

What does a graduation tower aerosol contain? Possibilities and limitations of the method for determining mineral salts in graduation tower aerosol

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Abstract: Graduation towers are structures that saturate the air with a brine aerosol, mimicking a coastal microclimate. They are increasingly attracting interest for both their therapeutic potential and recreational use. However, scientific evidence supporting the effectiveness of inhalation therapy using brine graduation towers is limited, as are data on methods for determining the mineral composition of the brine aerosol. The aim of this study was to evaluate an analytical method for determining the chemical composition of aerosols generated by brine graduation towers, in which water-soluble ionic species are extracted using a gas washing bottle equipped with a sintered glass disc. Water-soluble components were collected by drawing air through the gas washing bottle filled with deionised water. The method parameters were first optimised in a purpose-built chamber generating a controlled aerosol stream. Following laboratory experiments, the procedure was validated under field conditions to determine the major ionic constituents of the air at a health resort brine graduation tower. Collected aerosol samples were analysed using high-performance ion chromatography (HPIC), flame photometry, and inductively coupled plasma optical emission spectrometry (ICP-OES). The mean concentrations were 5.2 mg/m³ for sodium, 9.4 mg/m³ for chloride, and 0.37 mg/m³ for calcium. Concentrations of potassium, magnesium, arsenic, cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, antimony, sulphate(VI), nitrate(V), and phosphate(V) were below the detection limits.

Keywords: graduation tower, graduation aerosol, brine, aerosol sampling, health resort treatment

INTRODUCTION

The number of brine graduation towers in Poland is growing rapidly. Large graduation towers are being commissioned in health resort municipalities

and, under the 2024 regulation, are officially recognised as health resort treatment facilities (Burkowska-But et al. 2014, Rogula-Kozłowska et al. 2022, Rozporządzenie 2024, Chudy et al. 2026). Smaller facilities, also known as micro-graduation

towers, are being constructed outside health resort areas. Micro-graduation towers are typically small structures found in urban spaces, with a height not exceeding 4 m, a length of 8 m, and a width of 2 m. The surface area of the blackthorn (Latin: *Prunus spinosa*) used as the brine-dispersing medium in micro-graduation towers is often a dozen or several dozen square metres, whereas in health resort graduation towers, the corresponding area is measured in thousands of square metres (Engelhardt & von Borstel 2015, Kalwasinska et al. 2018). As a result, the efficiency of producing a brine air aerosol is lower for graduation towers located in non-health resort areas (Engelhardt & von Borstel 2015, Rogula-Kozłowska et al. 2022, Bodziacki & Wolny-Koładka 2023, Chudy et al. 2026). Poland's largest health resort graduation towers include structures in Ciechocinek (19th century), Inowrocław (2001), Gołdap (2014), Wieliczka (2014), and Busko-Zdrój (2020) (Burkowska-But et al. 2014, Kalwasinska et al. 2018, Rogula-Kozłowska et al. 2022, Kamińska & Krusiec-Świdergoń 2023, Toloczko et al. 2023, Ciuchciński et al. 2025, Chudy et al. 2026). Due to the beneficial effects of saline aerosol on respiratory system disorders (Cătălina et al. 2012, Burkowska-But et al. 2014, Zajac et al. 2020, Bralawska et al. 2022, Flores et al. 2022, Ciuchciński et al. 2025), visits to these towers are recommended for patients staying in sanatoriums or health resort hospitals, which results in a high number of users. In addition, the towers are popular with tourists and locals due to their distinctive appearance and specific microclimate (Kejna et al. 2022, Görden & Rossi-Schwarzenbeck 2023). Reliable data on the number of people using brine aerosol exposure in graduation towers are scarce, although the largest towers attract hundreds of thousands of visitors annually, e.g., the graduation tower in Ciechocinek is visited by more than 500,000 people per year (Ciechocinek city website n.d.).

The primary legal framework governing health resorts in Poland is the Act of 28 July 2005 on Health-Resort Treatment, Health Resorts, Health-Resort Protection Areas and Health-Resort Municipalities (Ustawa 2005, Art. 5). This legislation defines, among other aspects, the therapeutic directions of health resorts, the principles

of supervision of health resort treatment, and the criteria for granting the status of a health resort or a health resort protection area. Brine graduation towers are classified as health resort treatment facilities, which must comply with the requirements specified in the Regulation of the Minister of Health of 30 September 2024 (Rozporządzenie 2024). According to this regulation, health resort graduation towers should be located in zone 'A' of the health resort protection area; operate according to a functional instruction defining the method of aerosol production and the scope and frequency of monitoring sanitary, hygienic, and functional conditions, including the quality of water used and the technical condition of the equipment; and employ water recognised as therapeutic for the production of therapeutic aerosol.

The therapeutic properties of mineral waters are confirmed by certificates issued by bodies authorised by the Ministry of Health. In balneotherapy, chloride-sodium waters are used to produce saline aerosol, provided that they meet the requirements laid down in the Regulation of the Minister of Health of 13 April 2006 on the scope of tests necessary to determine the medicinal properties of natural therapeutic resources and the medicinal properties of the climate, the criteria for their assessment, and the model certificate confirming these properties, as amended (Rozporządzenie 2006). The definition of brine in hydrogeochemical and hydrogeological literature is closely related to the concentration of dissolved salts, particularly sodium chloride (NaCl), although the exact thresholds may vary slightly depending on the context (geological, environmental, or technological). In the classical hydrogeological approach presented by Heath (1983), groundwater is classified according to total dissolved solids (TDS). Brines represent the extreme end of this classification and are typically defined as waters with salinity exceeding 35 g/L, i.e., comparable to or higher than seawater (Kharaka & Hanor 2003). In practice, this means that NaCl is the dominant component responsible for most of the mineralisation. Similarly, according to the definition in the Geological and Mining Law, brine is groundwater containing dissolved mineral substances, including sodium chloride (NaCl), with a total mineralisation of not less than 35 g/L (Ustawa 2011). In

environmental and technological contexts, for example in the review by Panagopoulos et al. (2019) on desalination, brine is defined more operationally as a waste solution with a salinity higher than seawater, usually greater than 35 g/L NaCl or equivalent total dissolved salts. Geochemical studies emphasise that brines are waters with very high mineralisation, typically above 50 g/L (Tralka-Blachowicz et al. 2025). Institutional sources, such as materials from the Polish Geological Institute – National Research Institute, also indicate that brines are groundwater with very high mineralisation, typically above 35–50 g/L, dominated by sodium chloride (Państwowy Instytut Geologiczny – Państwowy Instytut Badawczy website n.d.). In Polish balneology, brines are defined as chloride–sodium waters with mineralisation at least 15 g/L (Ponikowska & Kochański 2017). Brines classified as medicinal mineral waters are characterised by chemical and microbiological purity as well as natural variability in their physicochemical properties (Rozporządzenie 2006).

