Regular Article

ANALYTICAL
AND BIOANALYTICAL
CHEMISTRY
RESEARCH

Published by the
Iranian Chemical Society

***Anal. Bioanal. Chem. Res.**, Vol. 11, No. 1, 51-67, January 2024.*

Determination of Biocide-type Disinfectants in Water Condensates Originating from Cleaning Chemicals with Capillary Electrophoresis

Markus O. Lehtonen[a] and Heli Sirén[b,*]

[a]*Origin by Ocean, Ystadinkatu, Tekniikantie 2, 02150 Espoo, Finland*

[b]*Department of Chemistry, University of Helsinki, A. I. Virtasen Aukio 1, P. O. Box: 55, FI-00014 University of Helsinki, Finland*

(Received 21 August 2023, Accepted 26 October 2023)

The commonly used polymeric disinfectants, didecyldimethylammonium chloride (DDAC) and polyethylene glycol monoalkyl ether (GEN) can drift with water vapor and aerosols, although they are not volatile. When they present in aerosols, their amines have justified to irritate the respiratory organs of humans. But, when they are not in breading air they stay on surfaces due to absorption and accumulation when temperature and moisture of the environment are low. The paper presents a new approach to determine disinfectants in alkaline tetraborate solution using capillary electrophoresis with direct UV detection. DDAC and GEN were investigated to move from surfaces to aerosol. The studied compounds were quantitatively analyzed at low concentrations (1-10 ng ml^{-1} with RSD 2%) to prevent micelle formation and to guarantee the method specificity. The limits of detection and quantification were 0.006 and 0.018 ng ml^{-1}, and 1.0 and 3.84 ng ml^{-1} for DDAC and GEN, respectively. They both were studied from 46 samples collected from two school environments which were daily washed with the commercial chemicals. The results showed that concentrations of these biocide-type disinfectants were between 2.5-1029 ng ml^{-1}.

Keywords: Disinfectants, Capillary electrophoresis, Complexation, Water condensates

INTRODUCTION

Chemicals, dust, pollen, mold, and microbe growth, among other materials, downgrade air quality [1,2,3,4,5,6]. It is recognized that *e.g.*, surfactants and disinfectants [7,8] stay on material surfaces due to their adsorptive and adherence properties aided by electrostatic forces and/or hydrophobic/hydrophilic interactions.

Likewise, many chemicals from personal care and household products [2], and disinfectants are very water-soluble and thus they can move with water aerosols from humid environments and surfaces to air when the relative humidity (RH) is between 50-70% [9]. On the contrary, when RH is below 40%, they stay on material surfaces [9]. The commonly used surfactants and disinfectants are non-ionic alcohol ethoxylates and cationic ammonium compounds. The latter compounds have molar masses of 100-600 g mol^{-1} with alkyl chains of 2 to 20 carbon atoms [67]. They are also the most examined in-industry-used surfactants [63], but their removal from locations with water vapor is hardly noticed.

Cleaning and disinfecting agents irritate respiratory organs in humans and have potential effects on continuous sniffles, allergies, and asthma. Cleaning agents and/or disinfectants used at work are diagnosed to cause asthmatic reactions in 39% of the participants who are exposed to these products [10,11]. Similar phenomena were evidenced with didecyldimethyl ammonium chloride (DDAC) when DDAC was used to expose rats by inhaling the contaminated air [12,13].

Disinfectants are bactericidal across a range of microorganisms. They are stable, long-lasting, and immobile as concentrates. Thus, it is unlikely that these chemicals are biologically degraded e.g., when absorbed in house dust [14]. Those biocide-type compounds may cause long-term

*Corresponding author. E-mail: helimmsiren@gmail.com

exposures since they are used as surfactants, detergents, and emulsifiers in cleaning products. Due to toxicity issues, high cumulative concentrations of disinfectants in clustering are significant for monitoring and measuring the individual minimum residue level (MRL), which for DDAC is 0.1 mg kg^{-1} [15,16,17,18].

At high concentrations, surfactants and disinfectants are not trouble-free, although they are useful chemicals on humid surfaces and in water devices to prevent microbial growth (microbes, bacteria, and viruses) by blocking the performance of biocompounds [8,9,13,19]. Biocides are widely used in the healthcare industry to control infections and microbial contamination [13]. For instance, DDAC is documented to prevent the action of canine coronavirus (CCoV), and both human's severe acute respiratory syndrome-coronavirus-2 (SARS-CoV-2, COVID-19) [20,21,22] owing to the formation of biocomplexes between the coronavirus structure and the amine part of DDAC.

Traces of quaternary ammonium compounds were detected in indoor air with ion chromatography (IC) [23]. Then, the compounds were sampled from the inside air of a hospital. The analysis results correlated insignificantly with the extracted DDAC. However, accidentally it was wrongly supposed that quaternary ammonium compounds are not able to contaminate the indoor atmosphere of the hospital during disinfection and thus DDAC was not detected as the target compound [24]. However, the research group took back their negative outcomes and informed that DDAC may contaminate the atmosphere when DDAC is in suspension of aerosols. Afterwards examined, the no-found DDAC would be detected, if detection sensitivity was enhanced [25].

Officially, today determination of organic compounds in indoor air is based on ISO standard instructions [26]. Anyhow, volatility of the compounds is expected when the standard obligates the analyses with gas chromatography (GC) at temperatures below 250 °C [27,28,29]. Most disinfectants, like DDAC and polyethylene glycol monoalkyl ether (GEN) in cleaning products are not inherently volatile. Also, the ISO standard for GC experiments permits the use of flame ionization detection which does not meet the accuracy of identification. For untargeted analyses (identification of unknown compounds), mass spectrometry (MS) is needed for accurate and reliable identification, but the methodology is not standardized [30]. Further, the

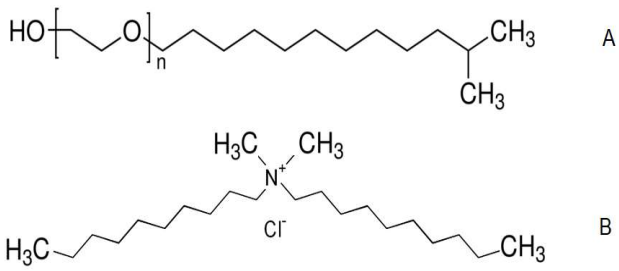

Fig. 1. The structures of (A) non-ionic polyethylene glycol monoalkyl ether (GEN, n = 9, alcohol ethoxylate) and (B) cationic didecyldimethyl ammonium chloride (DDAC).

weakness of the ISO standard method is that compounds like DDAC and GEN (Fig. 1) are not covered by the regulation [31,32].

As for GC, liquid chromatography (LC) is suited for the separation of both polar and non-polar disinfectants, but nevertheless MS is needed to justify their structures [17,33]. When uncharged non-polar or polar surfactants are concerned, capillary electrophoresis (CE) is a considerable choice for their simultaneous determination. Especially CE techniques can be performed using complexing agents added to electrolyte (buffer) solutions when the compounds are not sensitive enough for detection or when they cannot be separated from each other.

When the concentrations of disinfectants are high, both DDAC and GEN form micelles. But then micelle formation is prevented by complexation. The samples may also be diluted to prevent polymerization to allow monomer formation before the disinfectants are quantitatively complexed for one-peak detection [34,35,36,37,38,39]. If surfactants have chromophores as benzalkonium ions, complex forming agents are not needed for UV detection, and their direct identification may be a better choice [13,40,41,42]. Likewise complex formation, organic additives in CE electrolytes are known to prevent the formation of micelles when they are at high concentrations, and thus their selective complex formation with tetraborate is possible [7,43,44]. When organic solvents like tetrahydrofuran (THF) modified buffer solutions are used, THF is evidenced to prevent adsorption of cationic surfactants, such as N-benzyl-N-alkyl-N,N-dimethylammonium chlorides [42].

