CHEMICAL COMPOSITION OF AEROSOLS ON THE NORTH OF WEST SIBERIA

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Data on chemical composition of the atmospheric aerosols (AAs) on the north of West Siberia were presented. Multielemental and ion composition of AAs as well as concentration of some organic ecotoxicants have been determined. From the determined chemical composition and its time variations (factor analysis), possible sources of aerosols particles have been determined.

The present paper is connected with studying regional peculiarities of atmospheric aerosols (AAs) in Siberia. This investigation is a part of the project on Siberian Aerosols, the aims and problems of which are outlined in Ref. 1. Therefore, since 1991 in some regions of Siberia regular observations of different characteristics of atmospheric aerosols have been performed. By now the data on the dispersed and chemical composition of AAs have already been collected from different sites of the Siberian region.[2,3,5,7] These data show noticeable differences in concentrations and chemical composition of AAs sampled in different seasons of year and at different observation sites. The maximum differences in the AA concentrations are recorded in the north regions of Siberia. At the same time, investigations on the north of Siberia are of prime interest for problem of long-range aerosol transport from industrial regions of the Northern Hemisphere to the Arctics (the problem of the Arctic haze).[8–12] Although the Arctic region is extremely sensitive to any technogenic impact, now the data on the atmospheric aerosol characteristics are very limited. Especially this is true for north territories of Russia.

In the present paper we present results of determination of AA chemical composition on the north of West Siberia obtained during expeditions in the Purov region of Yamalo–Nenetskii National District in 1995 and 1996. In Fig. 1 the map of Russia and Purovskii Region is drawn on which the sites of aerosol sampling are indicated: Regional Center – Tarko-Sale settlement and Samburg settlement. The distance between the sampling sites was about 200 km. The samples were collected by scientists of the Institute of Cytology and Genetics of the SB RAS, during expeditions that have been organized in this region already for years. In the first expedition in March 1995 samples were collected on nuclepore filters during 24 hours with volume rate of air intake of 1 m^3/h. The samples were analyzed by the method of X-ray fluorescent analysis using the synchrotron radiation (XRFA SR) at the Institute of Nuclear Physics of the SB RAS and with a scanning electronic microscope equipped with microsonde at the Antverpen University (Belgium). Results of these analyses are presented elsewhere.[3,4,6,13]

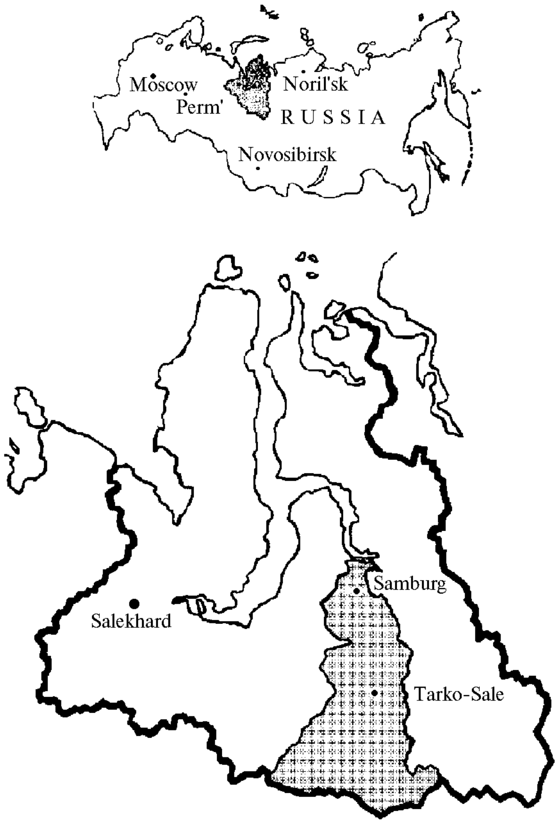

FIG. 1. Map of expeditions. At the top: Yamalo-Nenetskii Autonomous District is shown by hatching; at the bottom: Purovskii Region is shown by hatching; points indicate the sampling sites.

