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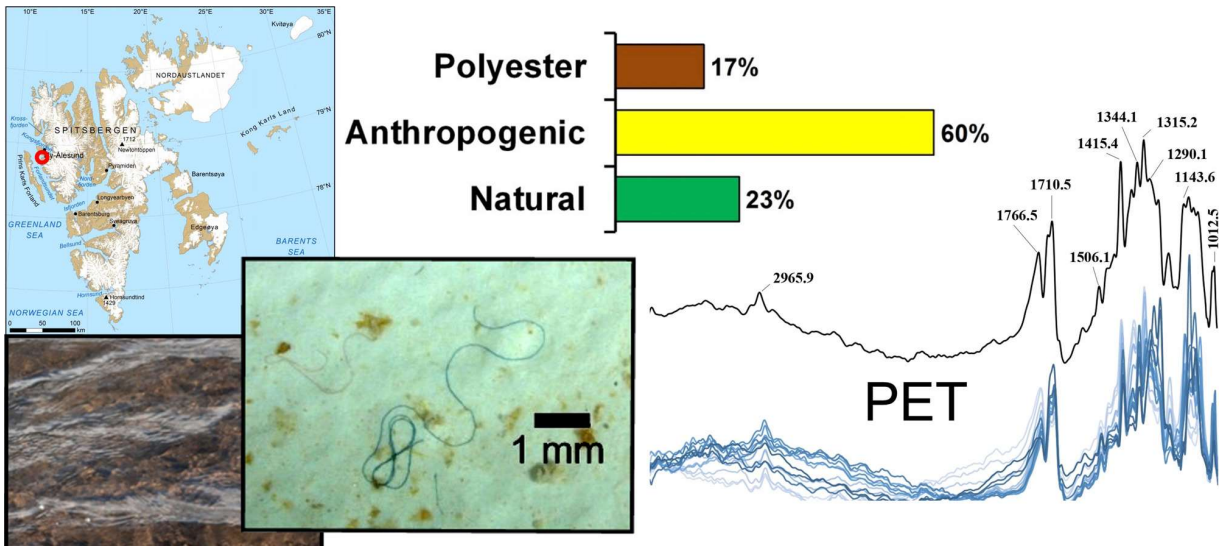

Fibers spreading worldwide: Microplastics and other anthropogenic litter in an Arctic freshwater lake

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Abstract

We investigated the presence of microplastics and other anthropogenic litter in the sediments adhered to rocks of an Arctic freshwater lake at Ny-Ålesund (Svalbard Archipelago, 78ºN; 11ºE). Most of the sampled microparticles were fibers (more than 90%). The identification of polymer types and additives was performed by combining three spectroscopic techniques, namely Raman Microscopy, Fourier-Transform Infrared microspectroscopy (μFTIR) and Synchrotron Radiation μFTIR (SR-FTIR). SR-FTIR confirmed the presence of poly(ethylene terephthalate) fibers, while RAMAN spectroscopy provided evidence of fibers containing industrial additives. Our results estimated an average concentration of 400 microparticles/m^2 of rocks identified as anthropogenic litter, which included an estimation of 90 microplastics/m^2 identified as polyester fibers; the rest are mostly natural fibers with evidence of anthropogenic origin. Taken together, the results proved the occurrence of anthropogenic pollutants in remote polar areas. Their probable origin is the long-range atmospheric transport.

Keywords: Microplastics, Additives, Anthropogenic fibers, Polar, Arctic lake

1. Introduction

Anthropogenic litter is recognized as a serious global environmental concern. Many studies have shown that it consists primarily of plastics (McCormick and Hoellein, 2016). In 2018, almost 360 million tonnes of plastics were produced globally (PlasticsEurope, 2019). The life span of plastic products may range between one to more than 50 years and, at the end of their life of use, plastic products become solid waste, which is normally treated through collection schemes. Wasted plastics without any proper post-use treatment pollute all the parcels in worldwide ecosystems and are further washed out through the freshwater networks into the Ocean, which acts as the final sink of most plastic debris (Thompson, 2006). According to Geyer et al. (2017) between 1950 and 2015, 6300 million T of plastic waste has been produced, and 79 % has been accumulated in landfills or the environment.

Following NOAA´s definition, plastics fragments below 5 mm are considered microplastics (Gago et al., 2016). Microplastic pollution in the environment greatly varies in shapes and sizes. Regarding shapes, microplastics may be found as spheres (microbeads or pellets), odd -shaped fragments of larger plastics or fibers. These synthetic fibers represent nearly 60% of world fiber consumption and the most common are those made of polyester, polyamide, acrylic polymers and polyolefins (FAO-ICAC, 2013; Cesa et al, 2017); they have a range of uses including packaging, textiles and fishing gear. Besides synthetic fibers, natural textile fibers (made of wool, cotton, or other cellulosic materials) eventually modified by anthropogenic processes may also end up in the environment. They may be a source of pollution as they may degrade faster and release compounds such as toxic additives and dyes used in their commercial production (Stanton et al, 2019; Cesa et al., 2017); thus, it should be considered that fibers present in the environment are not always made from plastic although they may have an anthropogenic origin. The wearing of clothes made from synthetic polymers has been recognized as an important source of environmental pollution. Polyester, acrylic, polyamides, and other synthetic fibers constitute most of the material clothes are made of nowadays. The laundry of synthetic fabrics releases a huge amount of fibers in the micron or tens of micron size range that reach the environment mainly through the effluent of wastewater treatment plants (Napper et al., 2016). Atmospheric fallout has also been identified as a source of microplastics from different origins (see e.g. Dris et al., 2016).

Microplastics have been found in surface waters, shorelines, continental waters, soils, sediments, and the

atmosphere at a global scale (Law et al, 2010; Cole et al., 2011; Browne et al, 2011; da Costa et al., 2016; Matsson et al, 2018; Allen et al, 2019; Zhang et al., 2019). Polar Regions besides its remoteness are no exception. In this context, microplastics have been detected in the Arctic in deep-sea sediments (Bergmann et al. 2017), seawater (Lusher et al 2015; Cincinelli et al 2017; Cozar et al 2017) and sea ice (Obbard et al., 2014, Peeken et al 2018). Up to our knowledge and despite freshwater lakes are one of the most important ecosystems in Polar Regions, the presence of microplastics has not been reported yet.

Freshwater bodies are the main features of surface waters in the Arctic. Arctic wetlands play an important role in ecosystem structure and function as well as in biogeochemical cycles. Those wetlands consist of shallow and perennial lakes and ponds that sustain most of the microbial biodiversity of the Arctic and provide natural refuges and feeding places for wildlife (Walseng et al., 2018). Therefore, even far-distant anthropogenic activities affect their ecological balance (Prowse et al. 2015; Emmerton et al. 2016; Walseng et al. 2018). In this sense, it has been proved that remote areas may receive a significant amount of microplastics due to atmospheric transport (Allen et al., 2019).

