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Development of nasal and throat spray based on Värskä mineral water from Gdov aquifer, Estonia

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ABSTRACT

The Värskä 6 well extracts mineral water from the sandstones of the Gdov aquifer at a depth of 575–595 m, with a total dissolved solids contents varying between 17 and 27 g/L. The aim of the work was to study 1) the market for nasal/throat sprays in Estonia; 2) the opinions of pharmacy employees and customers; 3) the composition and sterility of the mineral water; and 4) the possibility of developing a nasal/throat spray based on local mineral water. A total of 17 nasal sprays were marketed in Estonia in 2022, of which 12 were for nasal use only and 5 were nasal/throat sprays. Most (70%) were solutions with isotonic salt concentrations made from dissolved or undiluted sterilised seawater or ocean water. The indications for their use are diverse, ranging from daily nasal hygiene to the relief of colds. Värskä 6 water has a high salinity (20 g/L), a Na–Cl chemical type, and – compared to ocean water and nasal sprays made from ocean water – is enriched in lithophile elements (Fe, Mn, Li, Sr, and Ba) characteristic of deep groundwater. The hypertonic Värskä 6 mineral water is bacteriologically clean, and the temporal stability test confirmed that the mineral water does not promote bacterial growth but inhibits it. A new product was developed – the mineral water-based hypertonic nasal/throat spray Tsilk.

Introduction

Respiratory tract infections, such as the common cold, influenza, sinusitis, bronchitis, and tonsillitis, are very commonly experienced by most people; these illnesses are unpleasant and disruptive (Mayrhuber et al. 2018; Williamson et al. 2022). Their prevention and treatment include using rinses, nasal sprays, and irrigation. The action can be mechanical; they may either form a biological barrier or have a washing-out effect. Additionally, they may alter the environment of the nose and/or throat, reducing the viral load and the likelihood of the virus surviving/thriving (Bichiri et al. 2021; Kramer et al. 2021).

A survey of nasal spray users (n = 407) has shown that people who suffer from disruptive, frequent, or severe respiratory tract infections, or who are vulnerable to such infections, are interested in using a nasal spray for prevention. Among the factors preventing their use, side effects or fear of their occurrence are often mentioned (Williamson et al. 2022). For example, the administration of intranasal steroid preparations is problematic due to side effects, the risk of which decreases with lower doses. An increased risk of nosebleeds as an adverse effect of treatment at higher doses has been found (Chong et al. 2016a). The newer generation of intranasal steroid sprays should be used at the lowest effective dose (McDonnell et al. 2020).

Non-pharmacological therapies with good tolerability and safety profile are of interest to many patients with allergic rhinitis. The side effects of oral corticosteroids include hypothalamic–pituitary axis suppression, growth suppression in children, glaucoma, and cataracts (Bilstein et al. 2021).

A literature review on the efficacy of various topical therapies for chronic rhinosinusitis showed that topical saline and corticosteroids should be considered the first line of therapy. A hypertonic saline is recommended over an isotonic one if side effects are well tolerated (Huanga and Govindaraj 2013).

Several treatments involving nasal irrigation were discussed as early as 1902 in *The Lancet* (Wingrave 1902). Nowadays, various hypertonic or isotonic saline solutions, often prepared using seawater, are used as nasal washes (Wise et al. 2018). Nasal irrigation is a procedure that rinses the nasal cavity with saline solutions. This can be performed using low positive pressure from a spray, pump, or squirt bottle, with a nebuliser, or with gravity-based pressure using a vessel with a nasal spout (Chong et al. 2016b).

The application of different mineral waters in prophylactics, as well as in the treatment and rehabilitation of various diseases, has a long tradition. One of the most important fields of application is the treatment of nasal and throat problems. Rhinosinusitis is a group of disorders characterised by inflammation of the mucosa of the nose and paranasal sinuses, which affects patients' quality of life. Unfortunately, standard treatments for rhinosinusitis have known side effects and variable efficacy rates. Thus, complementary and integrative treatments such as crenotherapy, which utilises high mineral content water or vapour as an inhalation therapy, are of continued interest (Nimmagadda et al. 2022).

The mechanisms by which inhalation of mineral waters influences the treatment of lung and throat diseases have been studied in several papers (Sain et al. 2018; Wenchuo et al. 2020). It has been shown that the size, composition, and concentration of inhaled particles (liquids and gases) have a substantial influence on efficacy. Particularly efficient is the generation of mineral water aerosol using ultrasonic humidifiers and nebulisers.

During the last decades, much attention has been paid to the treatment of bronchial asthma, as it contributes significantly to global incidence and mortality rates. A recent review (Fesyun et al. 2023) examined the widespread treatment with mineral water inhalation and analysed the conflicting information regarding its effectiveness. The meta-analysis, drawing on 1266 sources and including 14 studies, concluded that patients with mild, moderate, and hormone-dependent bronchial asthma, after undergoing mineral water inhalation therapy, demonstrated a statistically significant decrease in the frequency and intensity of the cardinal symptoms of bronchial asthma, along with an improvement in forced expiratory volume (FEV1), compared with the control group.

Mineral waters from different geological sources may contain radon and other radioactive elements. Therefore, from the perspective of patient safety, radiological control of water intended for human consumption is necessary (Gümbür 2024). On the other hand, results also show that exposure to radon-containing mineral water leads to improved spatial perception, potentially improving age-related cognitive functions (Nikolaishvili et al. 2024).

Värška Sanatorium is located in southeastern Estonia and offers a range of wellness and medical treatments. The sanatorium was opened in 1980 and is known for the freshwater

mud and mineral water used there. The mud is mined from a local lake and is considered one of the world's most unique therapeutic muds. It is a fine-grained lake sediment with an organic matter content reaching 50%. The mud also has a high H₂S content (12–70 mg/100 g wet mud) and a high heat capacity (Ramst and Kask 1997).

The therapeutic mud bath is the most unique and popular mud therapy procedure at Värška Sanatorium. The mud, diluted with saline Värška mineral water at a temperature of 41–43 °C, acts on the body as a thermal, mechanical, and chemical irritant. Freshwater mud therapy is a procedure that treats chronic inflammation and relieves pain, is bio-stimulating (accelerating metabolism and cellular renewal), and removes excess fluids and toxins from the body. Thus, therapy baths help with joint and skin diseases, and positively affect the nervous system.

Värška Sanatorium also offers mineral water baths. Baths at a temperature of 37–39 °C have a calming and strengthening effect on the nervous system. Bromine-rich and highly salty Värška 6 mineral water has a soothing effect on the body and a healing effect on the skin.