Aerosol therapy is a commonly used method in health resort treatment, as inhalation provides a rapid and convenient route for introducing the therapeutic substance into the human body. Sodium-chloride aerosols can form naturally in coastal zones and underground salt pits as well as through the use of various technologies: in areas surrounding brine graduation towers, above-ground salt caves, or during nebulisation. Health resort graduation towers, which are health resort treatment facilities, function as structures that saturate the air with a brine aerosol, creating specific microclimatic conditions (Burkowska-But et al. 2014, Kalwasinska et al. 2018, Kejna et al. 2022). The air surrounding the towers is often cooler, especially during high temperatures in summer, when heat losses due to the evaporation of water from the brine flowing over blackthorn twigs are the greatest (Ostwald & Banning 2021, Kejna et al. 2022, Görgen & Rossi-Schwarzenbeck 2023, Chudy et al. 2026). Brine evaporation increases the relative humidity (Kejna et al. 2022, Görgen & Rossi-Schwarzenbeck 2023). Salt aerosol is formed as a result of brine flowing over the structure of the graduation tower fill, which is usually composed of blackthorn twigs. The flowing brine is broken up by wind into small droplets

that form an aerosol around the structure (i.e., droplets and crystals dispersed into the surrounding air) (Burkowska-But et al. 2014, Burkowska-But 2016, Kejna et al. 2022). Staying within the perimeter zone of the graduation towers allows inhalation under natural conditions. The aerosol dispersed around the towers primarily contains sodium chloride and, depending on the type of therapeutic water used, may also contain iodides, bromides, calcium, magnesium, or iron (Engelhardt & von Borstel 2015, Ostwald & Banning 2021, Schäffer et al. 2021, Tralka-Blachowicz et al. 2025). The therapeutic effect of the mineral waters used for aerosol therapy in health resorts is mainly based on hydration of the respiratory tract mucosa, supplementation of deficient elements such as sodium, potassium, calcium, and magnesium, mechanical cleansing of the mucosa from retained pathological secretions, and improvement of mucosal physiological function through the local osmotic effect induced by NaCl. These processes modify the physicochemical properties of the mucus covering the ciliated epithelium and enhance mucociliary transport, improving clearance of the respiratory tract (Ponikowska & Kochański 2017, Ziemska et al. 2020). Moreover, inhalation of saline aerosol has beneficial effects in various conditions, including improvement of respiratory system function and anti-inflammatory and immunomodulatory effects (Cătălina et al. 2012, Burkowska-But et al. 2014, Zając et al. 2020, Bralawska et al. 2022, Flores et al. 2022, Kamińska & Krusiec-Świdergoń 2023, Ciuchciński et al. 2025). Moreover, it has been reported that aerosol inhalation may reduce obstructive airway complications by decreasing inflammation in post-COVID-19 patients (Babov et al. 2024). However, there is still a lack of clinical studies on the health benefits of inhaling aerosols from brine graduation towers. Therefore, the salt content of the graduation aerosol can serve as a guideline for determining the recommended duration of patient exposure.

The quantity and distribution of brine aerosols depend on several factors, primarily meteorological conditions, including wind direction and strength, sunshine, and precipitation. Strong winds can lead to excessive dispersion of aerosol particles, whereas optimal conditions for high aerosol concentrations occur on sunny days with

moderate winds. The aerosol concentration varies depending on the distance from the graduation tower. The design and size of the tower are also important determinants of the amount and quality of the brine aerosol (Burkowska-But 2016, Görden & Rossi-Schwarzenbeck 2023).

Aerosol measurement methods differ substantially in terms of their performance, applicability, and inherent limitations (Chow 1995). Gravimetric techniques using filters are widely regarded as the reference standard due to their high accuracy and direct mass determination (Kim et al. 2004); however, they are time-consuming and do not provide real-time data (McMurry 2000). In contrast, real-time optical instruments, such as an optical particle counter, enable continuous monitoring with high temporal resolution, making them suitable for exposure assessment and field studies (Görner et al. 2012, Crilley et al. 2018). Nevertheless, their measurements are strongly influenced by the aerosol properties, including particle size distribution, refractive index, and humidity, often leading to systematic biases relative to gravimetric methods and necessitating instrument-specific calibration (Javed & Guo 2021, Moazami et al. 2023). Electrical mobility-based techniques, such as the Scanning Mobility Particle Sizer (SMPS), provide detailed particle number size distributions, particularly in the ultrafine range, but involve complex instrumentation and do not directly measure mass concentration. Additionally, electrostatic sampling approaches offer high sensitivity for fine and ultrafine particles and can be coupled with further physicochemical analyses, although their performance depends on particle charging efficiency and environmental conditions. Emerging optical methods, including photoacoustic spectroscopy, enable sensitive detection of light-absorbing aerosols, such as black carbon, but remain less standardised and are not yet widely applied in routine monitoring (Crilley et al. 2018, Molina Rueda et al. 2023).

