Spatial distribution and environmental assessment of heavy metals in the surface sediments of Kongsfjorden, Svalbard

Neelu Singh[1*], Rajan Sivaramakrishnan[1], Shabnam Choudhary[2], Krishnaiah Chikkamadaiah[3]

[1]*National Centre for Antarctic and Ocean Research, Headland Sada, Goa 403 804, India*
[2]*Department of Marine Science, Goa University, Goa 403 206, India*
[3]*Department of Marine Geology, Mangalore University, Mangalore 574 199, Karnataka, India*

Abstract

Spatial distribution and environmental assessment of heavy metals of the surface sediment samples collected from the Kongsfjorden system, Svalbard during summer months of 2011, 2012 and 2013 were studied using grain size, organic carbon and metal concentrations (Mn, Cr, Zn, Co, Cu, Ti and Pb). The Inner fjord was blanketed by clay-rich sediments while clayey silt or silty clay was found in the Outer fjord. High sedimentation rate and the water column turbulence resulted in the poor preservation of the organic matter in the Inner fjord while high concentration of TOC in the Outer fjord sediments indicated high primary productivity. Further, in the Outer fjord, the concentration of metals like Mn, Cr, Zn, Co, Cu and Pb were found to be higher near the mouth of the fjord while the lowest concentrations were at its distal end. The Inner fjord was also characterized by a concurrent enrichment of these heavy metals near the glacier outlets with the lowest values occurring near the shallow sill separating it from the Outer fjord. The significant positive correlation of all the heavy metals except Pb in the outer and inner part of the fjord, among themselves and also with Ti corroborated their terrigenous source possibly derived from the rocks present in the catchment area through the glacier melt waters. While, uniform negative correlation exhibited by Pb with all the other elements may point to its source from elsewhere, indicating its source to be anthropogenic.

Key words: Kongsfjorden, anthropogenic, enrichment factor, grain size, TOC (Total Organic Carbon)

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[*]Corresponding author: N. Singh <neelu.singh0387@gmail.com>
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Introduction

Presence of heavy metals such as cadmium, chromium, copper, mercury, lead, zinc, arsenic, boron *etc*. in the aquatic environment in concentrations above their natural background values can be a major cause of concern because of the risks they pose to humans and ecosystem on account of their toxicity. Unlike organic contaminants which are oxidised by microbially-mediated reactions in the water column or at or below the sediment-water interface, these metals do not undergo chemical or biological degradation leading thereby to an increase in their abundance over time, and subsequent bio-accumulation and bio-magnification through different trophic levels (UNEP/GPA 2004 - [WP 3]). Furthermore, the accumulation of these heavy metals in the sediments can ultimately lead to a situation where in the sediments themselves become a source.

Although still pristine compared to the rest of the globe, there is a growing concern about the increasing heavy metal fluxes to the Polar Regions, especially the Arctic. Many Arctic inland ecosystems have been documented to contain relatively high concentrations of heavy metals (*e.g.* Skotvold et al. 1997, AMAP 2005 - [1], Bidleman et al. 2005 and references therein). These may be further enriched by atmospheric deposition from distant anthropogenic sources, considering that long-range transport has been suggested to be the most important mechanism whereby many heavy metals reach the Arctic (Barrie 1986, 1992; AMAP 2005 - [1]). Heavy metals can also be transported to the Arctic by rivers and ocean currents. In addition, on a local scale, heavy metal inputs from glacier melt water and other terrestrial sources such as from the rocks and soil of the hinterland are likely to enter inland aquatic systems and ultimately reach the sediments and the ecosystems. Contributions from the latter sources may not be very large on a year-to-year scale; however, there is a real possibility that over a period of time their concentrations particularly in the sediments and ecosystems of enclosed/semi-enclosed aquatic systems can attain toxic levels through the processes of accumulation, bio-concentration and bio-magnification. The rates of accumulation of heavy metals in the sediments and in the ecosystem are characteristically different and would depend to a great extent on the environment of deposition (lakes, fjords, glacial ice *etc*.) and the metal bio-availability respectively. Thus whereas establishing the baselines for the various contaminants in the Arctic region is essential, it is equally important to realise that the baseline data so generated from a specific environment or a specific ecosystem would be unique for that environment or the ecosystem.

The present study focuses on heavy metal (Co, Cu, Pb, Cr, Zn and Mn) concentration in the surface sediments of the Kongsfjorden adjacent to the International Arctic Research Facilities at Ny-Ålesund (78° 55' 30" N, 11° 55' 20" E) on the Spitsbergen island of Svalbard Archipelago (Fig. 1). The investigations form a part of an ongoing long-term program of monitoring of the fjord as a reference site for Arctic climate variability, initiated by Indian scientists in 2009.

Although several studies have been carried out at Ny-Ålesund on the heavy metal concentrations in the atmosphere, on the lake sediments and biota (*e.g.* Berg et al. 2004, Sun et al. 2006, Evenset et al. 2007, Ahn et al. 2004, 2009, Sagerup et al. 2009, Jæger et al. 2009, Jiang et al. 2011, Samecka-Cymerman et al. 2011 and references therein), investigations on the sediments of the fjords in Svalbard and especially on Kongsfjorden, have been relatively few (*e.g.* Zaborska et al. 2006a,b; Grotti et al. 2013, Lu et al. 2013, Bazzano et al. 2014, Ardini et al. 2016). This is surprising considering the proximity of Kongsfjorden to Ny-

Ålesund, "the northernmost settlement in the world" and that it has been listed among the European flagship sites of biodiversity. Therefore, there is an imperative need for systematic monitoring of this fjord as well as for the establishment of proper baseline data on trace elements from an environmental point of view. Furthermore, Kongsfjorden occupies a unique niche in the Arctic climate system, maintaining a delicate balance between the advection of the warm, saline Atlantic water at its mouth and the glacial runoff from the large glaciers at its head. Short-term variations in this dual Atlantic/Arctic inputs marked by pronounced hydrographical and biotic changes in the fjord have been suggested to be climate-modulated (Hop et al. 2002, Svendsen et al. 2002, Cottier et al. 2005, and references therein; David et al. 2015, Bhaskar et al. 2016, David et al. 2016). Such variabilities can be expected to influence the heavy metal flux to the sediments from the ocean as well as from the catchment areas of the glaciers. This study thus has a three-fold objective: (i) augmenting the existing database on heavy metals in Kongsfjorden; (ii) examining the spatial trends in variability of the heavy metals in the surficial sediments of the fjord in terms of the anthropogenic influence if any, on the observed levels of contamination; and (iii) assessing the spatial and temporal variations in the concentrations of heavy metals in terms of the possible variations in their relative contributions from the Atlantic and the glacial melt waters.

Towards the above objectives, surface samples were collected and analysed from 12 pre-fixed stations along and across the fjord during July 2011, July 2012 and August 2013 (Table 1, Fig. 2). In addition to Co, Cu, Pb, Cr, Zn and Mn, the concentration of Ti in the samples was determined as a proxy parameter to characterize the terrigenous input to the fjord, as well as a reference element for estimating the enrichment factor of the heavy metal contamination in the sediments (*c.f.* Zabel et al. 1999, Wei et al. 2003, Lu et al. 2012). The weight % organic carbon and the grain size characteristics of the sediments were also determined to provide constraints on the concentration and distribution pattern of the heavy metals in the fjord.

