

Article

Characteristics of Anthropogenic Pollution in the Atmospheric Air of South-Western Svalbard (Hornsund, Spring 2019)

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Abstract: The character of atmospheric pollution and its impact on surface waters may vary substantially in space, and hence, we add a potentially important location for the studies of atmospheric air pollution to the map of the High Arctic. We have investigated the anthropogenic particle characteristics and selected persistent organic pollutant concentrations, in a priorly unmonitored location in the Arctic (Svalbard), exposed to a climatic gradient. Single-particle analysis of *PM* indicates that besides the prevailing natural aerosol particles, anthropogenic ones were present. The likely anthropogenic origin of some particles was established for spherical Fe-rich or aluminosilicate particles formed in high-temperature processes or metal-rich particles of the chemical composition corresponding to industrial products and atypical for natural minerals; soot, tar balls, and secondary sulfate were also likely of anthropogenic origin. Some of the observed anthropogenic particles could only come from remote industrial sources. *POP* concentrations indicated a background of LRAT, consistent with the Σ PCB concentrations and volatility profile. However, the Σ DDX composition indicating aged sources and an order of magnitude higher concentrations of both Σ DDXs and Σ HCHs than at other High Arctic monitoring stations indicate their potential source in two types of re-emission from secondary sources, i.e., from seawater and snowpack, respectively.

Keywords: particulate matter (PM); single-particle analysis; secondary sources of POPs; air quality; High Arctic; human impact



Citation: Pawlak, F.; Koziol, K.; Wilczyńska-Michalik, W.; Worosz, M.; Michalik, M.; Lehmann-Konera, S.; Polkowska, Ż. Characteristics of Anthropogenic Pollution in the Atmospheric Air of South-Western Svalbard (Hornsund, Spring 2019). *Water* **2024**, *16*, 1486. <https://doi.org/10.3390/w16111486>

Academic Editor: Yaoming Ma

Received: 15 April 2024

Revised: 8 May 2024

Accepted: 17 May 2024

Published: 23 May 2024



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1. Introduction

The Arctic has been gradually driven into the focus of political and economic interests and heavily impacted by climate change, which leads to unknown trajectories of pollution impact in its fragile environment [1]. Attention has been drawn especially to the changing characteristics of the Arctic haze in the winter and spring and the role of local sources in the summer [2]. Arctic haze, a landmark feature of Arctic air pollution [3–5], remains under close investigation currently (e.g., [6,7]), with a typical composition including particulate matter, sulfate and persistent organic pollutants (POPs) [8]. Chlorinated POPs, while typically connected to human activity, may represent either primary or secondary sources of pollution in the Arctic. Furthermore, the origin of other chemical species in the atmospheric air of the Arctic is considered still more ambiguous, with new sources occurring both due to the ongoing industrialization and urbanization of the Arctic [9,10], including shifting

of the shipping routes northwards [11]. The changing anthropogenic pollution sources include industry and transportation (e.g., [12–14]), yet anthropogenic pollution may be confounded with natural particles from newly exposed soil contributing rock dust with similar metallic components to anthropogenic particles [15]. Therefore, a catalog of features identifying pollution from anthropogenic sources in the Arctic needs to be documented more thoroughly, in search of robust markers of both near and far sources of such pollution. In this research, we provide a step towards identifying particles characteristic of anthropogenic pollution sources found in the composition of the atmospheric air at Hornsund, Svalbard, where local human activity is very limited on-site. This priorly unmonitored location is investigated here since characteristics of the Arctic aerosol largely differ between monitoring stations [16], and Hornsund is a site with a strong climate gradient, where mean temperature increases by 1.00 °C per decade, which is one the highest rates on Earth [17].

The Arctic has been largely free of primary sources of chlorinated persistent organic compounds such as industry and agriculture [18]; hence, most of its pollution has been delivered by long-range atmospheric transport (LRAT) from primary sources located at low latitudes [19]. More than 25 years of atmospheric monitoring in the Arctic allowed the determination of a downward trend of POP concentrations in the air [20–24]. However, climate change is expected to remobilize previously stored pollutants, increasing the share of secondary sources such as permafrost and soil [25–27], seawater [28–30] and glaciers [31,32]. Therefore, in this research, we explore the chlorinated POP concentrations at Hornsund, Svalbard, for potential markers of local and long-range pollution sources.

The aim of this work was to identify pollutants typical for human activity in the atmospheric air in Hornsund and search for their source on the basis of their composition characteristics, with support of backward air-mass trajectories. Through this procedure, we give a snapshot of how different characteristics of the Arctic atmospheric air may be captured by including chemical characterization of air and aerosol from Hornsund in the atmospheric monitoring conducted in Svalbard. As this is the first joint investigation of the chemical composition of two types of pollutants in that location, where no chemical air quality monitoring existed at the time, the data are limited to a qualitative analysis of atmospheric particulate matter (PM) including selected particle size, morphology and chemical composition, as well as the content of selected POPs in samples of atmospheric air collected during the spring season 2019 in Hornsund (Svalbard). However, we deem our data offer valuable insight since April and May 2019 have already been shown to be impacted by atypically high concentrations of chlorinated POPs in snowfall at Hornsund [33], while recent work suggests that the Arctic haze season may be shifting towards later months of spring [2,6].

2. Materials and Methods

2.1. Study Site, Air Sampling and Data Collection

Sampling was performed in the vicinity of the Polish Polar Station Hornsund, Svalbard (77° N, 15°30' E), in the environmental chamber facility, at a distance of approx. 750 m from the main station building (Figure 1). Next to the main station, a diesel power plant and a waste incinerator are located, in a close cluster of buildings, while the environmental chamber is placed away from them towards north-east—upwind from the local pollution sources in the most frequent easterly wind circumstances [34]. Samples of total suspended PM for single-particle characterization and combined air and PM for POP analysis were collected between 14 April and 11 May 2019.

Samples of total suspended PM were collected on polycarbonate membranes (pore size 0.1 µm or 0.2 µm, alternating; 47 mm in diameter) using a Life 1 One (Mega System) low-volume sampler. The sampler was sheltered inside the environmental chamber building, with the sampling head exposed on the rooftop. Samples were thus collected from approx. 3 m above ground level and approx. 14 m a.s.l., while the mean boundary layer height generated with HYSPLIT (see Section 2.4.2. for details and references) reached 274–456 m a.s.l. Samples were collected with increasing sampling time over the sampling

period, ranging from 8 to 24 h (the sampled air volumes ranged from 4.17 to 21.52 m³). The variable sampling period length was part of the study setup directed to ensure obtaining at least some filters that were not overloaded and did not contain too little material for analysis. We decided upon such a sampling setup due to a lack of prior information on particle loading in the area. As none of the samples were overloaded and all had sufficient material for analysis, we decided to include all the collected samples in the reported study. Air temperatures during PM sampling ranged from −6 to +2 °C, and there was either no precipitation or a trace of precipitation measured (on three occasions); wind speeds ranged from 1.2 to 9.7 m s^{−1}, blowing most frequently from the N-NE-E quadrant. The details of samples collected for PM are reported in Table 1.

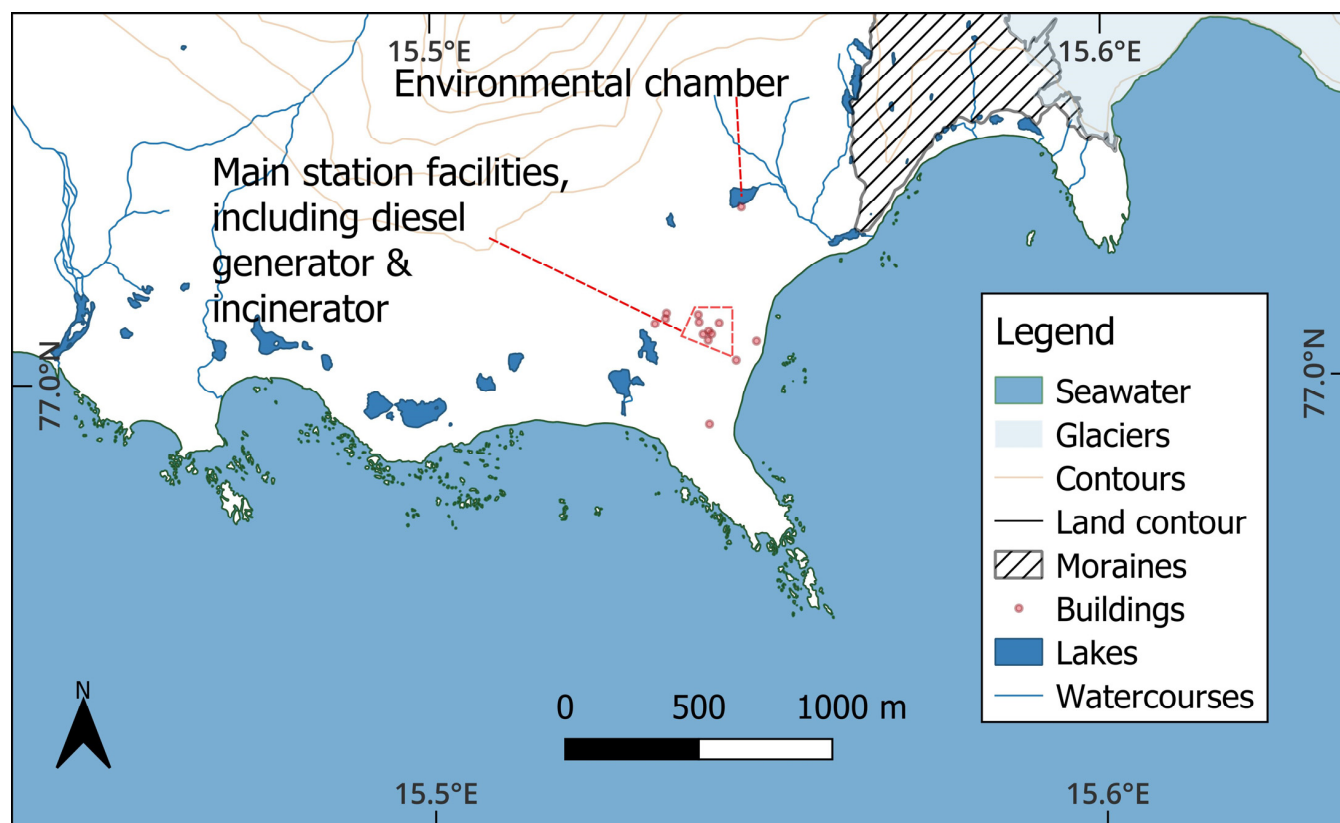


Figure 1. Location of the environmental chamber (sampling site) in relation to the potential local sources of anthropogenic pollution at Hornsund (diesel power generator and high-temperature waste incineration facility).