There is a limited number of literature reports concerning studies on graduation tower aerosols. Two main techniques for determining the chemical composition of brine graduation tower aerosols and salt chamber aerosols are worth highlighting. In the first technique, air flows through a gas

washing bottle equipped with a sintered glass disc. To increase the liquid–gas contact area, air is dispersed beneath the water surface into small bubbles by means of a sintered glass disc. This procedure makes it possible to extract substances that are readily soluble in water from the air and to determine their concentrations (Czajka et al. 2006, Kostrzon et al. 2017, Drobnik et al. 2018). In the second approach, aerosol particles are collected on a filter according to their aerodynamic particle diameter. The content of a given fraction in the air is determined from the difference in the mass of the filter weighed before and after the passage of the sample. The collected material can then be dissolved in water to determine the chemical composition (Rogula-Kozłowska et al. 2017, 2022). This filtration method, initially developed for atmospheric particulate matter monitoring in atmospheric air in zones categorised as rural areas, urban areas, traffic areas, and areas with impacts from industrial sources, has been adapted for brine graduation tower aerosols (Polski Komitet Normalizacyjny 2024).

The rapid increase in the number of brine graduation towers, and consequently the growing number of people using them, raises several questions: Do the technical solutions used in tower construction ensure efficient aerosol generation? Which factors influence the mineral composition of the aerosol? Are the inhalations microbiologically safe?

Addressing these questions requires properly designed studies. The air in graduation towers should be analysed both for its chemical composition and microbiological quality, highlighting the need to review and validate aerosol sampling methods.

The aim of the present study was to evaluate the suitability of the gas washing bottle method for determining the chemical composition of brine graduation tower aerosols. The first part of the study focused on testing the precision and absorption efficiency of the technique for extracting sodium chloride from aerosols in a purpose-built laboratory chamber. Following these preliminary tests, the analytical procedure was validated under field conditions through the determination of the dominant ions in the air of a health resort graduation tower.

METHODS

Laboratory studies were conducted first under controlled (in vitro) conditions, followed by field (in vivo) measurements.

Laboratory studies (in vitro)

An 80 cm × 50 cm × 50 cm glass chamber was designed for in vitro testing (Fig. 1). Within the chamber, an air jet was generated using a Planet Energy E125 fan (5 in Figure 1). The airflow was controlled by adjusting the size of the inlet opening (1 in Figure 1) located in the top wall of the chamber. To increase the length of the gas path, the chamber was divided by an internal wall measuring 60 cm × 50 cm. Stirring baffles (3 in Figure 1) were also installed to improve aerosol mixing. An RF7 Dual Speed Plus nebuliser (2 in Figure 1) was placed near the air inlet port and connected to a Hospineb Flaem Junior F2000 inhaler (Flaem, Poland) located outside the chamber. The adjustable airflow enabled the generation of an aerosol jet with sodium concentrations ranging from 4 to 78 mg/m³. At the end of the gas pathway, two sets of washing bottles (4 in Figure 1) – each set consisting of two bottles connected in series – were positioned upstream of the ventilator, with gas lines connecting the bottles to aspirators. Series

connection of the washing bottles made it possible to determine the absorption efficiency (recovery), while the use of two independent sets allowed the assessment of precision. The nebuliser was filled with a 6.5% aqueous sodium chloride solution (POCH S.A., Poland). This concentration was selected due to its similarity to the sodium chloride content in the brine supplying the graduation tower in which further studies were conducted.

Samples were collected using Air Sampling Pump 224-52MTX air aspirators (SKC Ltd., UK), and the airflow was monitored with a calibrated check-mate air flow rate meter model 375-00205 (SKC Ltd., UK). The sample air was passed through two PSB 4/100 washing bottles (100 mL capacity, 40 mm diameter) connected in series, equipped with a 34/35 grind and an inverted G0 sinter (Labit, Poland). The grinds were sealed with Teflon gaskets. Each washing bottle was pre-conditioned by filling it several times with deionised water and leaving it for several days to reduce the sodium background.

Before sampling, each washing bottle was filled with 100.0 mL ± 0.3 mL of deionised water (conductivity < 0.5 µS/cm) with the use of a balance (weight 99.8 g ± 0.3 g, Axis AD2000 balance, verification scale interval $e = 0.1$ g, readability $d = 0.01$ g).

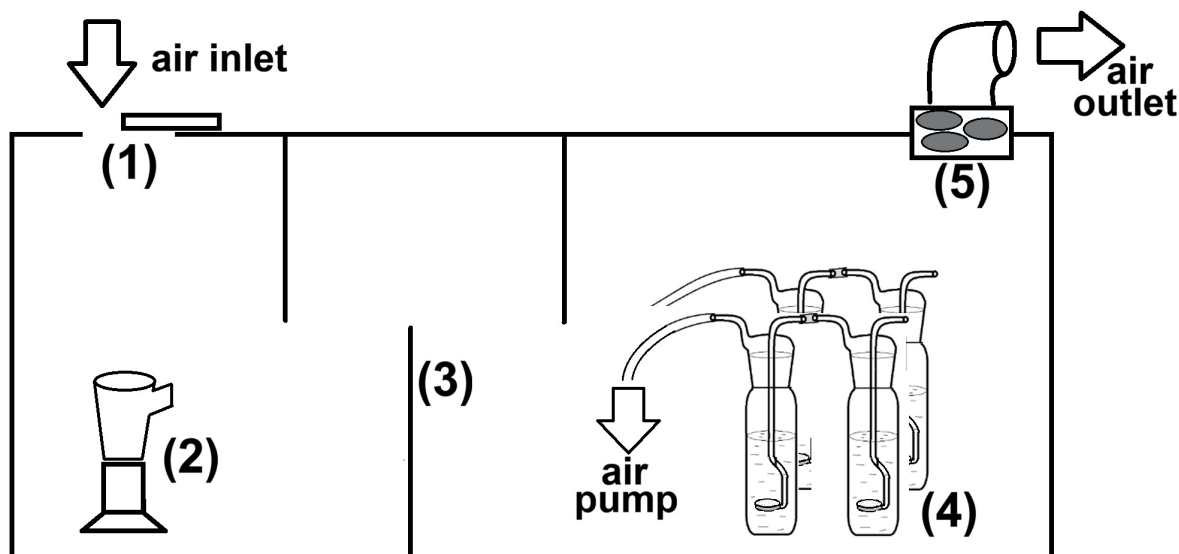


Fig. 1. Schematic diagram of the brine aerosol generation chamber (1 – air inlet, 2 – nebuliser, 3 – stirring baffles, 4 – washing bottles, 5 – air outlet with fan)

