AGU PUBLICATIONS

Water Resources Research

Supporting Information for

Microbial sulfate reduction (MSR) as nature-based solution (NBS) to mine drainage: Contrasting spatio-temporal conditions in northern Europe

Sandra Fischer[1*], Carl-Magnus Mörth[2], Gunhild Rosqvist[1], Sergey R. Chalov[3], Vasiliy Efimov[3], Jerker Jarsjö[1]

[1] Department of Physical Geography and the Bolin Centre for Climate Research, Stockholm University, SE-106 91 Stockholm, Sweden

[2] Department of Geological Sciences, Stockholm University, SE-106 91 Stockholm, Sweden

[3] Faculty of Geography, M.V. Lomonosov Moscow State University Leninskie Gory 1, 119991 Moscow, Russia

* Correspondence: sandra.fischer@natgeo.su.se

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Introduction

This supporting information provides details and sources to calculation of climatic variables as well as the complete record from the regional data synthesis used in the isotopic mixing scheme. It also contains output results from the Monte Carlo simulation and results from the stream water measurement campaign in Khibiny 2017.

S1. Temperature and precipitation data for the Khibiny massif

Temperature and precipitation data were taken from the CRU TS 4.02 grid-box data at 0.5° resolution produced by the Climatic Research Unit (CRU) at the University of East Anglia (available at https://crudata.uea.ac.uk/cru/data/hrg/ and described by Harris et al. (2014). The cells 67.75 N, 33.25–34.25 E (covering the Khibiny massif) had monthly data from 1901 to 2017 and was used for deriving long-term average temperature and precipitation.

S2. Details on regional parameter datasets for the Khibiny catchments

S2.1. Sulfur concentration in atmospheric deposition (c_{dep})

The sulfur concentration (S_{SO_4}) from atmospheric deposition (c_{dep}) was derived from a regional synthesis on previously published monitoring data (Arctic Monitoring and Assessment Programme, 2006; Äyräs et al., 1995; EBAS, 2021; Malinovsky et al., 2002; Reimann et al., 1997) from 13 stations in northeastern Fennoscandia (Fig. S1) that are detailed in Table S1.

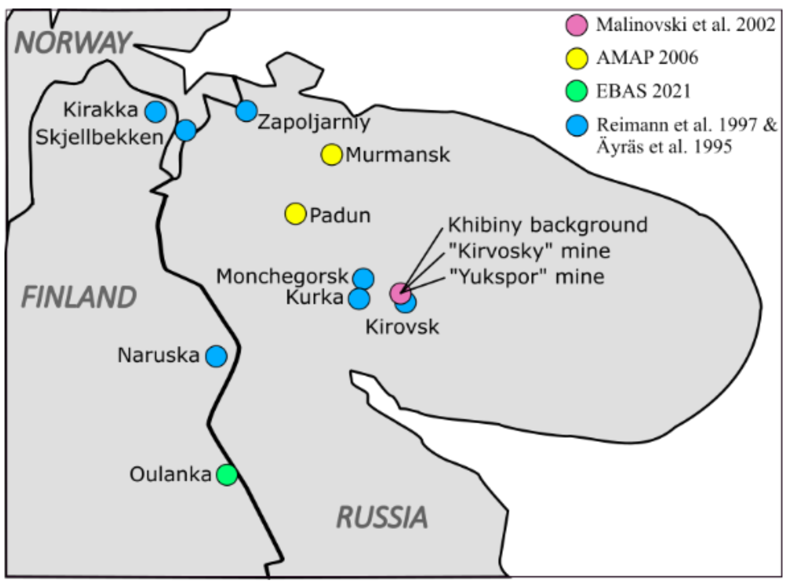

Figure S1. Map over stations used to derive average sulfur concentration (S_{SO_4}) in runoff originating from atmospheric deposition (c_{dep}) for the Khibiny catchments.

Table S1. Station details for the derivation of sulfur concentration in atmospheric deposition (c_{dep}).