A study on the Värška mud and Värška mineral water baths (Tuulik et al. 2022) showed a positive effect on musculoskeletal pain and muscle tension syndromes in the working-age population. Both the Värška curative mud bath and the mineral water bath demonstrated a positive effect and can be recommended as drug-free interventions for mild to moderate musculoskeletal pain syndrome and muscle tension, both for prevention and treatment.

This study was initiated by the desire to enhance the value of mineral water as a unique natural resource and to find a new and innovative way of using it. Thus, the aim of this project was to determine whether, and under what conditions, Värška mineral water is suitable for the production of a natural nasal/throat spray.

Materials and methods

Social pharmacy study

To gain an overview of current nasal spray-using practices in Estonia, an anonymous written pilot survey of community pharmacy employees (n = 100) and customers (n = 100) was conducted in January and February 2020. Pharmacists, pharmacy assistants, or customer service representatives who completed the questionnaire – either using a printed paper form or via the electronic survey platform eFormular.ee – worked in both urban and rural pharmacies (Tallinn, Tartu, Narva, Pärnu, and 15 Estonian counties). A written questionnaire was also offered to customers of three Tallinn pharmacies who expressed an interest in purchasing a solution containing sea salt. The synopsis of the nasal spray nomenclature marketed in Estonian pharmacies is based on the information available on the websites www.raviminfo.ee and www.apteegiinfo.ee.

Chemical study

Mineral water samples were collected from the Värška 6 borehole for chemical and microbiological analyses in 2018

and 2022. During sampling, the end of the sampling hose was disinfected, and the mineral water was allowed to flow freely for 15 minutes before collection. Sample bottles were transported to laboratories in a thermal box. The chemical compositions of nine natural nasal sprays sold in Estonian pharmacies were also analysed and compared with the compositions and chemical types of ocean water and Värskä 6 mineral water. Chemical analyses of the solutions (nasal sprays, mineral water) were carried out in the laboratories of the University of Tartu.

Mineral water samples for trace element analysis were filtered on site using 0.45 µm Teflon membrane syringe filters, and 9.1 mL of filtrate was collected into acid-washed polypropylene (PP) vials containing 0.6 mL of 1:1 HNO₃. Nasal spray samples were directly diluted prior to analysis using 2% HNO₃, and the same acid solution was used to prepare appropriate dilutions of acidified mineral water samples before analysis with inductively coupled plasma mass spectrometry (ICP-MS). ROTIPURAN® Supra 69% HNO₃ (Carl Roth GmbH + Co. KG, Germany) and 18.2 MΩ·cm water were used to prepare all acid solutions used in this study. All sample vials used for the collection and dilution of samples for ICP-MS analysis were 15 mL PP vials that were cleaned by soaking in 2% HNO₃ for 24 h, rinsed three times with 18.2 MΩ·cm water, and dried in a fume hood.

The main anions F⁻, Cl⁻, NO₂⁻, Br⁻, NO₃⁻, PO₄⁻, and SO₄²⁻ were quantified using a 930 Compact IC Flex ion chromatography system (Metrohm, Switzerland) from 200-fold dilutions of unacidified mineral water and nasal spray samples made with 18.2 MΩ·cm water.

HCO₃⁻ concentration was quantified with a TitroLine 6000 automatic titrator (SI Analytics GmbH, Germany) using 0.1 M HCl titration to pH 4.3. For the analysis, 50 mL of 10-fold diluted nasal sprays (made with 18.2 MΩ·cm water, due to the low sample volume) and 100 mL of undiluted mineral water were used. It was possible to measure two nasal sprays without dilution, and the calculated HCO₃⁻ concentrations from diluted and undiluted measurements deviated by less than 5%.

Na, Mg, K, Ca, and trace element concentrations were measured using an Agilent 8800 QQQ ICP-MS (Agilent Technologies, Inc., Canada). Indium, added online via a mixing 'T' in 2% HNO₃, was used as the internal standard element and measured on-mass as ¹¹⁵In in all acquisition modes. ⁷Li, ⁹Be, ¹¹B, ²³Na, ²⁴Mg, ²⁷Al, ⁵⁵Mn, ⁶⁰Ni, ⁶⁶Zn, ⁸⁵Rb, ⁸⁷Sr, ⁹⁵Mo, ¹¹⁸Sn, ¹²¹Sb, ¹³³Cs, ¹³⁷Ba, ²⁰⁸Pb, ²⁰⁹Bi, ²³²Th, and ²³⁸U were measured on-mass in NoGas mode. ⁴⁵Sc, ⁴⁷Ti, ⁵¹V, ⁵²Cr, ⁵⁹Co, ⁶³Cu, ¹⁰⁷Ag, and ¹¹¹Cd were measured on-mass using 6 mL/min of He in single-quad mode. ³⁹K and ⁴⁰Ca were measured on-mass using 8 mL/min of H₂ in MS/MS mode. ⁵⁶Fe, ⁷⁵As, and ⁸⁰Se were measured as mass-shifted ⁵⁶Fe¹⁶O, ⁷⁵As¹⁶O, and ⁸⁰Se¹⁶O using 0.6 mL/min of O₂ in MS/MS mode.

Calibration solutions were prepared by mixing Agilent multielement calibration standards 1, 2A, and 4, and 10 mg/L single-element standards of Bi, Sb, and Sn. Calibration standards were prepared in a concentration range of 0.1–250 µg/L using 2% HNO₃ for dilution. Detection limits for Na, Al, K,

Ca, Zn, and Fe were 5 µg/L; for As, 5 µg/L; for U and Th, 0.02 µg/L; and for other elements, 0.4 µg/L. Linear calibration using normalisation to ¹¹⁵In and batch-specific independent P/A factors were used. NIST SRM1643f, Trace Elements in Water, was used as a quality control standard, and the measured concentrations of all elements were within 5% of the reference value.

Microbiological study

Water samples for the microbiological study of the mineral water were collected in pre-sterilised glass bottles, placed in a thermal box, and transported to the laboratory on the same day. Analyses of microbiological quality were performed in three stages in the laboratory of AS Tartu Veevärk, according to the standards (EVS-EN ISO 9308-1:2014, EVS-EN ISO 7899-2:2002, and EVS-EN ISO 6222:2001). The first determinations were carried out immediately after water sampling, the second after one month, and the third after three months. Between analyses, the sample bottles were hermetically sealed and stored at room temperature.

Study area

The Värskä region is located at the outcrop of Middle Devonian terrigenous rocks in southeastern Estonia (Fig. 1). In addition to Devonian sandstones and siltstones, the bedrock sequence of the region includes Ordovician carbonate rocks with varying clay contents, as well as Cambrian and Ediacaran terrigenous rocks, such as sandstone, siltstone, and clay. Palaeoproterozoic metamorphic and igneous rocks form the crystalline basement at a depth of 600 m (Raukas and Teedumäe 1997). The bedrock is covered by a Quaternary sediment layer, which reaches a thickness of up to 25 m, based on geological cross-sections from boreholes at Värskä Sanatorium.