Preliminary studies established the optimum airflow through the washing bottles at 500 mL/min, which allowed the extraction of salts from 30 L of test air within one hour without spillage (monitored by weighing most of the bottles before and after sampling). The flow was measured immediately before and after sampling, under the conditions of the sampling process. After sampling, the bottles were sealed with parafilm, wiped both wet and dry, and weighed to determine water weight changes. Weight loss in the first of the two series-connected bottles ranged from 0.28 g to 0.47 g (0.28–0.47% of water volume), while the second bottle showed negligible changes (0.00–0.05 g).

The solution obtained after aerosol aspiration was poured out of the washing bottles via the air inlet tube, ensuring the transfer of salts deposited on the tube walls. The solution was stirred, re-circulated back into the bottle twice, and poured again through the same tube.

Eleven parallel sampling events were performed in the chamber. The absorption efficiency (recovery) was determined by analysing the sodium concentrations in the solutions from the sequential washing bottles. Using two sets of washing bottles for each sampling permitted the assessment of repeatability and measurement precision.

Field studies (in vivo)

Samples were collected under real-world conditions inside a brine graduation tower in Konstancin-Jeziorna, Poland. The graduation tower is shaped as an elongated closed circle with a circumference of 140 m and a height of 6 m (Fig. 2). Aerosol is produced by crushing drops of brine flowing down the tower and spraying brine through an atomiser, called a 'mushroom', located inside the circle. Brine is extracted from a borehole at a depth of 1,750 m near the graduation tower and contains 6.8% mineral matter.

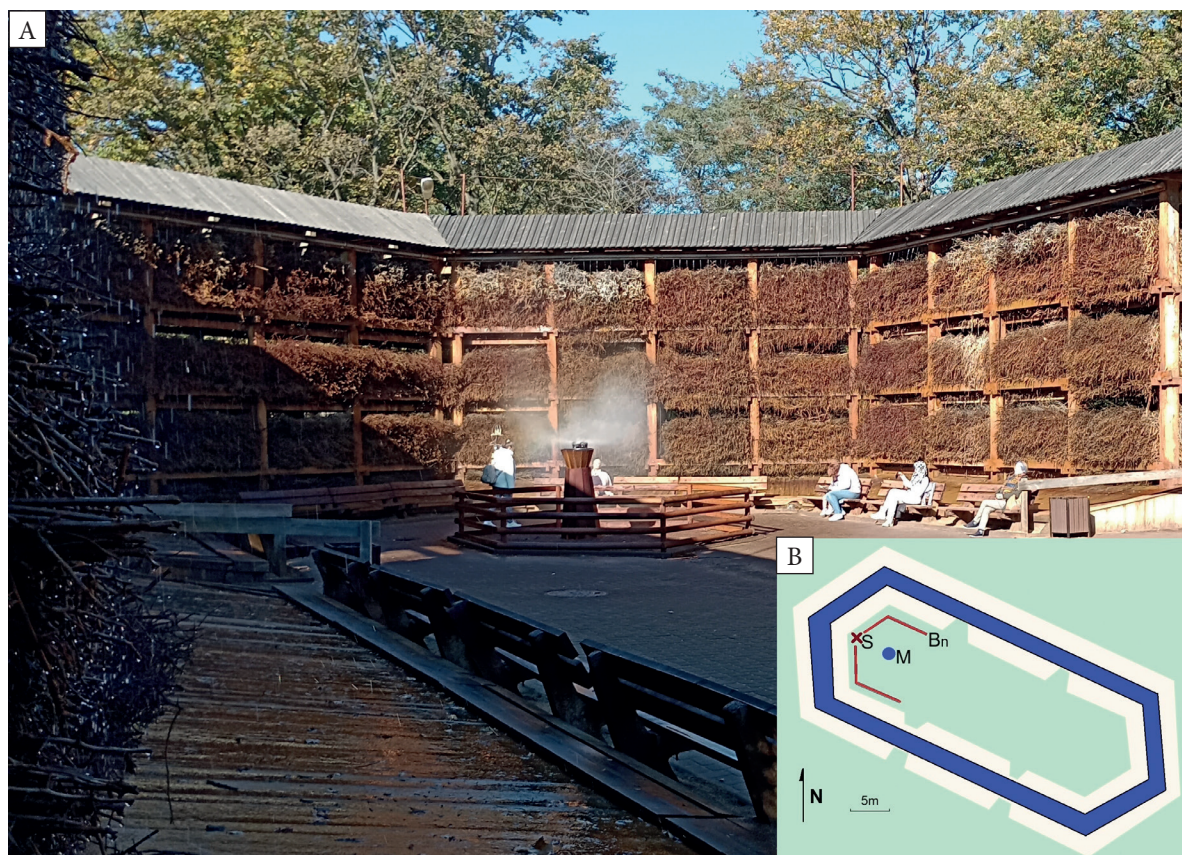


Fig. 2. Photo (A) and schematic diagram (B) of the graduation tower and sampling location (S – sampling point, M – atomiser, called a 'mushroom', Bn – benches most often occupied by visitors)

The physicochemical properties and chemical composition of the brine and circulating brine are presented in Table 1. The brine circulates in a closed circuit and is periodically replaced, while during operation it is fed directly from the intake to the sprayer.