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During the expedition in 1996 the samples were collected on AFA-Kh filters during 24 hours with a volume rate of air intake of 13 m^3/h. The increase of the aspiration rate by more then an order of magnitude permitted us to extend the number of components determining the chemical composition of the atmospheric aerosols. In addition to multielemental composition, we determined ion aerosol composition as well as the content of some toxic organic compounds, namely, polycyclic aromatic hydrocarbons (PAHs), some organochlorine pesticides (OCPs), and polyclorinated biphenyls (PCBs). Multielemental composition of AAs as in the last expedition, was determined by the XRFA SR method. The procedure of XRFA SR and its capabilities for measuring of multielemental composition of AAs were described in Refs. 14–17. Ion composition of water soluble fraction was determined by the method of ion chromatography at the Institute of Inorganic Chemistry of the SB RAS. The procedure for ion composition determination was described in Ref. 18. Organic ecotoxicants were determined by the chromatomass-spectrometry method at the Institute of Organic Chemistry of the SB RAS. Chromatomass-spectrometric method used for the determination of the organic ecotoxicants was described in Ref. 19.

RESULTS OF MEASUREMENTS OF AA CHEMICAL COMPOSITION

Mass concentrations of different components of the AA chemical composition are given in Tables I, V, and VI.

TABLE I. Multielemental composition of aerosols on the north of West Siberia (ng/m^3).

Element	03.13.95–03.26.95						Element	12.06.96–12.25.96					
	Samburg			Tarko-Sale				Samburg			Tarko-Sale		
	$<x_i>$	σ_{gi}	n_i	$<x_i>$	σ_{gi}	n_i		$<x_i>$	σ_{gi}	n_i	$<x_i>$	σ_{gi}	n_i
Cl							Cl	102	2.6	3			
K							K	102	1.6	6	106	1.1	2
Ca	780	2.7	6	1240	1.9	5	Ca	122	2.2	8	380	2.2	4
Ti							Ti	18	2.0	5	17	1.3	4
V				160		1	V	13	2.0	4	16	1.4	3
Cr	25	1.4	3	25	8.4	3	Cr		1.0	0	28	1.2	2
Mn							Mn	6.2	2.3	5	8.1	6.0	3
Fe	54	6.3	8	53	1.4	4	Fe	130	1.4	9	190	1.7	4
Co							Co				4.7	1.1	2
Ni	19	1.1	2	26	1.7	2	Ni	3.9	2.1	4	5.2	1.7	4
Cu	16	4.1	2	5	2.7	3	Cu	6.6	2.2	9	8.9	2.0	4
Zn	22	6.9	8	11	10.5	3	Zn	27	1.8	9	18	2.0	4
Ga							Ga	0.4	1.7	3	1.0	1.2	3
Ge							Ge	0.3	1.3	3	1.0	1.0	1
As							As	1.3	2.6	4	5.5	1.0	1
Se							Se	0.3	3.2	8	0.6	2.1	4
Br	5	1.9	6	10	1.5	6	Br	2.7	2.1	9	28	2.7	4
Rb							Rb	0.3	2.6	6	0.7	1.6	3
Sr	1	3.9	2				Sr	1.2	2.7	9	0.7	2.6	3
Y							Y	0.2	1.4	2	0.2	1.3	2
Zr	2	1.2	2				Zr	0.8	1.5	9	0.2	1.2	2
Mo	5	1.1	2	4	1.0	2	Mo	0.1	1.5	7	0.4	3.8	3
Ba							Ba	72	5.5	2	31	1.0	1
Lu							Lu				0.9	1.0	1
Hf							Hf				1.6	1.5	2
W							W	0.3	3.1	2	1.9	2.3	4
Hg							Hg				3.0	1.5	3
Pb	18	2.6	8	40	2.4	5	Pb	8.3	1.7	9	41	2.7	4
Bi							Bi	0.3	2.3	4	0.5	1.5	3
U							U	0.3	2.2	8			

In Table I the mass concentrations of different AA elements are tabulated, in Tables V and VI the average daily mass concentrations of different water-soluble AA ions and some organic ecotoxicants comprised in aerosol particles are tabulated. In these tables, $<x_i>$ denotes the geometric average values of daily mass concentrations of the ith (the subscript denotes different elements, ions, or organic compounds). The symbols σ_{gi}, and n_i denote the geometric average standard deviations of the average values and the number of experiments for which the quantities $<x_i>$ and σ_{gi} were calculated. Dimensions of the parameters given in the Tables are ng/m^3 for elements and organic compounds and ng-eqv/m^3 for ions. Table II gives the relative concentrations of different elements (x_{Fe}) normalized to the iron concentration

$$x_{i\text{Fe}} = C_i / C_{\text{Fe}} , \qquad (1)$$

where C_i is the mass concentration of the ith element in the examined sample, C_{Fe} is the mass concentration of iron in the same sample.