Similarly to DDAC contamination in the working atmosphere, airborne diffusion of GEN showed toxicity toward mammalian cells [45,46]. Furthermore, GEN could be diffused and precipitated at 1.0-10.0 ng ml^{-1} concentrations. The particle sizes of DDAC in contaminated aerosols and exhaled air were 0.63 µm, 0.81 µm, and 1.65 µm. Then, each concentration exposure from low (0.11 ± 0.06 mg m^{-3}), and middle (0.36 ± 0.20 mg m^{-3}) to high (1.41 ± 0.71 mg m^{-3}) could be detected [46].

Previous studies of indoor molds, however, showed that GEN had shockingly high lethal concentrations (>50 mg ml^{-1}) [47]. However, the toxicity data proved that 1/1000-fold smaller concentrations (*i.e.*, 50 µg ml^{-1}) already destroyed mammalian cells justified by *in vitro* bioassays. Naturally, it must be recognized that GEN has been precipitated on surfaces at carcinogenic magnitude over decades due to cumulative processes by small concentrations [47]. That study showed that 20% of indoor air samples from the studied office buildings contained pollutants that could be identified. As to the building-related symptoms, it was discovered that the findings mentioned above may be linked to the *in vitro* toxicity of indoor dust and airborne microbial population [48].

The present paper describes a method development for a cationic and a non-ionic biocide-type disinfectants and their determination in authentic samples as aerosol droplets, water condensates, and samples wiped from indoor surfaces from two elementary schools (*ex vivo* measured contaminations). This paper casts an assumption that cleaning chemicals being biocide-type disinfectants may be involved in peoples´ sickness due to three reasons which are: Pupils and teachers stay working days in the buildings and rooms, the cleaning chemicals are daily used, and the ventilation is low during night-time at the school.

MATERIALS AND METHODS

Chemicals

Disodium tetraborate ($Na_2B_4O_7$ hydrate, assay min 98%, analytical grade) was purchased from Merck (Germany). NaOH (0.1 M and 1.0 M) and HCl (0.1 M) were from FF-Chemicals (Finland). Genapol X-080 (GEN, CAS 9043-30-5; MW 552.78 g mol^{-1}, $HO(CH_2CH_2O)_n(CH_2)_mH$; assay 98%, Fig. 1) and didecyldimethyl ammonium chloride (DDAC, CAS 7173-51-5; MW 362.08 g mol^{-1}; $C_{22}H_{48}ClN$; analytical grade, Fig. 1), acetonitrile, and acetone were from Sigma-Aldrich (Finland). Methanol and 2-propanol (both HPLC grade) were from Fisher Chemicals (Finland). Didecyldimethyl ammonium chloride (DDAC, 50% solution in 2-propanol/deionized water (2:3, v/v) for synthesis) was from Merck (Schucharalt OHG, Germany). The standards were made into methanol (MeOH, 99.99%, HPLC-MS quality, Fisher Chemicals). In addition, Tricine (Sigma-Aldrich), CAPS (N-cyclohexyl-3-aminopropanesulfonic acid, Sigma-Aldrich), formic acid (Sigma-Aldrich), and sodium phosphate (Merck) were used for electrolyte buffers.

Methods

Instruments. A Hewlett-Packard 3D capillary electrophoresis instrument (Agilent, Waldbronn, Germany) equipped with a photodiode array detector (the wavelength range at 190-600 nm) was used with ChemStation programs (Agilent) for CE analyses and data handling.

The capillaries were made of fused silica with total lengths of either 48.5 cm or 40 cm, lengths to the detection either 40 cm or 31.7 cm (i.d. 50 µm, o.d. 362 µm) from CM Scientific (Silsden, UK) and Polymicro Technologies (TSP050375 and TSP075375, Phoenix, USA).

The new capillaries were conditioned by flushing consecutively with 0.1 M NaOH, deionized water, and the electrolyte solution at 13.634 p.s.i. (940 mbar) for 20 min, 10 min, and 20 min, respectively. Furthermore, the separation repeatability was assured by changing a new capillary between the analysis series (*i.e.*, after 50-100 analyses). More details are listed in Table 1.

Deionized water was made with a Millipore Direct-Q 3 UV instrument (18 MΩ). InoLab pH 2110 device with a combination electrode (WTW, pH-Electrode SenTix 42) was used for pH measurements. Three-point pH calibration line was made with commercial pH buffers (pH 4.00, 7.00, and 10.00).

The lab-made device, instrumentation, and experiments for collecting disinfectants in water vapor are described earlier [49]. Briefly, a 90-L aquarium chamber was used for tests of moving DDAC and GEN from the starting plate to the collector. The sensors used were for monitoring temperature, humidity, and total volatile organic compounds (TVOC). The indoor air quality was monitored with a special

Table 1. The Final Optimized Parameters Used in DDAC and GEN Analyses were Done with CE-UV

Parameters	Value
Capillary	
Length to the window and total length	Analyses: 40 cm or 48.8 cm, resp. Study on speciation: 31.7 cm or 40 cm, resp.
Conditioning	0.1 M NaOH for 10 min water (MQ quality) for 20 min 50 mM tetraborate (pH 8.5) for 20 min
Electrolyte solution (buffer)	50 mM sodium tetraborate (pH 8.5)
Washing between runs	20 mM NaOH for 10 min followed by the electrolyte solution for 4 min
Sample injection	At 35 mbar (0.5 p.s.i.) for 10 s followed by a waiting time of 0.75 min
Analysis	
Voltage	+17 kV for 40 cm capillary +20 kV for 48 cm capillary
Current	+90 µA created in the 48 cm capillary
Analysis time	55 min in 48 cm capillary 30 min in 40 cm capillary
Detection Wavelengths	UV: (200 +/- 16) nm, (214 +/- 20) (pilot signal), (254 +/- 10) nm, (280 +/- 10) nm, (320 +/ - 10) nm

indoor air quality sensor iAM-TVOC. Prior to each experiment, the baseline of the sensors was balanced with ambient air as blank and optimized with maximal humid air (RH 100%. The RH inside the chamber was adjusted to 75-80%. It was used to obtain reference samples.

Solutions

Electrolyte solutions. First, electrolyte solutions (buffers) in CE were made from two types of Tris buffers (Trizma® hydrochloride and Tricine) at pH 7.5 and N-cyclohexyl-3-aminopropanesulfonic acid (CAPS) at pH 9.0-11.0. Finally, many concentrations of sodium tetraborate, 0.1 M NaOH, and 0.1 M HCl were used to prepare several complex-forming electrolytes at various concentrations and pH. After experimental optimization, the optimal electrolyte composition was 50 mM sodium tetraborate at pH 8.5.

Compound solutions for calibration. Quantitatively GEN was quantified with stock solutions of 1000 µg ml^{-1} or 500 µg ml^{-1} by diluting with deionized water. In the case of DDAC screening, the standards were made from a 50% solution of DDAC (2-propanol-deionized water, 2:3 v/v). However, quantitative results were made by using 1000 µg ml^{-1} solutions in methanol to prevent micelle formation. In calibration, the concentrations of DDAC and GEN were between 1 ng ml^{-1} and 10 ng ml^{-1}. The calibration studies were also done with 50 ng ml^{-1}, 100 ng ml^{-1}, and 1000 ng μl^{-1} solutions to detect their micelle formation and to study the tetraborate concentration needed for complexation.

Washing solutions for electrodes in the CE instrument. Since the studied disinfectants have features to form polymers and their concentrations are unknown in real samples, it was important to clean the electrodes of the CE

instrument systematically. They were cleaned by washing consecutively with NaOH (0.1 M), HCl (0.1 M), and deionized water –MeOH solution (50:50, v/v) for 10 min each by using an ultrasonication bath. Especially, the cleaning was needed when sensitivity and electropherogram profiles collapsed due to too high a viscosity difference between samples and the electrolyte solution.