Sampling was conducted on ten selected days between 14 August and 30 October 2024 (Table 2). Two sets of washing bottles (two bottles connected in series per set) were positioned at the location with the highest inhalation activity, near the brine-spraying mushroom (point S in Figure 2, about 3 m distance from the tower). On each occasion, air was passed simultaneously through the two sets at a flow rate of 500 mL/min for 60 min using two aspirators to assess repeatability under field conditions. Washing bottles were filled with deionised water (conductivity <0.5 µS/cm) and handled as described for laboratory studies. Five filled washing bottles were transported to

the sampling site each time – no air was passed through the fifth bottle, and its solution was subjected to the same analyses as the solutions from the other bottles. The bottle inlets were positioned at a height of approximately 150 cm. All washing bottles were weighed both after filling with water and after sample collection. Water loss in the first bottles ranged from 0.05 g to 0.48 g, and in the second bottles from 0.00 g to 0.11 g, with slight increases in two of the second bottles (0.05 g and 0.07 g). The solution obtained after aerosol aspiration was poured out of the washing bottles via the air inlet tube, ensuring the transfer of salts deposited on the tube walls. The solution was stirred, re-circulated back into the bottle twice, and poured again through the same tube.

Solutions collected after aerosol aspiration were analysed for chloride, nitrate(V), sulphate(VI), and phosphate(V) anions as well as sodium and potassium cations.

Table 1

Physicochemical properties and chemical composition of the brine extracted from a borehole and circulating brine

Parameter	Brine from the borehole*	Circulating brine*
pH (measurement temperature)	6.8 (24°C)	6.5 (21°C)
Electrical conductivity [mS/cm] (measurement temperature)	88.2 (24°C)	69.7 (20°C)
Total dissolved solids [mg/L]	68,100	48,300
Sodium [mg/L]	22,860 ± 3,200	16,250 ± 2,280
Potassium [mg/L]	167 ± 23	116 ± 16
Lithium [mg/L]	1.9 ± 0.2	1.5 ± 0.2
Calcium [mg/L]	2,325 ± 349	1,750 ± 175
Magnesium [mg/L]	681 ± 150	482 ± 48
Strontium [mg/L]	110 ± 12	74 ± 9
Barium [mg/L]	0.35 ± 0.05	0.34 ± 0.05
Iron [mg/L]	23 ± 4	30 ± 5
Manganese [mg/L]	0.96 ± 0.12	0.76 ± 0.09
Ammonium ion [mg/L]	36.0 ± 6.8	0.3 ± 0.1
Chlorides [mg/L]	40,770 ± 4,077	28,930 ± 2,893
Fluorides [mg/L]	0.53 ± 0.05	0.39 ± 0.04
Bicarbonates [mg/L]	178 ± 18	67 ± 7
Sulfates [mg/L]	685 ± 69	501 ± 50
Nitrates [mg/L]	68 ± 10	16 ± 2
Nitrites [mg/L]	<1.0	2.0 ± 0.4
Phosphates [mg/L]	<1.0	<1.0

* Research results obtained from the operator of the graduation tower in Konstancin-Jeziorna; analyses conducted in 2023 and 2024.

Table 2

Characteristics of the conducted sampling of graduation tower aerosol, the measurements performed, and the prevailing atmospheric conditions

Date	Start of the sampling	Temperature* [°C]	Wind* [km/h], direction	Other conditions	Analyses of solutions after aerosol aspiration (analyzed substances)
14.08.2024	10:17	24–25	20–28, SE	cloudless, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K)
30.08.2024	11:02	29–30	7–13, E	cloudless, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K)
05.09.2024	10:57	24–26	18–22, E	partly cloudy, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K)
13.09.2024	10:55	16–18	11, NE	total cloudiness, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K)
17.09.2024	11:01	21–23	9–11, NE	partly cloudy, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K), ICP-OES (Na, Ca, Mg, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb)
10.10.2024	11:03	17–19	17–20, SE	cloudless, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K), ICP-OES (Na, Ca, Mg, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb)
17.10.2024	11:10	9–11	30–33, SE	cloudless, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K), ICP-OES (Na, Ca, Mg, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb)
18.10.2024	10:59	11–14	19–20, SE	cloudless, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K), ICP-OES (Na, Ca, Mg, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb)
23.10.2024	11:08	11–12	15–19, W	medium cloudiness, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K), ICP-OES (Na, Ca, Mg, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb)
30.10.2024	10:53	14	20–24, W	total cloudiness, without rain	HPIC (Cl^- , SO_4^{2-} , NO_3^- , PO_4^{3-}), photometer (Na, K), ICP-OES (Na, Ca, Mg, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb)

* Data obtained from WeatherSpark (n.d.) for Warsaw Chopin Airport, located approximately 13 km from the graduation tower.

Electrical conductivity was measured using a CX-710 multimeter (Elmetron) with a CDT-3 sensor designed for low-conductivity solutions. Anion concentrations were determined using a high-performance ion chromatograph (HPIC) with conductivity detection, ICS-2500 (Dionex, USA), equipped with a four-channel gradient pump GS50, a conductivity detector ED50A, and an ASRS ULTRA II (4 mm) ion suppressor. Anions were separated using an AS9-HC analytical column with an AG9-HC precolumn, and an aqueous sodium carbonate solution was used as the mobile phase. The apparatus parameters used for the determination of anions are shown in Table 3.

Table 3

Apparatus parameters used for the determination of anions using a high-performance ion chromatograph (HPIC) with conductometric detection

Parameter	Value
Eluent composition	0.009 mol/L Na_2CO_3
Eluent flow rate	1 mL/min
Injected sample volume	0.025 mL
Suppressor current	45 mA
Pressure on the analytical column	~2,000 psi
Column temperature	30°C
Analysis time	25 min
Number of repetitions per sample	3

Sodium and potassium were measured using an XP 2011 flame photometer (BWB Technologies, UK).

The solutions obtained after aerosol aspiration in the graduation tower over six measurement days (17 September–30 October) were also analysed for sodium, calcium, magnesium, arsenic, cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, and antimony using an inductively coupled plasma optical emission spectrometer (ICP-OES) with a solid-state Charge Injection Device (CID) IRIS Advantage Duo ER/S (Thermo Jarrell Ash, USA). The characteristics of the ICP-OES spectrometer sample introduction system and selected instrumental parameters used in the study are presented in Table 4.