We hypothesized that Arctic freshwater lakes might be relevant sinks of microplastics and other anthropogenic litter; thus, here, we investigated their occurrence in a shallow freshwater lake in the High Arctic in the surroundings of Ny-Alesund (Svalbard, 78ºN; 11ºE), a coastal and summer ice-free area at Kongsfjorden (Svalbard Archipelago). Svalbard is a High Arctic Archipelago whose total land area is ca. 63 000 km^2 with two-thirds of this region permanently covered by perennial ice and glaciers (Humlum et al. 2003). Svalbard is also far from major sources of human pollution due to its remoteness (Birks et al. 2004a, b; Jiang et al., 2011). The aim of the study was to increase the knowledge about the microplastic pollution that reaches remote areas, which have been traditionally considered unaffected by any anthropogenic activity, and to understand better the environmental impact of this global pollution issue.

2. Materials and Methods

2.1. Study area and sample collection

The study area is a shallow lake with ~0.75 m maximum depth located at the flat wetland of Knudsenheia. This lake is in the western shore of Kongsfjorden 1 km away from the nearby research town of Ny-Ålesund (Spitsbergen, Svalbard Archipelago; 78.94765 °N, 11.81299 °E; Fig. S1A and Fig. S1B, Supplementary Material, SM). The lake is in a hyporheic area fed by a complex freshwater network running along the tundra plain from upstream glaciers that freezes during winter. The benthic zone consists of hand-size rocks and pebbles (~10 x 10 x 5 cm) covered by cohesive sediments (Fig. S1C, SM).

To determine the anthropogenic litter in sediments, we started by separating all microparticles contained in sediments. For it, rocks covering a total surface of 553 cm^2 were carefully recovered with their characteristic thin, muddy sediment layer (Fig. S1C, SM). All samples were stored in clean aluminum containers at room temperature until their analysis (Fig. S2A, SM). Procedural blanks were taken during sample collection and manipulation for microplastic counting validation. To do so, glass Petri dishes containing wet glass microfiber pads remained open during sampling and manipulation (Fig. S2F, SM). Microplastics and fibers similar in color and composition to those found in samples were subtracted from the total counting.

2.2. Quantification of microparticles

The organic matter in the muddy sediment that covers the rocks was removed by digestion with KOH. For it, the rocks covered by sediments were put in contact with 10 % KOH solution in glass beakers covered by aluminum foil for 24 h at 50 °C (Fig. S2B, SM) (Kühn et al., 2017). The resultant suspension was sieved using a 0.5 mm opening size stainless-steel meshes (Fig.S2C, SM). Particles both retained (> 0.5 mm) and non-retained (< 0.5 mm) were filtered using 1.5 μm cutoff glass microfiber filters (Fig.S2D and E, SM) for further analyses.

All filters were preserved in closed glass Petri dishes until analyses. All microparticles were inspected counted and classified using a Leica S8 APO stereomicroscope fitted with a Carl Zeiss Axiocam ERc5s camera. The images were treated using the ZEN Blue Edition image analysis software. Microparticles were measured and classified by size according to their larger dimension (< 0.3 mm, 0.3-0.5 mm, 0.5-1 mm, 1-2 mm, 2-5 mm and > 5 mm) and shape (fragments, fibers, filaments, films and paint sheets). Procedural blanks during quantification consisted of glass top-open Petri dishes with glass microfiber pads kept open during manipulation (Fig. S2F, SM). A 1 L sample of water from the same location was taken and studied in the same way in the search for anthropogenic microparticles.

2.3. Identification of microplastics and other anthropogenic fibers

A sample of 30 fibers (representing one -third of the total number of recovered microparticles) was analyzed using spectroscopic techniques. A subsample of six fibers was analyzed by means of Raman micro - spectroscopy. A Thermo Scientific DXR Raman Microscope equipped with Omnic software was used to acquire Raman spectra. Samples were sequentially observed using 10x, 20x, and 50x objectives and the spectra were taken 780 nm laser with 400 lines mm-1 grating and power adjusted depending on the

fluorescence of each sample. The spectra taken were analyzed in the range of 200-3100 cm^{-1}, spectral data spacing 0.964 cm^{-1}, with acquisition time fitted depending on the signal-to-noise ratio (Edo et al., 2019). μFTIR analyses were performed on 22 microfibers using a Perkin-Elmer Spotlight 200 apparatus with mercury cadmium telluride detector using the following parameters in micro-transmission mode: spot 50 mm, 32 scans, and spectral range 550-4000 cm^{-1} with 8 cm^{-1} resolution. μFTIR spectra were compared with Omnic 9 database and with spectra from our own database. Both in Raman and FTIR spectra, 60 % matching was considered enough for positive identification following Frías et al. (2016).

A subsample of two fibers was selected to be inspected by Synchrotron-based μFTIR (SR-FTIR) operating transmission mode with IR synchrotron from ALBA Synchrotron (Barcelona, Spain). The equipment used a Bruker Hyperion 3000 microscope coupled to a Vertex 70 spectrometer (Bruker Optik, Ettlingen, Germany). For SR-FTIR analyses, the selected fibers were placed on CaF_2 windows (10 mm diameter, 1 mm thick) and embedded in KBr aqueous solutions. Spectra were taken with 2 cm^{-1} resolution. Baseline correction and unit vector normalization were applied, and plots were denoised by means using the Savitsky-Golay smoothing filter. Data analyses were performed with OPUS 7.5 (Bruker) and Unscrambler® X 10.3 software (Camo Analytics, Oslo, Norway) and identification was carried out with the help of IR spectra library DEMOLIB.S01 (minimum hit quality 100 in the 900-3000 cm^{-1} wavenumber range).

2.4. Contamination Control

The possible contamination of samples with external materials was checked and quantified in every step of the present study. All glass materials were carefully cleaned with ultrapure water to remove all materials fixed in surfaces and covered with aluminum foil previously heated to 300 ºC. The deposition of airborne particles was measured using procedural blanks during sample collection and manipulation. For it, glass Petri dishes containing wet glass microfiber pads were kept open during sampling and manipulation (Fig. S2F, SM). Whenever possible, cotton was used for clothes. Nitrile blue gloves were always used. The color and material of clothes worn by people manipulating samples were also checked. Microplastics and fibers similar in color and composition to those found in samples were subtracted from the total counting. Controls didn't account for more than 10% of the total number of microparticles. Plastic materials in KOH solution were tested using a negative control without sample but undergoing the same digestion process.

3. Results and discussion

3.1. Microparticles in Knudsenheia freshwater lake

Potentially anthropogenic microparticles were detected in all sediments from the freshwater Arctic lake. 1 L of water taken from the same place was filtered using 0.5 mm size opening mesh without finding pieces of evidence of anthropogenic contamination (< 1 particle/L). 93 microparticles (< 5 mm) and 19 particles (> 5 mm) were found in the six different samples taken from the muddy sediment. Fibers were the most abundant category, representing 93.8 % of the total number of particles (n = 105), followed by fragments (2.7 %; n = 3), paint sheets (1.8 %; n = 2), filaments (0.9 %; n = 1) and films (0.9 %; n = 1). Concerning fiber length distribution, the 25th, 50th and 75th percentiles were 0.35, 0.67 and 1.40 mm, respectively. Fragments, paint sheet and films were smaller and mostly below 1 mm. The rock surface covered by sediments was accurately measured (553 cm^2) and used to determine an average concentration of 1680 ± 440 microparticles/m^2. The error represents the standard deviation calculated from different subsamples.