Method Development and Optimization

Pre-conditioning of silica capillaries was made with NaOH (1.0 M) for 10 min, deionized water for 20 min, and sodium tetraborate electrolyte (50 mM, pH 8.5) for 20 min in that order before the capillaries were used in analyses. Sample injection was optimized, and finally, it was made at 35 mbar (0.5 p.s.i.) for 10 s. The pre-run conditioning at the start of the analysis sequence was made with NaOH (0.1 M) followed by the electrolyte solution for 2 min each.

Before analysis, the repeatability of separations was assured from sample to sample by prewashing the capillary for 10 min with NaOH (0.1 M). Then, the final CE method included the following steps: flushing for 2 min with NaOH (0.1 M), for 2 min with sodium tetraborate buffer (50 mM, pH 8.5), for 2 min with deionized water -2-propanol mixture (50:50, v/v), for 20.0 s with organic solvent at 0.5 p.s.i. (35 mbar), and for 10 s with the sample at 0.5 p.s.i. (35 mbar). The reason for using finally mere 2-propanol as the organic solvent was its capability to generate a solution plug before the sample and concentrate the compounds for stacking. Finally, before switching the electric field on, the action of movement by pressure was stopped for 0.75 min, which after the analysis was started by switching the electricity on.

Samples

The samples studied contained DDAC and/or GEN. They were chemicals in washing solutions used daily for school cleaning. As to instructions for cleaning the schools, the washing solutions were selected from many available and the decision was made by the Education Organization of Vantaa City in Finland.

In total, 46 water, aerosol, and wall surface samples were collected and studied. They were sampled from two schools which were the primary schools of Hämeenkylä (H) and Kannisto (K) in the subarea of Vantaa City. It was supposed that the schoolrooms suffered from mold problems. But it was not the reason, since anyhow staying in the buildings the indoor air caused health disorders and the school rooms suffered from severe air quality problems.

The experimentally studied samples were four types: a) artificial water samples collected inside a glass chamber to have reference samples (made in a laboratory), b) water condensates sampled from the schools, c) samples collected inside the school environments (rooms, kitchen, corridors, and lobby), and d) outdoor air samples from almost in the school entrance areas.

All samples were stored in a freezer (-20 °C) until analyses. Before the start of the analyses, the frozen samples were warmed to +40 °C to enable the dissolving of the wall-adsorbed compounds from the surfaces of the sampling devices (5 ml-volume glass tubes). However, several of the school samples needed filtration with PTFE membranes (0.20 μm), since they contained precipitates which was not resoluble after the melting.

Equations for Method Validation

The total mobility (μ_{tot}) is characteristic of each analyte. It is measured by using the electropherogram (migration time, sensitivity in detection) and instrumental parameters (capillary dimensions, voltage used for electromigration). Here it is measured by using DDAC and GEN standards, and methanol as the electroosmosis marker by the equation

$$\mu_{tot} = (L_{det}\, L_{tot})/(t_R \times V)$$

where L_{det} is the effective length of the capillary, L_{tot} is its total length, t_R is the absolute migration time of a compound to the detector, and V is the applied voltage during the analyses.

Where L_{det} is the effective length of the capillary (the length of the capillary from the inlet to the detector), L_{tot} is the total length of the capillary, t_R is the absolute migration time of the analyte to the detector, and V is the applied voltage during the analyses. Migration time (t_R) is measured using the maximum height of the analyte peak. The electrophoretic mobility of an analyte is calculated from the μ_{tot} and the mobility of electroosmosis ($\mu_{eo} = (L_{det}\, L_{tot})/(t_{eo}V)$ when the time of electroosmosis marker is detected in the electropherogram. Peak confirmation was done with spiked standards which also justified the correctness of the absolute

migration times of the disinfectants.

Limit of detection (LOD) and limit of quantification (LOQ) were experimentally measured with peak areas. Then the baseline noise around the peak was calculated (area) to use the noise value. All standards and samples were analyzed 3-6 times to evaluate the reliability of profiles and electrophoretic mobility. LOQ was also experimentally calculated using corresponding standards. Similarly, as LOD, the LOQ value for the blank sample was made by spiking with the analyte and for estimation of the minimum quantitative concentration for the study.

Since the methodology for LOQ was too time-consuming, for quantification of indoor samples we used calculation 10 × LOD = LOQ. In our tests, the experimentally measured values were lower than the calculated ones.

RESULTS

Disinfectants were complexed with TB anions during the analysis process and separated in TB buffer in CE-UV. Based on the literature, CE was not earlier used similarly for the determination of both cationic and neutral biocide-type disinfectants when they were at very low concentrations in the same sample.

Instrumental Parameters Needing Optimization

In CE sample injection plays a significant role in detection at extremely high sensitivity. Especially, the injection has specific importance when capillaries and sample volumes are 50 µm and 10-80 nl, respectively [36,38,42,50,51,52,53,54], and when detection concentrations depend on injection types [54]. Thus in the present study, the whole procedure needed adjustment to get reproducible volumes (concentrations) in a controlled manner. In this study, only hydrodynamic injection could be used, and the pressure and time were optimized. In the present work, they were validated to 35 mbar (0.5 p.s.i.) and 10 s, respectively (Table 1).

The electrolyte solutions were made of Tricine, CAPS, formic acid, sodium phosphate, and sodium tetraborate (TB) with various pHs. Then, the results showed that the TB electrolyte was superior to the other buffers due to low detection limits. Among alkaline TB buffers (10, 25, and 50 mM), the best was the 50 mM at pH 8.5.

Direct Identification

The targeted analytes DDAC and GEN are structurally challenging monomers since they are polymerized or form clusters like micelles in polar water solutions. Their structures are not UV-detectable since they lack chromophores. To minimize polymerization separation medium allowed complexation of analytes inside a capillary permitting UV-hypersensitivity and good separation efficiency for DDAC and GEN. Traditionally, disinfectants are studied with indirect UV detection in CE. But, then only micelle formation and derivatization improve the sensitivity [55,56,57].

A few papers show that water-soluble neutral analytes without chromophores could be studied in sodium tetraborate (200 mM, pH 9.5) or boric acid (100 mM) - potassium hydroxide (pH 10.0) solutions. [58,59,60] They were used as references to the present CE-UV analyses in TB buffer (50 mM, pH 8.5) and for sensitivity enhancement for non-ionic GEN.

Stability of Anionic Tetraborate Complexes

The species of DDTC and GEN vary from monomers to polymers. Their micelle formation and conjugation can be avoided *via* sample dilution, tetraborate complexation, and manipulation with organic solvents. The grounds of stability of non-ionic GEN [61] and cationic amine-type DDAC as TB complexes are due to interaction with the anionic ligands. In water, the ether oxygen structure of polyoxyethylene chains in GEN is involved in hydrogen bonding with the water hydroxides [62]. It is also possible that hydrogen bonds appear between hydroxylated glycol and ether oxygen of the structure, which may show as extra peaks in the electropherogram [63].

Complexation of analytes is evident since basic borate solutions (pH 8-12) contain tetraborate, triborate, $[B_3O_3(OH)_6]^{2-}$, and tetraborate octahydrate, $[B_4O_5(OH)_8]^{2-}$ anions. However, it is proved that only pH and the total borate concentration have significance [61]. In the present study, DDAC and GEN form 1:1 (mol:mol) -combinations with TB. The complexes contained TB at 0.05%, 0.26%, and 0.52% when analytes were at concentrations 10 pg ml^{-1}, 50 pg ml^{-1}, and 100 pg ml^{-1}, respectively. Then only 500 µM of 50 mM TB buffer was lost for the disinfectant complexes (calculated with the CeExpert Lite program). The loss in

ionic strength did not affect the velocity of electroosmosis, pH, or the electrical current. When the concentrations of DDAC and GEN were high, the complexes migrated faster than they were at low concentrations.