Table 1. Aerosol sample collection conditions (technical and meteorological).

Sample ID	Date of Sample Collection	Sample Collection Duration [h]	Volume of Air [m ³]	Filter Pore Size [μm]	Wind Direction (Sector)	Wind Speed [m/s], Averaged	Air Temperature at 2 m [°C], Averaged
SP1	14/15 April 2019	unknown *	1.002	0.1	W	9.7	1.23
SP2	17 April 2019	8	5.611	0.2	SW-W	7.0	1.83
SP3	23 April 2019	8	4.167	0.1	NE-E	3.0	−4.3
SP4	24/25 April 2019	12	8.912	0.2	W-NW	4.4	0.74
SP5	26/27 April 2019	12	6.642	0.1	E	4.8	−1.9
SP6	28/29 April 2019	15	5.864	0.1	SW-W, N, NE	1.2	−0.5
SP7	02/03 May 2019	15	13.447	0.2	SE, NW-N	3.5	−2.51
SP8	03/04 May 2019	24	21.503	0.2	NE-E	7.8	−5.94
SP9	04/05 May 2019	24	11.502	0.1	N-NE-E	6.0	−5.54
SP10	05/06 May 2019	24	21.522	0.2	N-NE-E	3.4	−4.25
SP11	06/08 May 2019	24	11.083	0.1	NE-E	5.3	−4.26
SP12	09/10 May 2019	24	21.267	0.2	NE-E	6.8	−5.19
SP13	10/11 May 2019	24	10.722	0.1	E	9.4	−3.61

* The sampling pump stopped automatically due to an error before the designated end time. Meteorological data: courtesy Institute of Geophysics, Polish Academy of Sciences (from the Polish Polar Station Hornsund).

Samples for POP determination were collected with a medium-volume air sampler (Genius 5 Instruments, Technospec, Kottlingbrunn, Austria), with a maximum flow rate of 15 m³/h. Air volumes of 1209–1520 m³ were thus sampled in three 7–10 day sampling periods. The air was passed through two polyurethane foam (PUF) cylinders sandwiched with Amberlite® XAD resin (Supelpak™-2SV purified Amberlite® XAD2, 90 Å mean pore size, Sigma-Aldrich, St. Louis, MO, USA); preceded with a quartz filter (Whatman QMA, precombusted 24 h in 450 °C, after [35]). These were stored in aluminum foil within a plastic bag, at −18 °C, until analysis.

2.2. Atmospheric PM Single-Particle Analysis

An S-4700 Hitachi field emission scanning electron microscope (SEM)(HITACHI, Tokyo, Japan) with a Noran NSS energy-dispersive spectrometer (EDS) (Thermo Fisher Scientific Inc., Waltham, MA, USA) was used for PM analysis. Particles were observed and analyzed at 20 kV accelerating voltage. Both secondary electron (SE) and backscattered electron (BSE) modes were used in the imaging of samples. The quantification of chemical element content was based on a standardless method. All results were recalculated to 100% (without carbon content). Fragments of filters were attached to carbon disc holders. Two fragments from each sample were prepared—for carbon and for gold coating. The thickness of the coating layer was not measured. Carbon-coated fragments of polycarbonate membranes were used mainly for chemical analyses (using EDS method) and imaging using backscattered and secondary electron signals. Gold-coated fragments were used mainly for imaging using the secondary electron signal.

The density of particles on polycarbonate filters was low enough to avoid the overlapping of X-ray signals from neighboring particles. All samples were dominated by soot particles, tar balls and natural mineral particles, taking into account particle numbers. The identification of soot was based on the morphology of aggregates supported by occasional verification of the chemical composition. Tar ball identification was based on morphology and chemical composition. The chemical composition of other particles was analyzed to distinguish natural and anthropogenic ones. In total, more than two thousand particles were examined in 13 samples, and the chemical composition of more than 350 considered to be anthropogenic was analyzed. The number of anthropogenic particles rich in metals (possible indicators of source area) identified in each sample was too low to consider differences between samples as statistically important.

2.3. POP Determination and Quality Assurance/Quality Control (QA/QC)

Prior to extraction, the PUF-XAD-PUF and quartz filter sets were spiked with isotope-labeled standards solutions of 100 ng mL^{−1} of *p,p'*-DDT-*d*8, PCB-28 ¹³C and PCB-180 ¹³C, in 10 µL dichloromethane. Each sample was subjected to triple ultrasonic-assisted extraction in 1 L volume of hexane/acetone mixture (4:1, *v:v*). The obtained extract was then reduced to 10 mL, using a rotary evaporator, and then further evaporated under a gentle flow of N₂ until almost dry. Finally, it was reconstituted in 0.2 mL isooctane.

The determination of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) was carried out with a gas chromatograph coupled with a triple-quadrupole mass spectrometer (GC-MS/MS) (7890B and 7000D, Agilent Technologies, Santa Clara, USA), with the following analytes being determined: seven PCBs (PCB 28, 52, 101, 118, 138, 153 and 180) and 10 OCPs (*α*-HCH, *γ*-HCH, *β*-HCH, aldrin, heptachlor epoxide, DDD, *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDT and *p,p'*-DDT). Other information on the analytical method is provided in the Supplementary Tables S1–S4. Between every 10 samples, a reference solution was analyzed, either of a certified reference material or of a standard solution used before, to control for chromatographic parameters.

As QC against contamination potentially introduced into the samples by the entire analytical procedure, including sample collection, we analyzed a field blank. All validation parameters (such as recovery; coefficient of variation, CV; limit of detection, LOD; and limit of quantification, LOQ) are given in Table S4. In brief, recoveries calculated from samples

prepared by adding standard solutions to a clean PUF-XAD-PUF sandwich ranged from 65 to 126%, while CVs ranged from 0.069 to 3.6. For the internal standards, recovery and CV were 86–109% and 1.0–2.1, respectively.

2.4. Factors for Data Interpretation

2.4.1. Shape and Chemical Composition of PM as a Particle Origin Criterion

Interpreting sources of atmospheric aerosols could be difficult. Both primary particles (directly emitted from sources) and secondary particles (formed in chemical reactions in the atmosphere with an important role of anthropogenic gaseous precursors) may indicate an anthropogenic impact. To clarify, we use the term “anthropogenic” to denote particles composed of material formed during technological processes, e.g., fuel combustion, metallurgy, industrial production of various materials, etc. In the case of secondary atmospheric aerosols, the interpretation of anthropogenic sources was based here mostly on their chemical composition and the analysis of sources of primary gaseous pollutants.

We applied the criteria of shape and chemical composition to distinguish natural and anthropogenic particles in the collected aerosol samples. Spherical particles are usually interpreted as a product of high-temperature processes, both technological and natural. They are common in fly ash produced in coal-fired power plants [36] but are also emitted from household heating installations [37]. Spherical Fe-dominated particles are emitted mainly from metallurgical installations [38,39]. Natural spherical particles (cosmic, volcanic, lightning-induced; e.g., [40–42]) are less abundant in comparison with anthropogenic ones. The distinction between natural and anthropogenic spherical particles requires a precise determination of the chemical composition and internal structure of the particles. The identification of soot (in various aggregates) also relies upon the morphology of particles. Tar balls are often identified using their morphology determined in SEM studies (without verification of their internal structure using a transmission electron microscope, TEM) [43–46]. We include soot and tar balls among anthropogenic particles since significant emissions of these particles originate from fuel combustion processes (while we are aware they may also originate from wildfires).

The chemical composition different from common minerals (but also mineral aggregates, intergrowths and rock weathering products) can be considered as an indicator of the anthropogenic origin of airborne particles [47]. This criterion is not very strict, yet it gives a possibility to include in the group of anthropogenic particles that are rich in several groups of metals in proportions rarely occurring in nature. Notably, particles formed during the production of cement and building materials and destruction of concrete constructions are not distinguishable from natural components due to similar main elemental components (e.g., Si, Al, Ca).

2.4.2. Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT)

The origin of the air masses during the sampling was determined using HYSPLIT model [48–50]. Three time periods were determined for the model, corresponding to POP samples, each period starting two days earlier than the beginning of air sampling and ending on its last day (Figure S3). Details concerning back trajectory analysis and its results are given in SM.S5 (cf. also [51]).

3. Results and Discussion

Both PM and POPs detected in the analyzed samples indicate an impact of both natural and anthropogenic sources and processes, and distinguishing the anthropogenic sources by their composition features is the focus of the discussion. For POPs, the division of primary and secondary sources is equally important, as the latter type typically occurs as a result of environmental change.

3.1. Atmospheric PM

3.1.1. Mineral Particles from Rocks and Soils

Natural mineral particles derived from soils, disintegrated rocks, moraine sediments or dust deposited on glaciers predominated, taking into account both the number of particles and their volume. Their chemical composition varies from typical rock-forming minerals such as quartz, feldspar, carbonates or rarely occurring minerals (e.g., monazite) to more complex mineral aggregates, rock fragments and material derived from weathering crusts.