3.2. Identification and quantification of microparticles

The subsample used for chemical identification consisted of 30 microparticles, all of them fibers, which was by far the predominant shape. We analyzed particles of all size categories (8 below 0.3 mm, 50th percentile 1.1 mm). As indicated before, the microparticles in this subsample were analyzed using μFTIR (sample size = 22), Raman microscopy (sample size = 6), and SR-FTIR (sample size = 2). These results, together with the size distribution of all sampled fibers are shown in Fig. 1

Anthropogenic materials as indicated in Fig. 1 comprise all microparticles not unambiguously identified as synthetic polymers, but that show evidence of anthropogenic origin due to their color or the presence of additives. This category includes processed natural fibers and refers to those made from natural sources like wool, cotton or cellulose derivatives, but that underwent industrial processing and became altered with the introduction of artificial additives like dyes, bleaching agents, softening or stiffening additives, flame-retardants or light stabilizers (O'Brien et al., 2015). There is also the possibility of finding composites between natural and synthetic fibers (see e.g. Kuranska and Prociak, 2012). Such additives may reach the environment together with the fibers giving rise to another type of anthropogenic pollutant. Therefore, manufactured natural polymers, even if they are not made of plastic materials, should be monitored in view of their possible chemical risk associated with their additives due to their delivery into the environment.

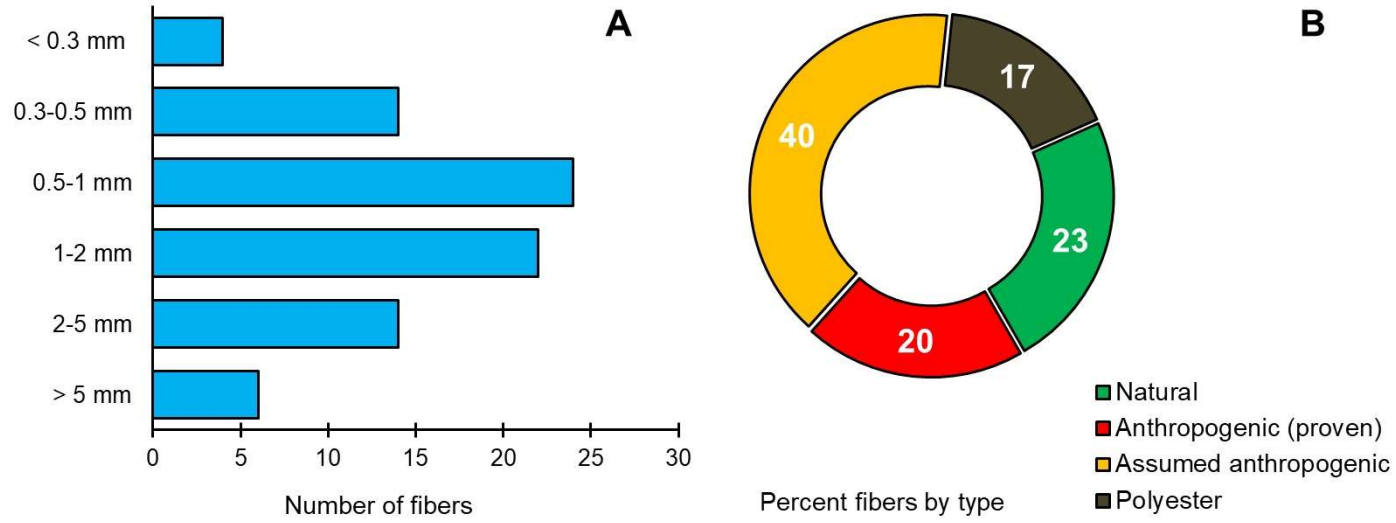

Figure 1. Size (length) distribution of all fibers recovered (A) and composition of the representative sample analyzed by FTIR or Raman spectroscopy (B). (The term anthropogenic refers to fibers not identified as synthetic polymers, but with evidence of anthropogenic origin.)

Raman spectra provided information about chemical additives in fibers. Fig. 2 shows the spectra of two of the fibers analyzed that, thanks to Raman spectroscopy, could be unambiguously identified as anthropogenic litter due to the presence of known industrial dyes. The indigo-colored fiber (Fig. 2A) revealed an indigoid dye by the main Raman peaks corresponding to the vibrations of the five-membered ring at 545 cm^{-1}, C=O at 600 cm^{-1} and that of C=C at 1574 cm^{-1} (Tatsch and Schrader, 1995). The blue dyed fiber is shown in Fig. 2B displayed a spectrum corresponding to copper phthalocyanine pigment as revealed by peaks at ~1530 cm^{-1} and ~1450 cm^{-1}, corresponding to vibrations originated by displacements of the C–N–C bridge bonds of the phthalocyanine macrocycle and benzene stretching vibrations respectively, among others (Basova et al., 2009). Van Cauwenberghe et al. (2013) also found copper -phthalocyanine pigment in microplastics in deep-sea sediments. μ-FTIR analyses (sample size = 22 fibers) allowed identifying three fibers as polyester (Fig. 3). These fibers displayed the characteristic carbonyl stretching peak at about 1700 cm^{-1}, the C–H stretching vibration in the region immediately below 3000 cm^{-1}. The peaks in the 1000-1300 cm^{-1} region corresponded to C–H bending of aromatic carbons and the C–O stretching of alkoxy ether, while the absorption at ~1012 cm^{-1} could be assigned to H-vibration attached to the aromatic ring (Shen et al., 1996).

The use of synchrotron radiation sources allowed high-resolution FTIR spectroscopy and the identification of polyester fibers as polyethylene terephthalate (PET) by differentiating from other polyesters with similar backbone. The reason behind the low resolution of μFTIR and Raman microscopy was the small size of fibers (Larkin, 2018). The spectra for the two fibers analyzed by SR -FTIR are shown in Fig. 4, which displays the characteristic carbonyl stretching peak at about 1710 cm^{-1} (Vijayakumar and Rajakumar, 2012). The maximum absorbance of this strong characteristic peak could appear shifted due to differences in the degree of crystallinity and chain orientation on deformation, which is the most probable reason explaining the difference between the two fibers shown in Fig. 4 (Chen et al., 2012). Other PET peaks were also clear in SR-FTIR including the C–H stretching