Location	E coordinate	N coordinate	Type	Time period	Source
Khibiny background	33.63845	67.66292	Snow	Mar-Apr 1998-1999	Malinovski et al. 2002
"Kirovsky" mine	33.73572	67.66259			
"Yukspor" mine	33.82030	67.63780			
Padun	31.80000	68.60000	Rain	1991-2004	AMAP 2006
Murmansk	33.08472	68.94759			
Oulanka	29.40167	66.32028	Rain	1990-2017	EBAS 2021
Zapoljarnij	31.06351	69.44975	Rain & Snow	May-Sept 1994	Reimann et al. 1997 & Äyräs et al. 1995
Monchegorsk	32.91323	67.84171			
Kirvosk	33.81488	67.54715			
Kurka	32.83729	67.69005			
Skjellbekken	29.45699	69.35677			
Kirakka	28.86278	69.58651			
Naruska	29.36816	67.36195			

The c_{dep}-dataset was assumed to approximate a lognormal distribution (Fig. S2a) based on their log-transformed fit to the normal distribution (Fig. S2b) and inset quantile-quantile plot (QQ-plot). QQ-plots compare the quantiles of the sample data (y-axis) to the theoretical quantiles of a normal distribution (x-axis) with a red line marking the 1:1 relationship.

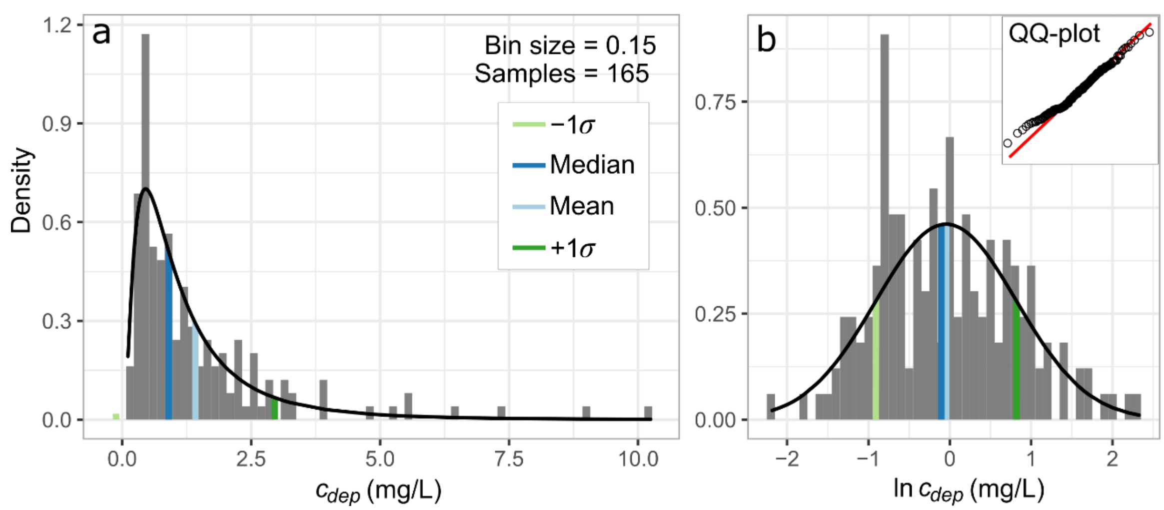

Figure S2. (a) Distribution of annual average sulfur concentration (S_{SO_4}) in runoff originating from atmospheric deposition (c_{dep}) based on 165 observations in northeastern Fennoscandia. The density distribution (black curve in panel a) was calculated with *dlnorm* in R based on the parameter mean and standard deviation.

S2.2. Synthesis of sulfur isotopic value of atmospheric deposition (δ_{dep})

The observed values of δ_{dep} (Fig. S3) were assumed approximating a normal distribution as seen in their respective inset quantile-quantile plot (QQ-plot). δ_{dep}-values represent the isotopic composition in epiphytic moss samples collected in 1994 in the Kola Peninsula (de Caritat et al., 1997).