Ny-Ålesund is host to fifteen permanent research stations run by institutions from ten countries (Kings Bay 2016 - [WP 2]). In addition, there are several institutions which carry out research activities without having their own stations. The population varies from a year-round 30 to 35, peaking to over 120 during the summer (Kings Bay 2016 - [WP 2]). But more importantly, several tourist cruise ships call at the harbour during the summer, with the footfalls ranging from about 25,000 during 2013 to a high of about 40,000 the year before (Kings Bay 2016 - [WP 2]). Therefore, the emissions from the cruise ships as well as the tourist traffic can be expected to add to the trace metal fluxes to the fjord. Our studies however, indicate that despite its proximity to the research base and being a summer tourist hub, the anthropogenic impact of heavy metals on the fjord sediments is negligible. The average concentrations of the various heavy metals are comparable to, or in some instances less than the concentrations reported from other Arctic locales (*c.f.* Naidu et al. 1997, AMAP Assessment 2005 - [1], Lu et al. 2013). Nonetheless, our studies show marked spatial and temporal variations in the concentrations of the heavy metals which are attributable to inter-annual variations in the supply of sediments from the Atlantic and the glacial meltwaters.

Study Area

Kongsfjorden is located along the western coast of Spitsbergen, the largest island of the Svalbard archipelago (Fig. 1). It is the southern arm of a twin fjord system "Kongsfjorden" and "Krossfjorden", which join into a submarine glacial trough to the NW and open onto the western Svalbard shelf. Kongsfjorden, orientated in a SE-NW direction, is ~20 km long and ranges in width from 4 to 10 km (Howe et al. 2003). Five tidewater glaciers (Kongsvegen, Kronebreen, Kongsbreen, Konwaybreen, and Blømstrandbreen) head the fjord (Fig. 2) greatly influence the fjord's hydrography. The water depth in the fjord ranges from 60 m at its head to around 400 m at the mouth (Trusel et al. 2010). Based on bathymetry, the fjord has been divided into an Outer and a Middle Zone with water depths between 200 and 400 m, and a Transitional and an Inner Zone with water depths below 100 m (Hop et al. 2002, Howe et al. 2003; inset in Fig. 2).

Fig. 1. Map of Svalbard archipelago, with the study area of Kongsfjorden, circled.

Because of the lack of a typical fjord sill at its mouth, the water column characteristics of Kongsfjorden are influenced to a great extent by the inflow of Transformed Atlantic water (TAW), which is a mixture of the warm and saline Atlantic water transported by the West Spitsbergen Current (WSC) flowing along the slope and the cold and less saline Arctic water (ArW) transported by coastal currents flowing northward around southern and western Spitsbergen (Svendsen et al. 2002). The characteristics of the TAW have been observed to fluctuate on a seasonal to inter-annual scale. In summer and early fall, the water column characteristics of the fjord are controlled by the increased freshwater flux from the glacier melt water and the thermo-haline stratification of the water mass primarily within the outer and central parts of the fjord. In contrast, during winter and early spring, the stratification is weak and the water masses

are almost homogeneous (Cottier et al. 2010). Consequent on a reduction of the freshwater input, the waters in the fjord also become more saline with only a small temperature variation between the surface and deeper waters (Elverhøi et al. 1983).

The sediment input into Kongsfjorden is dominated by the two glaciers, Kongsvegen and Kronebreen (Howe et al. 2003), with the suspended load from the TAW being much less. Sedimentation rates are very high where the glaciers terminate (0.6 m y^{-1} to > 1 m y^{-1}; Elverhøi et al. 1983, Trusel et al. 2010, Kehrl et al. 2011) but decrease away downstream of the fjord to as low as 0.4 mm y^{-1} in the Outer fjord. About 90% of the total sediment input is deposited within 400 m of the ice front (Elverhøi et al. 1983).

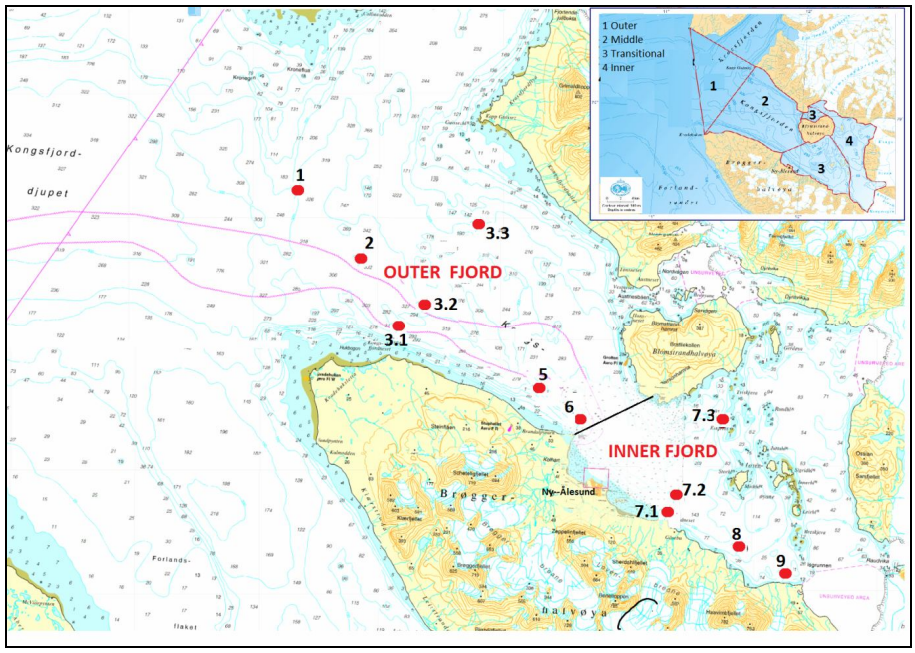

Fig. 2. Bathymetry and topography of the Kongsfjorden system showing the subdivisions of the fjord into Outer and Inner. (*http://toposvalbard.npolar.no/* - [WP 1]). The twelve locations of sampling are also marked. Inset shows the sub-divisions of the fjord by Hop et al.(2002). In this paper, the outer and middle zones of Hop et al. (2002) are grouped under "Outer fjord", while the Transitional and Inner Zones comprise the "Inner fjord ".

Material and Methods

Sampling and sample preparation

Surface sediment samples were collected from twelve pre-fixed locations along and across the length of the Kongsfjorden during three summer months of 2011, 2012 and 2013. The locations were so chosen as to cover uniformly the outer, central and inner part of the fjord and also representing the oceanic influx and glacial runoffs (Table 1, Fig. 2). The same locations were occupied during all the three years of sampling. The samples were collected onboard the research boat 'Teisten' belonging

to M/s Kings Bay AS, using a small stainless steel Van Veen grab. Extreme care was taken during the entire process of sampling to ensure that the samples obtained were undisturbed. At all locations, sub-samples were collected from the top (2-5 cm) of the grab samples brought on board, in acid-rinsed and pre-baked 100 ml glass bottles. Immediately after collection, the samples were stored at −20°C until analyses. Prior to analyses, the samples were allowed to thaw in a desiccator, all visible organisms and shell fragments were removed, the samples were sub-sampled for different analyses, and dried in a hot-air oven at 40°C. Samples for the elemental analyses were subsequently ground to a fine powder in an agate mortar.

Sample No.	*Location* (*Co-ordinates*)	*Water depth* [*m*]
1	79.0354° N: 11.2836° E	340
2	79.0099° N: 11.4245° E	290
3.1	78.9844° N: 11.5086° E	245
3.2	78.9931° N: 11.5547° E	320
3.3	79.0253° N: 11.6544° E	79
5	78.9587° N: 11.8224° E	320
6	78.9490° N: 11.8820° E	160
7.1	78.9170° N: 12.0741° E	87
7.2	78.9228° N: 12.0937° E	160
7.3	78.9536° N: 12.1755° E	59
8	78.9050° N: 12.2278° E	87
9	78.8951° N: 12.3201° E	57

Table 1. Locations of sampling in Kongsfjorden.