According to the hydrostratigraphic section of the region, the uppermost is the Quaternary aquifer system, followed by the Middle Devonian, Middle–Lower Devonian, Ordovician–Cambrian, and Cambrian–Vendian aquifer systems (Fig. 2).

The uppermost Quaternary aquifer system, consisting predominantly of glacial till and glaciolacustrine sandy loam, is a source of drinking water, mostly for private households. The groundwater level is typically 1–5 m below the surface, and the water is classified as Ca–Mg–HCO₃ type, with a total dissolved solids (TDS) contents of 200–500 mg/L (Perens and Vallner 1997).

The Middle Devonian aquifer system spreads throughout the entire southern Estonia and is the primary water supply source in this region. The aquifer-forming rocks are sandstones and siltstones interbedded with clay layers and lenses. The groundwater in this aquifer is fresh, of the Ca–Mg–HCO₃ type, with TDS typically ranging between 300 and 500 mg/L (Perens and Vallner 1997). The main water quality problems are associated with naturally high iron and manganese contents (Hiio and Karro 2012; Karro et al. 2020).

The Middle–Lower Devonian aquifer system is present at a depth of 250–300 m (Fig. 2) and is composed of fine-

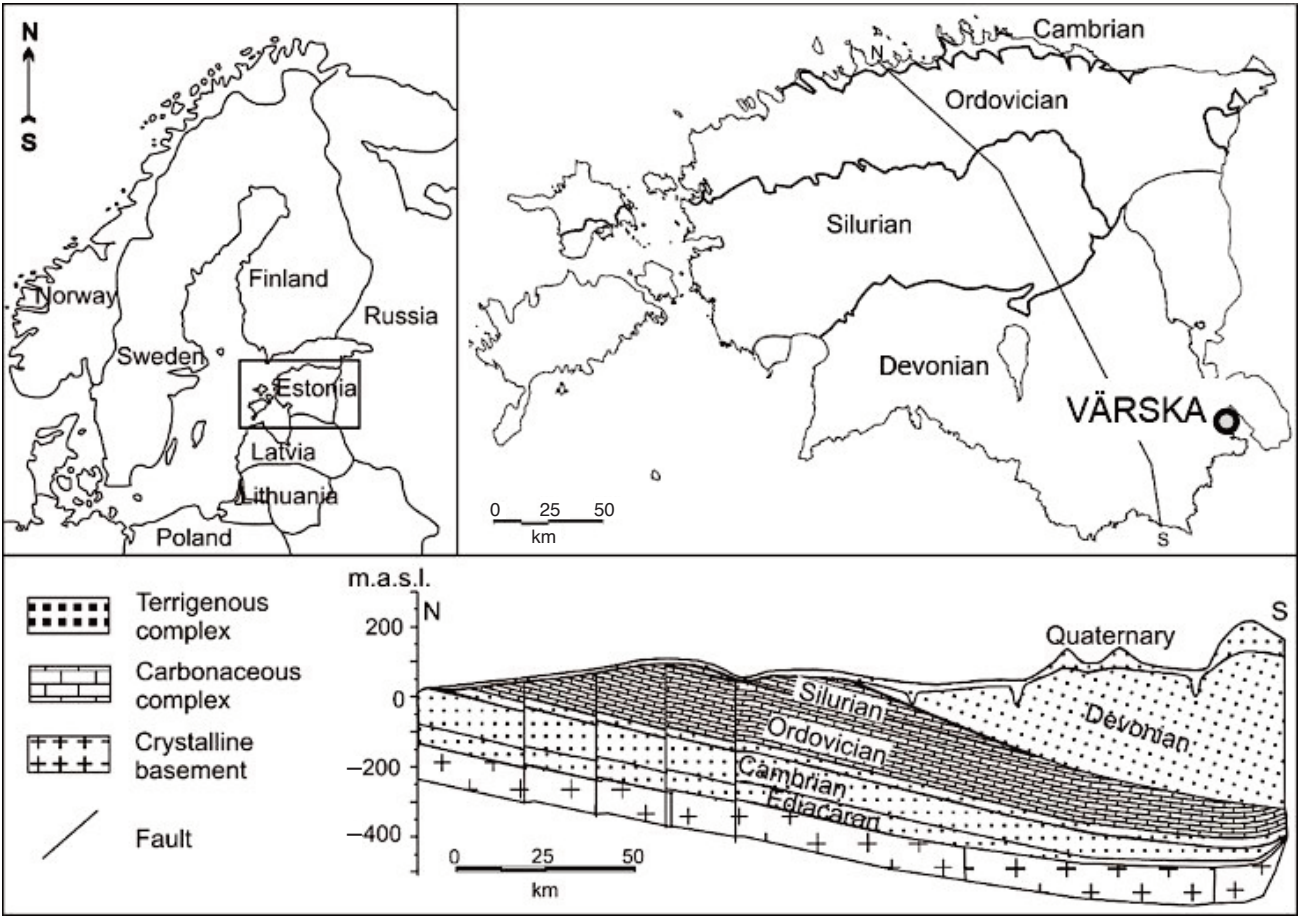


Fig. 1. Location of the study area (Värška) and the geological cross-section (North–South) of Estonia.

