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Serum concentrations of per- and polyfluoroalkyl substances (PFAS) among men from the Danish fire services and Armed Forces

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A R T I C L E I N F O

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A B S T R A C T

Background: Per- and polyfluoroalkyl substances (PFAS) have been used extensively in firefighting foams with resulting occupational exposure among firefighters.

Objective: To examine serum concentrations of PFAS among current and former employed and volunteer firefighters from the Danish fire services and Armed Forces.

Methods: During 2023–2024, 429 men from the Danish fire services and Armed Forces participated in the study. They were asked to provide a blood sample and fill in an online questionnaire. Concentrations of 15 PFAS were measured in serum. Measurements from the general population sampled in 2021 (the ENFORCE study) were used as reference. Associations between occupational factors and serum PFAS were assessed using multiple linear regression.

Results: Participants were from municipal fire services (n = 208), governmental fire services (n = 59), civilian airport fire services (n = 50), the air force (n = 98) and the navy (n = 14). Their median age was 50 years and median year of commencing service was 1999. While serum concentrations of PFAS among most participants were at level with those of the general population, civilian airport firefighters had higher serum concentrations of especially perfluorohexane sulfonic acid (PFHxS), perfluoroheptane sulfonic acid (PFHpS) and perfluorooctane sulfonic acid (PFOS). Age-adjusted geometric means were 1.42 ng/mL for PFHxS, 0.28 ng/mL for PFHpS and 6.92 ng/mL for total PFOS among civilian airport firefighters.

Conclusion: Higher serum concentrations of PFHxS, PFHpS and PFOS among civilian airport firefighters likely reflected past occupational exposure to firefighting foam. Findings emphasized the importance of regulatory measures and substitution.

1. Introduction

They are lurking in all the comforts of our modern lives. The versatile family of per- and polyfluoroalkyl substances covers more than 9000 individual chemicals found in a wide range of products (The Interstate Technology, 2023; Miljøstyrelsen. Kortlægning af brancher, 2016; National Institute for Occupational Safety and Health, 2021; Evich et al., 2022). Their fluorinated carbon structure yields an appealing combination of surface-active properties and exceptional stability (The Danish Environmental Protection Agency et al., 2015). As industries and consumers continue to rely on PFAS, concerns for environmental footprints and health impacts have gained significance. Especially the non-polymeric PFAS have attracted much scientific and regulatory attention due to their inevitable bioaccumulation (Hull et al., 2023). In

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humans, their half-lives for excretion are exceptionally long (5.3 years for perfluorohexane sulfonic acid (PFHxS), 2.7 years for perfluorooctanoic acid (PFOA), and 4.7 years for perfluorooctane sulfonic acid (PFOS)) (Rosato et al., 2024). The most common non-polymeric PFAS are epigeno- and immunotoxic (Zahm et al., 2024). These are characteristics of carcinogens and PFOA has recently been classified as *carcinogenic to humans* (group 1) by the International Agency for Research on Cancer (IARC) (Zahm et al., 2024). In addition, exposure to non-polymeric PFAS has been linked to changes in liver and kidney function, lipid metabolism, reproductive organs along with developmental effects (EFSA panel on contaminants in the food chain, 2020).