Enhance UV Detection Using a Stacking Method in Injection

TB complexation was done in the following order by injecting: TB buffer (high ionic strength, *I*), propanol (low *I*), and the sample (high *I*). The sequence allowed slow movement of the analytes for contacting analytes with TB ions before the complexes moved to the separation buffer. Complexation was also further improved by waiting for the pressure to decrease before complexes continued to migrate forward. All timings were optimized to control peak shifting and detection sensitivity. Then, electro-osmosis and current were stable in all analyses under the controlled method. In addition, methanol added to samples allows the enrichment of DDAC and GEN.

Differences in conductivity of consecutive solvents zones in capillary (sample, cond. ~0.5 µs cm^{-1}; buffer-modified sample, cond. > 80 ms cm^{-1}; deionized water, cond. 0.5 µs cm^{-1}; separation buffer, cond. ~80 ms cm^{-1}) aided disinfectants to form complexes, to focus the zones, and to minimize mobility differences. The sequence made of the TB electrolyte and the 2-propanol-deionized water allowed free movement of the anionic complexes and enrichment since then anions were occasionally stopped between the solvent barriers.

Organic solvents (acetonitrile, acetone, 2-propanol, and methanol) were used to concentrate the analytes and to decrease their electrical movement. When acetonitrile and acetone were used their sensitivity enhanced significantly (10-30-fold) compared to non-modified sample injection [64]. It was not logical because acetone forms anionic TB complexes and acetonitrile in a basic solution forms acetamide which forms TB complexes [65]. Since the solvent has a role, in the present case only 2-propanol was suitable for improving sensitivities. 2-Propanol followed by deionized water in stacking showed that DDAC and GEN complexes can be separated in the TB buffer when they are in the same cleaning product (Fig. 2). Then UV sensitivity of the complexes is enhanced to detect small amounts of substances with fast reaction of complex formation.

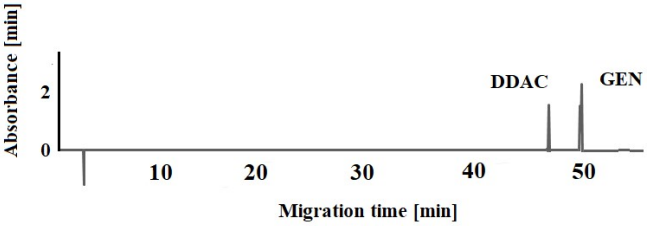

Fig. 2. Electroseparation of DDAC and GEN as TB complexes in the alkaline tetraborate buffer. Capillary: Ltot/Ldet 48.8 cm/40 cm; Voltage: +17 kV; UV detection at 214 nm. Electroosmosis marker (negative peak): methanol. Samples were injected in methanol.

Structural Effects for Complex Formation Stability

Usually, cationic disinfectants are studied using low pH (pH < 5) electrolytes to prevent interactions with the capillary surfaces since neutral and basic buffers prevent protonation-deprotonation of silanol and silanolate groups of the capillary [66]. In the present study, acids could not be used, and the alkaline TB electrolyte solution was used as noticed for non-ionic carbohydrates [67,68]. Then pH range was 9.0-9.7 [68] which was higher than pH 8.5 here.

TB complexes are stronger and more stable when having open-chain than cyclic structures [68]. In the present study, it was observed that linear non-ionic GEN absorbs UV light more intensively than DDAC (pKa < 8) amine with strong complex formation ability at pH 8.5. In addition, it may form ion pairs or micelles, too [69,70].

Earlier, it was discovered that non-linear DDAC has CMC of about 180 µg ml^{-1} which is higher than that of linear non-ionic GEN (CMC 19-150 µg ml^{-1}), meaning that GEN forms more easily neutral micelles [63]. In the present study, ion-pairing and micelle formation were excluded with organic solvents.

Repeatability of the Separation Profiles

In the present study, generally, DDAC and GEN were detected separately since they were not informed to be in the same disinfectant products that were used in the daily cleaning of the two schools. Their mobilities were very repeatable standardized to the intraday velocity of electroosmosis (RSD% < 1%). However, the migration speeds of the complexes depended on their concentrations studied at concentrations >10 ng ml^{-1} and 1 µg ml^{-1} shifted

slightly during the measurements. Unexpectedly, the GEN peaks in electropherograms were always doubled. Probably, the reason was the complex formation, since complexes could be formed with both the hydroxyl group and the ether oxygen. Details of the method quality are compiled in Tables 2 and 3.

Table 2. Parameters Calculated for DDAC and GEN from Data Obtained Using the Optimized Tetraborate Buffer Composition

Parameters*	Values
DDAC as the TB complex	
Retention factor	12.4
Tailing factor $T_f = (a + b)/2a$	1.3
Number of theoretical plates (31.7 cm L_{det}):	
$N = 5.545 \times (t_r/w)^2$	64100
Resolution: $R = \frac{1}{4}\sqrt{N} * \left(\frac{\Delta\mu}{\bar{\mu}}\right)$	0.259 (1 and 2 peaks)
Resolution between GEN and DDAC	15.5
GEN as the TB complex	
Retention factor	14.0
Tailing factor $T_f = (a + b)/2a$	1.2
Number of theoretical plates (31.7 cm L_{det}):	
$N = 5.545 \times (t_r/w)^2$	N_1 142000; N_2 334000
Resolution $R = \frac{1}{4}\sqrt{N} * \left(\frac{\Delta\mu}{\bar{\mu}}\right)$	1.38

*Equations from [71].

Table 3. Repetition Analyses of the Samples Containing DDAC and GEN. The Mixtures were Directly Introduced to the CE Analyses without Filtration

Disinfectant as TB complexes	DDAC	GEN	Electroosmosis (marker: methanol)
LOD (ng ml^{-1})	0.006	1.0	
LOQ (ng ml^{-1})	0.018	3.84	
Linearity	Y = 4.3429x + 187.39 (r^2 0.841)	y = 26.985x + 125.60 (r^2 0.837)	
Maximum concentration in calibration (μg ml^{-1})	35	50	
Repeatability of the method:			
t_{eo} (RSD %), (min)	1st and 2nd signals: 27.69, 28,19 (7)	1st and 2nd signals: 29.36, 29.95 (6)	3.2 (5)
μ_{eof} (RSD %), (m^2 V^{-1} s^{-1})	1st and 2nd signals: -2.07×10^{-8} (10), -2.05×10^{-8} (11)	1st and 2nd signals: -2.44×10^{-8} (12), -2.45×10^{-8} (11)	4.6×10^{-8} (13)
Performance of the method during 3 months:			
μ_{eof} (RSD %), (m^2 V^{-1} s^{-1})			$4.6 \cdot 10^{-8}$ (13)
Veritable and repeatable concentrations			
Low (ng ml^{-1}), (RSD%)	1-10 (2)	1-10 (2)	
High (ng ml^{-1}), (RSD%)	50-200 (10)	50-200 (10)	
Repeatability of concentrations in authentic samples (RSD%)	Sampled into ethanol: 9-38 Sampled into the water: 8-10	Sampled into ethanol: 9-15 Sampled into the water: 9-20	
Accuracy of indoor air and surface samples average value (%)	4	10	

The results of concentration calibration for DDAC and GEN complexes are listed in Table 3. The method is valid between 1-10 ng ml^{-1} and 50-200 ng ml^{-1}. The blanks and standards were conducted by dissolving the studied analytes in deionized water or methanol when the solvent peak was needed as the electroosmosis marker.