Elemental analyses

Approx. 30 mg each of the dried and homogenised sub-samples from all the locations were digested in a mixture of 5 ml HNO_3, 10 ml HF and 10 ml $HClO_4$ and evaporated to dryness on a hot plate, following the sample preparation technique proposed by Elik (2009). The process was repeated in each case till the dissolution of the sample was complete. The dry residue was then dissolved in 2% HNO_3 diluted to a standard volume of 50 ml with 2% HNO_3 (Elik 2009). The resulting clear solution was stored in an acid-washed plastic bottle for analysis.

Determination of the concentrations of Cr, Mn, Co, Cu, Zn and Pb in the digests was carried out through Inductively Coupled Plasma Mass Spectrometry (ICP-MS) on a Thermo elemental X7^{TM} ICP-MS. The concentration of Ti was also determined in all the samples as a reference element for assessing the terrigenous source of the sediments (Wei et al. 2003, Lu et al. 2012).

The following instrumental parameters were adopted for the analysis:

a. 1 l/min nebulizer gas flow;
b. 0.9 l/min auxiliary gas flow;
c. 15 l/min plasma gas flow;
d. 1400 W ICP RF power;
e. Measuring mode: peek hoping.

^{103}Rh was added as an internal standard and the percentage of recovery of the standard was between 90-100%. Accuracy of the measurements was confirmed by measurements of blanks and repeated measurements of international USGS standard (NIST1646a estuary sediment) digested along with the batches of sediment samples. Duplicate analyses of samples were carried out for ensuring data reproducibility. The range of accuracy obtained was between 1.3 and 5% for all elements while the percentage deviation was less than 5%. All results are reported in mg kg^{-1} (Table 2).

Grain size analysis

Grain size analysis of all samples was carried out by sieving through a 230 mesh (equivalent to 63 μm or 4 Φ on the Wentworth grain size scale) to separate the sand fraction (> 63 μm) from silt plus clay, followed by the conventional pipette method to obtain the weight percentage of silt and clay (Folk et Ward 1957, Carver 1971, Folk 1980). Prior to analysis, the samples were pre-treated with 10 ml of sodium hexametaphosphate (Calgon) to disaggregate the sediments followed by 30% H_2O_2 to oxidize the organic matter. The results are included in Table 2.

Total Carbon analyses

The determination of total organic carbon was carried out based on the Walkley-Black method (Walkey 1947, Jackson 1958). The oxidisable organic matter was oxidised by 1N $K_2Cr_2O_7$ solution with the aid of the heat generated by mixing two volumes of concentrated H_2SO_4 and Ag_2SO_4 with one volume of the dichromate. On completion of the reaction, the solution was treated with 200 ml Milli-Q water, 10 ml of 85% H_3PO_4 and 0.2 gm of NaF, and back-titrated with standard $(NH_4)_2Fe(SO_4)_2 \cdot 6H_2O$ solution using diphenyl amine as an indicator to a one drop end point (brilliant green). The titre is inversely related to the amount of organic carbon present in the sample.

Results

The results of the various analyses carried out on the surface sediments collected during the three summer months of 2011, 2012 and 2013 are provided in Table 2. Considering the sampling density and distribution, the results have been grouped and are discussed in relation to a bathymetric sub-division of the fjord into an Outer fjord, where the average water depth is of the order of >100 m, and an Inner fjord with average water depth less than 100 m (Fig. 2). The Outer fjord corresponds to the outer and middle zones of Hop et al. (2002), while the Inner fjord comprises the transitional and inner Zones (Fig. 2, inset). A shallow, approximately 30-m deep sill in the central part of the fjord between Blomstrandhalvøya and Brøggerhalvøya separates the Outer fjord from the Inner fjord.

	Stn. No.	Mn	Cr	Zn	Co	Cu	Pb	Ti	Weight %			Nomenclature of sediment **
									Silt	Clay	TOC	
1. Period of sampling: July 2011												
Outer fjord	1	758.68	134.51	110.16	16.68	22.95	18.96	5863.36	52	46	1.31	Clayey silt
	2	683.35	114.15	93.4	16.71	22.35	14.61	5818.36	56	43	1.28	Clayey silt
	*3.1	588.01	121.46	84.26	15.69	24.13	13.74	5221.69	39	59	0.98	Silty clay
	3.2	667.01	107.53	94.1	17.28	26.83	17.26	5378.36	37	62	1.19	Silty clay
	*3.3	718.35	97.25	96.03	17.56	22.16	13.19	5481.69				
	5	600.51	104.51	68.86	14.67	20.81	15.39	4971.69	28	70	1.11	Silty clay
	6	583.35	94.86	74.5	14.43	18.5	16.11	4368.36	29	69	1.14	Silty clay
Inner fjord	*7.1	564.18	67.56	97.58	14.08	19.38	7.26	4996.69	19	78	0.64	Clay
	7.2	843.18	122.56	167.31	20.01	29.46	13.26	5771.69	23	74	0.26	Silty clay
	*7.3	867.35	112.71	124.08	19.53	24.36	10.88	5775.03	16	82	0.13	Clay
	8	896.35	114.06	129.15	20.18	28.93	12.01	6073.36	13	85	0.16	Clay
	9	813.35	98.3	113.85	19.08	24.26	13.31	5810.03	6	90	0.20	Clay
2. Period of sampling: July 2012												
Outer fjord	1	561.06	66.59	71.8	17.44	19.84	23.18	4719.51	58	41	1.49	Clayey silt
	2	415.39	72.65	50.3	15.22	16.46	29.44	4004.51	45	54	1.17	Silty clay
	*3.1	602.73	84.94	94.02	22.14	27.86	27.84	5007.84	55	42	1.04	Clayey silt
	3.2	415.39	72.65	50.3	15.22	16.46	29.44	4004.51	43	55	0.84	Silty clay
	*3.3	445.89	69.32	95.57	15.79	22.26	29.14	4281.18				
	5	373.06	66.35	53.05	11.87	14.44	23.43	3651.18	35	61	1.28	Silty clay
	6	442.56	62.09	54.6	13.19	16.99	28.58	3932.84	35	63	1.23	Silty clay
Inner fjord	*7.1	295.89	60.47	32.22	9.7	11.24	20.11	3226.18	23	74	0.58	Silty clay
	7.2	482.56	57.52	87.67	13.35	14.36	20.46	4069.51	28	70	0.29	Silty clay
	*7.3	758.68	134.51	110.16	16.68	22.95	18.96	5863.36	17	79	0.11	Clay
	8	321.23	46.14	90.47	6.35	9.29	19.79	2539.51	19	79	0.19	Clay
	9	465.73	56.12	102.54	12.38	17.86	30.48	4212.84	12	87	0.23	Clay
3. Period of sampling: August 2013												
Outer fjord	1	538.73	67.14	63.62	16.17	16.99	22.88	4361.18	55	44	1.87	Clayey silt
	2	475.06	66.49	42.02	14.79	14.09	22.91	4462.84	54	45	1.78	Clayey silt
	*3.1	286.73	67.95	32.3	13.9	19.08	16.89	2962.84	37	60	1.11	Silty clay
	3.2	453.39	55.92	52.14	13.69	13.49	19.44	3914.51	39	59	0.93	Silty clay
	*3.3	512.73	60.34	44.1	14.05	15.38	22.54	4047.84				
	5	374.23	58.3	39.69	11.45	14.08	23.73	3376.18	36	63	1.34	Silty clay
	6	413.23	61.45	90.29	13.53	17.21	29.74	4009.51	32	67	1.43	Silty clay
Inner fjord	*7.1	349.73	53.97	42.49	10.51	15.81	27.33	3742.84	24	73		Silty clay
	7.2	446.06	54.47	54.42	12.18	13.78	26.49	4312.84	29	69	0.79	Silty clay
	*7.3	370.73	49.62	24.12	8.68	12.21	24.18	3662.84	19	77		Clay
	8	396.06	57.05	51.1	12.27	16.59	27.79	3812.84	21	78	0.17	Clay
	9	362.39	45.79	21.44	7.72	10.31	23.93	3094.51	9	87	0.30	Clay