3.1.2. Sea-Salt Chlorides and Sulfate Particles of Anthropogenic or Natural Origin

NaCl particles occur as crystals of different shapes, from cubic to strongly elongated (Figure 2A,B), and may form clusters of spherical forms resulting probably from their deposition in a hydrated form at the time of collection [52]. NaCl particles are considered to be of natural origin (sea salt). Some NaCl crystals were formed during seeded crystallization on aluminosilicate grains (Figure 2B). Mixed particles containing sea salt and silicates can be formed in different processes such as activation and coalescence in clouds [53]. Thus, the origin of these combined particles can be related to the evaporation of seawater droplets containing mineral grains.

Sulfates, predominantly of Ca, were noted (Figure 2C). Their origin was related to the SO_4^{2-} anion reaction with Ca^{2+} in the atmosphere. Secondary calcium sulfate particles in atmospheric pollution are encountered in urban and industrial centers, due to combustion of fuel containing sulfur (mostly coal) (e.g., [54]). The anthropogenic origin of SO_2 is probable as a sulfate anion precursor in analyzed samples and can be related both to LRAT and to local sources [55]. Biogenic SO_2 from the oxidation of dimethylsulfide (DMS) should also be considered [55,56]. According to [57], estimation of the source of sulfate in aerosols in Ny-Ålesund the anthropogenic source dominated over biogenic, sea-salt and crustal ones (especially in the spring). It is also suggested by Adachi et al. [52] that the contributions of anthropogenic sources of sulfates are higher in spring, whereas those of marine sources increase in summer. Particles of a more complex composition, dominated by sulfates but also containing Cl^- and various cations (Ca^{2+} , Na^+ , Mg^{2+} , K^+), are relatively common (Figure 2D–F) and likely represent a mixture of natural and anthropogenic components. The occurrence of Si and Al in salt crystal aggregates (Figure 2D, Table 2) could indicate that crystallization took place in a solution droplet containing fine grains of aluminosilicates. Sulfate particles can thus be considered secondary aerosol formed in the atmosphere via a reaction of SO_2 gaseous precursors related to anthropogenic emissions, and partially of biogenic origin.

3.1.3. Particles Related to Fuel Combustion

Soot particles form aggregates which are relatively common in the analyzed samples. Soot aggregates exhibit various morphologies. Lacy or branched-chain aggregates could be composed of several to numerous soot particles (Figure 2G,H). Other soot particle aggregates are compact, usually composed of numerous particles (Figure 3A,B). The diameters of compact aggregates vary from over 2 μm to below 500 nm. The formation of more compact aggregates is commonly interpreted as an effect of soot aging [58]. Soot particles were noted in the Arctic by numerous authors, e.g., [59–61]. Soot particles could be transported in the atmosphere from remote sources (anthropogenic or natural, e.g., wildfires), but local origin cannot be excluded (coal and diesel fuel combustion in Svalbard) [13,62].

Tar balls were noted as relatively abundant components in the studied samples (Figure 3C,D). Their size was usually below 500 nm. Tar balls are often interpreted as a product of biomass burning [59,63], but their formation during coal combustion is also described [45,64]. Tar-ball-type particles are commonly noted in the polluted urban atmosphere [46]. Tar balls were noted in Arctic PM by several authors (e.g., [52,65,66]).

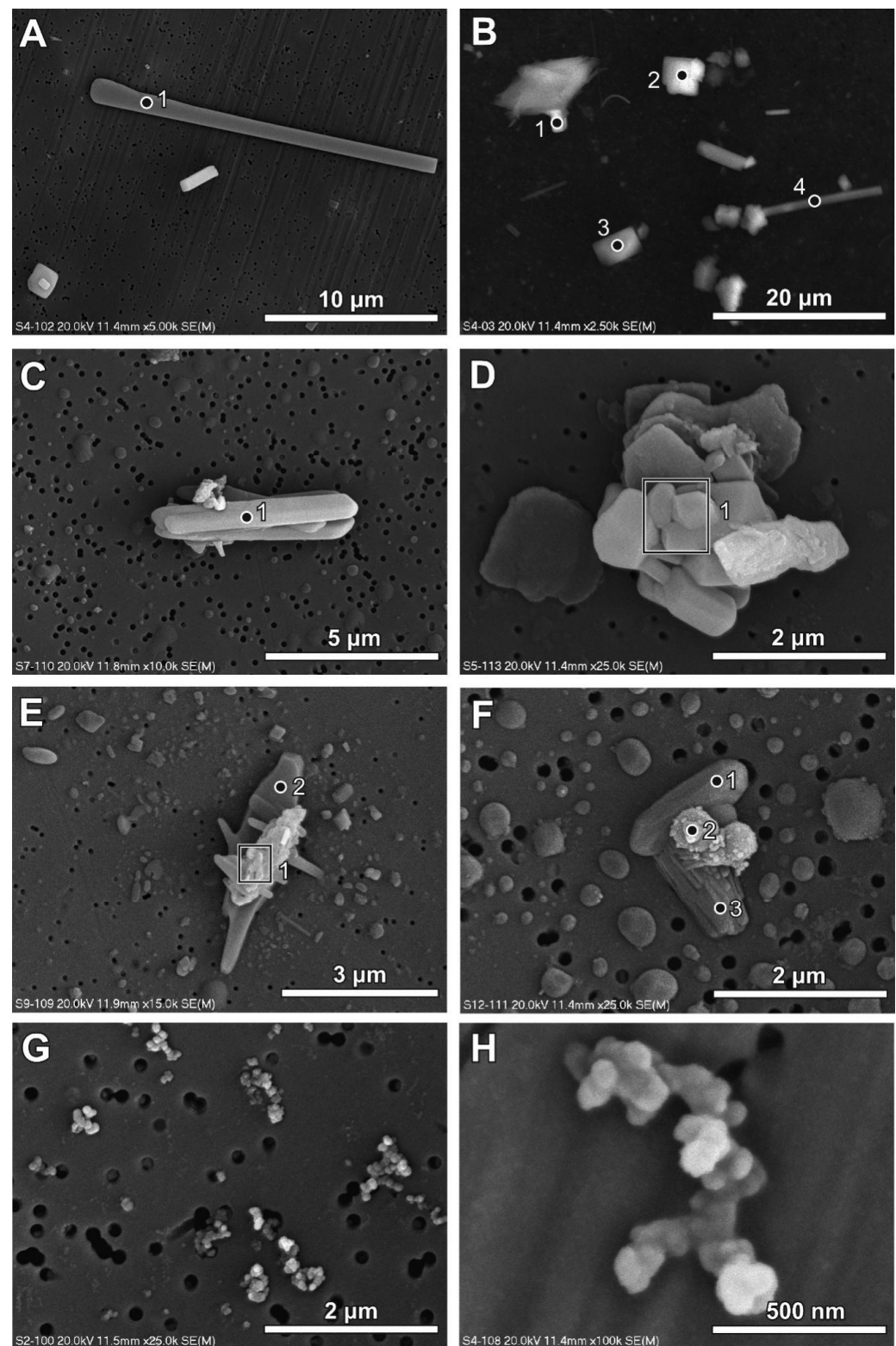


Figure 2. Aerosol particles (SEM; SE images; chemical composition for indicated spots and areas—see Table 2; numbers correspond to spots listed in Table 2). (A) NaCl particles of various morphologies; (B) NaCl particles seeded on aluminosilicate grains; (C) Ca sulfate particle; (D) an aggregate of salt crystals (mostly Ca and Na sulfate and Ca and Na chloride with dispersed aluminosilicates); (E) Ca, Na and Mg sulfates; (F) Na, Ca and Mg sulfates; (G,H) lacy soot particles.

Table 2. Chemical composition of aerosol particles (wt%) indicated in Figures 1–5; “-” = not determined.