Reference samples to mimic the evaporation and movement of the disinfectants were experimentally processed separately for DDAC and GEN. They were collected inside a glass chamber in a laboratory in a controlled manner. Water vapor contained the compounds from a departure plate. The vapor was relocated to an arrival plate when RH was 100% inside the glass chamber in a laboratory [9,49]. Before CE analyses the collected condensates were diluted (1/1000-1/100000-fold) with deionized water for reducing concentrations to fit between the linear ranges in calibration. Examples are presented in Fig. 3.

The results showed that the CE method was more sensitive compared to the earlier published IC method [72]. The LOD and LOQ values of DDAC were 2.97 µg m^{-3} and 8.92 µg m^{-3} in IC, respectively, with RSD 7.8% at a range of ~0-20 µg ml^{-1}. In the present study, the LOD and LOQ were 1.0 ng ml^{-1} and 3.84 ng ml^{-1}, respectively. The value of non-complexed DDAC (0.56 µg ml^{-1}) was lower than that analyzed by IC-MS/MS [24]. Using the equations published in the IC paper to calculate the correlating value from the CE data, the DDAC amount was 28 µg m^{-3} with an air volume of 100 l (in desorption volume of 5 ml). Unfortunately, they were not similar to the reference data available for GEN.

Determination of Indoor Samples

The chemicals studied are daily used for cleaning the school buildings in Finland. It was known that the sampling locations were not cleaned with the chemicals, since they were used for floors, tables, and chairs.

Very few recent studies about air contaminants in indoor buildings are reported despite the paper giving results about their existence in sports environments [73]. The contaminants stay in indoor air. Their discoveries are found due to requirements of energy savings for ventilation (the regulation of the Energy Performance of Building Directive 2010/31/EU (EPBD). The demands may result in increased amounts of indoor air pollution [74]. On account of that, the

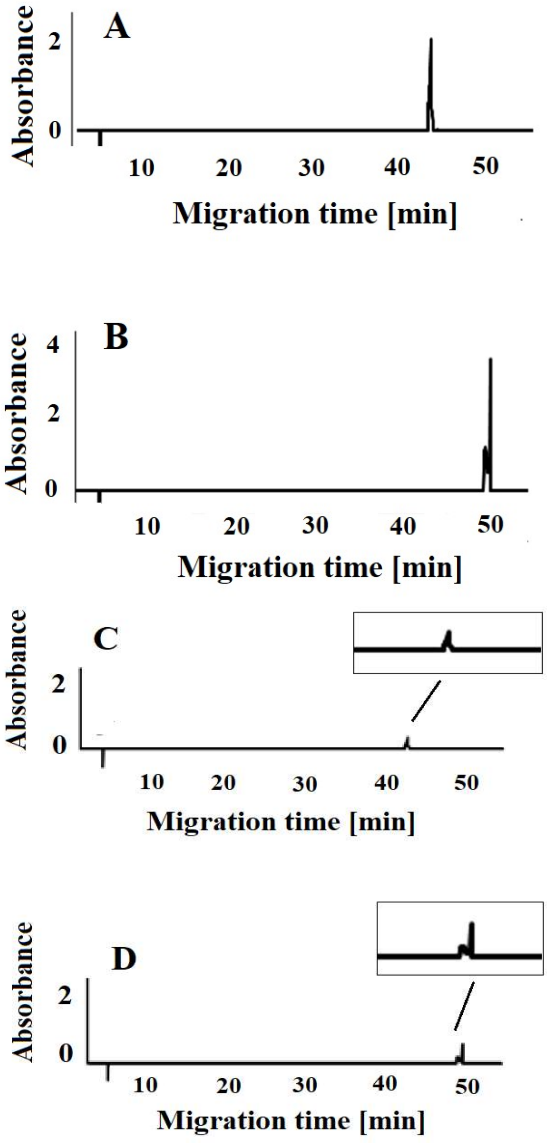

Fig. 3. The electropherogram of diluted condensed water samples (artificially made in the laboratory) containing (A and C) DDAC and (B and D) GEN. The condensed water samples were diluted three times with pure water; the first stock dilutions were tenfold; then A and B dilutions were 1/1000-fold; C and D dilutions were 1/100000-fold. Analytical conditions: capillary 48.8 cm/40 cm × 50 µm, detection 214 nm, voltage +17 kV; buffer $Na_2B_4O_7 \cdot 10H_2O$ (50 mM, pH 8.5); pre-run conditioning 2 min 0.1 M NaOH, 2 min buffer, water injection at 35 mbar for 20.0 s; sample injection (35 mbar, 10 s); run time 60 min. The current in these runs was approximately + 90 µA.

surfaces may contain harmful compounds which may be removed from indoor air when the humidity increases. Since DDAC and GEN are tensides they decrease the surface tension of water, and in indoor air, at very low concentrations they are toxic to human cells.

The present work showed selectively only the targeted DDAC and GEN disinfectants from indoor air and room surface samples of two elementary schools. The samples are listed in Table 4. The reasons to study the disinfections were: At first, it was believed that the buildings had mold and mildew problems, but the microbiological results were negative, and further investigations were needed. Secondly, despite renovations in the schools, the air problem was not extinct: the pupils and personnel suffered from asthma-like symptoms. Thirdly, washing agents used in cleaning were important targets and thus could be compared to published data by other researchers [12]. Thus, the disinfectants of the cleaning chemicals were studied to observe and identify them in authentic samples.

Table 4. Characterization of the Indoor Air and Surface Samples from the Schools of Hämeenkylä (H) and Kannisto (K)

Sample	School	Space	Solvent Volume (ml)	Observed GEN C (ng ml^{-1})	Observed DDAC C (ng ml^{-1})	Observed corresponding agent[a]
A1	H	HW 1062	Ethanol 5 ml	No disinf.	No disinf.	No disinf.
A4	H	C 2011	Condensed water 8.049 g	No disinf.	No disinf.	No disinf.
A5	H	C 1051	Condensed water 5 ml			x
A6	H	-	Condensed water 1.9 g	No disinf.	No disinf.	No disinf.
A7	H	HW 1062	Condensed water 5 ml	2.5		
B1	H	HW 1062	Ethanol 5 ml		214	
B3	H	C 2011	Ethanol 5 ml		399	
B5	H	C 1051	Condensed water 4.4 g			x
C1	H	C 1005	Ethanol 5 ml	12		
C2	H	C 1004	Ethanol 4 ml	18		
C4	H	C 2011	Condensed water 5 ml		223	
D1	H	C 1051	Condensed water 5 ml			x
D2	H	C 2011	Condensed water 5 ml	138		
D4	H	C 1051	Ethanol 5 ml	No disinf.	No disinf.	No disinf.
E1	H	C 1051	Condensed water 5 ml	No disinf.	No disinf.	No disinf.
E2	H	C 2011	Condensed water 5 ml	40		
E3	H	Lobby	Condensed water 5 ml	No disinf.	No disinf.	No disinf.