Table 5 shows the limits of detection and quantification of analytes in the solutions after aerosol aspiration for the mentioned analytical method and in air, calculated considering an air sample volume of 30 L.

Certified reference standards (ISO 17034) and environmental matrix reference material LETH-13 were used.

Table 4

Characterisation of the sample introduction system and apparatus parameters used for ICP-OES determination of metals

Parameter	Value
RF frequency	27.12 MHz
RF power	1,150 W
Torch type	Quartz Duo torch with horizontal geometry
Spray chamber type	cyclonic
Nebulizer	glass, concentric 26 psi
– type	
– pressure	
Argon flow rate:	15 L/min 1 L/min 4 L/min 3 L/min 80 units
– plasma	
– auxiliary	
– optical interface	
– detector purge	
– detector purge	
Pump speed:	110 rpm 110 rpm
– sample	
– waste	
Emission line integration time:	45 s (As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb) 30 s (Ca, Na, Mg)
– emission lines: 175–275 nm	
– emission lines: 275–780 nm	
ICP plasma observation system	axial (As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb) radial (Ca, Mg, Na)
Rinse time	60 s
Number of replicates per sample	3

Table 5

Limits of detection (LOD) and quantification (LOQ) of analytes in solutions after aerosol aspiration (analytical technique in brackets) and in air (V = 30 L)

Analyte (method)	LOD solution after aspiration	LOQ solution after aspiration	LOD aerosol (sample volume 30 L)	LOQ aerosol (sample volume 30 L)
Cl [−] (HPIC)	0.032 mg/L	0.065 mg/L	0.110 mg/m ³	0.220 mg/m ³
SO ₄ ^{2−} (HPIC)	0.047 mg/L	0.093 mg/L	0.150 mg/m ³	0.310 mg/m ³
NO ₃ [−] (HPIC)	0.05 mg/L	0.10 mg/L	0.17 mg/m ³	0.33 mg/m ³
PO ₄ ^{3−} (HPIC)	0.12 mg/L	0.24 mg/L	0.40 mg/m ³	0.80 mg/m ³
Na, K (photometric)	0.20 mg/L	0.50 mg/L	0.70 mg/m ³	1.70 mg/m ³
Ca (ICP-OES)	0.07 mg/L	0.14 mg/L	0.23 mg/m ³	0.47 mg/m ³
Mg (ICP-OES)	0.064 mg/L	0.128 mg/L	0.210 mg/m ³	0.430 mg/m ³
Na (ICP-OES)	0.056 mg/L	0.112 mg/L	0.190 mg/m ³	0.370 mg/m ³
As (ICP-OES)	4.5 µg/L	9.0 µg/L	15.0 µg/m ³	30.0 µg/m ³
Cd (ICP-OES)	0.48 µg/L	0.96 µg/L	1.60 µg/m ³	3.20 µg/m ³
Co (ICP-OES)	0.82 µg/L	1.64 µg/L	2.70 µg/m ³	5.30 µg/m ³
Cr (ICP-OES)	1.1 µg/L	2.2 µg/L	3.7 µg/m ³	7.3 µg/m ³
Cu (ICP-OES)	1.5 µg/L	3.0 µg/L	5.0 µg/m ³	10.0 µg/m ³
Fe (ICP-OES)	0.71 µg/L	1.42 µg/L	2.40 µg/m ³	4.70 µg/m ³
Mn (ICP-OES)	0.19 µg/L	0.38 µg/L	0.63 µg/m ³	1.26 µg/m ³
Ni (ICP-OES)	1.0 µg/L	2.1 µg/L	3.4 µg/m ³	7.0 µg/m ³
Pb and Sb (ICP-OES)	8.0 µg/L	25.0 µg/L	27.0 µg/m ³	83.0 µg/m ³

RESULTS AND DISCUSSION

In the present study, the parameters of the method using gas washing bottles with sintered discs to determine the concentrations of dominant ions in the brine graduation tower aerosol were optimised, and the method was subsequently tested under real conditions.

Range of cation and anion concentrations determined in aspirated aerosol from a brine graduation tower

Table 6 shows the concentrations of ions and elements determined in the brine graduation tower

aerosol. Sodium (ICP-OES) and chloride (HPIC) concentrations measured at the tower site were always above the limit of quantification. For sodium determined using the flame photometer, 80% of the samples exceeded the limit of quantification; two of ten measurements showed sodium concentrations slightly below 1.7 mg/m^3 , though still above the detection limit.

The determined concentrations of sodium, chlorides, and calcium in the graduation tower aerosol are shown in Table 7. The average mass ratio of chloride to sodium (Cl^-/Na^+) in the aerosol was 1.9, close to the ratio in the brine (1.8) (see Table 1) circulating in the tower.

Table 6

Average, minimum and maximum concentrations of ions and elements in the air from the tower perimeter zone

Concentration	Mean	Minimum	Maximum
Chlorides	9.4 mg/m^3	2.3 mg/m^3	35.3 mg/m^3
Sodium	5.2 mg/m^3	1.3 mg/m^3	21.0 mg/m^3
Calcium	0.37 mg/m^3 *	<LOD	0.45 mg/m^3 *
Magnesium, arsenic, cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, antimony, sulfates(VI), nitrates(V), phosphates(V)	below the limit of detection		

* Average of four measurements for which the calcium concentration exceeded the detection limit.