While progress has been made regarding our understanding of PFAS in various environmental matrices and general populations worldwide, documentation of occupational exposure remains scarce, and available measurements cover only a few industries (Lucas et al., 2022; Paris-Davila et al., 2023). Most frequently, occupational exposure has been studied in relation to firefighting (Burgess et al., 2022; Dobraca et al., 2015; Goodrich et al., 2021; Graber et al., 2021; Jin et al., 2011; Khalil et al., 2020; Laitinen et al., 2014; Leary et al., 2020; Tefera et al., 2023; Nilsson et al., 2020, 2022a; Rihackova et al., 2023; Rosenfeld et al., 2022; Rotander et al., 2015; Shaw et al., 2013; Tao et al., 2008; Trowbridge et al., 2020; Purdue et al., 2023; Nematollahi et al., 2023). Due to their flame-resistant and water-repellent profile, PFAS have long served as integral components of both protective equipment and firefighting foams (Rosenfeld et al., 2022). In particular, high concentrations of PFAS have been used in aqueous film-forming foam (AFFF) (NIRAS, 2021). Further, fire smoke and dust may also contain PFAS and contribute as sources of exposure (Rosenfeld et al., 2022; Tao et al., 2008; Young et al., 2021). Exposure may occur during actual fire incidents, training scenarios or activities in the hazardous zones of the workplace (i.e., decontamination area or apparatus bay). Inhalation is considered the main exposure route with potential contributions also from dermal absorption and incidental ingestion (Rosenfeld et al., 2022).

Previous studies measuring PFAS in relation to firefighting have shown striking variations in blood concentrations among employees and volunteers (Burgess et al., 2022; Dobraca et al., 2015; Goodrich et al., 2021; Graber et al., 2021; Jin et al., 2011; Khalil et al., 2020; Laitinen et al., 2014; Leary et al., 2020; Tefera et al. 2023; Nilsson et al., 2022a; Rihackova et al., 2023; Rotander et al., 2015; Shaw et al., 2013; Tao et al., 2008; Trowbridge et al., 2020; Purdue et al., 2023; Nematollahi et al., 2023). The contrasts in exposure may largely reflect differences in use of AFFF and related working conditions (Leary et al., 2020; Nilsson et al., 2022a; Rihackova et al., 2023). Globally, the quantities of AFFF used by military services far outweigh the amounts spent by civilian fire services (Rosenfeld et al., 2022; Ruyle et al., 2023). Despite the potential intensity of AFFF use in the military, the majority of existing studies cover PFAS exposures exclusively among regular firefighters (Burgess et al., 2022; Dobraca et al., 2015; Goodrich et al., 2021; Graber et al., 2021; Jin et al., 2011; Khalil et al., 2020; Laitinen et al., 2014; Leary et al., 2020; Tefera et al., 2023; Rihackova et al., 2023; Rotander et al., 2015; Shaw et al., 2013; Trowbridge et al., 2020; Nematollahi et al., 2023). Thus, the aim of this study was to examine serum concentrations of non-polymeric PFAS among different types of current and former employed and volunteer firefighters from both the Danish fire services and Armed Forces in relation to reference measures from the general population.

2. Materials and methods

2.1. Setting and study population

In Denmark, rescue and fire management primarily relies on municipal fire services with potential assistance from the governmental Danish Emergency Management Agency (DEMA). In addition, specific hazards require specialized fire services. Thus, airports and air bases have an aircraft rescue and firefighting (ARFF) response operated by either a civilian airport fire service or the Royal Danish Air Force (RDAF). Similarly, the Royal Danish Navy (RDN) contributes to the management of maritime rescue and firefighting. DEMA, RDAF and RDN are all a part of the Armed Forces in Denmark.

The study population consisted of current and former employees and volunteers from 27 municipal fire stations, three governmental emergency management centers, two civilian airport fire stations, three air force fire stations and two naval stations representing all regions of Denmark. Initially, we collected records on all staff with exposures related to firefighting from employers. The number of women affiliated with the selected workplaces was too limited for meaningful statistical analyses of occupational exposure to PFAS, and they were, thus, excluded. Men with a minimum age of 18 years and at least one employment or volunteer affiliation with a municipal, governmental, or specialized fire service or response during the years 2000 through 2024 were eligible for participation. We identified 1735 eligible male employees and volunteers from staff records with 21 records representing firefighters with additional affiliations at the selected workplaces. To ensure adequate representation of occupational exposure periods and durations, we selected 1535 men to be invited based on their period of employment. Between September 2023 and January 2024, each of these men were contacted through a secure digital mailbox system, e-Boks, held by all residents in Denmark. They were invited for at least one study information meeting at their current or former workplace (Supplementary Fig. S1). Among the 434 men attending an information meeting, five men were not enrolled in the study due to either inability to have a blood sample collected, lack of relevant exposure during the required time interval, or simply declining participation. All of the 429 participating men (participation rate 28%) received thorough oral and written information about the study prior to their enrollment. Each participant was required to give a blood sample and fill in an electronic questionnaire. We forwarded monthly reminders to participants failing to respond to the questionnaire. Despite these reminders, 40 participants (9%) provided no questionnaire data. A full overview of the recruitment process is shown in Fig. 1.