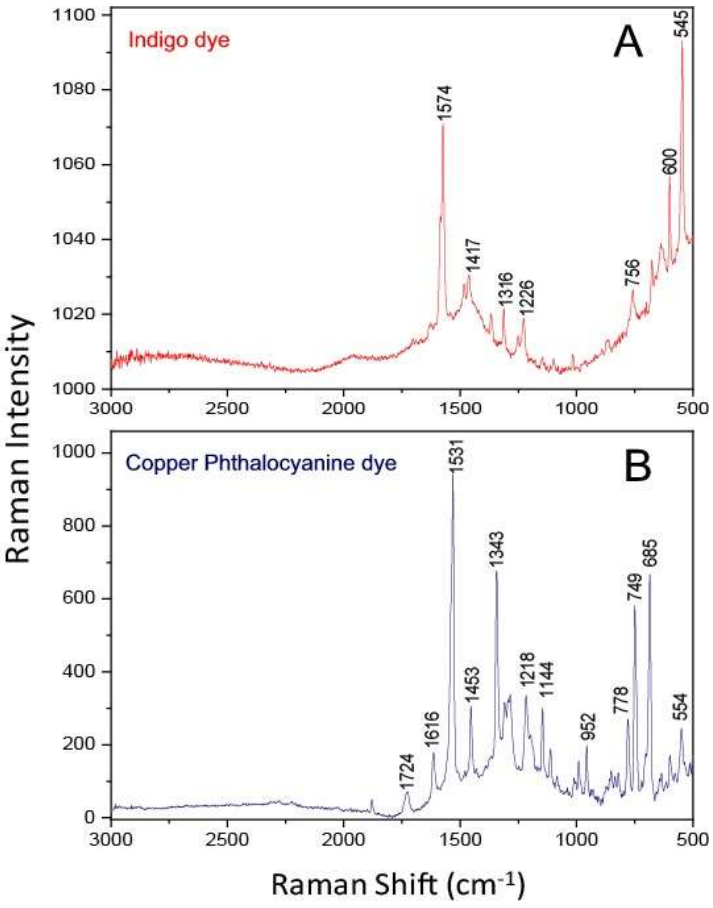

Figure 2. Raman spectra of two collected fibers with additives identified as indigo (A) and copper phthalocyanine (B) pigments.

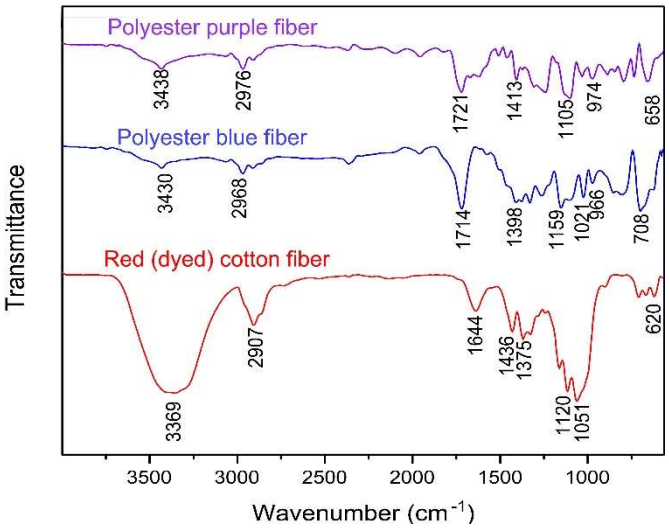

Figure 3. μFTIR spectra of three collected microparticles identified as polyester (two) and one fiber of artificially dyed cotton.

vibration in the region near 3000 cm^{-1}, the C–C stretching vibration in the benzene ring at about 1410 cm^{-1}, and the band at about 1015 cm^{-1}, which corresponds to the in plane bending vibration of the benzene C-H bonds (Wu et al., 2005). However, the most significant peak allowing a closer identification of the polyester as PET was the peak at about 1340 cm^{-1} that corresponds to O-C-H bending vibration and is typical form the PET trans conformer. This was the clearest difference with other polyester types that could be assessed due to the high resolution of SR-FTIR. This explains the high matching reached with PET, > 70 % for both fibers according to DEMOLIB.S01 spectra library.

Overall, our spectroscopic analyses found 5 polyester (PET) fibers, 6 anthropogenic fibers unambiguously identified in Raman spectra due to the presence of additives and 12 more assumed to be anthropogenic debris as they were natural fibers (wool, cotton or other cellulosic materials) with non -natural colors, which were not identified unequivocally due to the limited sensitivity of micro-FTIR to additives (Fig. 3). Our results showed that 17 % of the total number of collected fibers were polyester while an additional 60 % corresponded to other anthropogenic fibers. Micrographs of selected analyzed fibers are shown in Fig. 5.

Polar areas are not free of anthropogenic pollution and the Arctic is now considered a sink for microplastic particles (Cozar et al., 2017). Previously, the presence of microplastics has been reported in several marine environments in Arctic areas including surface water (Lusher et al. 2015), sub-surface water (Lusher et al. 2015; Kanhai et al 2018), snow (Bergmann et al., 2019), sea ice (Peeken et al 2018), deep-sea sediments (Bergmann et al 2017), and biota (Kühn et al 2018). Marine studies reported the presence of microplastics in Arctic waters with concentrations similar to other locations both in surface and sub-surface, which were attributed to long-distance transport (Obbard et al., 2014; Lusher et al., 2105). The types and shapes of microplastics found in Artic waters are generally coincident with studies in Southern seas, which is also suggesting transport from other latitudes. Contrary to sea ice or oceanic water column studies, we sampled a freshwater lake not directly connected with the Arctic Ocean. We found a limited typology of microplastics, with polyester as the only synthetic polymer, but a significant abundance of other anthropogenic materials, essentially fibers. Our results provide the first evidence of microplastics presence in Arctic freshwater bodies.

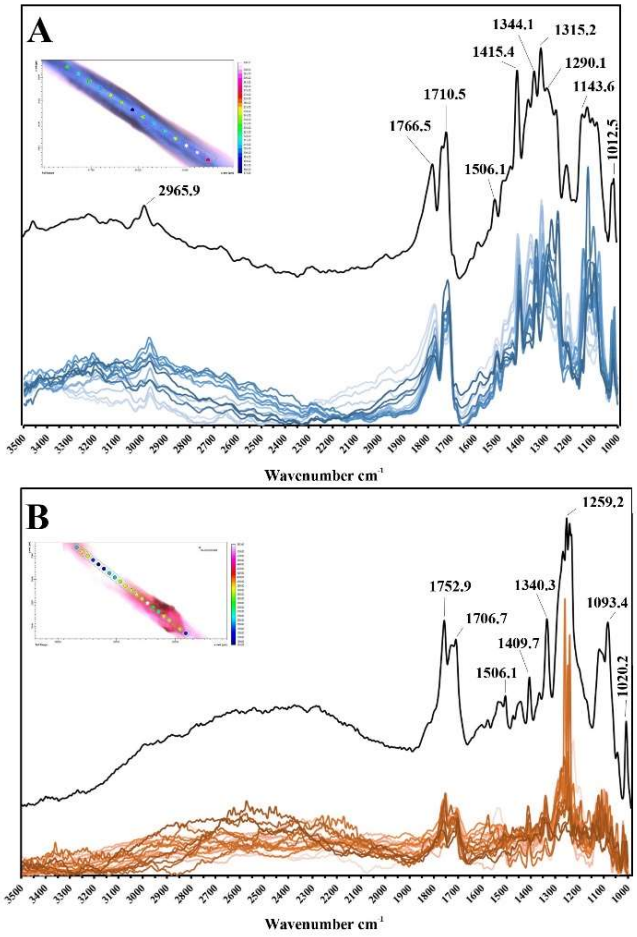

Figure 4. ST-FTIR spectra of the blue (A) and red fibers (B). Each panel shows compiled spectra and insets representing the spot location of IR-analysis within each specimen. The black lines represent mean spectra. The bars in each panel show the intensity scale for each group of spectra.