Figure	Spot	Element (wt%)																							Total (wt%)
		Na	K	Mg	Ca	Al	Si	P	S	Cl	Mn	Zr	Fe	Cr	Cu	Zn	Pb	Sn	Ti	Ni	Nb	Pt	Au	O	
Figure 2A	1	34.2	-	-	-	-	-	-	-	65.8	-	-	-	-	-	-	-	-	-	-	-	-	-	-	100.0
Figure 2B	1	29.1	0.3	-	0.2	0.5	1.5	-	0.2	59.9	-	-	0.5	-	-	-	-	-	-	-	-	-	-	7.8	100.0
Figure 2B	2	33.7	0.3	-	-	0.7	1.5	-	-	59.2	-	-	0.3	-	-	-	-	-	-	-	-	-	-	4.3	100.0
Figure 2B	3	33.1	-	-	-	-	-	-	0.3	62.5	-	-	-	-	-	-	-	-	-	-	-	-	-	3.5	100.0
Figure 2B	4	33.5	0.4	-	-	-	1.0	-	0.3	55.6	-	-	-	-	-	-	-	-	-	-	-	-	-	9.2	100.0
Figure 2C	1	1.3	-	-	27.2	-	-	-	-	23.0	-	-	-	-	-	-	-	-	-	-	-	-	-	48.5	100.0
Figure 2D	1	4.0	1.3	0.3	23.4	2.9	6.4	-	19.3	5.2	-	-	-	-	-	-	-	-	-	-	-	-	-	37.2	100.0
Figure 2E	1	14.1	-	5.4	17.0	-	-	-	25.9	-	-	-	-	-	-	-	-	-	-	-	-	-	-	37.6	100.0
Figure 2E	2	16.8	-	1.9	14.4	-	-	-	31.2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	35.7	100.0
Figure 2F	1	27.3	-	-	-	-	-	-	36.5	-	-	-	-	-	-	-	-	-	-	-	-	-	-	36.2	100.0
Figure 2F	2	23.9	-	2.1	-	-	-	-	42.9	-	-	-	-	-	-	-	-	-	-	-	-	-	-	31.1	100.0
Figure 2F	3	18.5	-	3.4	10.9	-	-	-	44.9	-	-	-	-	-	-	-	-	-	-	-	-	-	-	22.3	100.0
Figure 3E	1	4.9	3.4	0.9	0.7	6.0	40.5	-	-	-	-	-	17.1	-	-	-	-	-	-	-	-	-	-	26.5	100.0
Figure 3F	1	-	1.7	-	-	10.2	43.6	-	2.2	-	-	-	-	-	-	13.9	-	-	-	-	-	-	-	28.4	100.0
Figure 3G	1	-	-	-	-	-	0.8	-	-	-	-	-	83.7	-	-	-	-	-	-	-	-	-	-	15.5	100.0
Figure 3H	1	-	-	-	-	0.7	2.0	-	1.6	-	2.5	-	75.7	-	-	-	-	-	-	-	-	-	-	17.5	100.0
Figure 4A	1	-	2.7	-	-	5.3	4.4	0.4	-	-	-	-	72.9	-	-	-	-	-	-	-	-	-	-	14.3	100.0
Figure 4B	1	2.4	-	-	-	-	1.6	-	1.6	-	-	-	81.9	-	-	-	-	-	1.4	-	-	-	-	11.1	100.0
Figure 4C	1	-	-	-	-	-	-	-	52.5	-	-	-	42.5	-	-	-	-	-	-	-	-	-	-	5.0	100.0
Figure 4D	1	-	-	0.7	-	0.6	1.3	-	1.1	-	1.7	-	82.6	-	-	-	-	-	-	-	-	-	-	12.0	100.0
Figure 4E	1	7.0	0.5	-	2.1	0.9	1.0	-	3.3	3.6	-	-	59.5	-	-	5.5	-	-	-	-	-	-	-	16.6	100.0
Figure 4F	1	3.0	0.2	-	-	0.6	1.5	-	0.4	1.7	0.3	-	81.2	9.5	-	-	-	-	-	-	-	-	-	1.6	100.0
Figure 4G	1	-	-	-	-	-	1.7	-	1.3	-	1.7	-	66.1	15.3	-	-	-	-	-	11.5	-	-	-	2.4	100.0
Figure 4H	1	2.3	1.6	-	-	-	3.0	-	0.5	0.4	-	-	57.1	10.4	-	-	-	-	-	6.7	-	-	-	18.0	100.0
Figure 5A	1	-	-	-	-	7.4	1.7	-	8.5	1.6	-	-	33.6	-	4.5	17.2	-	-	-	0.6	-	-	-	24.9	100.0
Figure 5B	1	1.8	0.3	-	-	0.4	1.7	-	0.4	1.0	3.1	-	51.9	17.7	-	-	-	-	-	5.5	-	-	-	16.2	100.0
Figure 5C	1	-	-	-	-	3.5	2.2	-	17.6	-	-	-	3.1	-	2.4	41.7	-	-	-	-	-	-	-	29.5	100.0
Figure 5C	2	-	-	-	-	10.3	2.4	-	4.7	2.9	-	-	20.5	-	4.8	16.2	-	-	-	-	-	-	-	38.2	100.0
Figure 5D	1	-	-	-	-	1.2	0.8	-	6.4	-	-	-	-	63.5	-	-	-	-	-	-	-	-	-	28.1	100.0
Figure 5E	1	1.2	-	2.7	-	2.3	1.4	-	5.8	23.1	-	-	-	30.6	-	3.6	-	-	-	-	-	-	-	29.3	100.0
Figure 5F	1	-	-	-	-	-	2.5	-	21.2	-	-	-	0.4	-	-	50.7	-	-	-	-	-	-	-	25.2	100.0
Figure 5G	1	-	-	-	-	-	3.4	-	11.2	-	-	-	-	-	-	-	65.2	-	-	-	-	-	-	20.2	100.0
Figure 5H	1	-	-	-	-	-	0.5	-	-	-	-	-	-	-	61.6	37.8	-	-	-	-	-	-	-	0.1	100.0
Figure 6A	1	-	-	-	-	1.3	1.5	-	-	-	-	-	-	-	-	-	-	-	-	-	-	91.5	5.7	100.0	
Figure 6A	2	-	-	-	-	-	0.9	-	-	-	-	-	-	-	-	-	-	-	-	-	-	89.0	10.1	100.0	
Figure 6B	1	3.9	-	-	-	2.0	1.8	-	-	-	-	-	-	-	-	-	7.3	64.9	-	-	-	-	-	20.1	100.0
Figure 6B	2	3.0	-	-	-	1.7	2.6	-	-	0.4	-	-	2.5	-	-	-	7.4	60.6	-	-	-	-	-	21.8	100.0
Figure 6C	1	-	-	-	-	-	0.8	-	-	-	-	-	-	-	-	91.6	-	-	-	-	-	-	-	7.6	100.0
Figure 6D	1	0.5	-	-	-	-	-	-	-	1.0	-	89.1	-	-	-	-	-	-	-	-	-	-	-	9.4	100.0
Figure 6E	1	-	-	-	0.7	-	0.7	-	-	2.2	-	-	-	-	-	-	-	-	-	39.0	28.1	26.6	-	2.8	100.0
Figure 6F	1	-	2.9	-	4.3	-	1.2	-	-	1.9	-	-	-	-	-	-	-	-	-	27.6	20.6	18.6	-	21.4	100.0
Figure 6G	1	-	0.5	-	0.5	1.7	3.3	-	32.5	-	-	-	2.8	-	-	-	-	-	-	41.8	-	-	-	16.9	100.0
Figure 6H	1	2.6	-	-	0.5	34.3	1.2	-	3.5	4.0	-	-	-	-	-	-	-	-	-	8.0	-	-	-	45.9	100.0

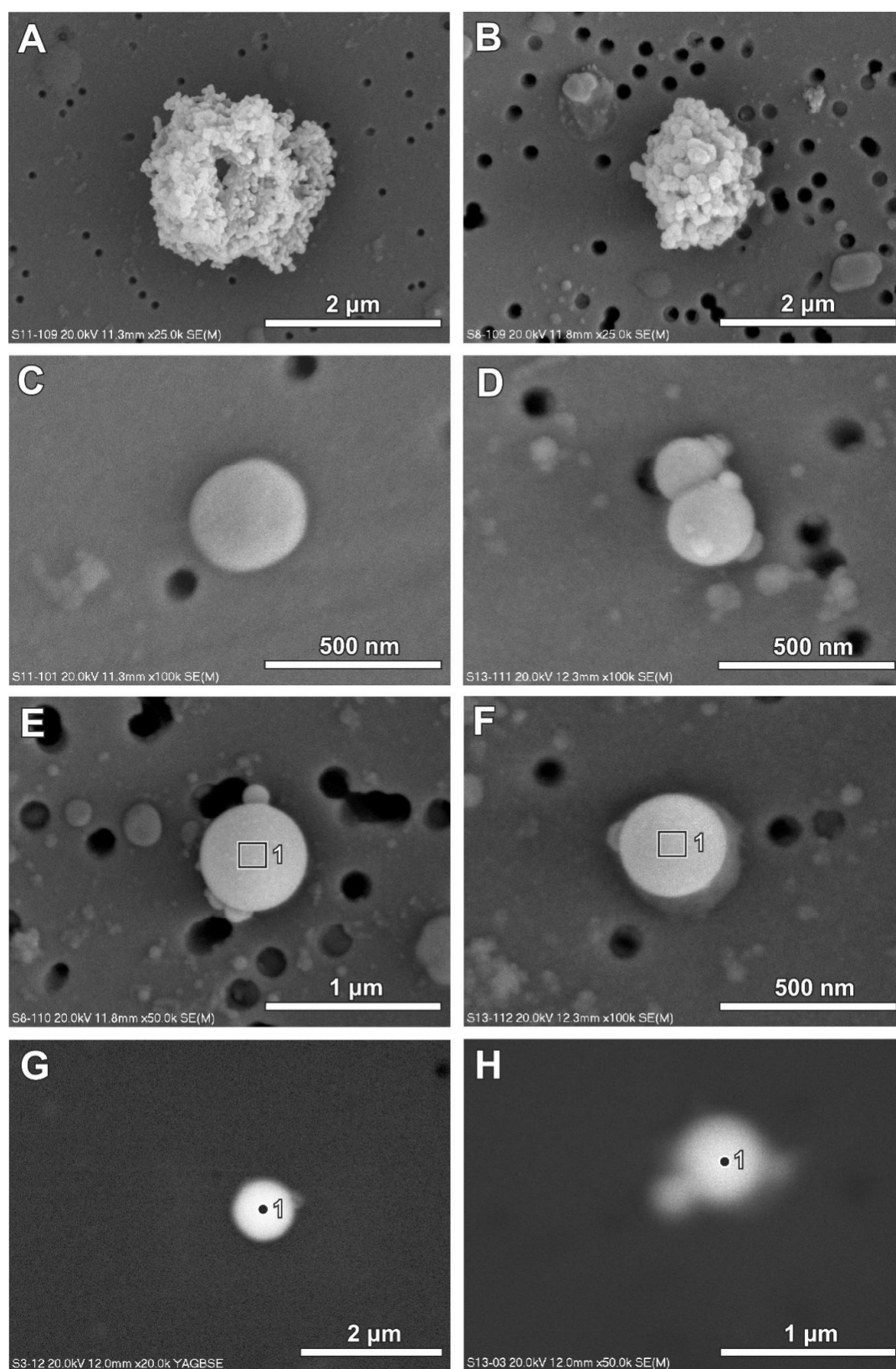


Figure 3. Aerosol particles (SEM; SE images except (G)—BSE image; chemical composition for indicated spots and areas—see Table 2; numbers correspond to spots listed in Table 2). (A,B) Compact soot particles; (C,D) tar balls; (E) spherical aluminosilicate particles; (F) spherical aluminosilicate particle enriched in Zn; (G,H) Fe-rich spherical particles.

