Russia's northern rivers is virtually unstudied. It is evident that our data cannot be used as unique characteristics of Ob and Yenisei waters. Since sampling stations were quite close to the estuarial zones of these rivers, we cannot ignore mixing of their waters with each other. However, our data are applicable for a general estimate of freshwater runoff source, because it is unlikely that unaltered or isolated river waters participate directly in desalinating Kara Sea water. Yenisei River waters showing a bimodal distribution of isotope parameters were collected at stations located on the western and eastern banks of the estuary and could be transformed to various degrees upon freezing or mixing with Ob River water. Ob River runoff to Yenisei River estuary was previously recorded by oxygen isotope data [7], which was interpreted by the authors as the predominance of northern winds during studies. However, we cannot ignore that river ice formation processes could change isotope parameters of water in the Yenisei River estuary. Figure 5 shows the trend line plotted through all points of samples collected on the Yenisei River (stations 25–28). The slope of trend (6.7) is close to that formed by freezing freshwater [20]. Intersection of this trend with that plotted through points of Novaya Zemlya glaciers occurs in the area of δ^{18}O ≈ –22‰, which is close to the average oxygen isotope composition of atmospheric precipitates in this region

Table 2. Isotope parameters and salinity of main sources of water in Kara Sea region

Component	Method of estimation	δD, ‰	δ^{18}O, ‰	*S*, PSU
Nondesalinated water of Barents Sea	Direct measurement, *n* = 44	+1.56 ± 0.4	+0.25 ± 0.1	34.9
Transformed waters of Yenisei River (stations 27 and 28)	Direct measurement, *n* = 2	–120.7	–15.7	0
Yenisei estuary (stations 25 and 26)	Direct measurement, *n* = 2	–134	–17.7	0
Ob estuary (station 18)	Direct measurement, *n* = 3	–131.4	–17.6	0
Novaya Zemlya glaciers (Rose, Serp i Molot)	Direct measurement, *n* = 9	–94…–123	–13.4…–17.0	0
Freshwater component coming to central part of Kara Sea	Calculation by isotope composition–salinity relationship (extrapolation for *S* = 0)	–119.4	–15.5	0

[18]. Most likely, a common component of local atmospheric genesis is included in the composition of both glacial deposits and river waters of estuarial zones. In addition, the Yenisei River estuary can contain waters coming from the Ob River (stations 25 and 26), as well as waters transformed upon freezing (stations 27 and 28). Interestingly, transformed waters of the Yenisei River with compositional deviations from the trend of meteoric waters matches most exactly the calculated composition of freshwater component for waters of the Yamal transect.

We applied a simple model of two-component mixing of nondesalinated waters of the Barents Sea and freshwater component with the composition obtained from isotope composition–salinity relationships for a quantitative estimate of freshwater content

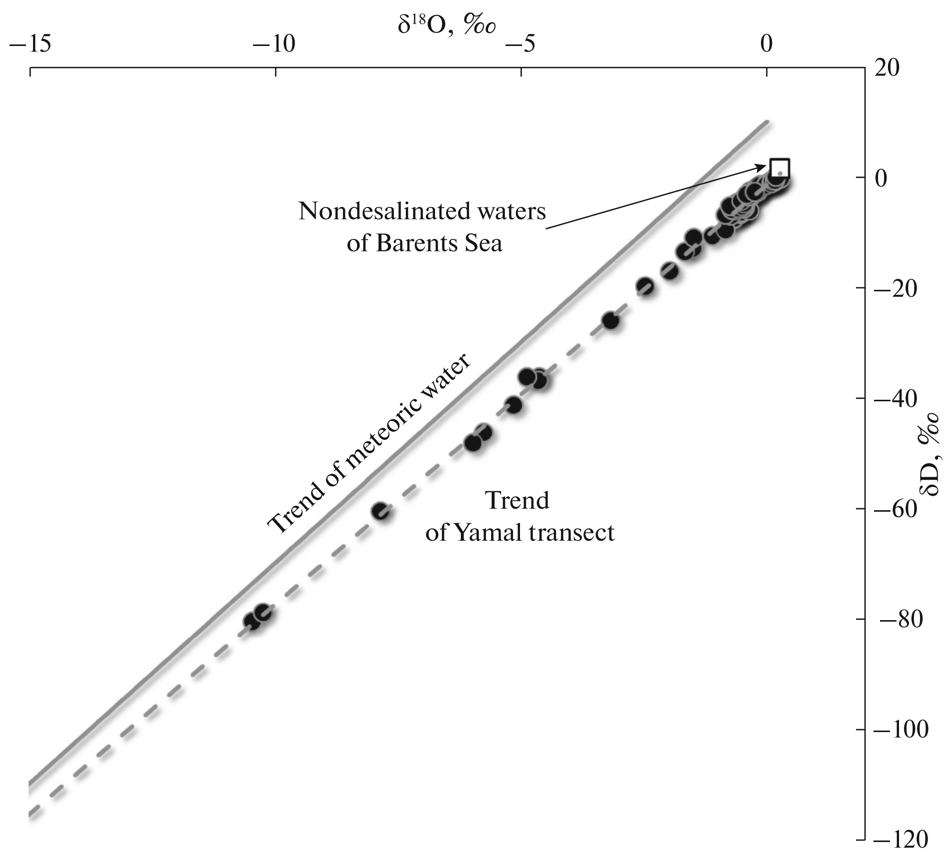

Fig. 3. Isotope parameters (δD, δ^{18}O) of Kara Sea waters along transect from Yamal Peninsula to Blagopoluchiya Bay (Novaya Zemlya).