2.2. Blood sample collection and PFAS analyses

A VACUETTE® Safety blood collection set with holder (Greiner-Bio-One Gmbh, Kremsmünster, Austria) was used to draw blood from an antecubital vein. Participants were non-fasting. Following 60 min of clotting time at room temperature, samples were centrifuged with subsequent separation of serum. Serum was stored in CryoPure® tubes (Sarstedt, Nümbrecht, Germany) at −80 °C until analysis.

Based on previous studies of occupational exposure among firefighters, we prioritized quantification of short- and long-chained perfluoroalkyl acids (six sulfonic acids and nine carboxylic acids)(Burgess et al., 2022; Dobraca et al., 2015; Goodrich et al., 2021; Graber et al., 2021; Jin et al., 2011; Khalil et al., 2020; Laitinen et al., 2014; Leary et al., 2020; Tefera et al., 2023; Nilsson et al., 2022a; Rihackova et al., 2023; Rotander et al., 2015; Tao et al., 2008; Trowbridge et al., 2020; Purdue et al., 2023; Nematollahi et al., 2023). The 15 PFAS selected for quantification were perfluorobutane sulfonic acid (PFBS), perfluoropentanoic acid (PFPeA), perfluoropentane sulfonic acid (PFPeS), perfluorohexanoic acid (PFHxA), PFHxS, perfluoroheptanoic acid (PFHpA), perfluoroheptane sulfonic acid (PFHpS), PFOA, PFOS, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluorodecane sulfonic acid (PFDS), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), and perfluorotridecanoic acid (PFTrDA). Total PFOS was calculated as the sum of branched isomers plus the linear isomer (nPFOS). A serum sample of 150 μL was prepared adding 20 μL of an internal standard solution containing 20 ng/mL carbon-13-labelled PFAS analogues (MPFAC-24ES from Wellington Laboratories, Guelph, Canada) and 120 μL methanol in a polypropylene tube. Subsequently, the sample was whirl mixed and centrifuged at 21,

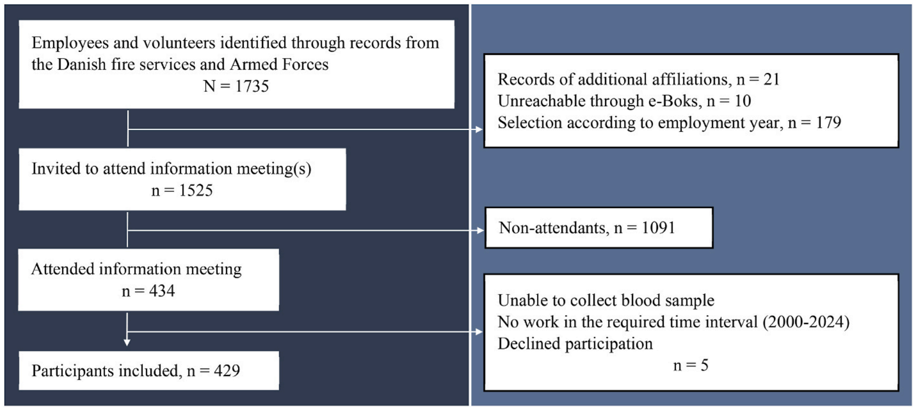

Fig. 1. Flowchart of the recruitment of participants from the Danish fire services and Armed Forces; 2023–24.