◄ **Table 2.** Concentrations (mg kg^{-1}) of trace elements and silt, clay and total organic carbon (weight %) in the surface sediments of Kongsfjorden collected during the summer of 2011, 2012 and 2013. *Note: The station locations are marked in Figure 2. * Sampling locations across the ford. ** Nomenclature after Shepard (1954).*

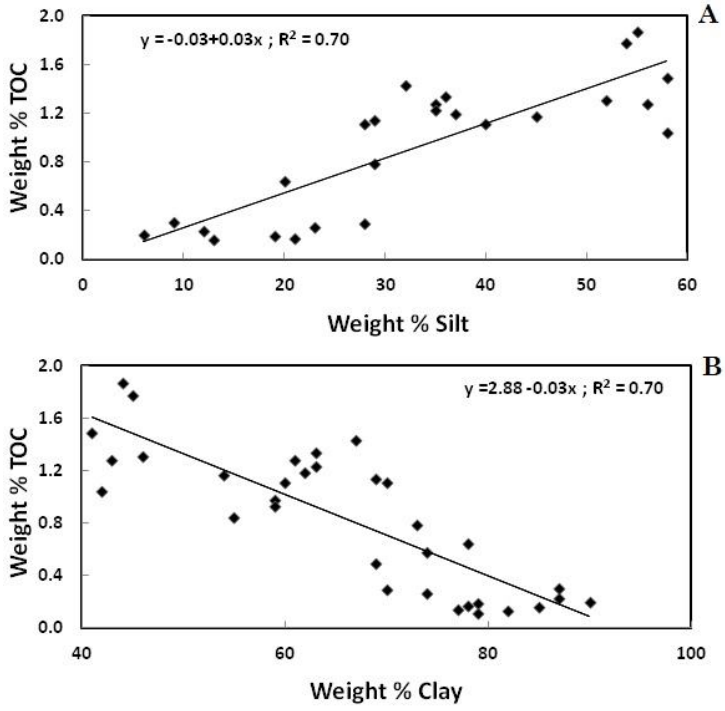

Fig. 3. Cross plots of (A) weight percent silt-TOC and (B) weight percent clay-TOC in the surface sediments of Kongsfjorden.

Element	(a) Outer fjord			(b) Inner fjord			(c) Entire fjord		
	Min	Max	Average	Min	Max	Average	Min	Max	Average
Mn	286.73	758.68	519.50	295.89	896.35	548.90	286.73	896.35	531.75
Cr	55.92	134.51	81.26	45.79	134.51	75.39	45.79	134.51	78.81
Zn	32.30	110.16	69.29	21.44	167.31	83.24	21.44	167.31	75.10
Co	11.45	22.14	15.31	6.35	20.18	13.51	6.35	22.14	14.56
Cu	13.49	27.86	19.16	9.29	29.46	18.05	9.29	29.46	18.70
Pb	13.19	29.74	21.83	7.26	30.48	19.75	7.26	30.48	20.96

Table 3. Range of concentrations of heavy metals in the surface sediments of Kongsfjorden, for (a) the Outer fjord, (b), the Inner fjord , and (c) for the Entire fjord, during the three years of study.

Year	Mn	Cr	Zn	Co	Cu	Pb	*Ti*	Remarks
I. Outer fjord								
2009	362.56	68.62	83.36	13.96	24.83	27.05	-	*Calculated from Lu et al. 2013*
2011	657.04	110.61	88.76	16.15	22.53	15.61	5300.50	*This study*
2012	465.15	70.66	67.09	15.84	19.19	27.29	4228.80	*This study*
2012	433.50	71.40	77.80	12.40	20.05	18.95	4015.00	*Calculated from Grotti et al.2013*
2013	436.30	62.51	52.02	13.94	15.76	22.59	3876.41	*This study*
II. Inner fjord								
2009	485.61	62.41	86.72	15.74	27.30	19.97	-	*Calculated from Lu et al. 2013*
2011	796.88	103.04	126.39	18.58	25.28	11.34	5685.36	*This study*
2012	464.82	70.95	84.61	11.69	15.14	21.96	3982.28	*This study*
2012	446.5	60.75	70.65	12.5	20.25	15.2	3860.00	*Calculated from Grotti et al.2013*
2013	384.99	52.18	38.71	10.27	13.74	25.94	3725.17	*This study*
III. Average for the Entire fjord								
2009	440.03	64.71	85.47	15.08	26.38	22.59	-	*Calculated from Lu et al. 2013*
2011	715.31	107.46	104.44	17.16	23.68	13.83	5460.86	*This study*
2012	440.00	66.08	74.23	12.45	20.15	17.08	3937.50	*Calculated from Grotti et al. 2013*
2013	414.92	58.21	46.48	12.41	14.92	23.99	3813.40	*This study*

Table 4. Average concentrations (mg kg^{-1}) of elements in the surface sediments of Kongsfjorden between 2009 and 2013.

(A) Inner fjord sediments

	Cr	*Mn*	*Co*	*Cu*	*Zn*	*Pb*	*Ti*	*Silt*	*Clay*	*TOC*
Cr	1.00									
Mn	**0.93**	1.00								
Co	**0.89**	**0.96**	1.00							
Cu	**0.89**	**0.95**	**0.96**	1.00						
Zn	**0.78**	**0.85**	**0.82**	**0.85**	1.00					
Pb	-0.63	**-0.71**	-0.64	-0.61	-0.64	1.00				
Ti	**0.90**	**0.96**	**0.97**	**0.95**	**0.78**	-0.64	1.00			
Silt	-0.18	-0.31	-0.16	-0.15	-0.07	0.13	-0.22	1.00		
Clay	0.22	0.35	0.23	0.28	0.22	-0.19	0.25	-0.98	1.00	
TOC	-0.40	-0.43	-0.30	-0.30	-0.38	0.09	-0.29	0.50	-0.47	1.00

(B) Outer fjord sediments

	Cr	*Mn*	*Co*	*Cu*	*Zn*	*Pb*	*Ti*	*Silt*	*Clay*	*TOC*
Cr	1.00									
Mn	**0.81**	1.00								
Co	0.45	0.64	1.00							
Cu	**0.72**	0.67	**0.79**	1.00						
Zn	**0.69**	**0.77**	0.63	**0.79**	1.00					
Pb	-0.63	-0.55	-0.08	-0.31	-0.22	1.00				
Ti	**0.84**	**0.96**	0.67	**0.72**	**0.81**	-0.47	1.00			
Silt	0.10	0.35	0.67	0.27	0.22	0.10	0.43	1.00		
Clay	-0.09	-0.35	-0.64	-0.22	-0.21	-0.13	-0.41	-1.00	1.00	
TOC	-0.23	0.04	-0.05	-0.26	-0.04	0.12	0.03	0.43	-0.40	1.00

Table 5. Correlation coefficients between the different elements, TOC and weight % Silt and clay in the surface sediments of Kongsfjorden during 2011 to 2013. (A) sediments from the Inner fjord, and (B) sediments from the Outer fjord.