Table 4. Continued

E7	H	KT 1062	Condensed water 5 ml			x
F1	H	C 2011	Water + Ethanol 5 ml			x
F2	H	C 1051	Ethanol 5 ml		40	
F3	H	Open air	Condensed water 5 ml		798	
F4	H	C 1004	Condensed water 4.055 g			x
F7	H	GYM	Ethanol	No disinf.	No disinf.	No disinf.
G1	H	C 1063	Ethanol			x
G2	H	Open air	Water + ethanol 5 ml			x
G3	H	C 1058	Condensed water 5 ml			x
G7	H	GYM	Condensed water 5 ml	6.6		
H3	K	C 2068	Water, pH 10	No disinf.	No disinf.	No disinf.
H4	K	C 2068	Ethanol 5 ml	No disinf.	No disinf.	No disinf.
H5	K	Lobby	Condensed water 3.9 ml	217		
H6	K	C 2042	Ethanol 3.54 ml	609		
H7	K	C 2039	Condensed water 5 ml			x
I3	K	C 2086	Condensed water pH 5.5			x
I4	K	C 2068	Condensed water 5 ml	No disinf.	No disinf.	No disinf.
I5	K	C 2039	Ethanol 5 ml	No disinf.	No disinf.	No disinf.
I6	K	C 2042	Ethanol 5 ml	No disinf.	No disinf.	No disinf.
I7	K	C 2039	Ethanol 5 ml	No disinf.	No disinf.	No disinf.
J3	K	-	Condensed water pH 10	No disinf.	No disinf.	No disinf.
J4	K	C 2086	Condensed water 3 ml			x
J5	K	C 2042	Ethanol 3 ml			x
J7	K	Open air	Ethanol 5.3 ml	66		
K3	K	C 2086	Ethanol 5 ml	64		
K5	K	C 2042	Condensed water 5 ml			x

Table 4. Continued

K6	K	Lobby	Condensed water 5 ml			x
K7	K	Open air	Condensed water 1 ml dil. with 0.5 ml ethanol			x
L6	K	C 2012	Condensed water 5 ml	No disinf.	No disinf.	No disinf.

Abbreviations: C = classroom, HW = hallway, O = outdoor, KT = kitchen, GYM = gymnastic hall, lobby = lounge; H (Hämeenkylä School) and K (Kannisto School). The compounds observed corresponding agents were also disinfectants complexed with tetraborate. The method was specifically developed to amine-type and neutral polymeric compounds. The observed compounds were different than those of DDAC and GEN (based on electro profiles). Because the method was targeted only to the studied compounds, the identification was not done. x) Other disinfectants.

Based on our laboratory experiments, DDAC and GEN move in aqueous vapor and aerosols. The samples are listed in Table 4. However, the samples needed dilution with deionized water before CE analyses, except for two of them (A7 H, hallway, and J7 K, open-air). From the 46 analyzed samples taken from different locations in the school buildings ten contained GEN and five DDAC at various concentrations. It was noticed that when the sampling places were near each other, the results were a similar order of magnitude (Table 4). The examples are samples from the room C 2011 (B3 H & C4 H). Then the twice sampled DDAC concentration was 399 ng ml^{-1} and 223 ng ml^{-1}, respectively. When the profiles were free from the targeted compounds, the analysis results were blank profiles (See the Kannisto School sample C 2068 in water, condensed water, and ethanol). However, the electropherograms showed unknown cationic surfactants which could not be identified.

The concentrations of the two disinfectants were quite low (2.5-270 ng ml^{-1}) in many samples, except in two classroom samples and an outdoor air sample, since the concentrations were above 600 ng ml^{-1} (Table 4). Overall, DDAC and GEN amounts in the condensates varied between 200-760 ng ml^{-1} and 2.5-1029 ng ml^{-1}, respectively.

GEN was determined in 11 samples at low amounts (Table 4). However, DDAC existed in a large concentration in samples from Hämeenkylä School. The cleaning chemicals used in Hämeenkylä School contained DDAC and GEN based on the results of the study which correlated with the documents of the purchased contract. On the other hand, in samples from Kannisto School DDAC was not observed. However, some unidentified cationic-type disinfectant was detected. As a final result, disinfectants make thin water surfaces on respiratory organs and human cells and thus decrease the hydration of potassium and sodium ions, which causes its relation to asthma disease [73].

DISCUSSION

The disinfectants DDAC and GEN as anionic tetraborate complexes allow their migration and direct UV detection with capillary electrophoresis. The studies show evidence that they both removed indoor air and stayed stabilized on low-humid or dry surfaces in the school rooms. High moisture percentages exist in indoor areas of school buildings when the students are inside and breathe the air in the classrooms: It is assumed that each pupil exhales several cubic meters of RH 100% air during school hours. Thus, the RH of the school rooms increases remarkably during teaching hours. The air of the classroom dries (even to less than RH 20%) during nights and weekends when the ventilation is kept at a minimal level causing the disinfectants to adsorb back on surfaces. The reason for the decrease in moisture is that the machines for incoming air are programmed to keep low performance during the hours when the schools are empty [75]. More recent information about the topic is obtained from a literature survey describing indoor air humidity. Versatile compaction was made by Wolkoff [76]. According to the literature, similar CE-UV studies (Table 4) have not been done to show profiles of the superior detectability of DDAC and GEN with tetraborate ions when the two compound-targeted screening technique is used. Naturally, surfactants and disinfectants have been studied with various kinds of chromatographic techniques, but there are not many methods developed with capillary electrophoresis.

Table 5. Selection Data Results of Disinfectants and Surfactants by Capillary Electrophoresis from Literature

Surfactants	Matrix	Sample preparation	Analysis method and conditions	LOD/LOQ	Ref.
FAE Alkyl polyglucosides	Laundry detergent Shower gel	Derivatization	CZE-UV Electrolyte: 150 mM tetraborate (pH 8-9) with 50% ACN	Not mentioned	[77]
APEO, alkylpyridinium salts, alkylsulphate, alkylsulphonates, LAS, quaternary alkylammonium salts	River water, toothpaste	SPE anions	CZE-UV Electrolyte *Aliphatic anions:* 20 mM salicylate pH 6 + 6 vol-% ACN, *LAS:* 100 mM phosphate, pH 6.8 *LAS isomer separation*: 100 mM phosphate, pH 6.8 +15 mM α-CD + 2 vol-% ACN *Alkylsulphonates*: 5 mM dodecylbenzene-sulphonate, 5 mM phosphate, pH 6.8 + 30 vol-% ACN *Non-ionic compounds*: 10 mM phosphate, pH 6.8, 35 vol-% ACN + 70 mM SDS Cationic compounds: 50 mM phosphate (pH 6.8), 50-55 vol-% ACN + 3 mM SDS	SDS: 84 ng l^{-1} CPC: 0.73 µg l^{-1}	[51]
Quaternary alkylammonium compounds: DTAB, TTAB, DDAC, ODAC, DDAB	Hand sanitizers Skin cleaners Laundry Detergents Wet wipes	Liquid samples: samples modified by adding 1.8 ml of buffer (MeOH/ACN, 20:80, v/v, containing 2 mM TFA) Wet tissue samples: Samples modified by adding 1.8 ml of buffer (MeOH/ACN, 20:80, v/v, containing 2 mM TFA)	NACE-IUD Electrolyte: MeOH/ACN (90:10, v/v) containing 2 mM sodium acetate, 2 mM trifluoroacetic acid (TFA) and 16 mM dodecyl dimethyl benzyl ammonium chloride	LOD 0.5 mg l^{-1}; LOQ 5.0 mg l^{-1} for all	[78]
Quaternary alkylammonium compounds	Industrial mixture of surfactants	Not any	CZE-UV electrolyte: 0.5 M phosphate, pH 4 + 50 vol% THF	LOD: 0.1 µM C12/14-benzyl-DMA: 5.6 mM C16/18-benzyl-DMA: 20 mM	[42]
AE, NPEO, OPPEO	Not applied to environmental samples	Not any	CZE-UV electrolyte: 10 mM phosphate, pH 6.8 + 70 mM SDS + 40 vol% ACN	Not mentioned	[50]