Most anthropogenic microparticles found in our study, either synthetic polymers or other materials with evidence of having undergone industrial transformations, corresponded to fibers, which have been found as the predominant shape in marine environments, particularly in sedimentary deep-sea bed habitats (Thompson et al, 2004; Woodall et al; 2014; Fischer et al., 2015). Bergmann et al. (2019) found high quantities of microplastics in Arctic deep-sea sediments but unfortunately they did not consider fibers. Woodall et al. (2014) calculated, as a conservative estimate

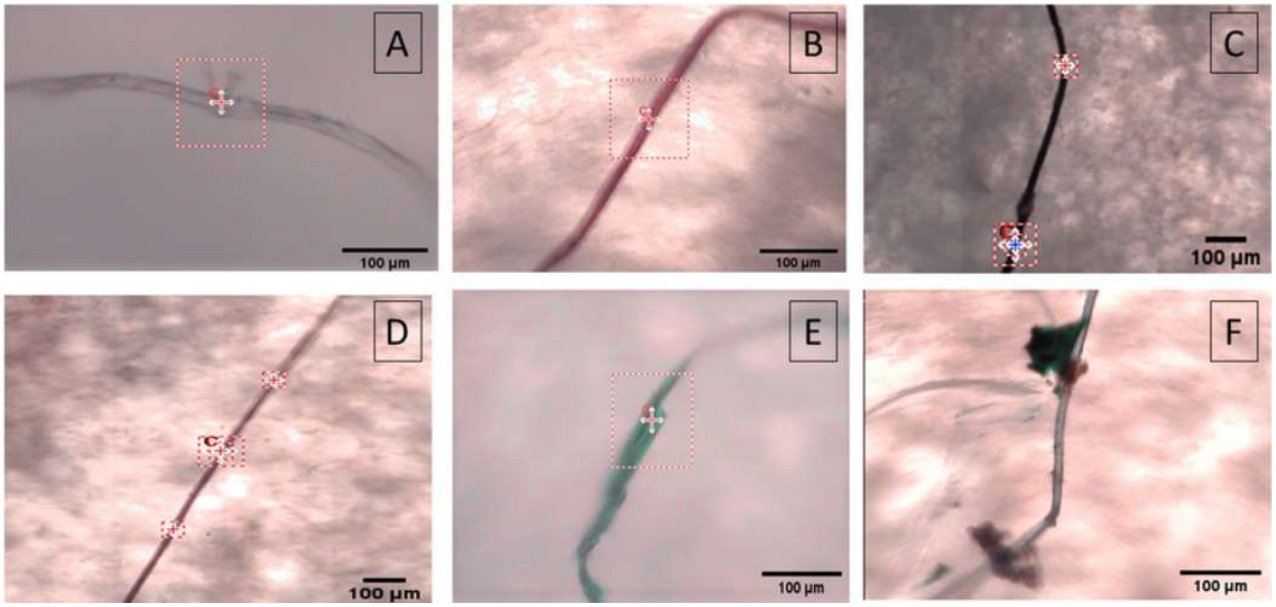

Figure 5. Examples of fibers found in sediments. A: cotton white fiber; B: wool red fiber; C: cotton black fiber; D: polyester violet fiber; E: cotton green fiber and F: polyester blue fiber.

4 x 10^3 fibers/km^2 in Indian Ocean sediments. Deep-sea sediments comprise a large repository of microplastics that might explain the missing fraction of marine debris in surface and subsurface marine habitats (Thompson et al, 2004). As compared to microplastics found in the water column such as polyethylene or polypropylene, which are buoyant, most fibers consist of polyester, polyamide and acrylic polymers, that sink to the sea floor due to their density higher than seawater (Cesa et al., 2017; Bergmann et al., 2019). Within synthetic fibers, polyester followed by acrylic fibers and polyamide dominated in sediments from the deep NE Atlantic, Mediterranean, SW Indian Ocean samples and subarctic sites; within non-plastic anthropogenic fibers, Rayon predominated (Woodall et al., 2014)

The majority of microparticles found in other polar studies (surface and subsurface waters and Arctic sea ice) were also fibers (Obbard et al., 2014; Lusher et al., 2015). Bergmann et al (2019) also identified a large number of fibers in their study of microplastics in snow from the Alps to the Arctic, although they did not provide information on the proportion of them being synthetic polymers. They discussed that atmospheric transport and deposition may be potential pathways for the substantial amounts of microplastic found in the snow, even in the Arctic. This agrees with other data reported in Polar locations, supporting the assumption that atmospheric transport, which is known to deposit in remote areas such as mountain lakes, may be responsible of their deposition (Free et al., 2014; Lusher et al., 2105). In this context, Dris et al, (2015) reported five times higher levels of microplastics after rain events and snow can clean aerosol particles to 50 times more efficiently than rain (Zhao et al., 2015; Bergmann et al., 2019).

The origin of the particles found in the lake is uncertain, but, given the absence of a tributary inlet from an inhabited area and the fact that fibers are the most abundant shape, wind-driven transport is hypothesized, either from local sources such as tourism and the nearby scientific station of Ny-Ålesund or from long-range transport (Zhang, 2019). In fact, polyester is one of the most used materials for manufacturing fleeces and other warm clothes (Carr, 2017).

In summary, from the total amount of 1680 microparticles/m^2, spectroscopic analyses allowed estimating a concentration of 400 microparticles/m^2 of anthropogenic litter. This included an average estimation of 90 microplastics/m^2 identified as solely polyester; the rest referring to fibers not identified as synthetic polymers, but with evidence of anthropogenic origin. Bergmann et al. (2017) found high quantities of microplastics in Arctic deep-sea sediments, reaching 2.47 x 10^5 microplastics/m^2 (retained by a 20 µm mesh) in one of their sampling stations in HAUSGARTEN observatory (Fram Strait). Lower amounts have been reported in sediments from the deep Atlantic Ocean and the Mediterranean Sea, with concentrations in the order of 200 microplastics/m^2 (retained by 35 µm sieve and converted from volume to area concentrations by Bergmann et al. (2017) (van Cauwenberghe et al., 2013). It is noteworthy that our figures, from a freshwater lake, are not too far from marine sediments, which emphasizes the large plastic pollution of Arctic lake sediments. Taken together, our results revealed that microplastics and other forms of anthropogenic pollution reach Arctic freshwater ecosystems and synthetic fibers appear together with a considerable number of natural fibers with evidence of anthropogenic processing. The ecological effects of anthropogenic pollution in Arctic areas are still uncertain, but their occurrence in relatively high amounts is a fact that needs further studies to assess the associated impacts.

4. Conclusions

We proved, for the first time, the presence of microplastics and other anthropogenic litter in an Arctic freshwater lake (located near Ny-Ålesund in the Svalbard Archipelago). The sediments covering rocks in the bed contained a large proportion (93.8 %) of fibers among sampled microparticles

The characterization of microparticles, performed by three spectroscopic techniques, namely Raman microscopy, μFTIR and SR-FTIR, allowed confirming the presence of poly(ethylene terephthalate) and a variety of other anthropogenic fibers that consisted of natural fibers with industrial additives and/or non-natural colors.