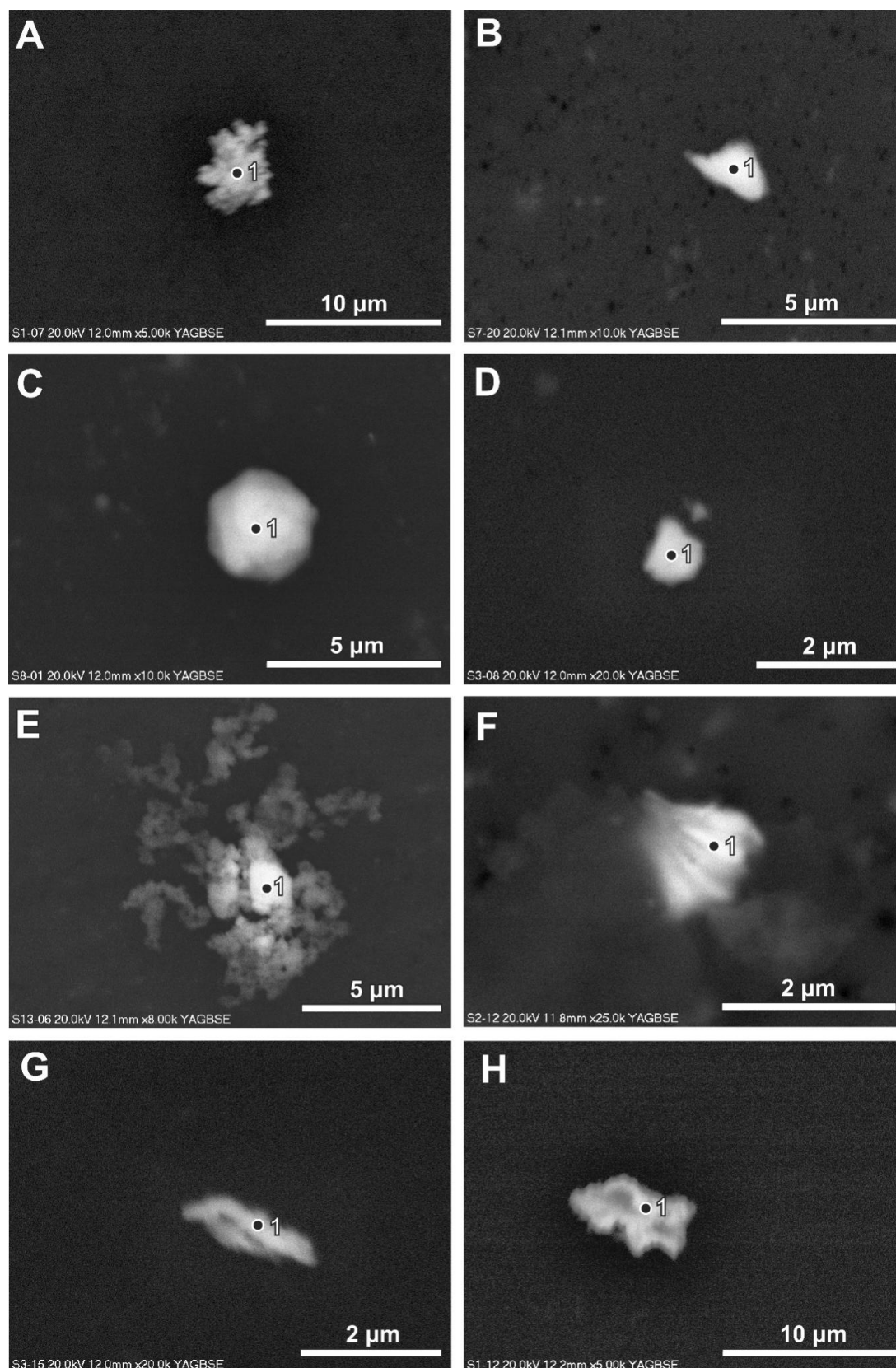


Figure 4. Aerosol particles (SEM; BSE images; chemical composition for indicated spots and areas—see Table 2; numbers correspond to spots listed in Table 2). (A–H) Fe-rich particles of irregular morphology ((C)—high content of S; (E)—high content of Zn; (F)—high content of Cr; (G,H)—high content of Cr and Ni).

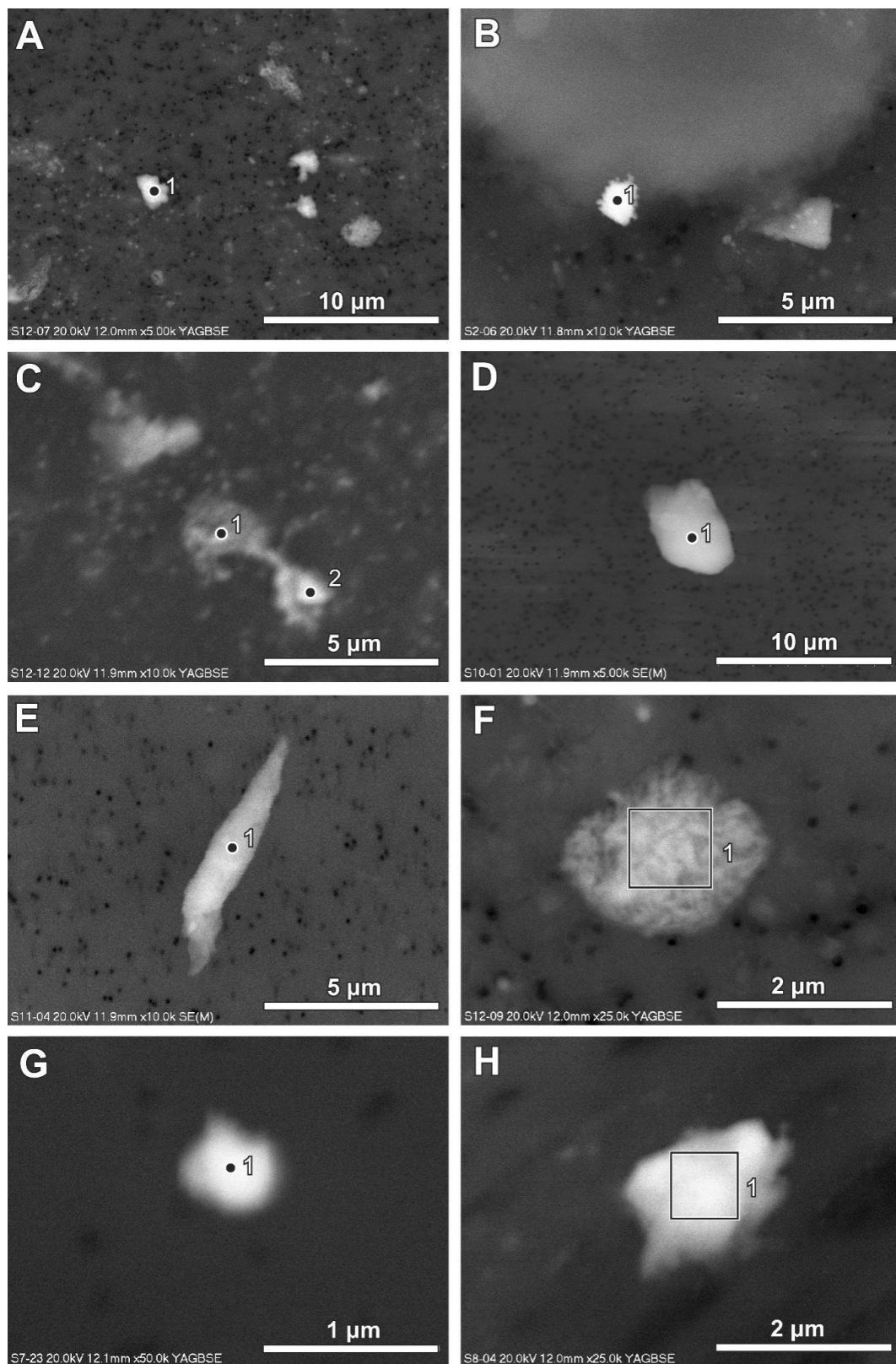


Figure 5. Aerosol particles ((A–C,F–H)—SEM; BSE images; (D,E)—SEM; SE images; chemical composition for indicated spots and areas—see Table 2; numbers correspond to spots listed in Table 2). (A) Particle rich in Fe, Zn with Cu and Ni; (B) Particle rich in Fe, Cr, Ni, Mn; (C) Zn-, Fe- and Cu-rich particles; (D) Cr-rich particle; (E) Cr- and Cl-rich particle containing Zn; (F) Zn-rich particle; (G) Pb-rich particle; (H) Cu- and Zn-rich particle.