000 g for 20 min. A volume of 160 µL supernatant was transferred to a new polypropylene tube and added 400 µL of formic acid 0.1 M. The sample was whirl mixed again and 400 µL was injected onto an online solid-phase extraction (SPE) column on a high-pressure liquid chromatography and triple quadrupole mass spectrometry (LC-MS/MS) system (Haug et al., 2009; Nielsen et al., 2024). The LC-MS/MS system consisted of an EQuan MAX ultra-high-pressure liquid chromatograph (UHPLC) connected to a TSQ Quantiva triple quadrupole mass spectrometer using Xcalibur v. 4.5 software (Thermo Fischer Scientific, San Jose, CA, USA). Serum samples were analyzed along with calibrators, solvent blanks and quality control (QC) samples (with low and high concentrations of PFAS). QC samples included serum from a previous Human Biomonitoring for Europe (HBM4EU) quality assessment program and in-house made samples. All QC samples were well within acceptable ranges and coefficients of variation (CV) ranged between 2.8% and 11.7% (Supplementary Table S1). The limits of detection (LODs) ranged from 0.03 to 0.1 ng/mL (Supplementary Table S1). Values below the LOD were assigned the value of the LOD divided by the square root of two. All analyses were performed at the Department of Clinical Pharmacology, Pharmacy and Environmental Medicine, University of Southern Denmark. Accuracy and reliability of PFAS analyses from the laboratory were ensured by regular participation in the German External Quality Assessment Program (G-EQUAS) organized by the German Society of Occupational Medicine (The German external quality assessment, 2024).

2.3. Questionnaire and covariates

A questionnaire was developed based on the existing literature on exposure to PFAS among firefighters, knowledge of specific working conditions in Denmark, and additional factors potentially contributing to either exposure to or excretion of PFAS (Burgess et al., 2022; Dobraca et al., 2015; Goodrich et al., 2021; Graber et al., 2021; Jin et al., 2011; Khalil et al., 2020; Laitinen et al., 2014; Leary et al., 2020; Tefera et al., 2023; Nilsson et al., 2020, 2022a; Rihackova et al., 2023; Rosenfeld et al., 2022; Rotander et al., 2015; Shaw et al., 2013; Tao et al., 2008; Trowbridge et al., 2020). Information on occupational history, work environment, educational level, age, height, weight, ethnicity, blood donation, health, diet and health behavior was collected in the questionnaire. The questionnaire was distributed using the secure browser-based tool, Research Electronic Data Capture (REDCap) under the Capital Region of Denmark (Harris et al., 2009).

Using information from the questionnaire, the following covariates regarding health and health behaviour were constructed: body mass index (BMI, weight in kg/(height in m)^2, continuous), intake of meat (≥4 days per week; *yes, no*), eggs (≥4 days per week; *yes, no*), dairy products (≥4 days per week; *yes, no*), fruit (≥4 days per week; *yes, no*), fish and shellfish (weekly; *yes, no*), takeaway (weekly; *yes, no*) and tea drinking (weekly; *yes, no*), total blood donations (continuous), and comorbidity (*yes, no*). Covariates related to occupational exposure included total foam usage (calculated as years of foam use multiplied by the frequency of foam use and categorized in tertiles as *low, medium and high*), dermal foam exposure during training and incidents (*never/rarely, regularly, most times/always*), function as instructor (*yes, no*), storage management (*yes, no*), and emergency vehicle technician or mechanic (*yes, no*), and civilian non-firefighting jobs (categorized according to db07 sector).

