Presence of nanoplastics in rural and remote surface waters

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Abstract

It is now established that microplastics are a pervasive presence in aquatic and terrestrial ecosystems. The same is assumed to be true for nanoplastics but data are lacking due to technical difficulties associated with sample analysis. Here, we measured nanoplastics in waterbodies at two contrasting sites: remote Siberian Arctic tundra and a forest landscape in southern Sweden. Nanoplastics were detected in all sampled Swedish lakes ($n = 7$) and streams ($n = 4$) (mean concentration = $563 \mu\text{g l}^{-1}$) and four polymer types were identified. In Siberia nanoplastics were detected in 7/12 sampled lakes, ponds and surface flooding, but only two polymer types were detected and concentrations were lower (mean $51 \mu\text{g l}^{-1}$). Based on back-calculation of air mass trajectories, we infer that nanoplastics arrive at both sites by aerial deposition from local and regional sources. Our results suggest that nanoplastics may be a near-ubiquitous presence even in remote ecosystems.

Introduction

Since the commercialisation of plastics polymers in the 1950s plastic usage has expanded rapidly, with ever-increasing yearly production, resulting in an estimate of >9300 million tons (Mt) produced globally to date ^{1,2}. However, 60% of all plastics ever produced have been discarded, either to landfills or the environment ². Much of the 360 Mt yr^{-1} of newly produced plastics are used in single-use items that are disposed of within a few years of production ^{1,2}. If deposited in the environment, plastics fragment to microplastics (pieces <5mm), and then fragment further to nanoplastics (<1 μm particles), particularly if exposed to air and UV radiation ³⁻⁵. This large plastic load has significant implications for aquatic and terrestrial ecosystem functioning, and for human health ⁶⁻⁹.

Microplastics have already been found in many environments, including marine waters, aquatic sediments, freshwater systems, soils, and the atmosphere. These reports span both urban and remote locations ^{6,10-16}. However, the research field is still relatively young and our ability to quantify ecosystem microplastic budgets, including major pools, sources and sinks, is still under-developed ¹⁷. Urban areas are primary sources and thus focal points for emissions ². Synthetic fibres and single-use products are a significant source of microplastics to the environment because microfibrils can be carried by water and air and can reach remote places including wilderness areas, mountaintops, and polar regions ^{10,12,18-20}.

Compared to microplastics, considerably less is known about the fate of nanoplastics in the environment and their abundance in different ecosystems. This is largely due to the technical difficulties associated with accurately analysing samples for nanoplastics. Only a handful of results have been reported to date but these limited measurements indicate the presence of nanoplastics in the ocean, in beach sediments, rivers, and high-altitude montane snow ^{19,21-24}. These early results suggest that nanoplastics can be subject to long-range transport and be present in the environment at high concentrations (i.e. concentrations of the same magnitude as microplastics) ^{19,23,25,26}. It is thought that the toxicity of nanoplastics to living organisms is greater than that of microplastics because nanoplastics can be

ingested easily (even crossing cell membranes) and thus can bioaccumulate^{7,27}. Nanoplastics have been shown to have various adverse effects on organismal biology: for example, they can denature proteins by changing the three-dimensional structure of them²⁸ and can inhibit microbial growth and metabolism²⁹. The consequences of nanoplastic ingestion on the organismal level include behavioural abnormality, inhibited development, and early mortality⁹. The effects of nanoplastics on human biology is largely unknown but it does appear that they can induce cellular responses, with immune system reactions being of particular concern²⁷. Thus, more research is urgently needed to address this novel environmental contaminant.

Here, we use Thermal Desorption – Proton Transfer Reaction – Mass Spectroscopy (TD-PTR-MS) to measure the mass-concentrations of nanoplastics in the surface waters from two contrasting sites; a remote tundra landscape in the far east Siberian Arctic, and a forested area close to population centres in Sweden. Our aim was to investigate the type and quantity of nanoplastics, both within and between sites, in order to make inferences about sources, deposition pathways, and removal processes.

Results And Discussion

Nanoplastics concentration and types

Sampling took place at two locations. The first was the Gårdsjön research catchment (58°03'35"N 12°01'18"E) in south west Sweden. Here, seven lakes and four streams were sampled in August 2017. The second sampling location was the Kytalyk Nature Reserve in the Indigirka River lowlands in north east Siberia (70°49'48"N 147°29'24"E). Here, samples were collected from three lakes, five ponds, overland flow from thawing permafrost and flooded tundra in July 2017. (Figures 1 and 2). Samples were analysed for polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polyethylene terephthalate (PET) and polystyrene (PS).