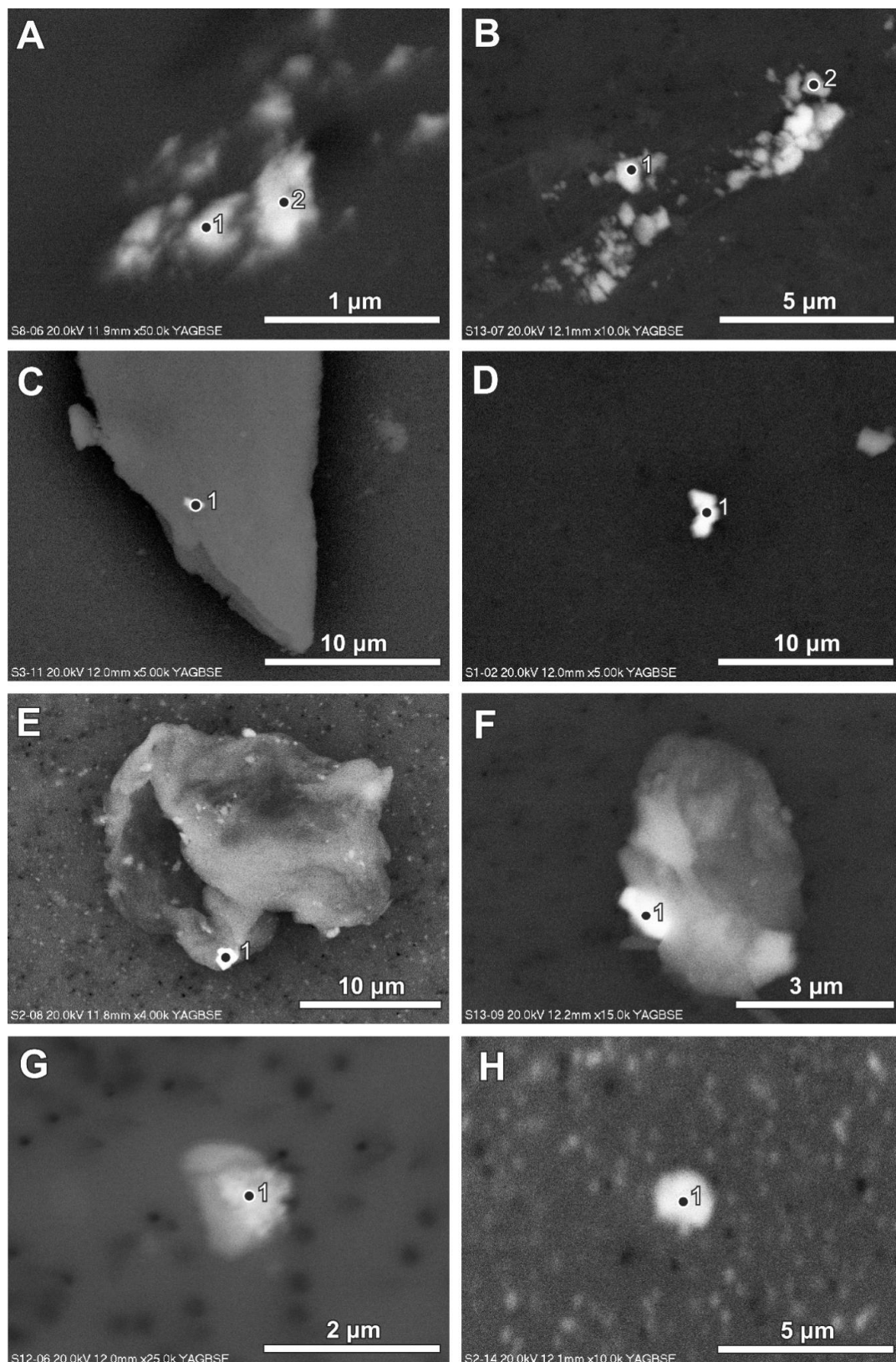


Figure 6. Aerosol particles (SEM; BSE images; chemical composition for indicated spots and areas—see Table 2; numbers correspond to spots listed in Table 2). (A) Au-rich particles; (B) Sn- and Pb-rich particles; (C) Zn-rich particle attached to carbonaceous grain; (D) Zr-rich particle; (E) Ni-, Nb-, Pt-rich particle attached to a carbon-rich grain; (F) Ni-, Nb-, Pt-rich particle; (G) Ni- and S-rich particle containing Fe; (H) Al- and Ni-rich particle.

Spherical aluminosilicate particles (Figure 3E,F) varying in size from $<10\text{ }\mu\text{m}$ to $200\text{--}300\text{ }\mu\text{m}$ could also be related to coal combustion in power plants [36]. In the studied samples, aluminosilicate spheres can be related to local [13] or remote sources. Similar particles are noted in soot formed during coal and other fuel combustion in household ovens [37]. Both local and remote sources of these particles could be considered. Natural sources (volcanic eruptions, micrometeorites) are less probable, and the chemical composition of particles is typical of so-called fly ash [36].

3.1.4. Metal-Rich Particles

Metal-rich particles occurred as discrete particles or in clusters (Figures 4F and 6A,B). The occurrence in clusters was related probably to their deposition on filters in the form of low-viscosity, likely hydrated material.

The group of metal-rich particles is strongly diversified both in terms of chemical composition and particle morphology and size. The interpretation of their origin (anthropogenic or natural) may be difficult because of variations in the chemical composition of PM; numerous possible sources (mining sites, industrial plants), often poorly described in the literature; and the rapidly increasing diversity of products (e.g., multicomponent alloys). For interpretation, it is necessary to keep in mind the recent mining (e.g., 144 Au mines, 58 Fe mines, 45 Cu mines, 39 Ni mines active in 2015) and smelting activities in the Arctic and sub-Arctic [67] as well as historical exploitation [68].

Spherical Fe-rich particles (Figure 3G,H) (Fe oxide) represent typical material related to high-temperature industrial processes [69,70], while such particles originating during in-cylinder melting of engine parts [71] or rail transport [72] usually fall in the nanoparticle size range. Interpreting the origin of irregular Fe-rich particles is challenging (Figures 4 and 5A–C). An opinion that usually spherical particles are formed in industrial processes is not supported by numerous observations (for a discussion, see, e.g., [73]). Fe-rich particles in the analyzed samples (Figure 4) were represented by mainly oxides (usually far from the stoichiometry of typical Fe-oxides), but in some particles, a high content of S was determined together with a very low content of O, indicating the presence of sulfides (Figure 4C). Some Fe-rich particles contain a significant content of other elements (e.g., Zn—Figure 4E; Cr—Figure 4F; Cr and Ni—Figure 4G,H and Figure 5B; Zn, Cu, Ni—Figure 5A; Zn, Cu—Figure 5C). The origin of Fe-rich particles devoid of other metallic elements could be either natural or anthropogenic. Their relatively high abundance may suggest an important role of an anthropogenic source. Particles containing Cr, Ni or Zn are most probably of anthropogenic origin related to Fe metallurgy and production of numerous alloys, e.g., Fe-Cr-Ni alloy (e.g., [74,75]). This suggestion is supported by the limited occurrence of natural compounds of similar composition.

Particles dominated by different metals (with or without Fe) (Figure 5C–H) are commonly interpreted as anthropogenic related to various sectors of industry, due to the rare occurrence of components of similar chemical composition in nature. The cluster of particles containing Fe, Zn and Al (Figure 5C; Table 2) exhibited a significant variation in the proportions of metal concentrations between analytical spots. The particle rich in Cr and devoid of other metals in Figure 5D (Table 2) contained S beside O (probably a mixture of Cr oxide and sulfate). The elongated particles rich in Cr and containing Zn (Figure 5E, Table 2) represent perhaps mixed material composed of oxide and chloride. Particles with predominant Zn (Figure 5F; Table 2) probably represent a sulfate phase. Pb-rich particles were scarce (Figure 5G; Table 2) and represented by oxide, probably with an admixture of sulfate. Particles containing Cu and Zn with a very low content of oxygen (Figure 5H; Table 2) originated probably from the production of a common alloy, i.e., brass (e.g., [76]).

Particles containing Au were rare (Figure 6A; Table 2). Atmospheric Au particles are most probably related to Au mining [67] or metallurgy. Other anthropogenic sources (e.g., dispersion of Au-containing products) are not probable, and neither are natural ones, e.g., from volcanic emissions [77]. Sn- and Pb-rich particles (Figure 6B; Table 2) were noted in the cluster. Sn-Pb alloys are commonly produced and used in different applications

(e.g., [78,79]). The metallic Zn (or slightly oxidized Zn) particle (Figure 6C) is most probably also anthropogenic since the natural occurrence of native Zn is very limited. The unique occurrence of a Zr-rich particle (Figure 6D; Table 2) has an uncertain form, most likely oxide (ZrO_2). However, the mineral baddeleyite (ZrO_2) occurs rarely in nature, and ZrO_2 is industrially produced for numerous applications that indicate the likely anthropogenic origin of this particle. Particles containing Ni, Nb and Pt can differ in O content (Figure 6E,F; Table 2), and they represent fragments of Ni-Nb-Pt alloys [80,81]. Particles containing Ni, S and a relatively low content of oxygen can represent slightly oxidized Ni sulfide (Figure 6G; Table 2). The Al- and Ni-rich particle (Figure 6H; Table 2) perhaps represents a Ni-Al alloy, a material with numerous applications (e.g., [82,83]). The relatively high abundance of Ni, Pt, Cu and other metals in PM samples from Spitsbergen, noted also by other authors [84] can be related to the exploitation and smelting of Ni and platinum group element (PGE) ores in northern parts of Eurasia and North America (e.g., Norilsk, Kola Peninsula and Sudbury industrial centers) [85–92]. Shevchenko et al. [93] consider Norilsk, Kola Peninsula, the Urals and the center of the European part of the former Soviet Union as possible sources of Ni in aerosols in the Russian Arctic (Franz Josef Land and Severnaya Zemlya).

Single-particle chemical analysis of particles considered to be anthropogenic is very useful in the interpretation of their sources. It can be also important in the attribution of chemical elements to different sources. This attribution can be different from an interpretation based on chemical analysis of whole aerosol samples for several elements (e.g., Al, Si, Fe). In [94], these elements are included in the group of crustal elements. An analysis of single particles indicates that these elements are, at least partially, of anthropogenic origin.

3.2. POPs in Hornsund Air

The determination of the content of selected POPs in air samples (Figure 7, Tables S5 and S6, cf. Figure S1 for station location) covered three periods: 4–24 April, 24 April–2 May and 2–11 May.