Table 7

Determined concentrations of sodium (photometer), chlorides (HPIC) and calcium (ICP-OES) in the graduation tower aerosol in parallel analyses and the spread between them

Date	Sodium concentration [mg/m^3]		Spread [%]	Chloride concentration [mg/m^3]		Spread [%]	Calcium concentration [mg/m^3]		Spread [%]
	Parallel	Average		Parallel	Average		Parallel	Average	
14.08.2024	19.6	21.0	13.6	30.2	35.3	28.7	n.d.*	n/a**	n/a
	22.5			40.3			n.d.		
30.08.2024	5.8	5.8	1.1	10.5	10.7	3.7	n.d.	n/a	n/a
	5.8			10.9			n.d.		
05.09.2024	3.1	3.0	6.6	6.2	6.1	5.0	n.d.	n/a	n/a
	2.9			5.9			n.d.		
13.09.2024	4.1	4.2	2.4	7.6	7.7	1.3	n.d.	n/a	n/a
	4.2			7.7			n.d.		
17.09.2024	4.2	4.1	4.5	8.1	8.0	3.8	0.46	0.45	8.9
	4.0			7.8			0.43		
10.10.2024	1.4	1.5	7.0	2.5	2.6	7.7	<0.23	n/a	n/a
	1.6			2.7			<0.23		
17.10.2024	3.6	4.3	31.9	6.8	8.1	31.1	0.34	0.41	35.6
	5.0			9.3			0.48		
18.10.2024	3.2	2.8	22.9	6.2	5.6	21.4	0.28	0.26	12.4
	2.5			5.0			0.24		
23.10.2024	3.8	3.8	2.6	7.2	7.3	2.7	0.37	0.36	3.2
	3.9			7.4			0.36		
30.10.2024	1.4	1.3	7.4	2.4	2.4	4.3	<0.23	n/a	n/a
	1.3			2.3			<0.23		
Average	n/a	5.2	10.0	n/a	9.4	11.0	n/a	0.37	15.0

* n.d. – not determined; ** n/a – not applicable.

In four of six measurements, calcium concentrations exceeded the detection limit, with an average value of 0.37 mg/m³ and an average Na⁺/Ca²⁺ mass ratio of 9.1, similar to the ratio in the brine (9.3). These results indicate that the spraying process and atmospheric conditions do not significantly alter the chemical composition of the brine, confirming the effectiveness of the aerosol collection method for the main mineral components.

In all the investigated solutions obtained after aspiration of the graduation tower aerosol, the concentrations of potassium, magnesium, arsenic, cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, antimony, sulphate(VI), nitrate(V), and phosphate(V) were below detection limits. Thus, the method and analytical techniques employed were only able to determine the sodium, calcium, and chloride concentrations.

Repeatability

Parallel air sampling with two sets of gas washing bottles was used to assess the repeatability, including subsequent solution analyses. Table 8 shows the mean, minimum, and maximum differences between the results obtained from parallel analyses of sodium concentrations (laboratory chamber) and sodium and chloride concentrations (field) in the graduation tower air.

Repeatability was significantly better under laboratory conditions than in the field, likely due to environmental variability. In the laboratory, airflow, temperature, and humidity were stable, whereas in the field, aspirators and collectors were exposed to fluctuating sunlight, temperature, and wind intensity. Apart from the sampling step, the other analytical steps were the same. These results indicate that the sampling stage is the critical factor affecting repeatability.

Salt absorption efficiency (recovery) in gas washing bottles connected in series

To evaluate the absorption efficiency during sampling, air was passed through two gas washing bottles connected in series. Due to the very high water solubility of the analysed salts, it was assumed that the mass of analytes breaking through both gas washing bottles is negligible compared to the mass retained within them and can therefore be disregarded. The recovery of salts in the first gas washing bottle was calculated using the following equation:

$$Rec[\%] = 100\% \cdot \frac{m.a._I - m.a._{bl}}{m.a._I + m.a._{II} - 2 \cdot m.a._{bl}} \quad (1)$$

where: $m.a._I$ – mass of analyte in the first washing bottle, $m.a._{II}$ – mass of analyte in the second washing bottle, and $m.a._{bl}$ – mass of analyte in the washing bottle through which no air was passed.

Calculations were carried out for sodium (chamber experiments) as well as for sodium and chlorides (graduation tower experiments). In all twenty-two chamber experiments, recovery exceeded 93%, with a mean value of 97.0%. Only two cases exhibited recovery below 95%. These results indicate that nearly the entire mass of salts was retained in the first gas washing bottle.

For sodium, recovery values obtained in twenty experiments conducted at the graduation tower were below 94% in two instances (68.1% and 81.0%), with a mean of 95.5%. For chlorides, the mean recovery was 96.7%, with values below 94% observed only once (66.9%). The reduced recovery is most likely attributable to contamination of the second solution during mixing and transfer from the gas washing bottle. This occurred at the highest recorded salt concentration (14 August 2024), when a visible salt layer was observed on the gas washing bottles following the sampling stage.

Table 8

Mean, minimum and maximum spread of the results of the determination of sodium concentration in the chamber and of sodium and chloride in the brine graduation tower air (parallel analyses)

Range	Mean [%]	Minimum [%]	Maximum [%]
Chamber / sodium (11 measurements)	1.3	0.3	3.7
Graduation tower/ sodium (10 measurements)	10.0	1.1	31.9
Graduation tower / chlorides (10 measurements)	11.0	1.3	31.1

Uncertainty

The combined uncertainty was calculated considering contributions from air sampling volume, recovery, collector background, water volume measurement, repeatability, and the analytical technique used for determining the ion in question (Na^+ or Cl^-). The last two factors contributed most to the overall uncertainty. After calculating the uncertainty using the sum-of-squares method, the resulting value was multiplied by a coverage factor of $k = 2$ to obtain the expanded uncertainty at a 95% confidence level. The resulting

uncertainty in the determination of sodium (flame photometry) and chloride (HPIC) was 20%.

Quality control

Quality control was ensured by:

- parallel sampling;
- determining the recovery of analytes in the first washing bottle when passing air through two bottles connected in series;
- analysis of certified reference standards, reference material, and blanks as part of quality control of ion determinations in the solutions obtained after aerosol aspiration.