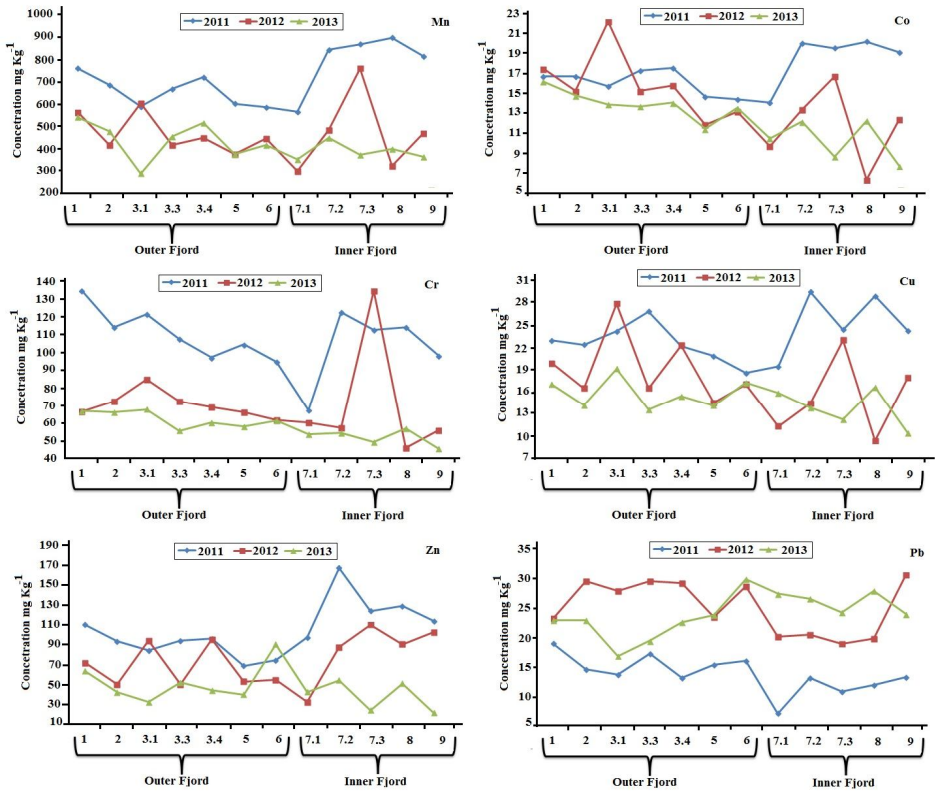

Fig. 4. Concentration of Heavy Metals in Kongsfjorden (Inner and Outer fjord).

The surface sediments of the fjord are by and large fine to very fine grained with varying proportions of silt (grain diameter between 0.0625 mm and 0.0039 mm) and clay (grain size smaller than 0.0039 mm). Sand fraction is negligible in all the samples analysed (less than 4 weight %). Based on the relative proportions of sand, silt and clay, the sediments have been classified as clay, silty clay or clayey silt following the nomenclature of Shepard (1954; Table 2).

The distribution pattern of sediments in the fjord is different in the Outer and Inner fjords. The sediments in the Outer fjord are mostly clayey silt or silty clay with the clay content ranging between 40 and 70%. In contrast, the sediments of the Inner fjord are more clayey, with the clay content in excess of 70%. There is also a perceptible difference between the Total Organic Carbon (TOC) concentrations in the outer and Inner fjord; the sediments of the Inner fjord are markedly low in TOC (average 0.3 wt %) as compared to those of the Outer fjord (average 1.3 wt %). A cross plot of the weight percent silt and TOC shows a strong positive correlation ($r^2 = 0.70$; Fig. 3A) pointing to the influence of grain size on the TOC content, with a preferential enrichment of TOC in dominantly silt-grade sediments. At the same time however, a strong negative correlation characterises the TOC-weight percent clay relationship ($r^2 = -0.70$; Fig. 3B) indicating that in addition to grain size, other factors control the distribution of TOC in the fjord. Similarly, the lack of any correlation between grain size or TOC and the heavy metals (Table 5) indicates that these characteristics of sediments are not significant variables that control the spatial distribution and concentration of heavy metals in the fjord. These aspects are discussed further, below.

The heavy metals in the fjord can be broadly grouped into two classes: (a) those which have a relatively high concentration, averaging over 70 mg kg^{-1} (Mn, Cr and Zn) and (b) those whose concentrations are on an average, less than 20 mg kg^{-1} (Co, Cu and Pb). Among these elements, the range of concentrations for the Entire fjord as well as for the outer and inner zones is shown in Table 3.

Table 4 provides the average concentration of heavy metals in the surface sediments during the study period. For a comparative evaluation, the elemental concentrations reported for the surface sediments of the fjord by Lu et al. (2013, sampling carried out during July 2009), Grotti et al. (2013, sampling during June 2012) and Ardini et al. (2016, sampling July 2014) are also provided. As can be seen, the values obtained by us broadly fall within the ranges previously reported. In particular, the average concentrations of heavy metals in the samples collected during the summer of 2012 are quite comparable with the corresponding values reported by Grotti et al. (2013) for samples collected during the same season in 2012, supporting the integration of the reported values in the comparative evaluations.

As can be seen from Tables 3 and 4, the Kongsfjorden is characterised by a fairly wide variation in the concentrations of the heavy metals both on a spatial and a temporal frame. The concentration of Mn is the highest in the Entire fjord, averaging over 400 mg kg^{-1} for all years excepting 2011, when the average concentration was over 700 mg kg^{-1}. The average concentrations of Cr and Zn are also relatively high for the sediments collected during 2011 as compared to 2009, 2012 and 2013. Next to Mn in decreasing order of abundance is Zn, Cr, Pb, Cu and Co. In general, along the Outer fjord, the concentrations of heavy metals are highest near the mouth and lowest near the farthest end, near the sill separating the Outer fjord from the inner. In contrast, the concentrations are quite variable in the Inner fjord.