The mean concentration of total nanoplastics for the Swedish sites was 563 $\mu\text{g l}^{-1}$ (Table 1). This value can roughly be translated to the number concentration (see material and methods) of 1.3×10^{11} nanoplastics particles l^{-1} . The highest concentrations were observed for PE (Figure 3, mean = 272 $\mu\text{g l}^{-1}$), which is the most common plastic in use in Europe¹. Mean concentrations of other detected polymer types were: PP 141 $\mu\text{g l}^{-1}$, PVC 117 $\mu\text{g l}^{-1}$, and PET 33 $\mu\text{g l}^{-1}$. PE was found in all samples, whereas PVC and PP were found in 7/11 samples, and PET was found in six. We did not detect PS in any Swedish samples.

At the Swedish site we were unable to discern any empirical relationships between plastic concentrations and waterbody properties (lake surface area, catchment area, water retention time) or chemistry (pH, dissolved organic carbon concentration) but some polymer-specific patterns in spatial distribution were observed. PVC was not found in any of the three headwater streams, which also showed relatively lower concentrations of PP and PE (Table 1, Figure 3): mean concentrations of these two polymers were only 63% and 45% of the mean of all samples, respectively. This may indicate that trees and soil in the

surrounding forest may have a role in retaining deposited plastics and limiting their subsequent transfer into streams. Intriguingly, PET was entirely absent from the chain of sampled lakes (Figure 1, sites 3, 4, 5, 9) despite PET being detected in three streams that enter the lake chain (sites 2, 6, 7). This could point towards burial of PET during lake sediment deposition, which can be an efficient sink process in aquatic sediments³⁰. Alternatively, removal could be via in-lake photodegradation which is capable of fully or partially photochemically oxidising PS (and therefore potentially other polymers) to carbon dioxide or dissolved organic carbon³¹. Spatial concentrations of PP were variable; although PP was present in much of the Gårdsjön catchment, it was not detected at the catchment outflow, nor the next lake downstream (Stora Bjurevatten) which also receives water from other lakes and rivers we did not sample. As for PET, this raises the possibility of degradation or burial of PP within lakes. PP was also absent from Västersjön, which is situated in an entirely separate watershed and, unlike the other sites, has a large number of dwellings in its immediate vicinity. Thus, direct local contamination is unlikely to be a pathway for PP to enter waterbodies in this area.

We detected PVC in four (mean = $109 \mu\text{g l}^{-1}$) and PS in five (mean = $15 \mu\text{g l}^{-1}$) of the 12 Siberian sampling sites – the average NP concentration was $51 \mu\text{g l}^{-1}$ (Table 1, Figure 3). The estimated number concentrations are 2.6×10^{10} NP particles l^{-1} for PVC and 3.6×10^9 NP particles l^{-1} for PS. No traces of PP, PE or PET were found. Both PVC and PS were present at very low concentrations ($< 10 \mu\text{g l}^{-1}$) in one lake (L10), and PS was also detected ($24 \mu\text{g l}^{-1}$) in the adjacent, but hydrologically unconnected lake, just 200 m away. One pond had very high concentrations ($354 \mu\text{g l}^{-1}$) of PVC (higher than any of the Swedish sites where the maximum was $264 \mu\text{g l}^{-1}$), whilst two other ponds had very low PS concentrations. The high-PVC pond formed following a plant removal experiment in 2007 where monitoring is ongoing^{32,33} so it may be that plastic contamination from long-term research activity has occurred. Similar contamination may explain the moderately high ($58 \mu\text{g l}^{-1}$) PVC concentration detected in a sample taken from flooded tundra. This flood washed through the nearby research campsite and also covered some infrastructure associated with a weather station and eddy covariance flux tower³⁴. Low concentrations of PVC ($16 \mu\text{g l}^{-1}$) and PS ($7 \mu\text{g l}^{-1}$) were also found in water draining a small thermokarst slump. The most probable explanation for the presence of nanoplastics in water directly draining from material that has previously been frozen for $>50,000$ years³⁵ is that overland flow is collecting plastics that have been deposited on the terrestrial surface rather than indicating the presence of nanoplastics within the permafrost.