The range of atmospheric concentrations for the sum of three HCHs (α -, β - and γ -HCH) was 40.6–100 $pg\ m^{-3}$, with the largest average share of γ -HCH being 57% (concentration range: 22.5–68.4 $pg\ m^{-3}$). The second largest share was α -HCH (41%, concentration range: 17.4–29.8 $pg\ m^{-3}$). The concentrations of β -HCH were much lower (0.18–0.78 $pg\ m^{-3}$). The concentrations determined by us were higher than those determined in neighboring areas of the High Arctic (Figure 7): at Zeppelin station in northern Svalbard, in 2017–2019, these ranged from <LOD to 5.8 $pg\ m^{-3}$; at Villum (N Greenland), in 2017–2019, they ranged from 1.76 to 9.7 $pg\ m^{-3}$; and at Alert (Canadian Arctic), in 2017, they ranged from 2.41 to 13.4 $pg\ m^{-3}$ “<https://ebas-data.nilu.no/>” (accessed on 31 July 2023). Moreover, the studies carried out in Ny-Ålesund in the period 2016–2018 showed a concentration range of 11.3–19.9 $pg\ m^{-3}$ [95]. In addition to the difference in concentration values in the air, differences can be shown in the ratio between α -HCH and γ -HCH, which is related to the fact that technical HCH was used initially; technical HCH is a mixture of isomers with the following percentages: α -, β -, γ - and δ -HCH with 60–70, 5–12, 10–12 and 6–10%, respectively [96,97]. This mixture was then replaced by lindane which consists of 99% γ -HCH [98]. An α -HCH/ γ -HCH ratio in the range of 3–7 indicates the influence of technical HCH, while lower values signify an increasing share of lindane [98]. In Hornsund, the average of three cases was 0.75, suggesting an influence of lindane in contrast to the [18,95] works where technical HCH prevailed.

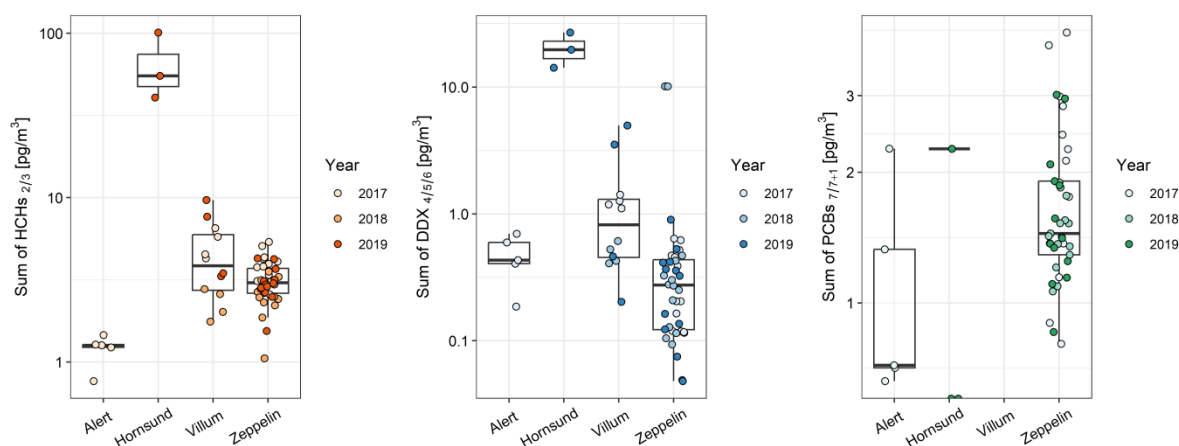


Figure 7. Comparison of concentration ranges of *HCH*, *DDX* and *PCB* sums in the High Arctic atmosphere: $\Sigma HCH = [\alpha-HCH] + [\beta-HCH] + [\gamma-HCH]$, except Zeppelin and Alert, where $[\beta-HCH]$ was not reported, most likely due to its low concentrations; $\Sigma DDX = [o,p'-DDD] + [p,p'-DDD] + [o,p'-DDE] + [p,p'-DDE] + [o,p'-DDT] + [p,p'-DDT]$, except Villum (no $[o,p'-DDD]$ reported) and Alert (no $[o,p'-DDD]$ or $[p,p'-DDD]$ reported in the study period); $\Sigma PCBs = [PCB28] + [PCB52] + [PCB101] + [PCB118] + [PCB138] + [PCB153] + [PCB180]$, except Alert, where the sum includes also *PCB-163*, due to joint value reporting with *PCB-138*. The differences in compound concentration sums due to listed differences (single unreported components) were unlikely to produce the differences in *HCH* and *DDX* concentrations between Hornsund and the other stations since the order-of-magnitude differences persist even if Hornsund data are reported for the minimal number of compounds available at another station. The box marks the Q1–Q3 span (i.e., the interquartile range [IQR]), with the median marked as a thick line. Whiskers reach no further than $1.5 \times IQR$ from the box hinge, where $IQR = Q3 - Q1$. Please note the logarithmic scale of concentrations. Data used for the chart come from the following sources: Hornsund—this study; Alert, Villum, and Zeppelin—EBAS “<https://ebas-data.nilu.no/>”, (accessed on 31 July 2023)” [22]; also see Acknowledgements. See Figure S1 for station location in the Arctic.

The range of atmospheric concentrations for the sum of *DDXs* was $14.2\text{--}26.9 \text{ pg m}^{-3}$, with the largest average share for *o,p'-DDE* (54%, concentration range: $8.4\text{--}12.6 \text{ pg m}^{-3}$) and second largest share for *p,p'-DDT* (18.4%; $2.4\text{--}5.2 \text{ pg m}^{-3}$). *DDD* contributed 16.5% ($<\text{LOD}\text{--}9.8 \text{ pg m}^{-3}$) to the *DDX* sum, while *p,p'-DDE* contributed only 11% ($1.6\text{--}3.7 \text{ pg m}^{-3}$). Again, the concentrations determined in Hornsund exceeded those determined elsewhere in the High Arctic, with the concentrations at Zeppelin, Villum and Alert available for the period 2017–2019 (EBAS) being $0.044\text{--}4.0 \text{ pg m}^{-3}$, $0.203\text{--}4.99 \text{ pg m}^{-3}$ and $0.208\text{--}1.36 \text{ pg m}^{-3}$, respectively. Furthermore, [95] reported $1.01\text{--}3.16 \text{ pg m}^{-3}$ of *DDXs* at Ny-Ålesund in 2016–2018. However, our data show a similarity of the predominant *DDX* compound to the cited data (*o,p'-DDE*). The predominance of *o,p'-DDE* may be due to its weaker ability to adhere to aerosols at low temperatures than other isomers [99]. The *p,p'-DDE/p,p'-DDT* ratio can be used to determine the source of *DDT*: a ratio >1 indicates aged *DDTs*, while ratios <1 indicate fresh inputs [100,101]. At Hornsund, the average ratio was 3.2, indicating aged sources of *DDT*, consistently with the previously cited studies [100,101].

The range of atmospheric concentrations for the sum of *PCBs* was $0.46\text{--}2.26 \text{ pg m}^{-3}$, where the largest share fell to the more volatile *PCB-28* and *PCB-52*; the remaining less volatile compounds were determined only in one of the three samples. The concentrations we found are lower than those determined in the vicinity of Ny-Ålesund: on Zeppelin Mt., in the period 2017–2019, similarly calculated ΣPCB concentrations ranged from 0.059 to 3.83 pg m^{-3} (Ebas), and in Ny-Ålesund town (in 2016–2018 [102]), ΣPCB concentrations reached $7.5\text{--}26.9 \text{ pg m}^{-3}$. On the other hand, at Alert in 2017, they were $0.41\text{--}1.93 \text{ pg m}^{-3}$, slightly less than in our study in 2019. In all cases, the more volatile *PCBs* also accounted

for the majority of Σ PCBs. This indicates the most likely source of PCBs in Svalbard is LRAT, as congeners with higher volatility have a higher transport potential [103,104].

3.3. POP Sources

The concentrations of OCPs determined by us were much higher than those determined elsewhere in the High Arctic during the spring. To explain this phenomenon, we first investigated the differences between individual POP concentrations in our three samples in relation to atmospheric conditions during each sampling period. The outstanding sample was the one taken in the period 24 April to 2 May, characterized by significantly higher contents of γ -HCH, Σ DDX and Σ PCB than other samples. During its collection, air temperatures were close to zero for the longest period, and an influx of air masses from the south was noted as a potential pathway for LRAT. Such increased temperature could have favored the re-emission of POPs from seawater, and the air trajectories then passed over sea without an ice cover (cf. Figure S2). This revolatilization of POPs from the sea is gaining importance as climate change progresses and sea ice cover shrinks [28,30], and the effects on atmospheric POP concentrations may be pronounced [105]. A period of warm temperatures may also have promoted a “spring maximum event”, consisting of specific chlorinated POPs being removed rapidly from the snow cover losing its grain surface area. Short-term high α -HCH and γ -HCH concentrations may increase to quadruple background levels during such events [106,107]. In a study conducted at Villum Research Station, [35] found a positive association between temperatures and α -HCH levels. This indicates that the presence of α -HCH at this location was significantly impacted by secondary processes like the release of the chemical from water through volatilization. On the other hand, the presence of γ -HCH in the air at the same station was not affected by temperature changes, implying that it probably originated from direct atmospheric transportation.

The sample taken in the period 17–24 April, unlike the sample collected in the period 24 April–2 May, during similar temperature and air-mass-inflow conditions, was characterized by the lowest POP concentrations in our study. This could have been caused by heavy precipitation which effectively washed POPs out of the air in the first four days of the sampling period (cf. Figure S4). These four days were also the only part of the sampling period with air mass inflow from the lower latitudes, typically interpreted as a source of anthropogenic contaminants (cf. Section S2.3, Supplementary Materials). Cappelletti et al. [108] referred to an LRAT event that took place in Hornsund between 14 and 21 April (cf. S.2.3 Figure S3A–C), associated with the influx of warm and humid air masses from Northern and Central Europe, which resulted in an increased content of ^{14}C radionuclide in snow. Since the event coincided with our sample of low concentrations, we surmise that precipitation (rainfall and snowfall) was effective in removing any POPs associated with it from the air.