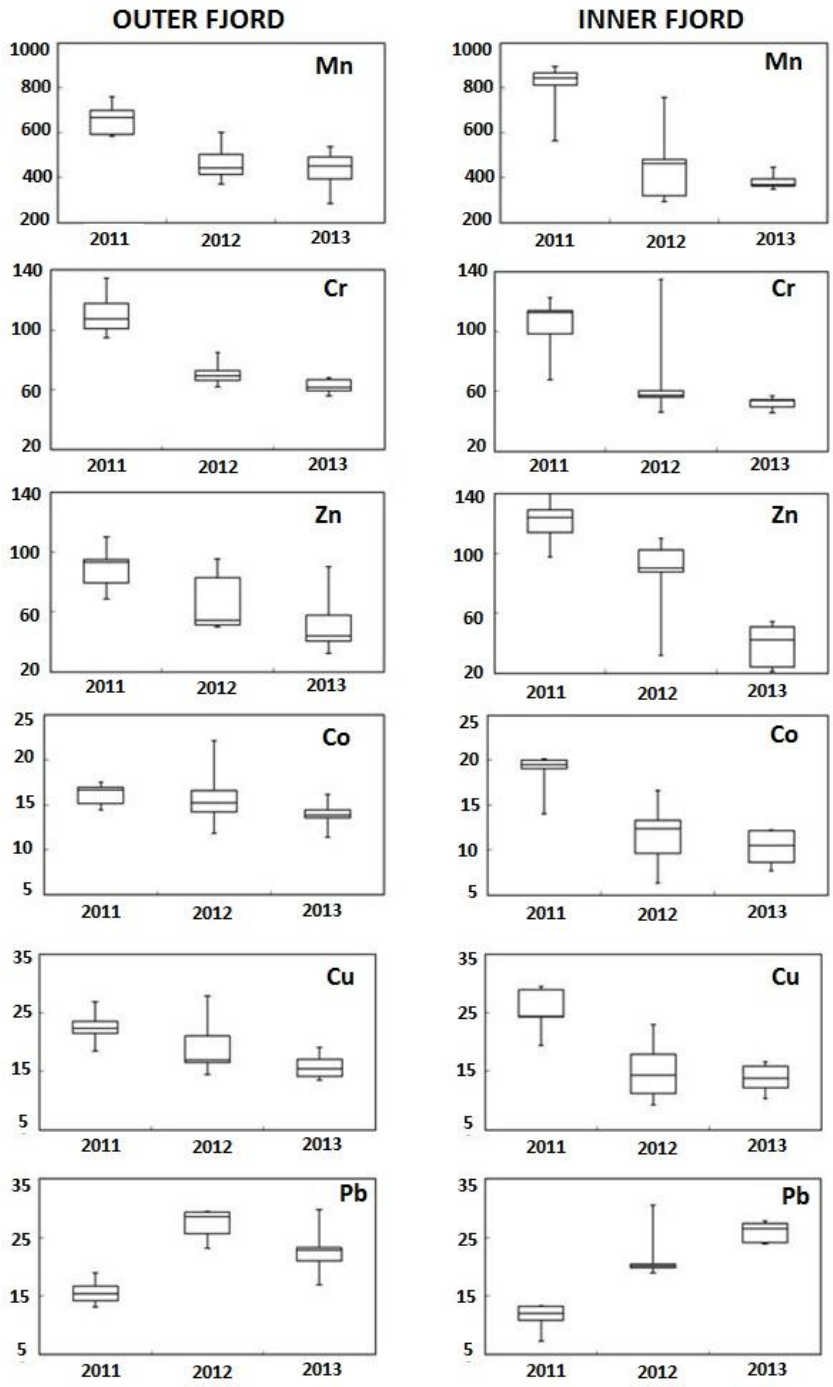

Fig. 5. Box and whisker plots showing the variations in the average concentrations of heavy metals in the surface sediments of the Outer and Inner Kongsfjorden, during the summer of 2011, 2012 and 2013. Concentrations (in mg kg^{-1}) along the Y-axis and the years along the X-axis.

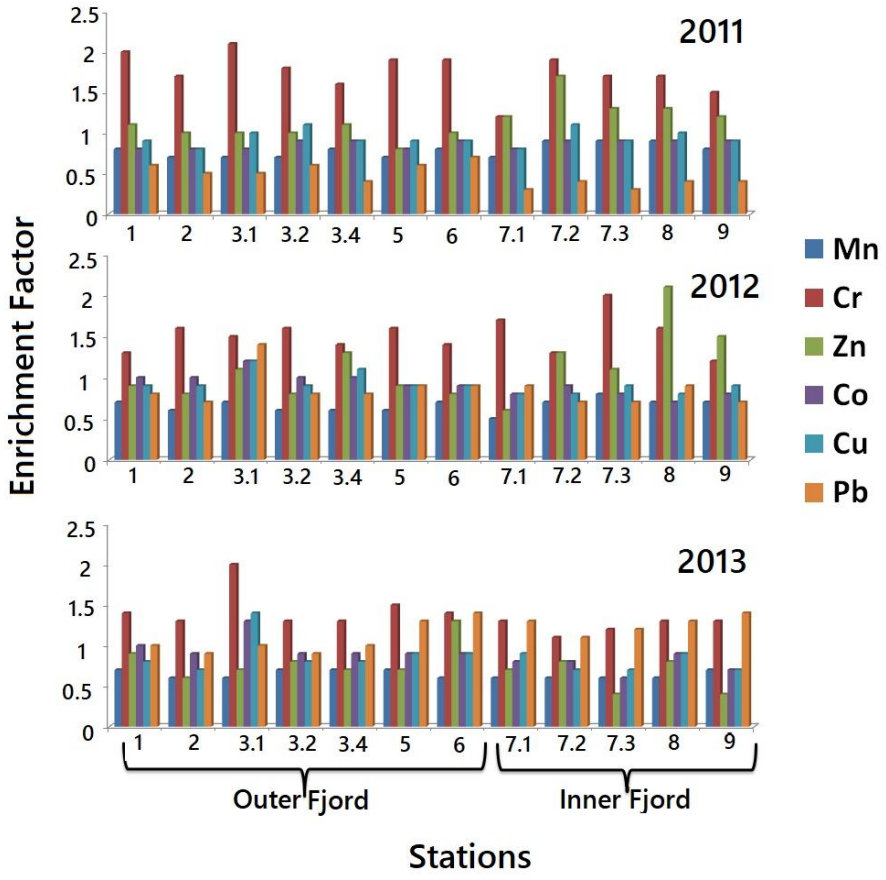

Fig. 6. Enrichment factors of the heavy metals in the surface sediments of Kongsfjorden during 2011, 2012 and 2013.

For instance, while for the years 2011 and 2013 the lowest concentrations of all the elements occur at Station 9 closest to the mouth of the glaciers Kongsvegen and Kronebreen, during 2012 the elements show enrichment at Station 9 relative to Station 8, away from the glacier mouth. The enrichment is to the tune of about 13% for Zn, over 50% for Pb, and to nearly as much as double in the case of Co and Cu.

Good positive correlation is observed between the concentrations of all the heavy metals excepting Pb in the Inner fjord sediments ($r > 0.80$), indicative of their common source of supply (Table 5A). The significant positive correlation of these elements with Ti also points to their terrigenous source, possibly through the glacier melt waters. Interestingly however, when the concentrations of these elements in the sediments of the Outer fjord alone are considered, the correlation is much weaker (Table 5B), indicative of the possible multiple source of sediments. The uniform negative correlation exhibited by Pb with the other elements may point to its source from elsewhere, and brought in by long-range atmospheric transport (Liu et al. 2012) or by the WSC (Lu et al. 2013, Bazzano et al. 2014).

Discussion

Sediment distribution pattern in Kongsfjorden

The primary source of sediments in Kongsfjorden has been suggested to be the material discharged directly from the glacier terminus by subglacial or englacial meltwater outlets. Contributions from iceberg-rafting and sea-ice are relatively small (Dowdeswell et Dowdeswell 1989). The glacier meltwater affects a large area in the fjord, up to 45 km distance from the glacier front and up to 30 m depth (Hop et al. 2002, Svendsen et al. 2002). However, as regards the deposition of sediments, the meltwater would be more significant in the inner part of the fjord as compared to the outer, primarily because of the rapid reduction of the current velocities. About 90% of the total sediment input from the glaciers has been suggested to be deposited within 400 m of the ice front, resulting in extremely high sedimentation rates in the inner part (Elverhøi et al. 1983, Svendsen et al. 2002, Kehrl et al. 2011). The finer grains would undergo sedimentation as flocs or aggregates while the remaining particles are transported down fjord in suspension. The aggregation and resultant preferential settling of fine grains in the proximity of glacier front would be aided by the water column turbulence due to the large suspended load (Gorlich et al. 1987). This would explain the dominance of clay-rich sediments in the Inner fjord.

Away from the glacier front, the sediment accumulation rates decrease sharply by an order of magnitude towards the sill separating the Inner fjord from the outer, and again by another order of magnitude in the Outer fjord (Elverhoi et al. 1983, Svendsen et al. 2002). Besides this sediment load from the glacier meltwater, the Outer fjord comprises material from the influx of Atlantic and TAW and ice-rafted debris. The sediments are also subjected to extensive reworking and redistribution by bioturbation and such physical processes as mass-transport events, geostrophic currents or iceberg-ploughing (Boulton 1990, Svendsen et al. 2002, Howe et al. 2003, Streuff 2013). Morphological differences between the outer and Inner fjord as well as enhanced biological productivity in the Outer fjord compared to the inner can also be expected to influence the nature, composition and distribution of sediments in the Outer fjord relative to the inner parts (Streuff 2013, Piquet et al. 2014). A combination of these factors would explain the observed differences in the textural and mineralogical characteristics of the sediments of the Outer fjord.