The use SR-FTIR technique was essential to confirm the presence of poly(ethylene terephthalate) fibers, which together with non-natural fibers allowed estimating a concentration of anthropogenic litter of 400 microparticles/m^2 of sediment. Our study indicates that Arctic lakes may be relevant sinks of microplastics, specially microfibers. A probable origin of sampled fibers is atmospheric deposition from local and distant sources

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References

Allen, S., Allen, D., Phoenix, V. R., Le Roux, G., Jiménez, P.D., Simonneau, A., Binet S., Galop, D. Atmospheric transport and deposition of microplastics in a remote mountain catchment. Nature Geosci. 12, 339-344, 2019.

Basova, T.V., Kiselev, V.G., Schuster, B.E., Peisert, H., Chassé, T. Experimental and theoretical investigation of vibrational spectra of copper phthalocyanine: Polarized single-crystal Raman spectra, isotope effect and DFT calculations. J. Raman Spectroscopy, 40, 2080-2087, 2009.

Bergmann, M., Wirzberger, V., Krumpen, T., Lorenz, C., Primpke, S., Tekman, M.B., Gerdts, G. High quantities of microplastic in Arctic deep-sea sediments from the Hausgarten observatory. Environ. Sci. Technol. 51, 11000-11010, 2017.

Bergmann, M., Mützel, S., Primpke, S., Tekman, M.B., Trachsel, J., Gerdts, G. White and wonderful? Microplastics prevail in snow from the Alps to the Arctic. Sci. Adv. 5, eaax1157. 2019.

Birks, H.J.B., Jones, V.J., Rose, N.L. Recent environmental change and atmospheric contamination on Svalbard as recorded in lake sediments–synthesis and general conclusions. J. Paleolimnology, 31, 531-546, 2004a.

Birks, H.J.B., Monteith, D.T., Rose, N.L., Jones, V.J., Peglar, S.M. Recent environmental change and atmospheric contamination on Svalbard as recorded in lake sediments–modern limnology, vegetation, and pollen deposition. J. Paleolimnology, 31, 411-431, 2004.

Browne, M. A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R. Accumulation of microplastic on shorelines worldwide: Sources and sinks. Environ. Sci. Technol. 45, 9175-9179, 2011.

Carr, S.A. Sources and dispersive modes of micro-fibers in the environment. Integr. Environ. Assess. Manage., 13, 466-469, 2017.

Cesa F. S, Turra A., Baruque-Ramos J. Synthetic fibers as microplastics in the marine environment: A review from textile perspective with a focus on domestic washings. Sci. Total Environ. 598, 1116–1129, 2017.

Cincinelli, A., Scopetani, C., Chelazzi, D., Lombardini, E., Martellini, T., Katsoyiannis, A., Fossi, M.C., Corsolini, S. Microplastic in the surface waters of the Ross Sea (Antarctica): occurrence, distribution and characterization by FTIR. Chemosphere, 175, 391-400, 2017.

Cole, M., Lindeque, P., Halsband, C., Galloway, T.S. Microplastics as contaminants in the marine environment: A review. Marine Poll. Bull. 62, 2588-2597, 2011.

Cózar, A., Martí, E., Duarte, C. M., García-de-Lomas, J., Van Sebille, E., Ballatore, T.J., Eguíluz, V.M., González-Gordillo, J.I., Pedrotti, M.L., Echevarría, F., Troublè, R. The Arctic Ocean as a dead end for floating plastics in the North Atlantic branch of the Thermohaline Circulation. Sci. Adv. 3, e1600582, 2017.

Chen, Z., Hay, J.N., Jenkins, M.J. FTIR spectroscopic analysis of poly(ethylene terephthalate) on crystallization. Eur. Polym. J. 48, 1586-1610, 2012.

da Costa, J.P., Santos, P.S., Duarte, A.C., Rocha-Santos, T. (Nano)plastics in the environment–sources, fates and effects. Sci. Total Environ. 566, 15-26, 2016.

Dris, R., Gasperi, J., Rocher, V., Saad, M., Renault, N., Tassin, B. Microplastic contamination in an urban area: A case study in Greater Paris. Environ. Chem. 12, 592–599, 2015.

Dris, R., Gasperi, J., Saad, M., Mirande, C., Tassin, B. Synthetic fibers in atmospheric fallout: A source of microplastics in the environment? Mar. Pollut. Bull. 104, 290-293, 2016.

Edo, C., Tamayo-Belda, M., Martínez-Campos, S., Martín-Betancor, K., González-Pleiter, M., Pulido-Reyes, G., García-Ruiz, C., Zapata, F., Leganes, F., Fernández-Pinas, F., Rosal, R., 2019. Occurrence and identification of microplastics along a beach in the biosphere reserve of Lanzarote. Mar. Pollut. Bull. 143, 220-227.

Edo, C., González-Pleiter, M., Leganés, F., Fernández-Piñas, F., Rosal, R., Fate of microplastics in wastewater treatment plants and their environmental dispersion with effluent and sludge. Environ. Pollut. 259, 113837, 2020

Emmerton, C.A., Louis, V.L.S., Lehnherr, I., Graydon, J.A., Kirk, J.L., Rondeau, K.J. The importance of freshwater systems to the net atmospheric exchange of carbon