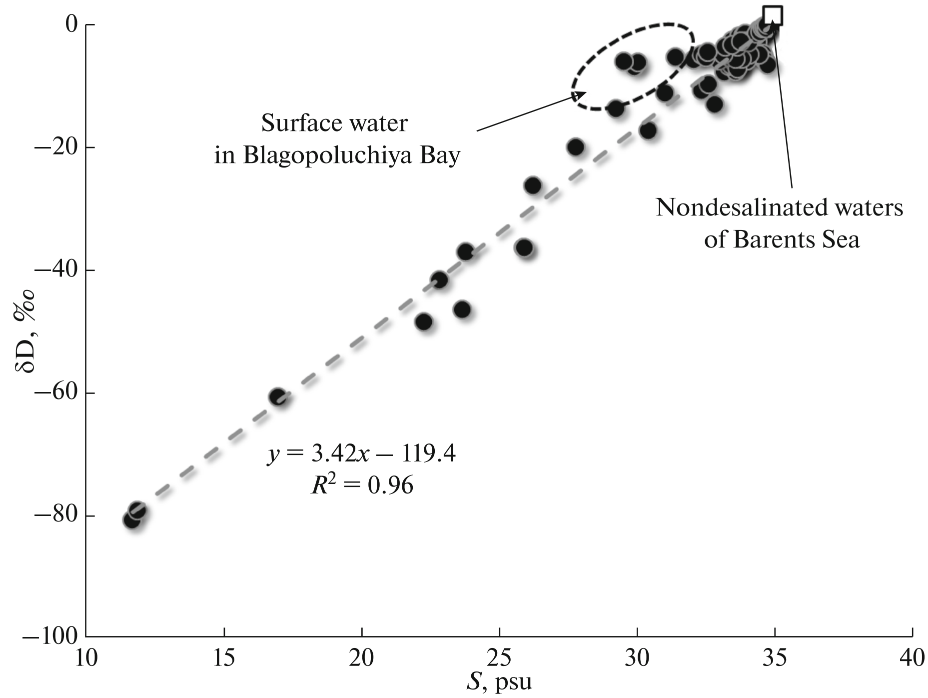

Fig. 4. Relationship between hydrogen isotope composition and salinity in Kara Sea waters collected along Yamal transect.

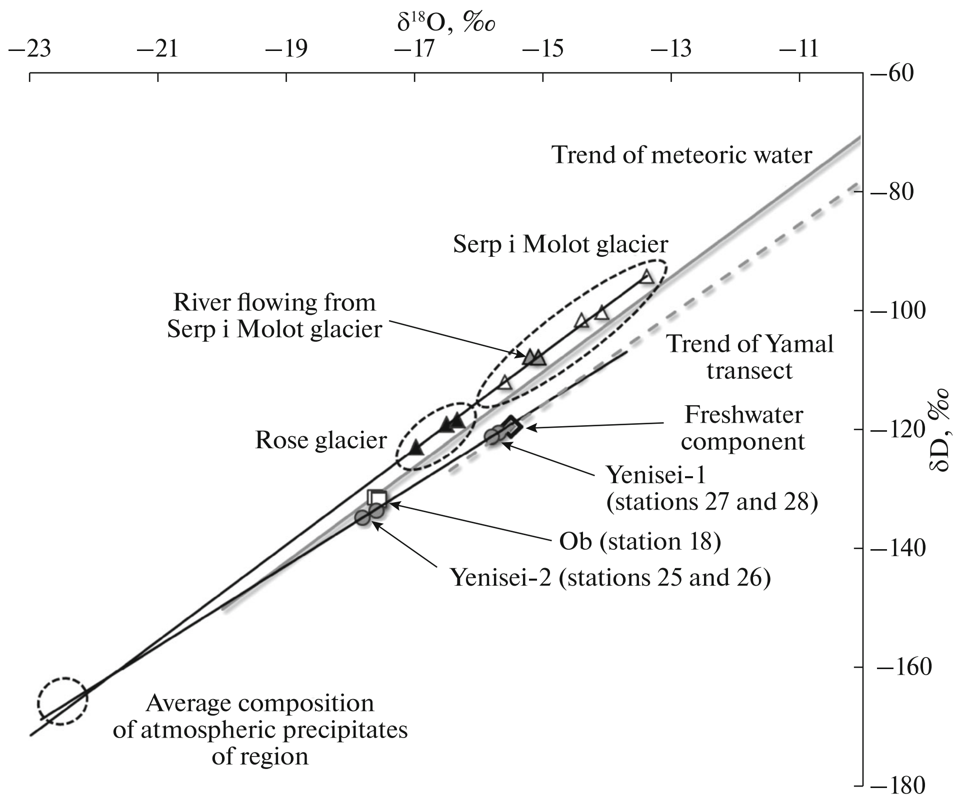

Fig. 5. Isotope (δD, $\delta^{18}O$) systematics of freshwater in Kara Sea region.