Table III gives the enrichment factors (EFs) of different elements calculated from the formula

$$EF_i = \frac{<x_{i\text{Fe}}>_{\text{aer}}}{<x_{i\text{Fe}}>_{\text{crust}}} , \qquad (2)$$

where $<x_{i\text{Fe}}>_{\text{aer}}$ is determined from formula (1), $<x_{i\text{Fe}}>_{\text{crust}}$ is Klarckis content of the ith element in the Earth's crust.

Table IV summarizes the results of factor analysis of time variations of multielemental and ion compositions of AA as well as the content of organic ecotoxicants in the aerosol particles. At the bottom of Table IV the results of factor analysis of all compounds determined by different methods are given.

TABLE II. Distribution of relative content of elements in aerosols on the north of West Siberia (x_{Fe}).

Element	03.13.95–03.26.95						Element	12.06.96–12.25.96					
	Samburg			Tarko-Sale				Samburg			Tarko-Sale		
	$<x_{\text{Fe}}>_i$	σ_{gi}	n_i	$<x_{\text{Fe}}>_i$	σ_{gi}	n_i		$<x_{\text{Fe}}>_i$	σ_{gi}	n_i	$<x_{\text{Fe}}>_i$	σ_{gi}	n_i
Cl							Cl	1.05	2.3	3			
K							K	0.77	1.3	6	0.43	1.9	2
Ca	20	4.6	6	21	1.4	3	Ca	0.93	1.8	8	2.0	1.3	4
Ti							Ti	0.14	2.2	5	0.089	2.1	4
V				2	1	1	V	0.10	2.0	4	0.11	1.8	3
Cr	0.19	2.4	3	0.071	1	1	Cr				0.096	1.7	2
Mn							Mn	0.045	1.8	5	0.035	3.9	3
Fe	1	1	8	1	1	4	Fe	1	1	9	1	1	4
Co							Co				0.016	1.7	2
Ni	0.39	1.2	2	0.21	1	1	Ni	0.038	2.6	4	0.028	1.5	4
Cu	3.0	2.7	2	0.052	2.8	2	Cu	0.052	2.9	9	0.047	1.4	4
Zn	0.40	9.9	8	0.24	13.2	3	Zn	0.21	1.9	9	0.095	1.4	4
Ga							Ga	0.0030	1.8	3	0.0048	1.8	3
Ge							Ge	0.0022	1.5	3	0.0070	1	1
As							As	0.0086	2.2	4	0.012	1	1
Se							Se	0.0023	3.3	8	0.0032	3.1	4
Br	0.049	2.2	6	0.17	1.44	4	Br	0.021	1.8	9	0.15	3.3	4
Rb							Rb	0.0018	2.5	6	0.0032	1.2	3
Sr	0.008	6.1	2				Sr	0.0095	2.4	9	0.0036	4.6	3
Y							Y	0.0016	1.5	2	0.0020	1.6	2
Zr	0.022	1.5	2				Zr	0.0061	1.4	9	0.0009	2.1	2
Mo	0.039	1.5	2	0.05	1	1	Mo	0.0009	1.7	7	0.0019	5.3	3
Ba							Ba	0.42	6.8	2	0.071	1	1
Lu							Lu				0.0021	1	1
Hf							Hf				0.0054	1.0	2
W							W	0.0035	3.7	2	0.0099	2.6	4
Hg							Hg				0.0145	1.2	3
Pb	0.33	2.8	8	0.74	2.06	4	Pb	0.065	1.5	9	0.22	3.3	4
Bi							Bi	0.0026	2.2	4	0.0026	1.8	3
U							U	0.0029	2.0	8			

TABLE III. Enrichment factor (EF) of different elements in aerosols on the north of West Siberia.