An aerial origin of nanoplastics

Nanoplastics could enter surface waters at the study locations via three pathways: soil, water or air. At the Swedish site, there are no obvious local sources that could directly discharge such a variety of plastics into the soils or waters. The land use is almost exclusively forest, with exceptions of a small number of dwellings (~ 50 and 80 , respectively) within the catchments of Stora Bjurevatten and Västersjön. There are no agricultural fields that, if present, could contribute large inputs of PE (e.g. via agricultural PE foil). Some PET could possibly arrive via local recreational activity and associated synthetic clothing¹⁸; a long-distance hiking trail (Bohusleden) runs through the Gårdsjön catchment, and

some local residents fish on Lake Gårdsjön. It is unlikely that these small-scale activities are responsible for the widespread and relatively high concentrations of aquatic nanoplastics. Thus, we suggest that the measured nanoplastics at the Swedish site arrive via atmospheric deposition. Supporting this is the fact that lower concentrations of PP and PE were found in the three headwater streams, alongside the complete absence of PVC. These streams are predominantly groundwater fed ³⁶. In forest ecosystems, it is conceivable that the high leaf surface area of the trees might act to remove nanoplastics from the air; a similar mechanism to the “forest filter effect” ³⁷. Nanoplastics deposited on the surfaces of trees will then be transferred to the soil (via rainfall, litterfall, etc) ³⁸ where some will likely be retained (either temporarily or permanently), then migrating into groundwater, and subsequently leaching into stream water ³⁹. This soil retention could partially explain the lower concentrations of nanoplastics, and absence of PVC, in our headwater streams. In contrast to the headwater streams, the larger areas of the lakes provide a substantial surface for direct atmospheric deposition; e.g., for the sampled lakes within the Gårdsjön catchment, lake surfaces occupy 20-30% of total subcatchment area.

The backward trajectory of air movements for the Swedish site indicates that the likely sources of the nanoplastics are within north west Europe (Figure 4). This is in agreement with research demonstrating that acidified lakes waters ⁴⁰ in the area were caused by moderately high levels of atmospheric acid deposition ⁴¹, the majority of which was the result of long-range pollutant transport from industry in the UK and central Europe ⁴². Long-range atmospheric transport of microplastics has been demonstrated over distances of hundreds and even thousands of kilometres, and modelling suggests that nanoplastics can be transported over greater distances ^{10,12,16,18,43–45}.

The wide variety of nanoplastics measured could have come from many point sources within the footprint area (e.g. urbanised and industrially developed western Europe). However, there is a waste-to-energy plant 40 km south of the catchment, on the boundary of the prevailing wind direction and within 90% of the wind trajectory, which could be acting as a regional point source. Although waste-to-energy plants have air filters, possible nanoplastic emissions have never been investigated.

PE is the dominant nanoplastic type at the Swedish sites, and was even found in the groundwater-fed streams. One explanation for this is that PE is the commonest plastic in use in Europe, and large volumes of it are used for single-use packaging material, which are subsequently disposed of. After disposal, PE might fragment into micro- and nanoplastics via various natural and industrial processes (incineration, UV light-induced fragmentation, weathering, mechanical and biodegradation etc.). The absence of PE in the Siberian samples and in other analyses from the remote Alps ¹⁹ suggests the possible dominance of a local source. In agreement with this idea, there is a major PE producer 12 km to the west of the Swedish sites, on the boundary of the prevailing wind direction.

In Siberia only PVC and PS were detected. The Siberian site is far away from large urban areas (~1200 km to the nearest city, Yakutsk, which falls outside of the HYSPLIT trajectory model footprint, Figure 5) which typically act as emission sources of atmospheric plastics (Figure 2 and 5). As discussed earlier, the

high-PVC concentrations observed in some locations here are plausibly the result of local contamination from regular visitors to the site over several years. However, the potentially contaminated sites showed only PVC, and no PS. Because PS was found in trace amounts in other ponds and lakes, at a distance of up to 4 km from the camp, it seems likely that direct local contamination is not the source of PS. Within the wider local area there are sparsely scattered hunting and fishing camps, and a settlement with a population of ~2000 is 30 km away. Additionally, within several hundred kilometers there are several more settlements of similar size, as well as abandoned mining stations, which could act as sources. We therefore assume that PS is transported to the site via aerial deposition; other studies have concluded that this is a viable transport pathway for plastics to reach remote sites^{10,12,46}. The presence of PS and PVC in water draining a thermokarst slump also lends weight to the atmospheric deposition hypothesis by suggesting that plastic is deposited to both terrestrial and aquatic surfaces. Although we assume that nanoplastics at the Siberian site originate from terrestrial sources, we cannot rule out an oceanic source. Marine waters are capable of ejecting plastics through a process of bubble burst and wave action, with the ejected plastics then being available for wind transport^{47,48}. Our site is just 150 km from the coast and the trajectory models show that air does arrive at the site from above the ocean (Fig. 5), and so we acknowledge that this is also a viable route for plastics to enter these waterbodies. An alternative explanation is that plastics arrive via aerial deposition following the burning of rubbish⁴⁶ at the research camp. Alongside the aforementioned flooding, this could also explain the presence of PVC in the sample taken from flooded tundra near the camp. However, several of the ponds closest (< 1 km) to the camp had no detectable nanoplastics in their waters. It could be that the prevailing wind carried smoke and associated plastics away from the ponds and lakes (which are west/north-west from camp), or that the combusted material does not form nanoplastics. The presence of PS in the Siberian, but not the Swedish, samples is unexpected, but we note a recent study that found PS microplastics in Arctic snow samples, but none in urban snow¹².