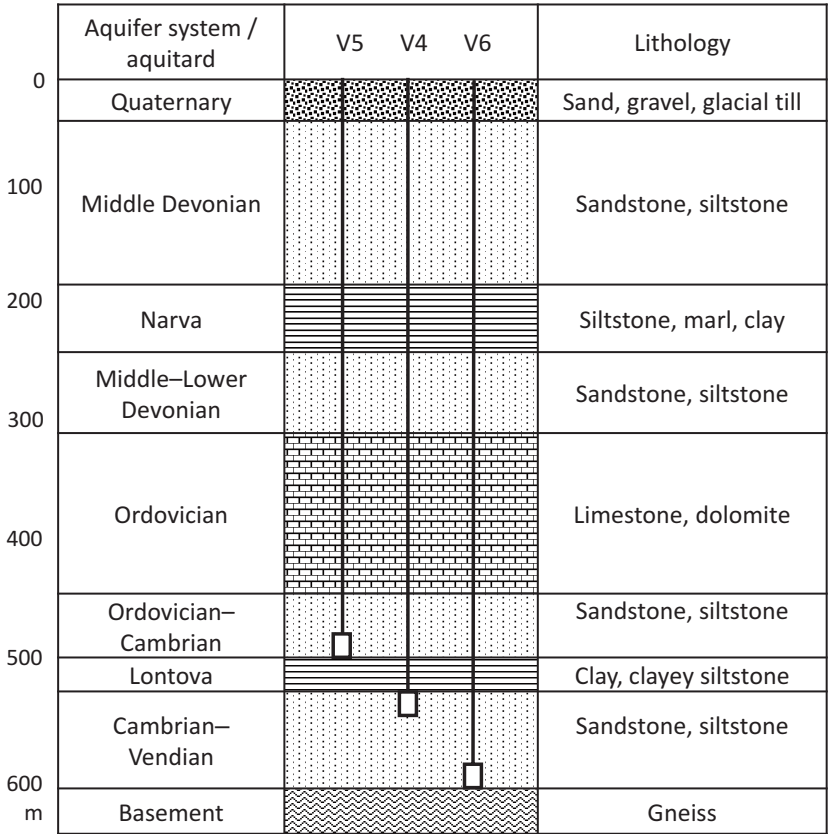


Fig. 2. Schematic hydrostratigraphy and lithology of the Värška area, based on the Estonian Nature Information System (EELIS 2024). The depths of the open sections of the wells are marked with rectangles: Värška 4 (V4) – 520–535 m, Värška 5 (V5) – 480–500 m, and Värška 6 (V6) – 575–595 m.

grained, weakly cemented sandstones and siltstones. The chemical types of the groundwater are Na–Ca–Cl or Ca–Na–SO₄–Cl, and TDS values reach up to 4 g/L in the Värška region (Karise et al. 2004).

The terrigenous Ordovician–Cambrian aquifer system lies below the Ordovician aquitard (Fig. 2), at a depth of about 500 m. The TDS values of this deep-seated aquifer exceed 2 g/L, and the chemical types of the groundwater are predominantly Na–Ca–Cl–HCO₃ and Na–Ca–Cl (Perens and Vallner 1997).

The water-bearing rocks of the deepest Cambrian–Vendian aquifer system are sandstones and siltstones of Cambrian and Ediacaran periods. The clays of the Kotlin Formation divide the aquifer system into two units: the upper Voronka and the lower Gdov aquifer. The groundwater belongs to the zone of passive water exchange, and thus the TDS values in Gdov aquifer reach 20 g/L (Karise et al. 2004), and the chemical types of water are Na–Cl and Na–Ca–Cl (Perens and Vallner 1997).

There are several deep wells in different aquifer systems for abstracting mineral water in southeastern Estonia. Värška's most popular bottled mineral water, used for drinking and treatment of chronic digestive tract diseases (hypoacidity), is Värška Original, which is derived from the 470 m deep Värška 7 borehole, drilled into the Ordovician–Cambrian aquifer system (Bitjukova and Petersell 2010).

Three of the mineral water wells – Värška 4, Värška 5, and Värška 6 – are located on the territory of Värška Sanatorium. The sanatorium uses mineral water with varying salinity and chemical compositions for different treatment procedures. The Värška 4 well extracts mineral water from the fine-grained sandstones of the Voronka aquifer, the upper part of the Cambrian–Vendian aquifer system. The groundwater at a depth of 520–535 m (Fig. 2) is saline, with a TDS content of up to 6.5 g/L. The shallowest well, Värška 5, is drilled into fine-grained sandstones, which form the Ordovician–Cambrian aquifer system at a depth of 480–500 m. The groundwater at this depth is slightly saline – the TDS values vary from 1.9 to 2.6 g/L. The Värška 6 well extracts mineral

water from the sandstones of the Gdov aquifer, the lower part of the deepest Cambrian–Vendian aquifer system. The depth of the open interval of the well is 575–595 m (Fig. 2). Värška 6 mineral water has the highest salinity among the Värška waters, with its TDS contents varying between 17 and 27 g/L.

Results and discussion

Overview of isotonic and hypertonic nasal sprays market in Estonia

The survey of the nasal spray nomenclature in Estonian community pharmacies revealed that a total of 17 nasal sprays were marketed, of which 12 were for nasal use only and 5 were nasal/throat sprays for universal use (Table 1). Most (70%) were solutions with isotonic salt concentrations, available in 5–150 mL bottles. In particular, all products are made from dissolved or undiluted sterilised seawater or ocean water. The majority of isotonic and hypertonic solutions contain only sea- or ocean water (67% and 60%, respectively); copper or manganese salts, aloe vera extract, or essential oils (eucalyptus oil, peppermint oil) have been used as additives to enhance the effect. Most isotonic solutions were also intended for children of different ages, starting from infants.

There is a relative lack of hypertonic solutions on the Estonian market. Most products are isotonic sea salt solutions, which can be used daily and, among other things, are suitable for children under one year of age. Isotonic solutions also have slightly more indications than hypertonic sprays. Usually, four to six indications are listed in the instructions for commercially available isotonic solutions. For example, the instructions for one product include cleaning and moisturising the nasal mucosa and acting as an additional treatment for nasal congestion. It can also be used to relieve irritation of the nasal mucosa, to moisturise the nasal mucosa in heated rooms with low humidity, to clean the nostrils during air travel, and in cases of increased air pollution (dust, pollen).

In contrast, hypertonic solutions, where the salt content is usually greater than 2% relative to NaCl, should not be used

Table 1. Isotonic and hypertonic nasal sprays* available in community pharmacies in Estonia in 2022

Characteristics	Isotonic water (0.9%)	Hypertonic water (2.3–2.6%)
Sea- or ocean water without additives	8	3
Sea- or ocean water with additives	4	2
IN TOTAL	12	5
Sea- or ocean water 0.9%	12	–
Sea- or ocean water 2.3%	–	1
Sea- or ocean water 2.6%	–	4
Nasal spray	7	5
Nasal/throat spray	5	–
Bottle volume 5–30 mL	5	3
Bottle volume 50–150 mL	7	2
For use on babies (less than 1 year)	4	1
For use by children from 2 years of age	4	0
For use by children from 3 years of age	–	1
For use by children in general	2	–

* number of products

continuously, but only as needed – for example, to relieve the symptoms or reduce swelling associated with a cold or allergic rhinitis. Some manufacturers add natural extracts or essential oils to their products to introduce new effects or strengthen existing ones. For example, aloe vera extract is added to moisturise the nasal mucosa (suitable for dryness or before nasal preparations), and eucalyptus oil is included to reduce nasal swelling more quickly, which in turn helps to ease breathing and open the nose. Individual age restrictions for children are primarily due to the long nozzle of the dispenser.