Element	03.13. 95–03.26. 95						Element	12.06. 96–12.25. 96					
	Samburg			Tarko-Sale				Samburg			Tarko-Sale		
	$<EF_i>$	σ_{gi}	n_i	$<EF_i>$	σ_{gi}	n_i		$<EF_i>$	σ_{gi}	n_i	$<EF_i>$	σ_{gi}	n_i
1	2	3	4	5	6	7	8	9	10	11	12	13	14
Cl							Cl	220	2.3	3			
K							K	1.4	1.3	6	0.8	1.9	2
Ca	26	4.6	6	27	1.4	3	Ca	1.2	1.8	8	2.6	1.3	4
Ti							Ti	1.3	2.2	5	0.8	2.1	4
V				740	1	1	V	38	2.0	4	42	1.8	3
Cr	94	2.4	3	36	1	1	Cr				48	1.7	2
Mn							Mn	2.4	1.8	5	1.8	3.9	3
Fe	1	1	8	1	1	4	Fe	1	1	9	1	1	4
Co							Co				34	1.7	2
Ni	280	1.2	2	150	1	1	Ni	27	2.6	4	20	1.5	4
Cu	3000	2.7	2	52	2.7	2	Cu	52	2.9	9	47	1.4	4
Zn	270	10	8	160	13	3	Zn	140	1.9	9	63	1.4	4
Ga							Ga	8.6	1.8	3	14	1.9	3
Ge							Ge	49	1.5	3	160	1	1
As							As	190	2.2	4	270	1	1
Se							Se	1800	3.3	8	2400	3.1	4
Br	800	2.2	6	2800	1.4	4	Br	340	1.8	9	2400	3.3	4
Rb							Rb	0.7	2.5	6	1.3	1.2	3
Sr	1	6.1	2				Sr	1.4	2.4	9	0.5	4.6	3
Y							Y	2.2	1.5	2	2.7	1.6	2
Zr	6	1.5	2				Zr	1.7	1.4	9	0.2	2.1	3
Mo	1300	1.5	2	1700	1	1	Mo	30	1.7	7	64	5.4	3
Ba							Ba	42	6.8	2	7	1	1
Hf							Hf				67.4	1.0	2
W							W	120	3.7	2	280	2.6	4
Hg							Hg				11000	1.2	3
Pb	1100	2.8	8	2600	2.1	4	Pb	220	1.5	9	740	3.3	4
Bi							Bi	630	2.2	4	630	1.8	3
U							U	52	2.0	8			

TABLE IV. Results of factor analysis of AA chemical composition on the north of West Siberia.

Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
		Multielemental composition		
Ca, Fe, Zn	Se, Br, Pb	Ni, Cu	–	–
		Ion composition		
$Ca^{2+} + Mg^{2+}$, Na^+, Cl^-	NH_4^+, K^+, SO_4^{2-}	H^+	–	–
		PAHs, OCPs, PCBs		
Fluoranthene,	acenaphthalene	naphtalene	–	–
pyrene, chrysene,	acenaphthene	–	–	–
benz(*a*)anthracene,	anthracene	–	–	–
benzo(*b*)pyrene,	–	–	–	–
dibenzo(*a,h*)anthracene,	–	–	–	–
benzo(*g,h,i*)perylene,	–	–	–	–
γ-HCCH, DDT	–	–	–	–
		Full chemical analysis		
Ca, Fe, $Ca^{2+} + Mg^{2+}$, Na^+, Cl^-, γ-HCCH	Se, Br, Pb, fluoranthene, pyrene, benz(*a*)anthracene, benzo(*g,h,i*)perylene, benzo(*b*)pyrene	Zn, NH_4^+, K^+, SO_4^{2-}, acenaphthene, DDT	H^+, F^-, naphtalene	PCB

TABLE V. Ion composition of water-soluble fraction of atmospheric aerosols on the north of West Siberia (December 1996).