dioxide and methane with a rapidly changing high Arctic watershed. Biogeosciences, 13, 5849-5863, 2016.
Fischer, V., Elsner, N.O., Brenke, N., Schwabe, E., Brandt, A. Plastic pollution of the Kuril-Kamchatka Trench area (NW pacific). Deep -Sea Res. II 111:399–405, 2015.
FAO-ICAC. Food and Agriculture Organization of the United Nations and International Cotton Advisory Committee. World Apparel Fiber Consumption Survey, Washington. Retrieved from: https://www.icac.org/cotton_info/publications/statistics/world-apparel-survey/FAO-ICAC-Survey-2013-Update-and-2011-Text.pdf. 2013
Free, C.M., Jensen, O.P., Mason, S.A., Eriksen, M., Williamson, N.J., Boldgiv, B. High-levels of microplastic pollution in a large, remote, mountain lake. Mar. Pollut. Bull. 85, 156–163, 2014.
Fuller, S., Gautam, A. A procedure for measuring microplastics using pressurized fluid extraction. Environ. Sci. Technol. 50, 5774-5780, 2016.
Frias, J. P. G. L.; Gago, J.; Otero, V.; Sobral, P. Microplastics in coastal sediments from Southern Portuguese shelf waters. Mar. Environ. Res. 2016, 114, 24–30.
Gago, J., Galgani, F., Maes, T., Thompson, R.C. Microplastics in seawater: Recommendations from the Marine Strategy Framework Directive implementation process. Frontiers in Marine Science 3, 2016.
Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made. Science advances 3, e1700782.
Gigault, J., Halle, A.T., Baudrimont, M., Pascal, P.Y., Gauffre, F., Phi, T.L., El Hadri, H., Grassl, B., Reynaud, S. Current opinion: What is a nanoplastic? Environ. Pollut. 235, 1030-1034, 2018.
Humlum, O., Instanes, A., Sollid, J.L. Permafrost in Svalbard: a review of research history, climatic background and engineering challenges. Polar Res. 22, 191-215, 2003.
Jiang, S., Liu, X., Sun, J., Yuan, L., Sun, L., Wang, Y. A multi-proxy sediment record of late Holocene and recent climate change from a lake near Ny-Ålesund, Svalbard. Boreas, 40, 468-480, 2011.
Kanhai, L.D.K., Gårdfeldt, K., Lyashevska, O., Hassellöv, M., Thompson, R.C., O'Connor, I. Microplastics in sub-surface waters of the Arctic Central Basin. Mar. Pollut. Bull. 130, 8-18, 2018.
Koelmans, A.A., Besseling, E., Shim, W.J. Nanoplastics in the aquatic environment. Critical review. In: Bergmann, M., Gutow, L., Klages, M. (Eds.), Marine Anthropogenic Litter. Springer International Publishing, Cham, pp. 325-340, 2015.
Kühn, S., van Werven, B., van Oyen, A., Meijboom, A., Bravo-Rebolledo, E.L., van Franeker, J.A. The use of potassium hydroxide (KOH) solution as a suitable approach to isolate plastics ingested by marine organisms, Mar. Pollut. Bull., 115, 86-90, 2017.
Kühn, S., Schaafsma, F.L., van Werven, B., Flores, H., Bergmann, M., Egelkraut-Holtus, M., Tekman, M.B., van Franeker, J.A. Plastic ingestion by juvenile polar cod (Boreogadus saida) in the Arctic Ocean. Polar Biology 41, 1269-1278, 2018.
Kuranska, M., Prociak, A. Porous polyurethane composites with natural fibers. Compos. Sci. Technol. 72, 299-304, 2012.
Larkin, P.J. Instrumentation and Sampling Methods. In: Peter J. Larkin (Ed.) Infrared and Raman Spectroscopy, 2nd Edition, 29-61, Elsevier, 2018.
Law, K. L., Morét-Ferguson, S., Maximenko, N. A., Proskurowski, G., Peacock, E.E., Hafner, J., Reddy, C.M. Plastic accumulation in the North Atlantic subtropical gyre. Science 329, 1185-1188, 2010.
Lusher, A.L., Tirelli, V., O'Connor, I., Officer, R. Microplastics in Arctic polar waters: The first reported values of particles in surface and sub-surface samples. Sci. Rep. 5, 14947, 2015
McCormick, A.R., Hoellein, T.J.2. Anthropogenic litter is abundant, diverse, and mobile in urban rivers: Insights from cross -ecosystem analyses using ecosystem and community ecology tools. Limnology and Oceanography 61: 1718-1734, 2016.
Napper, I.E., Thompson, R.C. Release of synthetic microplastic plastic fibres from domestic washing machines: Effects of fabric type and washing conditions. Mar. Pollut. Bull. 112, 39-45, 2016.
Obbard, R.W. Microplastics in Polar Regions: The role of long range transport. Curr. Opin. Environ. Sci. Health 1, 24–29, 2018.
Obbard, R. W., Sadri, S., Wong, Y.Q., Khitun, A.A., Baker, I., Thompson, R.C. Global warming releases microplastic legacy frozen in Arctic Sea ice. Earth's Future, 2, 315-320, 2014.
O'Brien, J.W., Thai, P.K., Brandsma, S.H., Leonards, P.E.G., Ort, C., Mueller, J.F. Wastewater analysis of Census day samples to investigate per capita input of organophosphorus flame retardants and plasticizers into wastewater. Chemosphere 138, 328-334, 2015.
Peeken, I., Primpke, S., Beyer, B., Gütermann, J., Katlein, C., Krumpen, T., Bergmann, M., Hehemann, L., Gerdts, G. Arctic sea ice is an important temporal sink and means of transport for microplastic. Nature Comm., 9, 1-12, 2018.
PlasticsEurope, 2019. Plastics– the Facts 2019: An analysis of European plastics production, demand and waste data. PlasticsEurope: Association of Plastics Manufacturers, Brussels.
Prowse, T., Bring, A., Mård, J., Carmack, E., Holland, M., Instanes, A., Vihma, T., Wrona, F.J. Arctic Freshwater Synthesis: Summary of key emerging issues. J. Geophys. Res.: Biogeosci. 120, 1887-1893, 2015
Shen, F.W., McKellop, H.A., Salovey, R. Irradiation of chemically crosslinked ultrahigh molecular weight polyethylene. J. Polym. Sci. Part B, 34, 1063–1077, 1996.
Stanton T., Johnson M., Nathanail P., MacNaughtan W., Gomes R.L. Freshwater and airborne textile fibre populations are dominated by 'natural', not microplastic, fibres. Sci. Total Environ. 666, 377–389, 2019.
Tatsch, E., Schrader, B. Near-Infrared Fourier Transform Raman Spectroscopy of Indigoids. J Raman Spectroscopy, 26, 467-473, 1995.
Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., Mcgonigle, D., Russel, A.E.. Lost at Sea: where is all the plastic? Science 5672:838, 2004.
Thompson, R. C. Plastic debris in the marine environment: Consequences and solutions. In J. C. Krause, H. Nordheim, & S. Bräger (Eds.), Marine nature conservation in Europe. Stralsund, Germany: Bundesamt für Naturschutz. pp. 107–115, 2006

van Cauwenberghe, L.; Vanreusel, A.; Mees, J.; Janssen, C. R. Microplastic pollution in deep-sea sediments. Environ. Pollut. 182, 495−499, 2013.
Vijayakumar, S., Rajakumar, P.R. Infrared spectral analysis of waste pet samples. Int. Let. Chem. Phys. Astron. 4, 58-65, 2012.
Walseng, B., Jensen, T., Dimante -Deimantovica, I., Christoffersen, K.S., Chertoprud, M., Chertoprud, E., Novichkova, A., Hessen, D.O. Freshwater diversity in Svalbard: providing baseline data for ecosystems in change. Polar Biology, 41, 1995-2005, 2018.
Woodall, L.C., Sánchez -Vidal, A., Canals,M., Paterson, G.L.J., Coppock, R., Sleight, V., Calafat, A., Rogers, A.D., Narayanaswamy, B.E., Thompson, R.C. The deep sea is a major sink for microplastic debris. R. Soc. Open Sci. 1 (140317):1–8, 2014.
Wu, T., Li, Y. Wu, Q., Song, L., Wu, G. Thermal analysis of the melting process of poly(trimethylene terephthalate) using FTIR micro-spectroscopy. Eur. Polym. J. 41, 2216-2223, 2005.
Zhang, Y., Gao, T., Kang, S., Sillanpää, M. Importance of atmospheric transport for microplastics deposited in remote areas. Environ. Pollut. 254, 112953, 2019.
Zhao, S., Yu, Y., He, J., Yin, D., Wang, B. Below-cloud scavenging of aerosol particles by precipitation in a typical valley city, northwestern China. Atmos. Environ. 102, 70–78, 2015.