In this study, the sales statistics of nasal sprays from three Estonian pharmaceutical wholesalers – Tamro Eesti OÜ, Baltfarma OÜ, and Loodustoode OÜ – were used for the period from 1 January 2018 to 30 June 2019. A total of 128 443 units of relevant products were sold during this period. Hypertonic nasal spray (30 mL) was the most sold in pharmacies and larger stores, followed by isotonic nasal sprays for children and adults. Brand preference often plays an important role in product selection, which certainly also depends on product prices and sales work. It is noticeable that buyers prefer the so-called pocket variants, i.e. smaller bottles with a volume of up to 50 mL, because these are more con-

venient to carry. It is also likely that each family member purchases their own bottle, which is advisable from a hygienic point of view. Nasal sprays with additives are chosen relatively rarely; their sales remain modest and generally minimal.

The use of sea salt solutions in nose and throat prophylaxis and treatment in Estonia

Answers from pharmacy staff

A total of 92 women and 8 men participated in the survey of community pharmacy employees, reflecting the fact that there are many more female than male pharmacists in Estonian pharmacies. Regarding age, respondents aged 30–39 formed the largest group (approximately 50% of respondents), followed by those aged 20–29, 40–49, and 50–59; there were a few respondents in the 60–69 age group. This distribution corresponds to the age structure of Estonian pharmacists. Of those surveyed, 20% were customer service personnel, 50% pharmacy assistants, and 30% pharmacists. Pharmacies were mainly represented by those in Tallinn (58%), followed by other larger cities (19%) and pharmacies in rural areas (23%). Table 2 shows that pharmacy employees are frequent consumers of sea salt solutions (72%), supported by their professional knowledge (86%) as an important factor when

Table 2. Results of surveys of pharmacy employees and customers

Aspect	Respondents, %	
	Employees	Customers
<i>How often do customers use sea salt solutions?</i>		
Never	–	7
Rarely (2–3 times a year)	–	36
Occasionally (1–2 times a month)	–	13
Often (3–6 times a month)	–	21
Constantly (more than 10 times a month)	–	23
<i>For whom do customers buy sea salt solutions?</i>		
For themselves	–	80
For a child	–	35
For their spouse	–	19
For other family members	–	16
It depends	–	4
Other	–	1
<i>For what reasons do customers buy them?</i>		
Common rhinitis (common cold)	90	61
Rhinorrhea ('runny nose')	59	15
Allergic rhinitis	79	21
Non-allergic rhinitis (medications, environmental factors, hormones, etc.)	72	6
In addition to antibiotics (e.g. for rhinosinusitis)	42	11
Prophylactically to prevent viral diseases	63	25
To clean the nose before nasal medications	74	21
For nasal hygiene	95	40
Other	5	3
<i>Do these products solve a specific health problem that the pharmacy employee or customer thinks they have?</i>		
Yes, definitely	12	39
Mostly	57	40
Rather not	5	5
Definitely not	1	1
It depends	21	13
Other	3	2

Continued on the next page

Table 2. Continued

Aspect	Respondents, %	
	Employees	Customers
<i>On what basis do pharmacists recommend sea salt solutions?</i>		
Professional knowledge	86	–
Feedback from customers or patients	46	–
Personal experience	72	–
Recommendations given by the producer	68	–
Customer's own wish	69	–
Other	2	–
<i>Which criteria, according to pharmacy employees, affect the purchase of sea salt solutions?</i>		
Price	91	–
Package volume	62	–
Company reputation	21	–
Pharmacist's recommendation	90	–
Recommendation from a friend/others	46	–
Personal experience	83	–
Advertising	53	–
Attractiveness of packaging	7	–
Other	0	–
<i>Which criteria affect the decision of a customer when buying a sea salt solution?</i>		
Doctor's recommendation	–	44
Pharmacist's recommendation	–	61
Recommendation from a friend/others	–	24
Advertising	–	12
Internet	–	9
Other	–	5
<i>What is the maximum amount a customer is willing to spend on a sea salt solution?</i>		
Less than 5 euros	–	19
5–10 euros	–	60
10–15 euros	–	21
15–20 euros	–	0
More than 20 euros	–	0

recommending these products to customers. When choosing a product, the most important factors, in pharmacists' experience, are price (91%) and package size (62%). The majority (60%) of buyers are willing to pay 5–10 euros for the product, with about 20% of respondents indicating a preference for products costing less than 5 euros or 10–15 euros, respectively.

Responses from customers

Most of the surveyed customers were also women (73%). More than half of the respondents (57%) were aged 20–39; the number of respondents in their 40s decreased, and 2% were younger than 20. This provides an indicative profile of typical buyers of these preparations. Most respondents purchase such products two to six times a year, mainly for themselves (80%) or for a family member (70%), more often for children (35%) (see Table 2). The majority buy sea salt solutions for colds (61%) and for nasal hygiene (40%). The purchase decision is usually based on the recommendation of a pharmacist or doctor (61% and 44%, respectively). Regarding the effectiveness of products, users tend to express an overall more positive opinion of the products' effectiveness than pharmacists do: 39% and 40% of customers responded 'yes, definitely' and 'mostly', respectively, compared with 12% and 57% of pharmacy employees.

The study concludes that there are quite a few users of sea salt solutions, and the indications for their use are diverse – from daily nasal hygiene to relief of colds. According to both pharmacy employees and customers, the results of their use are predominantly positive. However, buyers are quite price-sensitive. It is clear that sea salt solutions are valued products on the pharmacy market, and their development could be based on local natural resources, especially mineral water. This would help simplify production and reduce costs, while ensuring medicinal potential comparable to sea salt solutions.

Chemical compositions of nasal sprays and mineral water

There are mainly two types of nasal sprays currently in production and available on the market: sprays based on natural ocean water (seawater) and nasal sprays produced by mixing chemical compounds (such as xylometazoline, oxymetazoline, etc.). This study analysed and compared the chemical compositions of nine nasal sprays available in pharmacies with the chemical compositions and chemical types of ocean water and Värkska 6 mineral water (Table 3).

The salinity of ocean water (essentially the sum of the contents of the main ions) is high, with the amount of dissolved salts reaching 34.4 g/L. The salinity (TDS value) of ocean water-based nasal sprays is lower, varying between

Table 3. Chemical parameters and total dissolved solids (TDS) values (mg/L) of nasal sprays, Värška mineral water (samples: (1) – 28.02.2018, (2) – 27.08.2018, (3) – 18.10.2022, (4) – mean value of 18 determinations from the period 1986–2014), and ocean water (Cotruvo 2005)