Components	NH_4^+	$Ca^{2+} + Mg^{2+}$	Na^+	K^+	H^+	Cat	F^-	Cl^-	NO_3^-	SO_4^-	An
Samburg											
$<x_i>$	7.1	7.0	3.1	0.9	0.4	20.9	1.2	0.7	1.5	19.0	23.8
σ_{gi}	3.9	1.4	1.7	2.5	1.6	2.0	1.3	1.5	2.8	2.3	2.1
n_i	9	9	9	9	9	9	9	9	9	9	9
%eqv	33.7	33.5	14.7	4.6	2.1	100	5.4	3.2	6.6	79.9	100
ng/m^3	127.4	119.7	71.0	37.6	0.4	356.2	24.4	27.0	96.7	912.9	1061
%	35.7	33.6	19.9	10.5	0.1	100	2.3	2.5	9.1	86.0	100
Tarko-Sale											
$<x_i>$	8.1	9.6	10.7	1.8	0.7	32.4	1.2	2.7	2.6	19.5	29.9
σ_{gi}	1.1	1.6	1.7	1.4	1.8	1.3	1.5	3.2	2.3	1.7	1.5
n_i	4	4	4	4	4	4	4	4	4	4	4
%eqv	25.1	29.7	33.2	5.5	2.2	100	4	8.9	8.7	65.0	100
ng/m^3	146	163	248	70	1	628	23	94	162	933	1213
%	23.3	26.0	39.4	11.1	0.13	100	1.9	7.8	13.4	76.9	100

ANALYSIS OF THE DATA. Multielemental analysis

The data in Table I show that the increase of the volume pumping rate leads to the increase of the number of elements simultaneously detected in AAs from 12 to 30. Although the mass concentrations of different elements recorded during the expedition in 1996 in some cases differed strongly from those measured in 1995, at present it is difficult to indicate possible reasons for these differences. However, in most cases within the limits of the spread of the available experimental data the mass concentrations of different elements in aerosol samples collected in Tarko-Sale and Samburg are close to each other. For significant number of elements, the average mass concentration in samples collected in Tarko-Sale were 2–3 times larger than in samples, collected in Samburg. For Pb and Br the concentrations in Tarko-Sale exceeded those in Samburg 5–10 times. In no one of 5 samples of AAs from Tarko-Sale we detected noticeable quantity of U. At the same time in 9 of 10 samples from Samburg U was detected in measurable amounts.

From Table II it can be seen that except for Br, Sr, Zr, Ba, W, and Pb the relative concentrations of elements $<x_{Fe}>$ are within the limits of spread of the experimental data. This indicates the similarity of multielemental composition in this region. For Br and Pb, $<x_{Fe}>$ in Samburg were 3–6 times lower than in Tarko-Sale. To the contrary, the aerosols in Samburg were enriched from 2 to 8 times with Sr, Zr, and Ba in comparison with their contents in aerosol particles of Tarko-Sale.

Such elements as V, Cr, Ca, Ni, Ca, Zn, Ge, As, Se, Br, Mo, Hf, W, Pb, Bi, and U (as can be seen from Table III) have significant enrichment factors EF > 20. This likely means that in the Purov Region the aerosol particles are transported, sources of which are enterprises of non-ferrous metallurgy and motor transport. In Tarko-Sale, in addition to the increase of Pb and Br concentrations in comparison with Samburg the concentrations of these elements were highly correlated with the correlation coefficient close to 100%. Conclusion about close correlation between these elements was also confirmed by the results of factor analysis (Table VI). For Samburg the increase of content of Sr and U in aerosol particles was accompanied by fairly high correlation between variations of their concentrations. A correlation between Pb and Br content in aerosol particles in Tarko-Sale may be due to technogenic impact of local level caused by heavier traffic. High value of EF for U and EF for Sr close to unity together with close correlation between mass concentrations of these elements in Samburg may, in particular, be explained by their soil-erosive origin. It is likely that in addition to mineral fraction, soil-erosive particles comprise plant residues, which may be polluted by emissions comprising U. Therefore, in future investigations it is necessary to consider these facts.

Ion composition

In Tarko-Sale and Samburg, as can be seen from Table V, predominant anion is sulfate. The content of NO_3^-, Cl^-, and F^- in water-soluble AA fraction is less by an order of magnitude. In Tarko-Sale: cation fraction is equally represented by NH_4^+, $Ca^{2+} + Mg^{2+}$, and Na^+. Approximately by an order of magnitude lower is the K^+ content. Total percentage of water-soluble ion fraction in Tarko-Sale and in Samburg is, on average, about 40%. In winter this is within the limits of variations characterizing the Siberian Region.[5] The total concentrations of anion and cation compounds of water-soluble AA fraction were closely correlated. This is illustrated by Fig. 2.