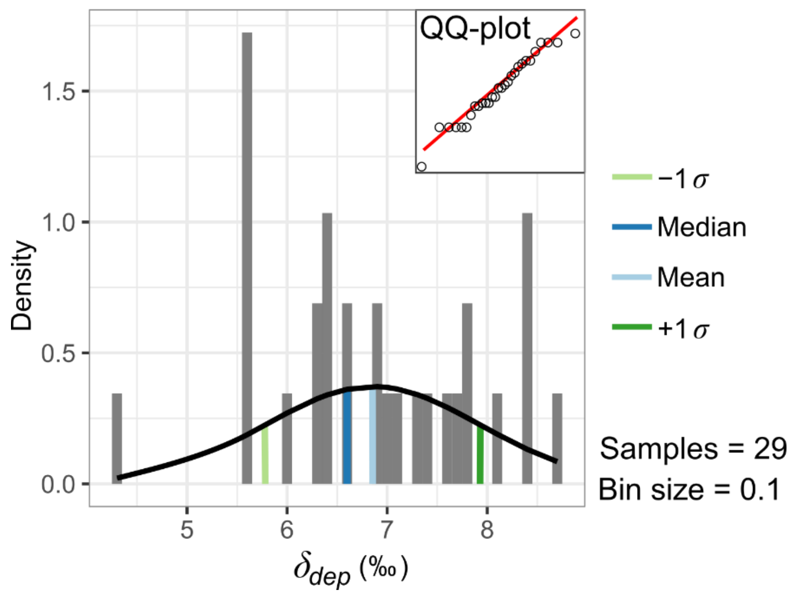

Figure S3. Distribution of sulfur isotopic values in atmospheric deposition ($\delta^{34}S_{SO_4}$) from de Caritat et al. (1997). The density distribution (black curve) was calculated with *dlnorm* in R based on the parameter mean and standard deviation.

S3. Stream water sulfur concentrations (c_{stream}) and isotopic values (δ_{stream}) for Imetjoki

Table S2. Measured sulfur concentration (total S; c_{stream}) and sulfur isotopic value ($\delta^{34}S_{SO_4}$; δ_{stream}) within the Imetjoki catchment in 2017 from the spring (May; this study) and the summer (August; from Fischer et al., 2022)). No measurement is marked with "-".

			c_{stream} (mg/L)		δ_{stream} (‰)	
Name	**ID**	**Zone**	**Spring**	**Summer**	**Spring**	**Summer**
Upstream Lake	1	Upstream	-	0.14	-	16.67
Upstr. Dagny mine	2	Upstream	-	0.50	-	6.87
Northern Lake	3	Mine	1.05	1.74	3.08	5.54
Imetjärvi inlet	4	Mine	0.51	0.50	5.17	7.97
Maria Lake	5	Mine	1.05	1.68	3.38	3.60
Imetjärvi outlet	6	Mine	0.66	0.82	5.25	5.97
Max mine	7	Mine	-	4.04	-	2.11
Industrial area 1	8	Mine	47.54	17.32	2.58	3.60
Industrial area 2	9	Mine	20.53	22.77	5.70	6.02
Downstream 1	10	Downstream	1.03	1.76	5.04	5.78
Downstream 1b	11	Downstream	-	1.65	-	5.78
Downstream 2	12	Downstream	0.65	0.61	6.89	8.08
Ref. Nietsajoki	13	Reference	0.37	0.40	10.52	12.40
Downstr. Fredrik mine	14	Downstream	2.40	-	4.35	-
Ref. Lina River	15	Reference	1.86	-	4.71	-

S4. Details on the calculated microbial sulfate reduction (MSR) from the Monte Carlo simulations

Table S3–5 corresponds to Figure 3 in the main text.

Table S3. Calculated MSR (%) for the Imetjoki catchment in the spring 2017 showing summary statistics: minimum, first quantile (Q1), median, mean, third quantile (Q3), maximum, and standard deviation (StD). Negative values are interpreted as 0% MSR. Catchment-scale MSR is calculated both for all spring-observations as well as for the selected IDs (*) that were sampled both in the spring and the summer.

Name	ID	Min	Q1	Median	Mean	Q3	Max	StD
Upstream Lake	1	-	-	-	-	-	-	-
Upstr. Dagny mine	2	-	-	-	-	-	-	-
Northern Lake	3*	-70.6	-2.1	5.3	10.9	14.6	100.0	23.6
Imetjärvi inlet	4*	-75.9	-1.0	4.4	13.7	17.0	100.0	25.2
Maria Lake	5*	-71.2	-0.7	7.1	13.1	16.9	99.9	23.9
Imetjärvi outlet	6*	-53.2	1.6	10.0	17.9	23.2	100.0	25.0
Max mine	7	-	-	-	-	-	-	-
Industrial area 1	8*	-4.8	11.6	17.4	26.2	31.9	100.0	22.5
Industrial area 2	9*	7.6	23.1	31.7	40.3	51.5	100.0	22.8
Downstream 1	10*	-48.8	8.6	16.0	23.8	30.5	100.0	24.3
Downstream 1b	11	-	-	-	-	-	-	-
Downstream 2	12*	-30.4	10.4	18.8	27.3	35.1	100.0	24.2
Downstr. Fredrik mine	14	-19.3	13.8	20.4	29.3	36.8	100.0	23.2
Catchment – all		**-75.9**	**5.7**	**15.1**	**22.3**	**30.5**	**100.0**	**25.5**
Catchment – selected*		**-75.9**	**4.6**	**14.1**	**21.4**	**29.5**	**100.0**	**25.6**
Ref. Nietsajoki	13	10.1	21.0	29.0	36.9	45.4	100.0	22.0
Ref. Lina River	15	-35.8	13.7	20.5	29.2	36.7	100.0	23.5