As the content of POPs in our samples could be influenced by the effects of LRAT or local emissions, they should be considered for individual compound groups, as the impacts may be different. The PCB content was above the detection limit only in two samples, characterized by above-zero temperatures during collection and southern air-mass inflow. The concentration level of PCBs in the air at Hornsund was lower than that encountered at a southern Norway station, Birkenes “<https://ebas.nilu.no/>” (accessed on 31 July 2023), and was not exceptional among the measured High Arctic concentrations from a similar period (Figure 7, cf. Figure S1 for location). These factors support the greater influence of LRAT on PCB content, although the impact of revolatilization cannot be excluded. Since the only sample with significant south-westerly wind influence during sampling was the first one (Table 1), which was characterized by a relatively low level of PCB concentrations, and the measured PCB concentration range did not differ significantly from the range measured at Ny Ålesund (Figure 7), we interpret the potential impact of the waste incinerator in Hornsund on the collected samples to be unimportant in the study period.

For OCPs, characteristic concentration ratios were considered with respect to the origin of pollutants. The average α -HCH/ γ -HCH ratio of 0.75 suggested a source connected to

lindane, typically associated with LRAT (as the low ratios indicate newer *HCH* sources) rather than re-emission from the long-term storage in the Arctic environment. However, revolatilization from snow may be the last stage in such an LRAT event. Consistently with this interpretation, the lower value of the α -*HCH*/ γ -*HCH* ratio in the first two air samples coincided with air mass inflow from the south. On the other hand, the p,p' -*DDE*/ p,p' -*DDT* ratio yielded an average of 3.2, indicating older emissions, i.e., likely re-emission from secondary sources in this case. Revolatilization from the sea may be such a source.

The arguments given above confirm that the main source of *PCBs* is LRAT, while LRAT does not explain the high levels of *HCH* and *DDX* concentrations determined in the atmospheric air at Hornsund. In their case, the meteorological conditions were conducive to the revolatilization of these compounds; however, the evidence is not conclusive. More detailed studies with better time resolution of sampling are needed to explain this phenomenon, yet the high concentrations detected point to an important problem for the Arctic air quality, which would not be noted at the regular monitoring site located further north in Svalbard (Zeppelin).

Consistently with the partial origin of *POPs* deduced to be from LRAT, several particles found in the SEM-EDS analyses have also unequivocally been assigned to distantly located human activities, and further particles likely had anthropogenic sources. This is also in agreement with elevated concentrations in late April and early May of several typically anthropogenic components at the more southerly located stations (in the sub-Arctic), such as Storhofði (sulfate and heavy metals in aerosol and precipitation) and Jäniskoski (nitrate and sulfate in precipitation), as well as the even more southern Birkenes (sulfate and heavy metals, elemental carbon and levoglucosan in atmospheric particulate matter) “<https://ebas-data.nilu.no/>” (accessed on 31 July 2023”).

3.4. Study Limitations

The study limitations include the relatively small number of samples collected, especially in the case of *POPs*, and the difficulty in comparing elemental analysis to bulk analyses, since the latter were not performed. Thus, total mass concentrations of elements could not be considered here.

Furthermore, a limitation pertains to the representativeness of individual particle sources because of the small sample included for single-particle analysis. As natural aerosol particles predominated in the studied samples, and soot and tar ball particles occurred relatively often among anthropogenic components, which could be from either local or distant sources, the relative contribution of the most characteristic particles for source attribution was too small for statistically meaningful information. Metal-rich particles could be considered a remote source indicator, yet the number of particles identified in individual samples was too low to interpret the differences between samples as statistically important (cf. [108]). Because of this reason, the potential sources of single particles found in the collected samples were not analyzed using the backward trajectory method.

4. Conclusions

Besides the prevailing natural aerosol particles, anthropogenic ones were identified in aerosol samples collected at Hornsund. The anthropogenic origin of some particles was established with high likelihood, especially for the spherical particles formed in high-temperature processes (Fe-rich or aluminosilicate particles) or metal-rich particles of the chemical composition corresponding to alloys or other industrial products but not typical for natural origins (rocks, minerals, weathering products, etc.). Carbonaceous particles formed in combustion processes (soot, tar balls) and secondary sulfate, both most likely anthropogenic, aligned with typical components of Arctic haze events. For a select group of anthropogenic particles, only remote industrial sources could be accepted, supporting the interpretation of the Arctic haze type of background pollution at the time of sampling and corroborating the recently proposed shifting of the Arctic haze season towards late spring [2,6].

The concentrations of Cl-POPs, whose original anthropogenic origin is beyond doubt, could have come from both long-distance transport and local sources, especially secondary sources. It seems that only in the case of compounds from the PCB group can we assume a dominant influence of LRAT on their content in atmospheric air, because during the sampling in which they were detected and marked, atmospheric conditions suitable for this phenomenon occurred, such as the inflow of warm air from the southern areas. Moreover, the concentrations of PCB compounds found in the atmospheric air in Hornsund were similar to the concentrations determined at other year-round monitoring stations in the Arctic. However, the concentrations of ΣHCH and ΣDDX determined in this study were an order of magnitude higher than those at year-round monitoring stations in the High Arctic, and we would attribute such high concentrations to the remobilization from the ocean or snowpack, thus highlighting the role of water in atmospheric pollutant transport into the Arctic. Therefore, further research and the expansion of the atmospheric pollution research infrastructure at Hornsund should be encouraged, as processes related to POP re-emission may be occurring there at a previously unprecedented scale.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/w16111486/s1>, Figure S1. Map of the relative location of the compared air monitoring stations within the Arctic: (a) northern part, (b) southern part. Background: Google Earth; Figure S2. Sea ice situation on May 6th in the area surrounding Svalbard; Figure S3. Air mass trajectories arriving at Hornsund during each of the sample collections (labeled with the last day of sampling): left—automatic mid-boundary layer height; middle—500 m above mean sea level (AMSL); right—1000 m AMSL; Figure S4. Precipitation (bars) and average air temperature (dots) measured at the PPSH station in April and May 2019; Table S1. Analytical parameters; Table S2. Temperature program; Table S3. Mass/charge ratio and retention time; Table S4. Validation parameters such as recovery, coefficient of variation (CV) for ten samples with known POP content, limit of detection (LOD) and limit of quantification (LOQ); Table S5. The content of seven PCBs in atmospheric air [pg/m^3]; Table S6. Content of selected organochlorine pesticides in atmospheric air [pg/m^3] [109].

Author Contributions: Conceptualization, F.P., K.K., W.W.-M., M.M. and Ź.P.; methodology, F.P., K.K., W.W.-M. and M.M.; validation, F.P., K.K., W.W.-M. and M.M.; formal analysis, F.P., K.K., W.W.-M. and M.M.; investigation, F.P., K.K., W.W.-M., M.W. and M.M.; resources, K.K. and Ź.P.; data curation, F.P., W.W.-M. and M.M.; writing—original draft, F.P., K.K., W.W.-M., M.M. and S.L.-K.; writing—review and editing, F.P., K.K., W.W.-M., M.W., M.M., S.L.-K. and Ź.P.; visualization, F.P., K.K., M.M. and S.L.-K.; supervision, W.W.-M. and Ź.P.; project administration, K.K.; funding acquisition, K.K. All authors have read and agreed to the published version of the manuscript.

Funding: This study was funded by the National Science Centre, Poland project No. NCN 2017/26/D/ST10/00630, RiS ID 11108. The Ministry of Science and Higher Education statutory tasks funds for the Faculty of Geography and Biology, Pedagogical University and Faculty of Geography and Geology, Jagiellonian University, funded the research of W.W.-M. and M.M., respectively. The study was included in “The Anthropocene as the Epoch of Natural Environment Transformation” project at the Pedagogical University. At Jagiellonian University, the study was performed within the “Anthropocene” Priority Research Area under the “Excellence Initiative—Research University” program. We also thank for the support from the Polish Ministry of Education and Science Project No. DIR/WK/201805 for the Polish Polar Station Hornsund. Air monitoring data from Zeppelin (Norway) are provided by NILU (PI Pernilla Bohlin-Nizzetto) for the atmospheric contaminant monitoring program funded by the Norwegian Environment Agency. Air monitoring data from Alert (Canada) are provided by Environment and Climate Change Canada (ECCC) (PI Hayley Hung) with funding from the Northern Contaminants Program (Crown-Indigenous Relations and Northern Affairs Canada). Air monitoring data from Villum Research Station were provided by Rossana Bossi (Aarhus University) with funding from the Danish Environmental Protection Agency (DANCEA funds for environmental support to the Arctic Region).

Data Availability Statement: The raw data supporting the conclusions of this article will be made available by the authors on request.

Acknowledgments: The staff of the Institute of Geophysics PAS and the Polish Polar Station Hornsund (PPSH) are thanked for establishing and maintaining the meteorological monitoring and support

during fieldwork, and so is the station personnel from the 41st expedition, K. Kosek, C. Larose, K. Jankowska and S. Henningsen for fieldwork support. The authors gratefully acknowledge the UNECE CLRTAP (EMEP), AMAP and NILU for the foundation of the EBAS database which contains, among others, data on the content of POPs in the atmospheric air of the Arctic. The authors are also grateful to the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and the READY website (<https://www.ready.noaa.gov>) used in this publication. W. Obcowski is acknowledged for the preparation of figures presenting aerosol particles.

Conflicts of Interest: The authors declare no conflict of interest.

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