Sample	TDS	Na	K	Ca	Mg	Cl	SO ₄	HCO ₃	Water type
Nasal spray 1	10 164	3429	116	123	424	5375	645	53	Na–Cl
Nasal spray 2	10 370	3465	120	128	418	5513	660	65	Na–Cl
Nasal spray 3	10 414	3475	117	123	435	5549	663	53	Na–Cl
Nasal spray 4	9337	3073	115	131	388	4998	592	40	Na–Cl
Nasal spray 5	27 233	9519	301	323	1203	13 988	1773	126	Na–Mg–Cl
Nasal spray 6	21 400	6472	167	372	1453	10 463	2225	248	Na–Mg–Cl
Nasal spray 7	11 328	2151	47	348	1257	5070	2329	126	Mg–Na–Cl–SO ₄
Nasal spray 8	24 507	7647	203	389	1453	12 359	2331	126	Na–Mg–Cl
Nasal spray 9	23 178	7842	263	276	969	12 195	1519	114	Na–Cl
Värška 6 (1)	15 222	4565	67	955	371	8960	166	138	Na–Cl
Värška 6 (2)	16 965	5480	72	1026	422	9633	192	140	Na–Cl
Värška 6 (3)	20 000	5200	75	1000	95	11 500	280	142	Na–Cl
Värška 6 (4)	20 666	5134	58	943	403	10 144	248	140	Na–Cl
Ocean water	34 367	10 556	380	400	1262	18 980	2649	140	Na–Cl

9.3 g/L (nasal spray 4) and 27.2 g/L (nasal spray 5). Depending on salinity, the nasal sprays can be divided into two groups. The first group consists of sprays with lower salinity values (9.3–11.3 g/L), which are isotonic or slightly hypertonic solutions in terms of salt content. The second group includes sprays with salinity ranging between 21.4 and 27.2 g/L, which are clearly hypertonic solutions. Since the salinity of the analysed nasal sprays is lower than that of average ocean water, it can be assumed that the ocean water was diluted during the production of the sprays or that certain additives were included to lower the salinity of the solution. Na and Cl ions, which are also the main components of ocean water, contribute most significantly to the TDS values of the sprays.

The saline Värška 6 mineral water originates from the Gdov aquifer of the Cambrian–Vendian aquifer system (Fig. 2). Water analyses performed previously (during 1986–2014) on samples from the Värška 6 well (Table 3) show significant variation in TDS values (17–27 g/L), with a mean value of 20.7 g/L (Karro et al. 2018). There may be several reasons for such variation: groundwater exchange due to pumping and natural flow, changes in laboratory analytical methods over decades, and possible differences in water sampling quality. The most recent analysis prior to this project (TDS = 19 300 mg/L), performed by AS Tallinna Vesi in 2014, is in good agreement with the salinity measurement performed in this study in 2022 (Table 3).

In terms of trace elements, the studied nasal sprays are characterised by high contents of Sr and B (Table 4), but they also contain smaller quantities of Al, Fe, Cu, Mn, Zn, and Li. These elements are typically present in ocean water as a result of long-term rock weathering and river runoff (Chester 1990).

In addition to salinity and the contents of main ions, trace elements were determined in Värška 6 mineral water to compare their content with the corresponding values in commercially available nasal sprays. Several chemical elements are

present in both Värška mineral water and the nasal sprays at very low concentrations or in amounts below the analytical detection limit (Table 4). However, all analysed solutions are rich in boron. Compared to commercially available nasal sprays, Värška 6 mineral water is characterised by its higher content of Fe, Mn, Li, Sr, and Ba. These elements are so-called lithophilic elements (White 2013; Chauvel and Rudnick 2016), suggesting their geological origin.

The open section of the Värška 6 well extends to a depth of 595 m, reaching sandstones that lie directly on the bedrock (Fig. 2). Chemical weathering of different types of feldspars is the main geological source of the aforementioned chemical elements in groundwater. The reducing conditions prevailing in deep-seated aquifers retain several elements (e.g. Fe and Mn) in dissolved form, resulting in their high concentrations in groundwater, as seen in the case of the Värška 6 well (Table 4). The mineral water has a naturally high iron content (4700 µg/L). Consequently, iron compounds may precipitate from the water due to the action of atmospheric oxygen, potentially clogging the spray dispenser and affecting the transparency of the spray solution. Suspended iron can be removed from water through filtration. Results from a study of water supply systems in southern Estonia (Hiiob and Karro 2012) showed that iron removal is most effective when preceded by aeration in treatment facilities.

Chemical types of mineral water, ocean water and nasal sprays

According to the manufacturers, the active and main ingredient of all the natural nasal sprays is ocean water. Therefore, it is appropriate to compare the chemical types of the sprays with that of average ocean water. Chlorides dominate among the anions and Na, followed by Mg, among the cations in the compositions of the analysed nasal sprays (Table 3). The only exception is nasal spray 7, in which Mg is the dominant cation, and SO₄ is also present among the anions.

Table 4. Trace element concentrations ($\mu\text{g/L}$) in the studied nasal sprays and Värška 6 mineral water samples: (1) – 28.02.2018, (2) – 27.08.2018

Sample	Li	Be	B	Al	Sc	Ti	V	Cr	Mn
Nasal spray 1	60	<0.4	1517	168	0.4	12	2.2	<0.4	18
Nasal spray 2	59	<0.4	1488	344	0.5	10	1.2	0.6	17
Nasal spray 3	61	<0.4	1552	295	0.6	9	1.7	0.6	22
Nasal spray 4	52	<0.4	1359	172	<0.4	4	1.8	1.4	170
Nasal spray 5	175	<0.4	4295	793	0.9	31	4.4	<0.4	13
Nasal spray 6	120	<0.4	5322	378	2.2	13	3.1	3.5	18
Nasal spray 7	49	<0.4	4598	260	<0.4	6	1.6	4.8	11
Nasal spray 8	143	<0.4	5139	452	<0.4	11	2.7	1.1	18
Nasal spray 9	139	<0.4	3470	599	0.9	29	2.7	<0.4	14
Värška 6 (1)	609	<0.4	5070	32	0.5	<0.4	<0.4	<0.4	1130
Värška 6 (2)	607	<0.4	4878	<5	<0.4	<0.4	<0.4	4.2	1257
Sample	Fe	Co	Ni	Cu	Zn	As	Se	Rb	Sr
Nasal spray 1	81	<0.4	<0.4	37	82	<1.0	27	35	2333
Nasal spray 2	161	0.4	<0.4	352	54	1.1	29	37	2372
Nasal spray 3	135	0.4	1.9	36	106	1.5	30	36	2333
Nasal spray 4	107	0.6	<0.4	36	9	0.9	26	37	2288
Nasal spray 5	405	0.8	<0.4	75	<5	<1.0	74	94	6204
Nasal spray 6	184	1.0	4.7	66	7	<1.0	31	53	7562
Nasal spray 7	124	0.6	21.7	25	50	1.8	8	16	6725
Nasal spray 8	187	1.0	13.0	75	220	2.3	41	63	7598
Nasal spray 9	287	0.8	<0.4	80	66	<1.0	63	80	5196
Värška 6 (1)	4705	<0.4	2.1	<0.4	104	<1.0	<0.4	37	21 527
Värška 6 (2)	4764	<0.4	<0.4	<0.4	<5	<1.0	<0.4	40	23 474
Sample	Mo	Cd	Sn	Sb	Cs	Ba	Pb	Th	U
Nasal spray 1	3.7	<0.4	<0.4	0.2	0.6	56	<0.4	0.04	0.76
Nasal spray 2	3.9	<0.4	<0.4	0.2	0.7	51	0.2	0.06	0.95
Nasal spray 3	4.0	<0.4	<0.4	0.1	0.7	76	<0.4	0.04	0.95
Nasal spray 4	3.6	<0.4	<0.4	0.1	0.7	31	<0.4	0.03	0.05
Nasal spray 5	9.9	<0.4	<0.4	0.2	1.4	11	193.0	0.08	2.39
Nasal spray 6	12.0	<0.4	<0.4	0.3	1.1	14	<0.4	0.09	3.19
Nasal spray 7	12.0	<0.4	<0.4	0.3	0.9	12	<0.4	0.04	3.33
Nasal spray 8	12.0	<0.4	<0.4	0.3	1.2	27	0.8	0.08	3.03
Nasal spray 9	7.3	<0.4	<0.4	0.4	1.5	10	<0.4	0.19	2.17
Värška 6 (1)	<0.4	<0.4	<0.4	0.1	0.8	201	0.9	<0.02	0.26
Värška 6 (2)	<0.4	<0.4	<0.4	0.1	0.1	204	<0.4	<0.02	0.29