Conclusion

Nanoplastics are ubiquitous in the streams and lakes of a forested catchment in south west Sweden. We detected PVC, PP, PE and PET in lakes with dwellings in their catchments, and in those without any habitation in the vicinity. We also detected nanoplastics in remote Siberian waters, although concentrations were lower, only PVC and PS were present, and for one lake and two ponds we detected no nanoplastics. Our data support the assumption that, although plastic particles can be carried over long distances from their source¹⁸, concentrations at a particular location are predominantly derived from diverse regional and local sources. If this assumption is correct, then atmospheric nanoplastic concentrations may be subject to sizeable spatial and temporal variation locally, regionally and globally. Therefore, our data suggest that nanoplastics could be present in a very large number of global waterbodies, including urban waters, rural waters, and even remote waters; the sources of these plastics could be deposition from distant sources in addition to local contamination. Due to analytical challenges, quantitative measurements of aquatic nanoplastics are scarce in the literature. For example, a concentration of 241.8 $\mu\text{g l}^{-1}$ (very similar to our concentrations) was reported for PS in a UK peri-urban

river²³. The implications of a potentially ubiquitous global nanoplastics load for aquatic ecosystem functioning remain largely unexplored, but nanoplastics have different effects when compared to larger plastic fragments^{9,27}. A full understanding of how nanoplastics affect aquatic environments is needed, alongside more concentration measurements, in order to assess the impact of this hidden anthropogenic pollutant.

Materials And Methods

Swedish Sampling

Sampling took place on August 19th 2017 in the Gårdsjön research catchment and surrounding area in southwest Sweden. The catchment altitude is 100-170 m ASL, and the climate is maritime temperate with an annual precipitation for 2017 of 1145 mm (data from SMHI's Komperöd site⁴⁹, 600 m from Lake Gårdsjön). The catchment has been extensively studied, particularly in relation to the effects of acid deposition on ecology and biogeochemistry^{50,51}, and both the terrestrial and aquatic components of the system have been exceptionally well-characterised^{40,52}. It is 12 km from the coast, and thus has been subject to moderately high levels of atmospheric acid deposition⁴¹, the majority of which is the result of long-range pollutant transport from industry in the UK and central Europe⁴². This deposition resulted in acidified lake waters⁴⁰ that have historically been managed by liming⁵³. All lakes within the catchment are oligotrophic and land use is forest with *Picea abies* (Norway spruce) and *Pinus sylvestris* (Scots pine) being dominant. Soils are thin and predominantly podzols, with outcrops of bedrock, and approximately 10% peat cover⁴⁰.

There are no settlements within the Gårdsjön catchment, and only two dwellings: one holiday cottage with no road access 70 m from Lake Gaffeln (a headwater lake) and the research cabin at the outflow of the catchment, with no permanent residents. Our project also considered two other lakes just outside the Gårdsjön catchment: Stora Bjurevatten and Västersjön. The surrounding land of Stora Bjurevatten is forested, but in the catchment, there are approximately 50 dwellings plus a negligible amount of farm land (~10 ha), whilst there are approximately 80 dwellings scattered immediately around Västersjön at a distance of 50-200 m from the lake, as well as a small road along one shoreline. The nearest town is 10 km to the west (Stenungsund, population = 10,000) and the city of Gothenburg (population = 600,000) is 40 km to the south.