Grain size plays a significant role in determining the content and distribution of organic carbon in sediments. Therefore, the factors which control the grain size distribution are also likely to be important as regards the type and amount of organic matter preserved in sediments. The surface sediments of Kongsfjorden are not particularly rich in organic carbon (*see* Table 2). The TOC contents vary from a low of 0.1 (wt %) to a maximum of 1.9 (wt %). There is also a perceptible difference between the organic carbon concentrations in the Outer and Inner fjord; the sediments of the Inner fjord are markedly low in TOC (average 0.3 wt %) as compared to those of the Outer fjord (average 1.3 wt %). We contend that the low concentrations of organic matter in the Inner fjord are related to a combination of three factors: (i) limited primary and secondary productivity on account of the poor transparency of water caused by an abundance of suspended particulate matter close to the glaciers (Zaborska et al. 2006a,b; Piquet al. 2014); (ii) dilution of the organic matter by the detrital matter on account of the high sedimentation rates in the Inner fjord; and (iii) proximity of the Inner fjord to the glaciers, which appears to limit phytoplankton biomass even during

particularly high biomass seasons (Piquet et al. 2014). This is further corroborated by the observed increase in organic matter concentration with increasing distance from the glaciers (Table 2), lower sedimentation rates in the Outer fjord, and the evidences for bioturbation in the outer part of the fjord (Zaborska et al. 2006a). Enhanced biological productivity in the Outer fjord is also consequent on an increased availability of nutrients through an enhanced inflow of TAW (Hegseth et Tverberg 2013, Piquet et al. 2014).

Despite the above, what may at the first sight be intriguing about the Kongsfjorden sediments is the relationship between grain size and the TOC. Two prominent trends are observed (Fig. 3): (a) a strong positive correlation between the weight % silt and TOC ($r^2 = 0.70$); and (b) an equally strong negative correlation between the weight % clay and TOC ($r^2 = -0.70$). In general, a positive correlation between TOC and finer particles has been considered characteristic of soils and sediments in most environments (*e.g.* Buchanan et Longbottom 1970, Mayer 1994, Tyson 1995). The close association of these two components has been explained by the larger surface area of finer particles which provides good binding sites for organic matter (Keil et al. 1994, Mayer 1994). Nonetheless we contend that the negative correlation observed between TOC and the clay fraction in the Kongsfjorden sediments as well as the preferential retention of organic carbon by the silt-grade fractions is not an aberration but is more reflective of the contrasting conditions of sedimentation and hydrodynamics of the two segments of the fjord, as described above. The spatial and temporal variations in the concentrations of heavy metals in the sediments further corroborate this argument.

Spatial distribution of heavy metals in the surface sediments

The varying concentrations of the heavy metals in the surface sediments of Kongsfjorden on a spatial and temporal frame bring out the sensitivity of the fjord to fluctuations in the influx of the glacial meltwater and the warmer Transformed Atlantic water (TAW) from the Western Spitsbergen Current (WSC). In the Outer fjord, with few exceptions, the concentrations of Mn, Cr, Zn, Co, Cu and Pb are by and large highest near the mouth of the fjord while the lowest concentrations are at its distal end. Similar higher concentrations of these metals in the surface sediments as well as in the suspended particulate matter near the mouth of the fjord have been attributed to the influence of the West Spitsbergen Current (WSC), which can bring sediments enriched in trace elements from low and middle latitude to the Arctic (Grotti et al. 2013, Bazzano et al. 2014, Ardini et al. 2016). While we subscribe to this conclusion, we also contend that the observed spatial variations in the concentrations of these heavy metals up-fjord as well as their high concentrations in the Inner fjord cannot entirely be ascribed to the influx of the watermass contained in the WSC (*i.e.* the TAW). Our analytical results show a concurrent enrichment of the heavy metals in the Inner fjord sediments as well, especially in the samples from near the glacier outlets (Stations 8 and 9; Fig. 2; Table 2). At the same time, the concentrations of the heavy metals are also the lowest at the distal ends of the inner and Outer fjords, near the shallow sill separating the two fjord segments (Fig. 4). These observations highlight the relative roles of a dual source for the heavy metals in the Kongsfjorden- the WSC and the glacial meltwater. The meltwater discharge of sediments would be more significant in the inner part of the fjord as compared to the outer, primarily because of the rapid reduction of the current velocities away from the glacier outlets. As noted earlier, about

90% of the total sediment input from the glaciers has been suggested to be deposited within 400 m of the ice front, and only the remaining material is transported with surface waters and deposited in the central and outer parts of the fjord. The very good positive correlation observed among the different heavy metals barring Pb ($r > 0.78$) as also with Ti in the Inner fjord sediments corroborates the possibility of the sediment flux from the glacial melt water discharge being the primary source of these elements.

In contrast to the Inner fjord, the sediments brought in by the TAW would be more important in the Outer fjord. Furthermore, since the horizontal extent of TAW is practically limited by the sill between the inner and outer basins (Svendsen et al. 2002), most of the sediment flux from TAW can be expected to be deposited in the Outer fjord itself. The Outer fjord sediments are subjected to extensive resuspension and redistribution, mass transport events *etc*. as discussed earlier. The weak positive correlation observed among the different elements can be ascribed to the interplay of these factors. The textural and mineralogical characteristics of the Outer fjord sediments would thus be different and largely independent of those of the Inner fjord, as susbstantiated by our results. Depending on the intensity of the influx of the TAW or the glacial melt water, the textural and mineralogical characteristics of the surface sediments especially in the central parts of the fjord would tend to approach the end-member characteristics of either the TAW or the glacial discharge respectively.

Temporal variations in the distribution of heavy metals in the surface sediments

Systematic temporal variation in the concentrations of all the elements between 2009 and 2013 is one of the most conspicuous features of the Kongsfjorden surface sediments (Table 4, Fig. 5). As can be seen, the average concentrations of the various elements, excepting Pb, show a consistent decrease from 2011 to 2013 for both the outer and Inner fjord. In contrast, Pb shows a reverse trend. The change from 2011 to 2013 is more pronounced in the case of the Inner fjord as opposed to the outer. Thus, whereas the decrease in the average concentration from 2011 to 2013 for Mn, Cr, Zn, Co and Cu ranges from a low of 45% for Co to about 70% for Zn, the corresponding decrease in the Outer fjord is between 14 and 40%. Similarly, while the average concentration of Pb in the Outer fjord shows an approximately 45% increase between 2011 and 13, the increase is almost 2.5 times in the sediments of the Inner fjord.

A comparison of the average concentrations of the heavy metals between the Outer and the Inner fjord sediments for the three years also shows two distinct trends: (i) a higher average concentration of all elements barring Cr and Pb, for the year 2011 in the Outer fjord sediments relative to those of the Inner fjord; and (ii) a lower average concentration of all elements except Pb in the Outer fjord sediments for 2013 relative to the Inner fjord (Table 4). The intervening year of 2012 shows a mixed result, with the average concentrations Mn and Zn showing comparable concentrations for both the segments of the fjord while Cr shows a marked increase from outer to inner. In contrast, the average concentrations of Co, Cu and Pb show a decrease from the outer to Inner fjord (Table 4).

The observed temporal variations in the concentration of the heavy metals in the fjord as a whole and also between its two segments can be ascribed to the variations in the inflow of the TAW and glacial meltwater into the fjord. Several studies have brought out the seasonal fluctuations in the freshwater input primarily in the inner part of the fjord and of the TAW in the outer

and central parts (Hop et al. 2002, Svendsen et al. 2002, Cottier et al. 2005, 2010; Piehl Harms et al. 2007, Hegseth et Tverberg 2013). Such short-term variations can be expected to result not only in changes in the water column characteristics of the fjord as a whole, but also in the composition of the suspension settling. On a longer timeframe, observational data also shows an inter-annual increase in the intrusion of Atlantic/TAW and resultant variability in the watermasses in the fjord during the summer through Fall of 2011-2014 (David et Krishnan 2015, David et al. 2016). Another feature noted during the period has been a sharp decline in the freshwater volume in Kongsfjorden over the years (David et Krishnan 2015). Such an increase in the inflow of the TAW and probable concomitant decrease in the freshwater volume can lead to a marked change in the nature of the sediment flux in the central and inner parts of the fjord. This would explain the observed decrease in the concentrations of the heavy metals in the fjord over the three-year period corresponding to an increased dominance of the particle flux from TAW.