The Piper diagram (Fig. 3) shows that the chemical types of most nasal sprays are similar to that of ocean water. However, nasal spray 7 deviates from the others due to its relatively high Mg and low chloride content. The scatter of points on the diagram is mainly caused by the Mg content, which is also reflected in the chemical compositions of some other nasal sprays (Table 3). Despite some variation, the chemical types of the nasal sprays are similar to that of ocean water, suggesting that manufacturers have indeed used natural ocean water as the main component of their products. While nasal sprays generally contain purified water in addition to ocean water, the results indicate that such dilution with freshwater has not altered the

chemical type, as the samples are still located near the point representing ocean water composition on the diagram (Fig. 3).

Värška 6 mineral water is of the Na–Cl chemical type, similar to ocean water and most of the studied nasal sprays. Although mineral water is richer in Ca and poorer in SO_4 than ocean water and the nasal sprays, its chemical type remains Na–Cl. On the Piper diagram, the point representing its composition is located near the one marking the composition of ocean water. The similarity between the chemical type of Värška 6 mineral water and those of commercially available nasal sprays suggests the potential for using mineral water as the main component of a nasal spray.

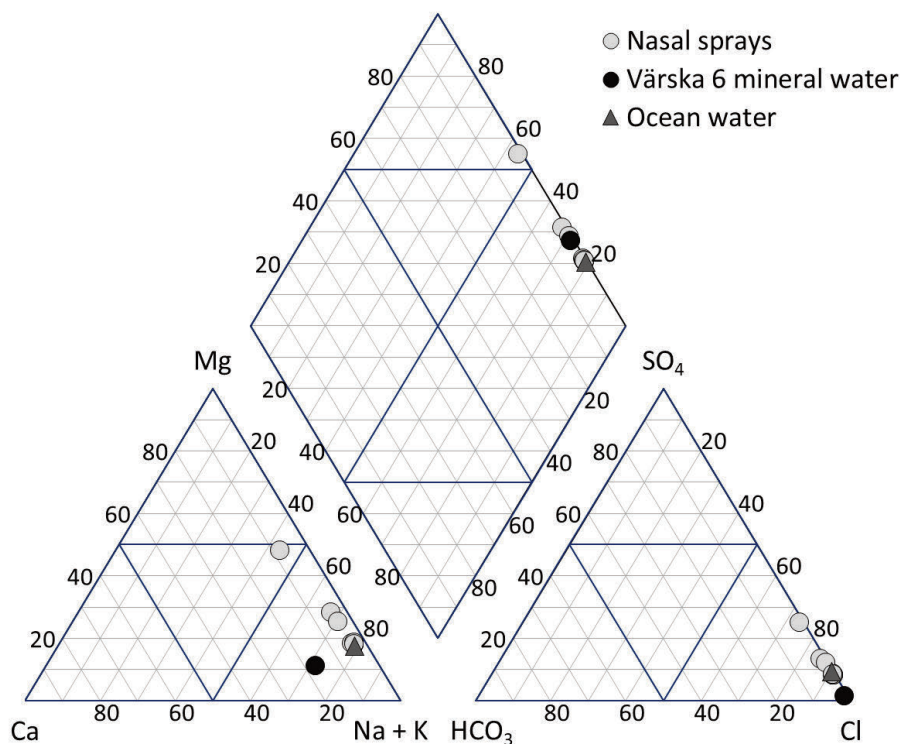


Fig. 3. Piper diagram illustrating the chemical compositions of nasal sprays, ocean water, and Värška 6 mineral water.

Microbiological quality indicators of mineral water

Microbiological analyses were conducted in three series at different times (Table 5). The first determinations were performed immediately after water sampling, the second series of analyses were conducted after one month, and the third after an additional three months. During the intervals between analyses, the bottles containing the mineral water samples were tightly sealed and stored at room temperature. This approach allowed the monitoring of potential changes in the microbiological composition of the solution under normal temperature and pressure.

The first analysis already indicated that Värška 6 mineral water is microbiologically pure, with a colony count of 22 °C of 5 CFU/mL. After one month, the colony count had decreased to 2 CFU/mL, and four months after sampling, it was

zero (Table 5). The content of the other microbiological quality indicators (Coliforms, *Escherichia coli*, and Enterococci) remained zero throughout the whole experiment. This simple test of temporal stability demonstrated that the highly saline Värška 6 mineral water does not promote bacterial growth but rather inhibits it.

The main component of the natural nasal sprays used as reference material during this project is seawater or ocean water. In order to ensure microbiologically pure ocean water for product manufacturing, it is necessary to treat the water beforehand.