Table S4. Calculated MSR (%) for the Imetjoki catchment in the summer 2017 showing summary statistics: minimum, first quantile (Q1), median, mean, third quantile (Q3), maximum, and standard deviation (StD). Negative values are interpreted as 0% MSR. Catchment-scale MSR is calculated both for all summer-observations as well as for the selected IDs (*) that were sampled both in the spring and the summer. Summary statistics for both seasons combined are also presented.

Name	ID	Min	Q1	Median	Mean	Q3	Max	StD
Upstream Lake	1	28.2	39.5	47.9	53.4	62.1	100.0	17.8
Upstr. Dagny mine	2	-23.2	7.8	14.5	23.4	28.7	100.0	23.5
Northern Lake	3*	-11.6	16.7	24.1	32.8	41.3	100.0	23.5
Imetjärvi inlet	4*	-3.6	12.7	20.2	28.8	35.9	100.0	23.3
Maria Lake	5*	-45.1	7.8	13.5	21.2	25.9	100.0	22.8
Imetjärvi outlet	6*	-27.6	9.5	17.6	25.5	32.9	100.0	24.2
Max mine	7	-51.4	6.3	11.0	18.4	21.1	100.0	21.1
Industrial area 1	8*	0.2	15.3	22.0	30.8	38.0	100.0	22.5
Industrial area 2	9*	9.7	24.5	33.6	41.5	52.2	100.0	22.6
Downstream 1	10*	-9.7	17.9	25.4	34.1	42.7	100.0	23.3
Downstream 1b	11	-11.5	17.4	24.8	33.5	42.1	100.0	23.4
Downstream 2	12*	-2.6	15.6	23.8	32.2	41.6	100.0	23.7
Downstr. Fredrik mine	14	-	-	-	-	-	-	-
Catchment – all		**-51.4**	**13.3**	**22.5**	**30.5**	**40.1**	**100.0**	**24.3**
Catchment – selected*		**-45.1**	**14.5**	**22.9**	**30.8**	**39.7**	**100.0**	**23.9**
Ref. Nietsajoki	13	16.2	29.0	38.3	45.4	56.3	100.0	21.6
Ref. Lina River	15	-	-	-	-	-	-	-
Both seasons								
Catchment – all		**-75.9**	**10.0**	**19.4**	**26.8**	**36.9**	**100.0**	**25.2**
Catchment – selected*		**-75.9**	**9.5**	**19.0**	**26.0**	**35.0**	**100.0**	**25.2**

Table S5. Calculated MSR (%) for the Khibiny catchments in 2017 showing summary statistics: minimum, first quantile (Q1), median, mean, third quantile (Q3), maximum, and standard deviation (StD). Negative values are interpreted as 0% MSR. Catchment-scale MSR are presented for the Yuksporiok and Vuonnemiok catchments.

Name	ID	Min	Q1	Median	Mean	Q3	Max	StD
Gakman 1	16	-89.8	3.4	8.6	19.6	22.8	100.0	27.6
Gakman 2	17	-89.0	1.7	7.0	18.1	21.4	100.0	28.6
Gakman 3	18	-86.6	2.1	7.5	18.4	22.1	100.0	27.9
Yuksporiok 1	19	-87.1	1.4	7.5	18.6	24.2	100.0	28.6
Yuksporiok 2	20	-99.6	1.5	5.9	10.6	12.7	100.0	20.2
Yuksporiok 3	21	-93.1	-0.5	4.5	8.3	10.8	100.0	20.3
Catchment – all		**-99.6**	**1.6**	**6.6**	**15.5**	**17.8**	**100.0**	**26.1**
Belaya River	22	-65.8	7.6	14.2	20.7	26.3	100.0	24.0
Vuonnemiok 1	23	-84.7	3.3	8.8	19.9	24.2	100.0	27.7
Vuonnemiok 2	24	-99.8	-4.2	0.6	1.7	5.5	99.6	18.1
Vuonnemiok 3	25	-98.5	-6.4	-1.2	-1.0	3.8	100.0	18.4
Catchment – all		**-99.8**	**-3.1**	**2.3**	**6.6**	**9.1**	**100.0**	**23.6**
Umba River	26	-87.6	-11.8	-1.2	1.3	8.6	100.0	26.7
Both catchments*								
Catchment – all		**-99.8**	**-0.2**	**5.2**	**12.5**	**14.5**	**100.0**	**25.6**

*IDs 16-21, 23-25

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S5. Results from field measurement campaign in Khibiny in August 2017

Table S6. Results in base chemistry of surface water samples from the 2017 measurement campaign at Khibiny. Coordinates are in WGS 1984.