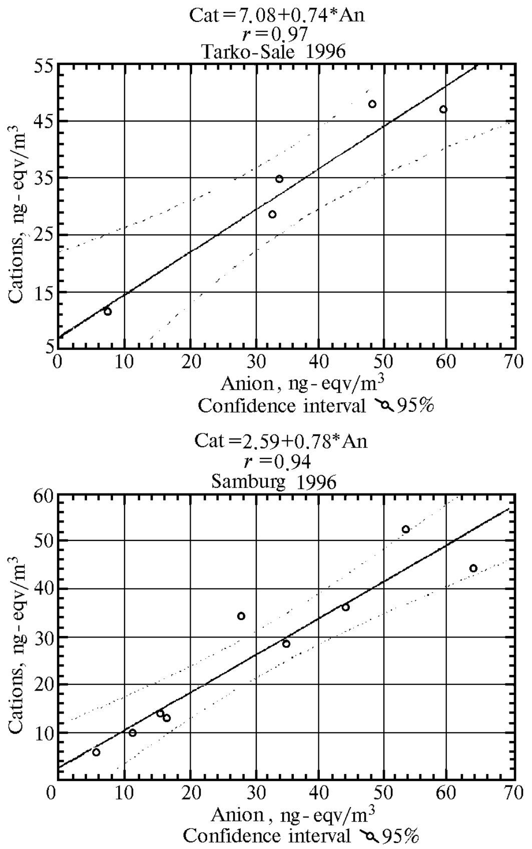

FIG. 2. Correlation between the total concentrations of anions and cations. Cat *is for the total concentration of cations:* An *is for the total concentration of anions;* ∘ *are experimental quantities; line is correlation dependence.*

Content of organic ecotoxicants

The results of mass-spectrometric determination of PAHs, OFPs, and PCBs given in Table IV show that up to 18 organic ecotoxicants of different types are comprised in AAs of investigated region. In connection with the fact that such measurements were made by us for the first time we restricted ourselves only factor analysis, results of which are given in Table IV.

As can be seen from Table VI, each component of the AA chemical composition comprises up to 3 different factors. At the bottom of this Table, the results are given of factor analysis of all data on AA chemical composition on the north of West Siberia. It can be seen that total chemical composition is determined already by 5 factors.

CONCLUSIONS

1. For the first time, the data on the AA chemical composition on the north of West Siberia have been obtained.

2. Different chemical compounds of AAs in this region have been analyzed. This has allowed us possible sources of aerosol particles and their regional peculiarities.

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*TABLE VI. Contents of polycyclic aromatic hydrocarbons, organic fluoride pesticides, and polyclorinated biphenyls in the atmospheric aerosols on the north of West Siberia (*ng/m^3*)*

Compound	12.06.96–12.25.96					
	Samburg			Tarko-Sale		
	$<x_i>$	σ_{gi}	n_i	$<x_i>$	σ_{gi}	n_i
Acenaphthalene	0.0086	2.09	9	0.0102	2.09	3
Acenaphthene	0.0061	2.09	9	0.0099	1.40	3
Naphthalene	0.055	1.29	9	0.057	1.37	3
Fluorene	0.031	2.31	9	0.031	2.08	3
Phenanthrene	0.084	3.24	9	0.030	10.3	4
Anthracene	0.036	3.03	8	0.0069	6.25	4
Fluoranthene	0.048	3.83	9	0.032	5.69	4
Pyrene	0.026	4.06	9	0.021	4.34	4
Benz(a)anthracene	0.012	2.24	8	0.005	3.97	4
Chrysene	0.046	1.84	7	0.011	6.79	4
	Samburg + Tarko-Sale					
Benzo(b)pyrene	0.043	2.00	12	–	–	–
Benzo(a) pyrene	0.036	2.63	9	–	–	–
Dibenz(a,h)anthracene	0.0069	1.74	11	–	–	–
Benzo(g,h,i)perylene	0.10	2.41	12	–	–	–
γ-HCCH	0.032	1.59	12	–	–	–
DDE	0.0055	2.93	11	–	–	–
DDT	0.0063	2.59	13	–	–	–
PCB	0.044	4.28	13	–	–	–

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