SUPPLEMENTARY MATERIAL

Fibers spreading worldwide: Microplastics and other anthropogenic litter in an Arctic freshwater lake

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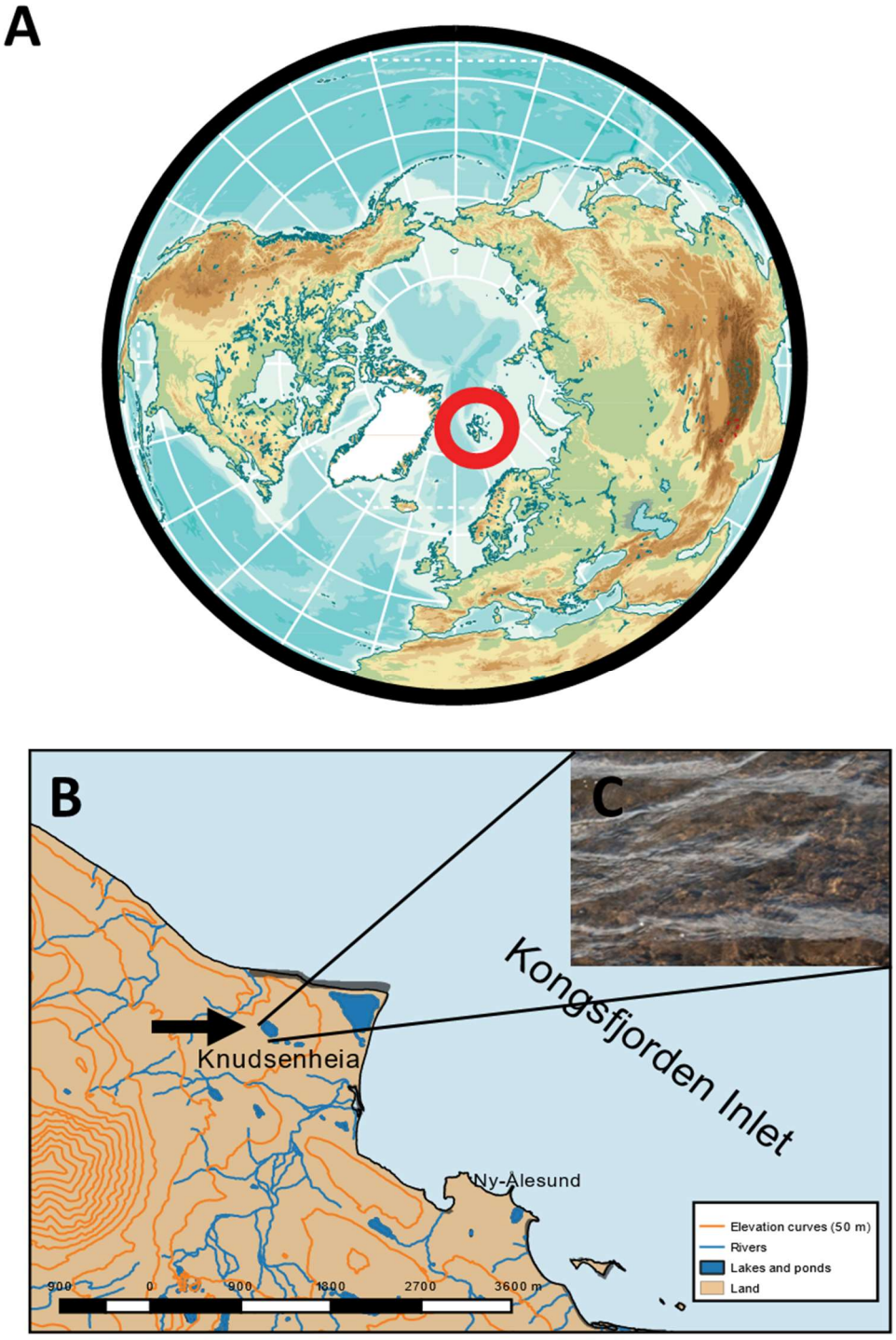

Figure S1. Location of the study area. A) General view of the study area: the red circle shows Svalbard Archipelago. B) Lake location (black arrow) on Spitsbergen Island. C) The lake bed consists of rocks covered with a thin layer of sediment.

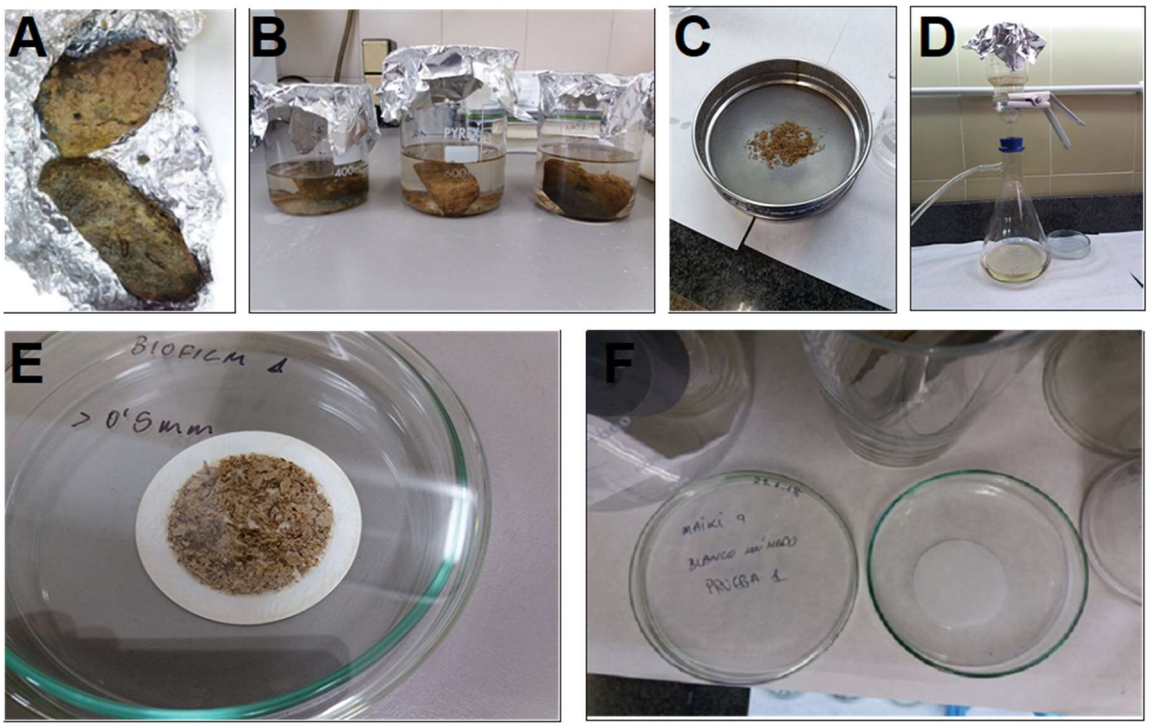

Figure S2. Scheme of the experimental procedure. A) Rocks covered with a thin layer of sediment collected at the bottom of the lake. B) Sediments on the rocks digested with KOH 10% (m/v) solution at 50 ºC for 24 hours. C) Sediments retained in > 0.5 mm sieve. D) Filtration device. E) Glass microfiber filters with sediments. F) Procedural blanks: Glass Petri dishes with glass microfiber filters that remained open during the experimental procedure.