Within the Gårdsjön catchment we collected ten surface water samples: four from streams and six from lakes. These sites comprised two locations within a headwater lake (Stora Hästevatten, which is divided into two separate waterbodies by a causeway); the other headwater lake (Gaffeln) and the stream draining it; the main inflow to Lake Gårdsjön (draining these lakes plus one additional lake, Lilla Hästevatten); the other three permanent streams flowing into Gårdsjön; and the lake outflow. We also sampled the next lake downstream of Gårdsjön, Stora Bjurevatten, and one lake in an adjacent catchment (Västersjön) with the intention of extending the trophic gradient of sampling sites to include mesotrophic

systems. Lake samples were collected at outflows (including the culvert separating the divided northern and southern basins of Stora Hästevatten) where access permitted; exceptions were Lake Gaffeln, where water was sampled from an accessible shoreline location approximately 100 m from the outflow, and Stora Bjurevatten where we sampled from a point on the eastern shore. The Gårdsjön inflow sample was collected next to a bridge over the short section of channel between Gårdsjön and Lilla Hästevatten and can thus also be considered representative of the outflow from this lake. The three small inflow streams (F1, F2 and F3) were sampled at established v-notch weirs and the remaining stream sample (Gaffeln Stream) was taken from the lowest point with a defined channel before flow enters a wetland downstream of Stora Hästevatten.

All analyses on the Swedish samples were originally conducted for an experiment investigating dissolved organic matter (DOM) composition and reactivity⁵⁴. Water samples were filtered *in-situ* with 0.45 µm filters (pre-rinsed with lake/stream water) and stored in 100 ml glass bottles in the dark at 4°C until analysis

Siberian Sampling

Sampling took place on the 16th and 17th of July, 2017 in the Kytalyk Nature Reserve in the Indigirka River lowlands in north east Siberia. The site is 10-40 m ASL and the climate is Arctic, with annual precipitation of 232 mm and mean annual temperature of -13.8°C⁵⁵. The landscape is oligotrophic tundra with thaw-induced drained lake basins and a Yedoma ridge³⁵. The site has been well studied in relation to greenhouse gas emissions, vegetation communities, and soil organic matter^{32,35,55}.

There are no settlements in the area. There is a research station/camp that has 10-30 visitors per year. The camp consists of four cabins (two for local hunters and fishing, two for research visitors) and a small educational centre associated with the nature reserve. Rubbish and human waste are disposed of by burning with petrol. Samples were collected from the shorelines of three lakes (one large thermokarst lake and two small lakes), five ponds, runoff from thawing permafrost, and flooded tundra. At the largest lake (51 ha), three samples were collected, each from a different shoreline, but all other sites were sampled at only one location.

Siberian samples were filtered *in-situ* using 0.7 µm pre-combusted GF/F filters and stored in 15 ml Polypropylene tubes with PE caps. Note that no PP/PE was detected in any of these samples, thus no plastic contamination occurred from the tubes. The filter housing was made of Polysulfone (which our method does not detect), PP, and silicone, and therefore no contamination occurred during filtering. Water samples were acidified to pH < 2 with HNO₃ and stored in the dark at 4°C until analysis. DOC concentrations were measured on an Aurora 1030 TOC Analyzer (OI Analytical, Texas, USA).

TD-PTR-MS analysis

For the nanoplastic analysis, aliquots of sample were filtered with 0.2 µm PTFE syringe filters (syringes were made of PE with rubber stoppers). The procedural blanks were exposed to the same potential

impurities as the samples: i.e. the same vials and the same syringes and filters were used. Samples (0.5 ml) were then subjected to thermal desorption proton transfer-reaction mass spectrometry (TD-PTR-MS) using the method described in Materić et al. (2017 and 2020) ^{56,56}. This method provides detailed information on the molecular composition of volatile and semi-volatile dissolved organic matter in the sample; it gives both the molecular weight and the concentration of molecules up to a size of ~500 m/z. Briefly, a low-pressure evaporation/sublimation process was used to remove the sample water, leaving behind residues of organic matter. Samples were then thermally desorbed by ramping the temperature up from 35°C to 350°C (at a rate of 40°C per minute) and measured on a PTR-TOF 8000 instrument (IONICON Analytik, Innsbruck, Austria). This assures complete thermal desorption of the sample as shown in thermograms of our method paper ¹⁹. Raw data of all the measurements in this work are available in the permanent repository (see the SI). During the optimisation of the system, blanks were analysed to minimise contamination: system blanks (clean vials), dry blanks (clean vials exposed to the low-pressure evaporation/sublimation process) and ultrapure water blanks (vials containing 0.5 ml of HPLC water, filtered using the same filters used for the samples, then exposed to the low-pressure evaporation/sublimation process). Following optimisation all samples were analysed, interspersed with ultrapure water blanks (procedural blanks) every 3-4 samples. The mean signal generated from the blanks was subtracted for each ionic mass detected in the samples. The detection limit was calculated as 3-sigma of the ultrapure water blanks and ion signals below the detection limit were excluded. Thus only reliable ions were considered for further analysis. For the analysis of the Swedish samples, five samples were analysed in duplicate, whilst for the Siberian samples all samples were analysed in triplicate.