Table 5. Continued

Quaternary alkylbenzylammonium compounds	Experimental disinfectant product	Not any	CZE-UV (direct, indirect) electrolyte: direct UV: 44 mM phosphate + 57.5 vol% THF indirect UV: 8 mM phosphate, 3 mM SLS, 3 mM C12 Benzyl + 57.5 vol% THF	Estimated to be less than mg l^{-1}	[79]
ALA, GLU, HAB, LAB, PEA, SAR, TAL	Commercial cosmetic products	Ultrasonication centrifugation filtration	HPCE-UV electrolyte: 80 mM borate, pH 9.2 + 20 mM NaOH	Not mentioned	[52]
Quaternary alkylbenzylammonium compounds	Not applied to environmental samples	Not mentioned	CZE-UV electrolyte: 20 mM phosphate, pH 5.0 + 30 vol% ACN/ 40vol% THF/ 50 vol% acetone	between 0.40-0.85 µM	[53]
Quaternary alkylbenzylammonium compounds	Not applied to environmental samples	Not mentioned	CZE-UV 60 vol% MeOH or 30 vol% ACN to sample solution electrolyte: 20 mM phosphate, pH 5.0 + 30-40 vol% ACN	Not mentioned	[80]
LAS	Riverwater	SPE	CZE-UV electrolyte: 100 mM phosphate, pH 6.8 + 30 vol% ACN + 10 mM α-CD (isomer separation), β-CD (total LAS), or γ-CD (homologue separation)	Average 4.8 mg l^{-1}	[77]
LAS	Wastewater from industry and sewage sludge	SPE	CZE-UV electrolyte: 250 mM borate, pH 8.2 + 30 vol% ACN	1 mg l^{-1}	[36]
Polyhexamethylguanidine (PHMG)	Not applied to environmental samples	Not any	CZE-UV electrolyte: 20 mM phosphate, pH 6.86 + ACN	0.002 mg ml^{-1}	[81]
Alkanesulphonates, alkylsulphates, LAS	Laundry detergents	Filtration	CZE-UV (indirect) electrolyte: MeOH/ACN + 10 mM PSA, 5 mM PSAH	Alkylsulphates: 9.6-13 µM alkanesulphonates: 12-75 µM	[82]
Benzethonium (BAC) and cetylpyridinium (CP) ions	Artificial samples	Not described	75 mM phosphoric acid and 50% acetonitrile electrolyte at pH 2.5.	1.47 and 4.30 µg ml^{-1} for benzethonium and cetylpyridinium	[83]
Alkyltrimethyland dialkyldimethylammonium compounds (ATMACs and DADMACs	Hair conditioners and liquid fabric softeners	Diluted with methanolic solution (90%, v/v)	10 mM phosphate buffer with 57.5% tetrahydrofuran and 3 mM sodium dodecyl sulfate (SDS) at pH 4.3.	0.05-0.1 and 0.1-0.5 µg ml^{-1}	[84]
Benzalkonium chloride (BAC) and cetylpyridinium chloride (CPC)	Industrial (for hospitals) and household formulations	Not described	Electrolyte: 80 mM borate (pH 8.5), 50 mM sodium deoxycholate and 30% ethanol	a few mg ml^{-1}	[85]

CONCLUSION

Biocide-type disinfectants used as cleaning agents are difficult to remove from indoor surfaces due to their adsorption and adherence to dry surface materials. However, under a moist environment, they move with water vapor and aerosols to various targets. Persons staying in the chemical-cleaned rooms may be exposed frequently. It was proved that the capillary electrophoresis method with tetraborate complexation was accurate for cationic didecyldimethyl ammonium chloride and non-ionic polyethylene glycol monoalkyl ether. Both analytes were detected as hypersensitive complexes which allowed their determination in multi-component water condensates of two elementary schools. Since they were used daily for cleaning, water-evaporated disinfectants were shown to have a possible correlation with the negative health symptoms of humans in the classrooms.

ACKNOWLEDGMENTS

The assistance of emer. prof. M. Salkinoja-Salonen and the members of her research group are highly appreciated for sampling and giving the samples to the capillary electrophoresis investigation.

REFERENCES

[1] U. Lignell, Doctoral Dissertation, University of Kuopio (2008). http://www.ktl.fi/portal/4043

[2] I. Senta, S. Rodríguez-Mozaz, L. Corominas, A. Covaci, M. Petrovic, Sci. Total Env. 845 (2022) 157309.

[3] H.M. Keskinen, I. Ylivinkka, L. Heikkinen, P.P. Aalto, T. Nieminen, K. Lehtipalo, J. Aalto, J. Levula, J. Kesti, L.R. Ahonen, E. Ezhova, M. Kulmala, T. Petäjä, AMT 2020 (2020) 1.

[4] K. Axelrod, V. Samburova, A.Y. Khlystov, Sci. Total Env. 799 (2021) 149254.

[5] T. Verdier, M. Coutand, A. Bertron, C. Roques, Building and Environment 80 (2014) 136.

[6] Z. Andualem, Z. Gizaw, L. Bogale, H. Dagne, Multidiscip. Respir. Med. 14 (2019) 1.

[7] B. Kronberg, K. Holmberg, B. Lindman, in B. Kronberg, K. Holmberg, B. Lindman (eds.). Surfactants and polymers. In aqueous solution, John Wiley & Sons Inc, 2014.

[8] M. Andersson, J. Salo, O. Kedves, L. Kredics, I.S. Druzhinina, J. Kurnitski, H. Salonen, Microorganisms 8 (2020) 1.

[9] P. Harmo, J. Selkäinaho, M. Salkinoja-Salonen, H. Sirén, M.L. Riekkola, A. Visala, SIY Report 36 (2018) 413.

[10] S. Prasad, J.C. Lipszyc, S.M. Tarlo, Lympho. Sign. J. 5 (2018) 121.

[11] O. Vandenplas, V. D'Alpaos, G. Evrard, J. Jamart, J. Thimpont, F. Huaux, J.C. Renauld, BMJ Open 3 (2013) e003568.

[12] W.Y. Kim, S.B. Hong, Tuberc. Respir. Dis. (Seoul) 80 (2017) 351.

[13] I.A. Jones, L.T. Joshi, Molecules 26 (2021) 2276.

[14] A. Friedle, J. Thumulla, K. Snepvangers, Indoor Air Congress Abstract book (2008) 17. Copenhagen, Denmark – Paper ID: 332.

[15] EFSA supporting publication, EN-483 (2013) 1.

[16] European Food Safety Authority, EFSA Journal 12 (2014) 3675.

[17] K. Slimani, A. Féret, T. Pirotais, P. Maris, J.P. Abjean, D. Hurtaud-Pessel, J. Chrom. A 1517 (2017) 86.

[18] W.A. Rutala, D.J. Weber, Guideline for disinfection and sterilization in healthcare facilities, Centers for Disease Control and Prevention, U.S. 2019.

[19] M. Mannerström, T. Toimela, J. Ahoniemi, A.S. Makiou, T. Heinonen, Appl. in vitro Toxicol. 6 (2020) 120.

[20] A.M. Pratelli, Zoon. Publ. Health 54 (2007) 383.

[21] C.I. Schrank, K.P.C. Minbiole, W.M. Wuest, ACS Infect. Dis. 6 (2020) 1553.

[22] P.I. Hora, S.G. Pati, P.J. McNamara, W.A. Arnold, Env. Sci. Tech. Lett. 7 (2020) 622.

[23] H.M. Dewey, J.M. Jones, M.R. Keating, J. Budhathoki-Uprety, ACS Chem. Health Saf. 29 (2022) 27.

[24] G. Vincent, M.C. Kopferschmitt-Kubler, P. Mirabel, G. Pauli, M. Millet, Env. Monit. Ass. 133 (2007) 25.

[25] R. Saito, M.A. Virji, P.K. Henneberger, M.J. Humann, R.F. LeBouf, M.L. Stanton, X. Liang, A.B. Stefaniak, Am. J. Ind. Med. 58 (2015) 101.

[26] ISO 16000-6, Indoor air - Part 6 (2011).