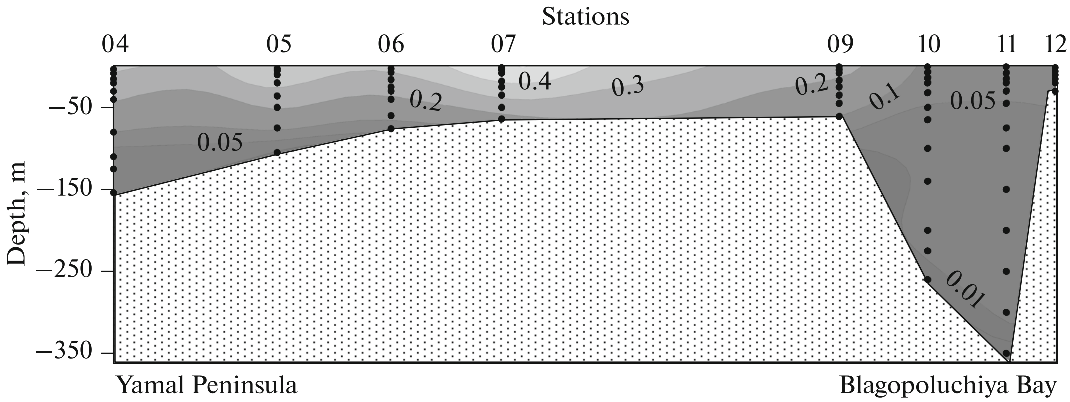

Fig. 6. Portion of freshwater component in Kara Sea water. Calculation for two-component mixing of nondesalinated water of Barents Sea with transformed water of Yenisei River (stations 27 and 28).

in waters of the Kara Sea. Since the composition of this component corresponds to the composition of transformed waters of the Yenisei River (stations 27 and 28), we can suggest that the proportion of transformed waters of this river was calculated for the profile. The results of calculations are shown in Fig. 6 and Table 1 (last column). It is evident that there is no seawater without admixture of freshwater component along entire studied section. Its concentration ranges from 5 to 10% in the main part of the water column, does not exceed 5% at depths of >100 m, and does not decrease to zero even in the deepest zones of the studied profile.

The maximum content of the freshwater component is observed in the surface water layer of the central part of the sea (67–68% in surface water of station 07, Table 1). The content of the freshwater component decreases from 67% at a depth of 4 m to 51% at a depth of 8 m, and then to 12% at a depth of 18 m. Similar sharp gradients are observed at all other stations (Table 1). Thus, our measurements and calculations support the presence of a sharp gradient of isotope parameters and salinity in the surface layer of the Kara Sea within a depth of 0–20 m and provide evidence for stable stratification of Kara Sea waters formed due to river runoff.

CONCLUSIONS

In this paper, we report the first systematic data on the distribution of isotope parameters (δD and δ^{18}O) in waters of the Kara Sea and surrounding water reservoirs of this region. The systematics allowed us to distinguish the main sources of components forming the water column and surface desalinated layer of the sea. Nondesalinated water of the Barents Sea ($S = 34.9$) is the main source for Arctic seawater in the Kara Sea; water from the Yenisei River estuary with isotope parameters transformed upon freezing is the source for freshwater. Most likely, runoff from the Ob River contributes to freshwater source, but results currently available do not allow us to support or reject this assumption. The contribution of glacial water to the formation of surface desalinated layer in the center of Kara Sea is unlikely. Probably it will be found directly near the Novaya Zemlya coast, but revision of this is a subject of further studies.

The desalinated layer of surface water in the central part of the sea is characterized by sharp vertical gradients of salinity and isotope parameters, uneven in space. We can state that, basically, the thickness of the desalinated layer is 5–10 m and it lenses out completely at a depth of 20 m. The desalinated layer is clearest in the center of the studied Yamal transect, where the proportion of the freshwater component on the sea surface can reach 67–68% (station 07). As a whole, waters of all stations in the entire depth range of the profile do not reach salinity and isotope parameters typical of unaltered waters of the Barents Sea, which leads to the conclusion on the global character of desalination in the Kara Sea.

ACKNOWLEDGMENTS

We thank head of cruise 128 of the R/V *Professor Shtokman* M.V. Flint and all members of the expedition. This study was supported by the Russian Science Foundation (project no. 14-17-00764).

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Translated by A. Bobrov