Heavy metal contamination in Kongsfjorden

As noted earlier, the proximity of Kongsfjorden to the International Arctic Research Facilities of Ny-Ålesund, the increasing importance of the latter as a hub of summer-time tourist activities, and the unique niche the fjord occupies in the Arctic climate system make it a prime area for assessing the presence and effects of anthropogenic (human-related or induced) contaminants in the Arctic. Our present study of the heavy metal content in the sediments indicates that the average concentrations of Mn, Cr, Zn, Co and Cu are comparable to the concentrations reported from other documented, relatively pristine Arctic locales, such as for instance, from the Northeast Chukchi Sea (*c.f.* Naidu et al. 1997 and references therein). Our studies also indicate that the observed spatial and temporal variations in the concentrations are attributable to inter-annual variations in the supply of sediments from the Atlantic and the glacial meltwaters, rather than to any anthropogenic influence.

To assess the degree to which the various heavy metals in the Kongsfjorden surface sediments is enriched relative to their natural background values, we calculated the Enrichment Factor (EF) for the various elements using the following equation (Ardini et al. 2016):

$$\text{Enrichment Factor (EF)} = \frac{\left(\frac{X}{R}\right)\text{sample}}{\left(\frac{X}{R}\right)\text{Background}} \qquad \textit{Egn.}\ 1$$

where $(X/R)_{sample}$ is the ratio of the concentration of a heavy metal to a reference element in the same sample, and $(X/R)_{Background}$ is the ratio between the same elements in the upper continental crust. We used Ti as the reference element for normalising the heavy metal concentrations and the world average continental crustal values by Wedepohl (1995) for the background ratios (Fig. 6).

Our calculations show that except for Cr, the average EF values of the elements are mostly less than 1.5, which according to the classification terminology of Holtan et Rosland (1992, cited in Skotvold et al. 1997) would qualify these elements as "Not or slightly enriched". Cr can be said to be moderately enriched. Although the EF is not strictly a measure of anthropogenic pollution, the slight to moderate enrichment of the heavy metals relative to the background might be indicative of their long-range transportation through the atmosphere or by the Atlantic waters from sources elsewhere.

Conclusions

The distribution pattern of surface sediments in the Kongsfjorden and the heavy metals present in them reveals the spatial and temporal variabilities in sediment flux to the fjord from a dual source- the Transformed Atlantic waters input through the mouth and the glacier meltwater discharge at the head. The fjord comprises two segments; a deeper, Outer fjord and a shallower Inner fjord, separated by a shallow sill. The hydrodynamics of the two fjord segments is markedly different. Whereas glacial meltwater discharge and high sedimentation rates are significant as regards the Inner fjord, suspension settling, and resuspension and redistribution of material brought in from the Transformed Atlantic water dominate the Outer fjord region. The Inner fjord is blanketed by relatively more clay-rich sediments as opposed to the more silty variants such as clayey silt or silty clay in the Outer fjord. The preferential enrichment of the clay fraction in the Inner fjord is the result of rapid settlement of finer particles as aggregates or flocs aided by the water column turbulence brought about by the large suspended load near the glacier outlet. The water column turbulence and high sedimentation rates in the Inner fjord also foster lower levels of primary production and poor preservation of the organic matter due to dilution by large loads of inorganic material. This is reflected in the low levels of TOC content in the Inner fjord sediments as compared to the Outer fjord.

In contrast to the Inner fjord, the sediments brought in by the Transformed Atlantic Water (TAW) are more important in the Outer fjord. Suspension settling is the dominant mechanism of sedimentation consequent on a decrease in the current velocity. This would explain the relatively higher silt contents in the Outer fjord sediments particularly near the mouth, and the increase in the clay fraction towards the distal end of the Outer fjord. The observed higher concentrations of TOC relative to the Inner fjord sediments are reflective of enhanced levels of biological productivity consequent on an increased availability of nutrients through an enhanced inflow of TAW, presence of a thicker euphotic zone and lower rates of sedimentation, all promoting the increased production and preservation of organic matter sediments.

The hydrological and hydrodynamic differences between the two segments of the Kongsfjorden are also reflected in the spatial distribution pattern of Mn, Cr, Zn, Co, Cu and Pb in the surface sediments. Based on their concentrations, the heavy metals in the fjord can be broadly grouped into two classes: (a) those which have a relatively high concentration, averaging over 70 mg kg^{-1} (Mn, Cr and Zn) and (b) those whose concentrations are on an average, less than 20 mg kg^{-1} (Co, Cu and Pb). Variations in grain size and TOC contents do not appear to exercise any control in the spatial distribution of these elements. Instead, the distribution pattern is dictated solely by the variations in the source and the hydrodynamic conditions controlling the sedimentation in the two segments of the fjord. In the Outer fjord, the concentrations of Mn, Cr, Zn, Co, Cu and Pb are by and large highest near the mouth of the fjord while the lowest concentrations are at its distal end. The Inner fjord is also characterised by a concurrent enrichment of these heavy metals near the glacier outlets with the lowest values occurring near the shallow sill separating it from the Outer fjord. These observations bring out the relative roles of a dual source for the heavy metals in the Kongsfjorden- the Transformed Atlantic water (TAW) from the Western Spitsbergen Current (WSC) and the glacial meltwater. The meltwater discharge of sediments is more significant in the Inner fjord as opposed to the TAW in the Outer fjord. The significant positive correlation of the heavy metals in the Inner fjord sediments among themselves as also with

Ti corroborates their terrigenous source through the glacier melt waters.

In contrast to the Inner fjord, the weak correlation exhibited by the heavy metals of the Outer fjord indicates that these sediments, although primarily sourced from TAW, have however, been subjected to extensive resuspension and redistribution, mass transport events, iceberg-ploughing etc. The observed variations in the grain size and TOC contents of the sediments also support this contention. The mineralogical characteristics of the Outer fjord sediments would thus be different and largely independent of those of the Inner fjord, as substantiated by our studies. Depending on the intensity of the influx of the TAW or the glacial meltwater, the textural and mineralogical characteristics of the surface sediments especially in the central parts of the fjord would tend to approach the end-member characteristics of either the TAW or the glacial discharge respectively.

Systematic temporal variation in the concentrations of the heavy metals during the time period between 2009 and 2013 is one of the most conspicuous features of the Kongsfjorden surface sediments. The average concentrations of the various elements, excepting Pb, show a consistent decrease from 2011 to 2013 for both the outer and Inner fjord. In contrast, Pb shows a reverse trend. The change from 2011 to 2013 is more pronounced in the case of the Inner fjord as opposed to the outer. These variations are consistent with the observed inter-annual increase in the influx of TAW and the resultant variability in the water-masses in the fjord during the summer through fall of 2011-2013. Such an increase in the inflow of the TAW with concomitant decrease in the glacier meltwater flux can lead to a marked change in the distribution pattern of sediments in the central and inner parts of the fjord. This would also explain the observed decrease in the concentrations of the heavy metals in the fjord over the three-year period corresponding to an increased dominance of the particle flux from TAW.

Our studies also indicate that despite the proximity of Kongsfjorden to the International Arctic Research Facilities of Ny-Ålesund as well as the growing importance of Svalbard as a summer tourist hub, the anthropogenic impact of heavy metals on the fjord sediments is negligible. The average concentrations of Mn, Cr, Zn, Co and Cu are comparable to, or in some cases even less than, the concentrations reported from other relatively pristine Arctic locales. Calculations of the Enrichment Factor of these elements also show that by and large, these elements are not or only slightly enriched relative to corresponding average world crustal values.

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