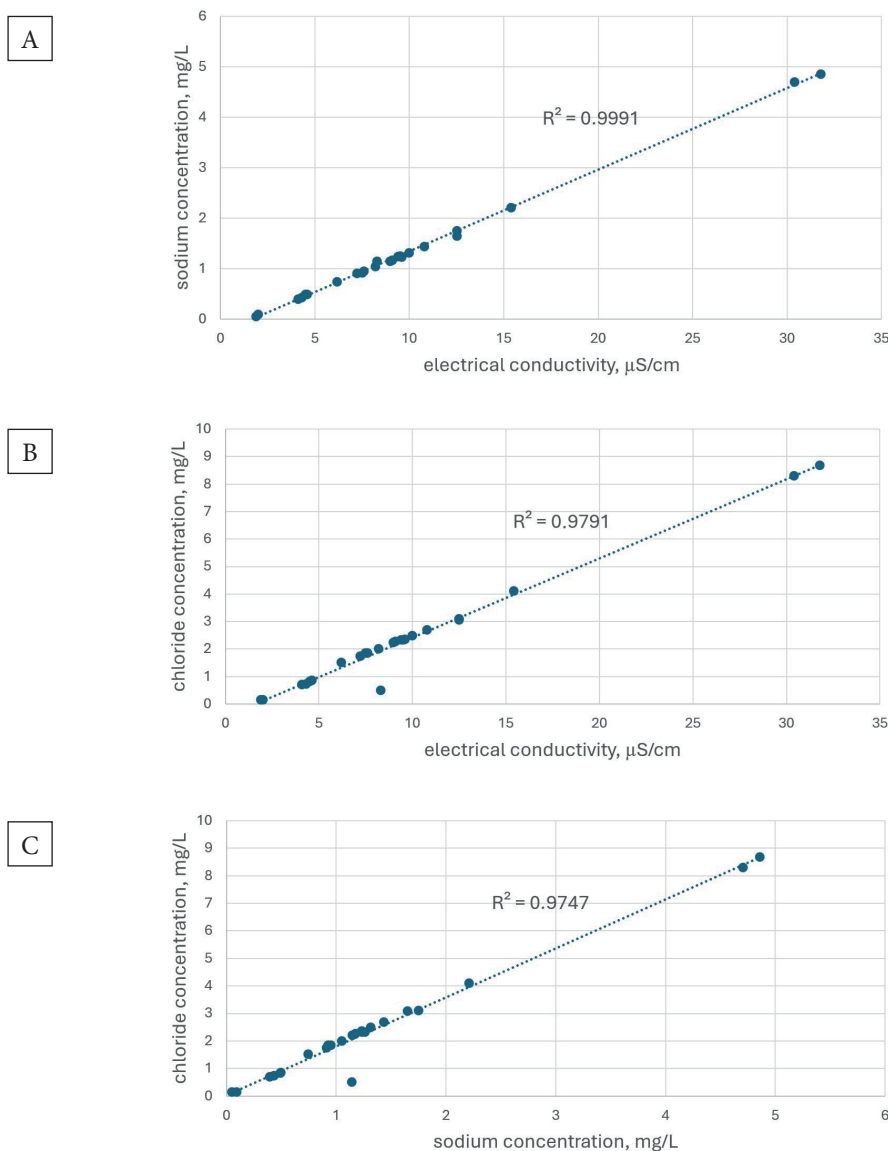


Fig. 3. Correlation between sodium concentration and electrical conductivity (A) and between chloride concentration and electrical conductivity (B); sodium and chloride concentrations (C) in solutions formed after the aspiration of a graduation aerosol

Correlations between sodium and chloride concentrations and between ion concentrations and solution electrical conductivity were also assessed. For correctly performed analyses, the relationships described should be linear due to the constant composition of the brine used to feed the graduation tower. These relationships are shown in Figure 3. The coefficients of determination (R^2) are only slightly less than unity.

CONCLUSIONS

Historically, brine graduation towers were primarily used for salt production. Today, their main application is outdoor inhalation therapy, as the high concentration of brine creates a coastal-like microclimate in their surroundings. The present study evaluated the suitability of the gas washing bottle method for determining the chemical composition of the brine graduation tower aerosol and enabled the determination of cations and anions in the aspirated aerosol. Aerosol sampling using gas washing bottles has several advantages, but also some limitations, which are listed below. An unquestionable advantage is its simplicity – substances initially dissolved in the brine tend to dissolve in the water filling the washing bottle. The resulting solution is an easy-to-analyse matrix that does not contain interfering substances. The technique does not require complex equipment, and an additional advantage is the good repeatability demonstrated in parallel analyses.

The main disadvantage is the high detection limit – the results obtained show that, using this technique for isolating mineral substances from air, only the major ions present in the brine in the highest concentrations can be determined (e.g., Na^+ , Ca^{2+} , Cl^-). Moreover, the results do not provide information about the particle size distribution of the aerosol. Minor limitations of the technique include difficulties in field handling and the transport of glassware, as well as the need for long-term conditioning of the washing bottles to eliminate the sodium background.

The following conclusions can be drawn:

1. The method enables the measurement of cation and anion concentrations in ambient air, with limits of quantification ranging from $1.3 \mu\text{g}/\text{m}^3$ for manganese (using ICP-OES) to $1.7 \text{ mg}/\text{m}^3$ for sodium (using a flame photometer).
2. Chloride, sodium, and calcium were detected and quantified in the air of the health resort graduation tower in Konstancin-Jeziorna. The mean concentrations were $5.2 \text{ mg}/\text{m}^3$ for sodium, $9.4 \text{ mg}/\text{m}^3$ for chloride, and $0.37 \text{ mg}/\text{m}^3$ for calcium. The concentrations of potassium, magnesium, arsenic, cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, antimony, sulphate(VI), nitrate(V), and phosphate(V) were below the detection limits.
3. The Cl^-/Na^+ and $\text{Na}^+/\text{Ca}^{2+}$ mass ratios in the aerosol closely correspond to those in the brine, indicating that the spraying process and ambient conditions do not alter the chemical composition of the brine.
4. The calculated expanded uncertainty of the method for determining sodium and chloride in the brine graduation tower aerosol is 20% ($k = 2$).
5. The method enables the determination of the total mass of selected anions or cations in the air passing through the washing bottles. However, the results do not provide information on the size distribution of the aerosol particles containing these salts, which determines their deposition location in the human respiratory system. Further research is required to characterise the composition of salt aerosol in graduation towers of various designs, including the analysis of particle size in the tower perimeter air.

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