All the steps of the data analysis (including all the mass spectra, subtractions, detection limit calculation, and final mass spectra) are provided in the permanent repository (see the SI). Further details on the analysis are provided in Peacock et al (2018) ⁵⁴, where the PTR-MS data were used to investigate the molecular composition of DOM.

Plastic fingerprinting and quantification

Nanoplastics detection and quantification was performed as described in Materić et al. (2020) ¹⁹. TD-PTR-MS data were analysed using PTRwid ⁵⁷, where the organic ion signals were integrated for 10 minutes starting when the TD oven reached a temperature of 50°C. Plastics polymers show a specific ion signal when heated up above the boiling point. PTR-ToF-MS is used to monitor these signals in real-time and at high resolution, and measures the quantity of each organic ion arising from the thermal desorption process. The ion list for each plastics type analysed in this work together with the concentrations for each ion produced are available in the permanent repository (see SI).

A mass spectrum is generated for each sample and the ions signals from the sample are compared with the signals coming from pure plastic standards, as described in previous work ¹⁹. The analysis software used 40 ions in the range m/z >100 from the library mass spectra (e.g. PET). The mass range of >100 Da is chosen to exclude lower volatility dissolved organic matter so as to increase the analytical

performance in preselecting the volatility range for target polymers. The high number of ions in the fingerprint (40) assures a specific fingerprint, further improving the analytical performance of the technique.

A fingerprinting algorithm scores the similarity between the mass spectra of the sample and that of a plastic standard material. A positive fingerprint was assigned for samples with the fingerprint score significantly higher than the mean score of 1000 randomly generated spectra plus 2s (z-score > 2, $p < 0.02275$, one tail distribution). The fingerprint scripts used in this work, together with the outputs (scores) are available in the SI.

The concentration of the detected plastics was then calculated based on the known kinetics of the reaction chamber^{58,59}, as explained in our previous work^{19,54,56,60}.

In short, the concentrations of organic ions are calculated according to the following^{59,61,62}:

$$[C_{ppb}] = \frac{1}{kt} \times \frac{[M \cdot H^+]}{[H_3O^+]} \times \frac{\sqrt{(m/z)_{H_3O^+}}}{\sqrt{(m/z)_{M \cdot H^+}}}$$

where $[C_{ppb}]$ is the molar ratio of ions in gas phase in ppb, k is the reaction rate coefficient, t is the residence time of the primary ions in the drift tube (corrected for the ToF transmission efficiency⁶³), $[M \cdot H^+]$ and $[H_3O^+]$ are ion counts representing the protonated analyte and primary ions, respectively, $(m/z)_{H_3O^+}$ and $(m/z)_{M \cdot H^+}$ represent the mass-to-charge ratio of protonated water and the protonated analyte M , respectively. This step is carried out using PTRwid software⁵⁷, and the actual script is included in SI.

From deduced molar ratio values (in ppb), the concentration of each ion (in $ng\ ml^{-1}$) was calculated as follows;

$$[C_i\ ng] = \frac{[C_{ppb}] \times m_{z_i}}{MV \times L} \times F \times It$$

Where $[C_i\ ng]$ is the concentration of the ion i in $ng\ ml^{-1}$, m_{z_i} is the molecular mass of the particular ion in Da (given as mass-of-charge in our TOF-MS output), MV is molar volume, L is volume loaded in ml, and F is the flow of the TD unit in $L\ min^{-1}$, and It is the integration time. After this, the concentrations of all the ions i released from the plastics were summed giving the final concentration. This step of data processing was included in the fingerprinting script, and the code is available in the SI.

Such calculated concentrations represent the amount of organic matter that is actually ionised in the system. The PTR ionization efficiency of organics is not 100% (e.g. there are losses in neutral molecular

fragments and CO₂ – not detected by the PTR-MS), so the values are considered as a minimum amount of the analyte ¹⁹.

Number concentrations of NPs particles are calculated from the mass concentrations, assuming 200 nm diameter (the mesh size of the filter used), spherical shape and density of 1 g/cm³, which is 238732.4 particles per ng of nanoplastics.