[27] J.M. Boyce, Antimic. Res. Inf. Cont. 5 (2016) 1.
[28] ISO/DIS 16000-6:2011, Indoor air - Part 6 (2011).
[29] ISO/DIS 16000-6:2020, Indoor air - Part 6 (2020).
[30] A. Onigbinde, B. Munson, B. Amos-Tautua, Res. J. App. Sci. Eng. Tech. 5 (2015) 2332.
[31] Commission Directive 98/8/EC, Off. J. Eur. Communities L123 (1989) 1.
[32] Commission Regulation/EU No 528/2012OF, Off. J. Eur. Union L167 (2012) 1.
[33] M. Danaher, K.N. Jordan, Irish J. Agr. Food Res. 52 (2013) 173.
[34] D. Paek, Y. Koh, D.U. Park, H.K. Cheong, K.H. Do, C.M. Lin, S.J. Hong, Ann. Am. Thorac. Soc. 12 (2015) 1813.
[35] K. Heinig, C. Vogt, G. Werner, Fresenius J. Anal. Chem. 358 (1997) 500.
[36] K. Heinig, C. Vogt, Analyst 123 (1998) 349.
[37] R.A. Wallingford, Anal. Chem. 68 (1996) 2541.
[38] P. Mukerjee, K. Mysels, P. Kapauan. J. Phys. Chem. 71 (1967) 4166.
[39] P. Warszynski, W. Barzyk, K. Lunkenheimer, H. Fruhner, J. Phys. Chem. B 102 (1998) 10948.
[40] W.P. Gardner, J.E. Girard, J. Chem. Educ. 77 (2000) 1335.
[41] L. Jiang, J. Wang, Y. He, J. Ye, Q. Chu, J. Chrom. Sci. 48 (2010) 584.
[42] E. Piera, P. Erra, M.R. Infante, J. Chrom. A 757 (1997) 275.
[43] J. Sukhapan, P. Brimblecombe, Sci. World J. 2 (2002) 1138.
[44] M. Abdel-Salam, J. Aerosol Med. 19 (2006) 434.
[45] E. Castagnoli, Doctoral dissertation, Aalto University (2020). ISBN: 978-952-64-0003-7.
[46] Y.S. Kim, S.B. Lee, C.H. Lim, Toxicol. Res. 33 (2017) 7.
[47] E. Castagnoli, M. Andersson, R. Mikkola, L. Kredics, T. Marik, J. Kurnitski, H. Salonen, A. Indoor Seminar in Helsinki. Conference paper: Sisäilmastoseminaari Volume: SYI report 35 (2017) Helsinki, Finland. www.google scholar
[48] J.T. Salin, M. Salkinoja-Salonen, P.J. Salin, K. Nelo, T. Holma, P. Ohtonen, H. Syrjälä, Env. Res. 154 (2017) 234.
[49] J. Selkäinaho, P. Harmo, M. Salkinoja-Salonen, J. Luukkaa, H.M. Siren, M.L. Riekkola, A. Andersson, R. Mikkola, H. Salonen, J. Kurnutski, A. Visala, In R. Kosonen, M. Ahola, J. Narvanne (Eds.), Roomvent & Ventilation 2018: Excellent indoor climate and high performing ventilation, Track 1 - Personal conditions: Air quality (2018) 91-96. www.google scholar
[50] K. Heinig, C. Vogt, G. Werner, Fresenius J. Anal. Chem. 357 (1997) 695.
[51] K. Heinig, C. Vogt, G. Werner, J. Chrom. A 745 (1996) 281.
[52] W.G. Lin, S.T. Lin, S.L. Shu, J. Surf. Deterg. 3 (2000) 67.
[53] C. Lin, W. Chiou, W. Lin W, J. Chrom. A 722 (1996) 345.
[54] J.W. Robinson, E.M. Skelly Frame, H.G.M. Frame II, Chromatography with liquid mobile phase in undergraduate instrumental analysis (2005) 797.
[55] S.J. Angyal, D. Greeves, V.A. Pickles, Carbohydrate Res. 35 (1974) 165.
[56] S. Chapelle, J.F. Verchere, Carb. Res. 191 (1989) 63.
[57] H.S. Isbell, J.F. Brewster, N.B. Holt, H.L. Frush, J. Res. Nat. Bur. Stand. J. Res. 40 (1948) 128.
[58] S. Hoffstetter-Kuhn, A. Paulus, E. Gassmann, H.W. Widmer, Anal. Chem. 63 (1991) 1541.
[59] E. Hurtado-Fernández, P.K. Contreras-Gutiérrez, L. Cuadros-Rodríguez, A. Carrasco-Pancorbo, A. Fernández-Gutiérrez, Food Chem. 141 (2013) 3492.
[60] R. Sekar, S.K. Kailasa, W.S. Li, H.C. Wu, H.F. Wu, Chin. Chem. Lett. 24 (2013) 833.
[61] V. Kochkodan, N.B. Darwish, Boron separation processes (2015) 35.
[62] Y. Furukawa, M. Horiuchi, T. Kakegawa, Orig. Life Evol. Biosph. 43 (2013) 353.
[63] L. Marzhall, Int. J. Pharm. 6 (1980) 253.
[64] Z.K. Shihabi, J. Chrom. A 1066 (2005) 205.
[65] N. Rajana, K.M. Yarbagi, K. Balakumaran, M.V. Madhubabu, J. Mosesbabu, K. Basavaiah, D.R. Devi, J. Chrom. Sci. 57 (2019) 769.
[66] B.M. Lowe, C.K. Skylaris, N.G. Green, J. Col. Int. Sci. 451 (2015) 231.
[67] Z. El Rassi, Electrophoresis 18 (1997) 2400.
[68] S. Honda S, J. Chrom. A 720 (1996) 337.
[69] S. Hiltunen, H. Sirén, Anal. Bioanal. Chem. 405 (2013) 5773.

[70] P. Lukkari, J. Jumppanen, T. Holma, H. Sirén, K. Jinno, H. Elo, M.L. Riekkola, J. Chrom. A 608 (1992) 317.
[71] T.A. Maryutina, E. Yu. Savonina, P.S. Fedotov, R.M. Smith, H. Siren, D.B. Hibbert, IUPAC Recommendations (2018).
[72] J.S. Yang, S.B. Choi, S.Y. Park, S.B. Lee, Anal. Sci. Techn. 25 (2012) 307.
[73] H. Salonen, T. Salthammer, L. Morawska, Indoor Air 30 (2020) 1109.
[74] REHVA. Energy Performance of Building Directive (2010). https://www.rehva.eu/eu-policy/energy-performance-of-buildings-directive-epbd
[75] J. Toivanen, MSc thesis, Aalto University (2017). www.aalto.fi
[76] P. Wolkoff, Int. J. Hyg. Env. Health 221 (2018) 376.
[77] C. Vogt, K. Heinig, B. Langer, J. Mattusch, G. Werner, Fresenius J. Anal. Chem. 352 (1995) 508.
[78] K. Yao, R. Jiang, P. Wang, J. Zhang, B. Shao, X. Ding, Separations 10 (2023) 387.
[79] C.S. Weiss, J.S. Hazlett, M.H. Datta, M.H. Danzer, J. Chrom. A 608 (1992) 325.
[80] C. Lin, W. Chiou, W. Lin, J. Chrom. A 723 (1996) 189.
[81] A.V. Rudnev, T.G. Dzherayan, J. Anal. Chem. 61 (2006) 1002.
[82] H. Salimi-Moosavi, R.M. Cassidy, Anal. Chem. 68 (1996) 293.
[83] N. Oztekin, F.B. Erim, J. Pharm. Biomed. Anal. 37 (2005) 1121.
[84] H.Y. Liu, W.H. Ding, J. Chrom. A 1025 (2004) 303.
[85] J.M. Herrero-Martínez, E.F. Simó-Alfonso, C. Mongay-Fernández, G. Ramis-Ramos, J. Chrom. A 895 (2000) 227.