Basin		Sampling zone	ID	Name	Latitude	Longitude	Temp. (° C)	pH (-)	EC (µS cm-1)	Alkalinity (ppm CaCO3)	Turbidity (NTU)	$\delta^{34}S_{SO_4}$ (‰)	S (mg/L)
Belaya	Gakman	Upstream	16	Gakman 1	67.66833	33.83322	5.8	6.8	1	0	2.61	7.95	0.42
		Upstream	17	Gakman 2	67.66367	33.81176	5.8	6.8	4	1	0.34	7.60	0.45
		Upstream	18	Gakman 3	67.65054	33.78993	6.2	7.0	8	-	3.34	7.67	0.48
	Yuksporiok	Upstream	19	Yuksporiok 1	67.6531	33.8251	4.2	7.3	10	8	1.54	7.40	0.62
		Mine	20	Yuksporiok 2	67.64291	33.78008	6.0	10.6	457	126	154	1.04	15.4
		Mine	21	Yuksporiok 3	67.63295	33.71846	4.9	10.2	323	88	121	1.02	11.2
		Downstream	22	Belaya River	67.58552	33.58551	9.9	9.2	161	44	10.1	3.36	5.58
Umba	Vuonnemiok	Upstream	23	Vuonnemiok 1	67.6648	33.86147	3.8	7.4	5	11	1.62	7.90	0.47
		Mine	24	Vuonnemiok 2	67.60687	34.16578	9.6	8.4	392	59	5.04	0.08	17.7
		Downstream	25	Vuonnemiok 3	67.61901	34.18064	5.3	8.0	197	31	4.81	0.07	11.3
		Downstream	26	Umba River	67.50352	34.22515	10.4	7.8	57	20	3.79	2.36	2.28

Table S7. Results of total element concentrations in surface water samples from the 2017 measurement campaign at Khibiny. All concentrations are in µg/L and represent the triplicate median.

Basin		Sampling zone	ID	Name	Al	Ba	Cr	Cu	DOC	Fe	Mn	P	SO_4	Sr	Ti	Zn
Belaya	Gakman	Upstream	16	Gakman 1	40.0	0.83	0.16	0.86	1180	8.90	0.58	7.33	1040	22.6	0.96	0.73
Belaya	Gakman	Upstream	17	Gakman 2	35.8	0.66	0.15	0.48	1080	7.31	0.40	5.05	1240	37.2	0.82	0.70
Belaya	Gakman	Upstream	18	Gakman 3	234	1.75	0.31	0.51	1000	84.3	2.71	11.9	1230	42.4	6.18	1.01
Belaya	Yuksporiok	Upstream	19	Yuksporiok 1	13.0	0.64	0.13	0.34	1130	4.15	0.14	6.25	1740	49.7	0.61	0.54
Belaya	Yuksporiok	Mine	20	Yuksporiok 2	4950	114	2.90	23.2	3070	2200	97.6	8840	50800	944	457	24.2
Belaya	Yuksporiok	Mine	21	Yuksporiok 3	3680	78.3	2.01	16.5	2200	1363	70.9	5908	35700	730	299	14.3
Belaya		Downstream	22	Belaya River	220	4.21	0.71	2.04	2420	59.0	3.72	409	17600	163	6.26	1.59
Umba	Vuonnemiok	Upstream	23	Vuonnemiok 1	21.2	0.67	0.11	0.18	1000	3.79	0.08	4.12	1220	55.2	0.69	0.56
Umba	Vuonnemiok	Mine	24	Vuonnemiok 2	201	8.59	0.29	3.70	2340	74.5	4.31	49.1	57600	331	10.6	1.52
Umba	Vuonnemiok	Downstream	25	Vuonnemiok 3	108	4.20	0.20	1.80	1730	43.9	2.04	145	36500	188	4.20	0.73
Umba		Downstream	26	Umba River	32.0	1.74	0.15	0.71	3190	21.0	1.86	7.02	7270	77.5	1.19	0.53