HYSPLIT model

Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) modelling was used to calculate footprint (trajectory frequencies) ⁶⁴. We used a 48-hour backward trajectory of air movement with a frequency grid of 1.0 x 1.0 degree. We choose to use the air parametrisation rather than particle parametrisation as the nanoplastics measured by our method have an upper size of 200 nm, which we assume behave more like a gas than a PM10 particle. We used monthly footprints for the entire year prior to the sampling dates and the trajectories are available in the Supplement.

Quality control and data limitations

In this work, we took particular caution to provide conservative results of the plastic concentrations that we report. Thus several steps were taken to ensure that our concentrations are conservative: 1) We had several types of blanks to assess the potential contaminating (explained in details in the TD-PTR-MS analysis section, all mass spectra available in SI). Measured nanoplastics were in the range of 6 to 508 ng/mL, well above the detection limit, e.g. of <0.34 ng/mL (based on a PS nanoplastics standard) ¹⁹. 2) We used a strict fingerprinting method including 40 ions from the library of each plastic to assure the presence of the polymer in question; 3) the low-pressure evaporation/sublimation system was used to remove volatile organic matter from the organic-rich matrix; 4) slow thermal desorption was used for boiling point separation of the matrix, where only organic molecules of the volatility range similar to the plastics we measure in real-time and high resolution, and 5) the postprocessing quantification algorithm preserves the ion signal ratio of the plastics library, thereby preventing the overestimation caused by single or group of ions coming from the matrix. Further details are provided our methods paper ¹⁹.

The original sampling campaigns were planned for studies investigating dissolved organic matter; i.e. they were not designed with plastic analyses in mind. Thus, no field blanks (FB) were collected at the Swedish site. Two FBs were collected at the Siberian site, using the same filtering and storage procedures as for the field samples. However, the location is remote and off-grid, and there was no access to 'fresh' Milli-Q water. Deionized water was used instead and transported in available commercial 5L PET drinking water bottles. Each FB was analysed in duplicate. No plastics were detected in either duplicate of FB2 but one duplicate of FB1 gave a positive return for PS, with a concentration of 43 µg l⁻¹. The PS algorithm match for this positive return was very low; just 27, compared to the matches for the field samples which were 30-65, with a mean and median of 53 and 55. We therefore cannot be entirely sure whether the signal in the FB1 duplicate is PS, or some other contaminant.

Where replicate samples were taken, agreement between them was consistently good for the Swedish samples, as shown by low standard deviations. Furthermore, if the presence or absence of a specific polymer was detected in any sample, the result (presence or absence) was the same for both replicates (Table 1). In contrast, standard deviations were larger for the Siberia samples, and there was a lower consistency between replicates; e.g. presence and absence of specific polymers was sometimes found for different replicates of the same sample.

Declarations

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Author contributions

DM and MP conceived the paper. MP and MF planned and carried out the sampling in Sweden. JD planned and carried out the sampling in Siberia. FM provided wind data for the Swedish site and logistical support for the Swedish sampling. DM, MP and JD analysed the samples. DM performed the plastic fingerprinting. DM and MP wrote the manuscript with ideas and feedback from all co-authors.

Competing interests

Authors declare that they have no competing interests.

Data and materials availability

All data needed to evaluate the conclusions in the paper are present in the paper and in the Supplementary Data (including raw files, all the scripts and processing stages of the data analysis) and it is available in the YODA public repository. [For the review process, this data is available at <https://surfdrive.surf.nl/files/index.php/s/SgXhMyLoJRsfzIN> (readme.doc has the file descriptions). Upon the revision we will upload the data to YODA - <https://public.yoda.uu.nl>]

Supplementary Materials

All Supplementary Data and Material (including raw files, all the scripts and processing stages of the data analysis) are available at <https://surfdrive.surf.nl/files/index.php/s/SgXhMyLoJRsfzIN> (readme.doc has the file descriptions). Upon the revision, we will upload it to the permanent repository YODA (<https://public.yoda.uu.nl>), and it will be assigned a DOI.

Data availability

All data (including raw files, all the scripts and processing stages of the data analysis) are available at <https://surfdrive.surf.nl/files/index.php/s/SgXhMyLoJRsfzIN>. Upon the revision, data will be stored at the permanent repository YODA (<https://public.yoda.uu.nl>), and it will be assigned a DOI.

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Table

Table 1: Overview of sampling sites, showing site type or lake area (if applicable), and the concentrations of all nanoplastics. Errors are standard deviations, for the samples where replicate analyses were performed.