On the other hand, deep-seated groundwater contains very few or no pathogenic microorganisms (Krauss and Griebler 2011; Ferrer et al. 2020). The results of the current study confirm this and allow us to state that the groundwater of the Cambrian–Vendian aquifer system in Värška is microbio-

Table 5. Microbiological quality indicators of Värška 6 mineral water

Date	Parameter	Colony-forming unit (CFU)	Result
05.03.2018	Coliforms	CFU / 100 mL	0
	<i>Escherichia coli</i>	CFU / 100 mL	0
	Enterococci	CFU / 100 mL	0
	Colony count 22 °C	CFU / 1 mL	5
05.04.2018	Coliforms	CFU / 100 mL	0
	<i>Escherichia coli</i>	CFU / 100 mL	0
	Enterococci	CFU / 100 mL	0
	Colony count 22 °C	CFU / 1 mL	2
06.07.2018	Coliforms	CFU / 100 mL	0
	<i>Escherichia coli</i>	CFU / 100 mL	0
	Enterococci	CFU / 100 mL	0
	Colony count 22 °C	CFU / 1 mL	0



Fig. 4. Nasal/throat spray Tsilk. Photo: A. Raal.

logically clean and can be used as the main component of a nasal spray. In the case of Värška 6 mineral water, the time and resource costs related to the microbiological treatment of water would be avoided – an important advantage in the production of nasal spray.

Natural mineral water-based nasal/throat spray Tsilk

Our research described above led to the development of a nasal and throat spray – the first analogue based on mineral water produced in Estonia. The product, Tsilk (meaning ‘a drop’ in southeastern Estonian dialect), is packaged in a 30 mL glass bottle enclosed in a cardboard box (Fig. 4). The bottle includes an antibacterial pump system that prevents the entry of germs during the total period of use.

Composition: 100% natural, sterile, unpurified hypertonic mineral water from a 595-m-deep borehole No. 6 in Värška, Estonia. The mineral content averages 1.5–2.1 g/100 mL. Tsilk is intended for adults and children over two years of age. The saline mineral water relieves nasal congestion and runny nose, clearing the nasal cavity of excess mucus and protecting it from infections. It also relieves throat irritation and dryness.

Produced by: OÜ Värška Vara, Väike-Rõsna village, Setomaa Parish, Võru County, Estonia.

Conclusions

The survey of the nasal spray nomenclature in Estonian community pharmacies in 2022 revealed that a total of 17 nasal sprays were marketed, of which 12 were for nasal use only and 5 were nasal/throat sprays for universal use. Most (70%) were solutions with isotonic salt concentrations made from dissolved or undiluted sterilised seawater or ocean water. Thus, the study also revealed that there is a relative lack of hypertonic solutions on the Estonian market.

There are quite a few users of sea salt solutions, and the indications for their use are diverse, ranging from daily nasal hygiene to the relief of colds. According to both pharmacy employees and customers, the results of their use are predominantly positive. However, buyers are quite price-sensitive. Thus, it is clear that sea salt solutions are valued products on the pharmacy market, and their development could be based on local natural resources, especially mineral water. This would help simplify production and reduce costs, while ensuring medicinal potential comparable to sea salt solutions.

The Värška 6 well extracts mineral water from the sandstones of the Gdov aquifer at a depth of 575–595 m. Värška 6 mineral water has the highest salinity among the Värška waters, with its total dissolved solids (TDS) content varying between 17 and 27 g/L.

The chemical compositions of nine nasal sprays sold in pharmacies were analysed and compared with the chemical compositions and chemical types of ocean water and Värška 6 mineral water. Depending on salinity, the nasal sprays can be divided into two groups. The first group consists of sprays with lower TDS values (9.3–11.3 g/L), which are isotonic or slightly hypertonic solutions. The second group comprises sprays with salinity varying between 21.4 and 27.2 g/L, which are clearly hypertonic solutions. Although the investigated spray solutions show differences in concentration, their Na/Cl ratios are very similar, which confirms their oceanic origin.

The results of laboratory analyses showed that Värška 6 water has a high salinity (20 g/L), a Na–Cl chemical type, and – compared to ocean water and nasal sprays made from ocean water – is enriched in lithophile elements (Fe, Mn, Li, Sr, and Ba) characteristic of deep groundwater. Based on salinity (TDS value), mineral water is a hypertonic solution, which is suitable for the production of a nasal spray. The results of microbiological analyses showed that Värška 6 mineral water is bacteriologically clean (no Coliforms, *Escherichia coli*, or Enterococci detected), and the temporal stability test confirmed that mineral water with high salinity does not promote bacterial growth but rather inhibits it.

As a result of this project, a new product was developed: the mineral water-based nasal/throat spray Tsilk. The spray is produced by OÜ Värška Vara and sold in Estonian pharmacies.

Data availability statement

All the data used here are published in this article. Additional information will be made available on request.

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Värskas mineraalveel põhineva nina- ja kurgusprei väljatöötamine

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Eesti apteekides turustatavate ninaspreide nomenklatuuri uuringust selgus, et kokku oli saadaval 17 nina-kurguspreid, millest enamik (70%) olid isotoonilise soolakontsentratsiooniga, mere- või ookeaniveest valmistatud steriliseeritud lahused. Nende kasutusnäidustused on mitmekesised alates igapäevasest ninahügieenist kuni külmetushaiguste leevendamiseni. Nii apteegitöötajate kui ka -külastajate hinnangul on spreide kasutamise tulemused valdavalt positiivsed. Looduslikud ninaspreid on apteegiturul oodatud tooted, kuid neid võiks valmistada ka kohalikest loodusvaradest, eelkõige mineraalveest. See aitaks lihtsustada tootmist ja hoida hinna madalamana, tagades samas meresoolalahustega võrreldava ravipotentsiaali.

Uuringu käigus analüüsiti üheksa ninasprei keemilist koostist ning võrreldi seda ookeanivee ja mineraalvee Värskas 6 keemilise koostise ning tüübiga. Soolsuse põhjal jagunevad ninaspreid kahte rühma: isotoonilised või kergelt hüpertoonilised lahused (9,3–11,3 g/L) ja hüpertoonilised lahused (21,4–27,2 g/L). Kõigi analüüsitud spreilahuste Na/Cl suhe oli väga sarnane, mis kinnitab nende ookeanilist päritolu.

Puurkaevust Värskas 6 ammutatakse mineraalvett 575–595 m sügavuselt Gdovi veekihist. Tegu on kõrge soolsusega (20 g/L), Na–Cl keemilist tüüpi mineraalveega, mis erineb ookeaniveest ja sellest valmistatud ninaspreidest sügaval lasuval põhjaveele iseloomulike elementide (Fe, Mn, Li, Sr ja Ba) poolest. Soolsuselt on puurkaevu Värskas 6 vesi hüpertooniline lahus, mis sobib hästi ninasprei valmistamiseks. Mikrobioloogiliste analüüside tulemused näitasid, et mineraalvesi on bakterioloogiliselt puhas, ning ajalise stabiilsuse katse kinnitas, et kõrge soolsusega mineraalvesi ei soodusta bakterite kasvu, vaid pigem pärssib seda. Projekti tulemusena töötati Värskas sanatooriumis välja uus mineraalveepõhine toode – nina- ja kurgusprei Tsilk, mida turustatakse Eesti apteekides.