Site name	Site code	Site type/lake area (ha)	PVC ($\mu\text{g l}^{-1}$)	PS ($\mu\text{g l}^{-1}$)	PP ($\mu\text{g l}^{-1}$)	PE ($\mu\text{g l}^{-1}$)	PET ($\mu\text{g l}^{-1}$)	Total ($\mu\text{g l}^{-1}$)
Sweden								
Stora Bjurevatten	10	30	122	0	0	276	61	459
F1	6	Headwater stream	0	0	59	86	39	184
F2	7	Headwater stream	0	0	85	122	41	248
F3	8	Headwater stream	0 (± 0)	0 (± 0)	123 (± 8)	155 (± 9)	0 (± 0)	278
Gårdsjön inflow	5	3.1	374	0	514	699	0	1588
Gårdsjön outflow	9	31	152 (± 7)	0 (± 0)	0 (± 0)	254 (± 9)	0 (± 0)	407
Gaffeln	1	6.2	264 (± 1)	0 (± 0)	392 (± 1)	508 (± 1)	116 (± 1)	1280
Gaffeln stream	2	Stream draining a lake	148	0	0	282	77	507
Stora Hastevatten north	3	3.4	0 (± 0)	0 (± 0)	155 (± 8)	211 (± 9)	0 (± 0)	365
Stora Hastevatten south	4	5.1	176 (± 8)	0 (± 0)	227 (± 5)	294 (± 8)	0 (± 0)	696
Västersjön	11	28	50 (± 1)	0 (± 0)	0 (± 0)	100 (± 1)	29 (± 1)	180
Siberia								
Lake 1, east shore	L01	51	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0
Lake 10	L10	4.2	8 (± 11)	6 (± 9)	0 (± 0)	0 (± 0)	0 (± 0)	42
Lake 1, north shore	L02	51	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0
Lake 1, south-west shore	L04	51	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0
Lake 8	L08	14.1	0 (± 0)	24 (± 5)	0 (± 0)	0 (± 0)	0 (± 0)	24
Pond 1	P01	0.010	354 (± 7)	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	354
Pond 11	P11	0.006	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0
Pond 2	P02	0.004	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	0
Pond 3	P03	0.016	0 (± 0)	21 (± 15)	0 (± 0)	0 (± 0)	0 (± 0)	31
Pond 8	P08	0.014	0 (± 0)	11 (± 16)	0 (± 0)	0 (± 0)	0 (± 0)	33
Tundra 5	T5	Flooded tundra	58 (± 41)	0 (± 0)	0 (± 0)	0 (± 0)	0 (± 0)	87
Thermokarst 2	TK2	Thawing permafrost	16 (± 11)	7 (± 10)	0 (± 0)	0 (± 0)	0 (± 0)	45

Figures

Figure 1

The Swedish sampling site. Left panel: the Gårdsjön research catchment with lake (blue circles) and stream (red circles) sampling locations marked; numbers refer to the results in table 1. Centre panel: the Gårdsjön catchment (red rectangle) shown in the wider landscape, with the two extra lake sampling locations marked. Satellite images: @2021 CNES/Airbus, Landsat/Copernicus, Lantmäteriet/Metria, Maxar Technologies. Right panel: map showing the Gårdsjön catchment (red circle) and major cities that trajectory models (Fig. 4) show as potential sources of atmospheric deposition. Yellow squares represent cities with urban populations > 1 million. Orange square represents Gothenburg, which has a population of 0.6 million and is the nearest city to the Gårdsjön catchment.

Figure 2

The Siberian sampling site in the Kytalyk Nature Reserve. Left panel: the sampling area showing small lakes (L, blue circles), thermokarst lakes (L, grey circles), thawing permafrost (TK, grey circle), ponds (P, white circles), and flooded tundra (T, yellow circle). Right panel: location of the Kytalyk Nature Reserve (red circle) in the Russian Far Eastern Federal District, showing large towns throughout the region (orange squares, population 100,000 – 300,000) and small settlements closest to the study site (green squares, population 1,400 – 3,500). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning

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Figure 3

Nanoplastic concentrations in Swedish (left panel) and Siberian (right panel) samples. Different polymer types are represented by different coloured bars. Note that “Tundra” is flooded tundra.

Figure 4

HYSPLIT trajectory model and wind rose of seasonal variation in air regimes at the Swedish site. (a) December 2016, b) June 2017 c) wind rose for the year 2016/17 Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 5

HYSPLIT trajectory model of seasonal variation in air regimes at Siberian site (a) December 2016, b) June 2017 Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.