2.4. Reference measurements

As a reflection of PFAS serum concentrations in the general population in Denmark, we applied measures from samples collected in 2021 through the Danish national cohort study of effectiveness and safety of SARS-CoV-2 vaccines (ENFORCE) (Staerke et al., 2022). ENFORCE included a total of 6943 participants from all five administrative regions of Denmark (Staerke et al., 2022). We sampled serum from 496 men (median age 64 years, 5th percentile 43 years and 95th percentile 82 years) receiving the mRNA-1273 vaccine. Serum was collected before the first vaccination and analyzed for the same PFAS as in our current study using liquid chromatography triple quadrupole linear ion trap mass spectrometry (LC/MS/MS, QTRAP 5500, AB Sciex, Framingham, MA, USA) (Supplementary Table S2) (Petersen et al., 2022). Analyses were performed at the Division of Occupational and Environmental Medicine, Lund University, Sweden. The laboratory participated in the Erlangen Round Robin inter-laboratory control program and qualified as a European Human Biomonitoring Initiative (HBM4EU) laboratory for analyses of PFAS.

2.5. Statistical analyses

All participants were assigned a primary occupational exposure group (*municipal, governmental, airport, air force and navy*) and employment type (*full-time, part-time or volunteer*) based on their longest held employments or volunteer affiliations. The most recent employment or affiliation was chosen, when durations of time spent were equal between categories. Initially, we examined the distribution of covariates and serum concentrations of PFAS among the men according to their primary occupational exposure groups and employment types. We decided *a priori* to include only PFAS with measured concentrations above the LOD in samples from at least 40% of the participants in analyses (Supplementary Table S1). Correlations between serum concentrations of PFHxS, PFHpS, PFOA, PFOS, PFNA, PFDA and PFUnDA were analyzed using Spearman's ρ.

We calculated age-adjusted geometric means with 95% confidence intervals for serum concentrations of PFAS among male participants from the ENFORCE study and the participants from the fire services and

Armed Forces according to their primary occupational exposure group and employment types. In a separate analysis, age-adjusted geometric means of serum PFAS were calculated exclusively for participants commencing service from 2011 and onwards, when PFOS was banned for use in firefighting foam in Denmark (NIRAS, 2021).

Subsequently, we examined potential associations between primary occupational exposure group and employment type and serum concentrations of PFAS using multiple linear regression analyses. The following covariates were included in the main analyses:

- Age was calculated as the interval from date of birth to date of participation and included as a continuous covariate.
- Years of service was counted for all non-overlapping employments with the fire services and Armed Forces and categorized based on 10-year intervals.
- The longest completed education was categorized according to the Danish version of the International Classification of Education, DISCED-15 (v1: 2024) with three main aggregates (*short, medium and long*) (Statistics Denmark, 2024; Eurostat - Statistics Explained, 2023).
- The total number of blood donations was calculated based on information on years and frequency of donating and included as a continuous covariate.

These covariates were selected *a priori* based on the existing literature (Graber et al., 2021; Rihackova et al., 2023; Rotander et al., 2015; Richterova et al., 2023). Bivariate correlations (Spearman's $\rho < 0.8$) and variance inflation factors (VIF) were assessed to limit potential issues with multicollinearity in analyses. We also tested for interactions between covariates. PFAS concentrations were skewed in their distributions and a natural log transformation was applied to secure adequate model fit. Results were back-transformed to ease interpretation. The adjusted analyses included only the 389 men providing questionnaire information.

An expanded multiple linear regression model was used to examine the importance of additional factors both within and outside of the working environment. We adjusted for the same covariates as in the main model with the addition of BMI, intake of meat, eggs, dairy products, fruit, fish and shellfish, takeaway and tea drinking, comorbidity, total foam usage, dermal foam exposure, function as instructor, storage management, and emergency vehicle technician or mechanic, and jobs outside the fire services and Armed forces. Finally, we repeated our main analyses with added adjustment for the most influential factors according to the expanded model.

To ensure adequate anonymity and compliance with national data protection regulations, all estimates were based on information from at least five individuals. Statistical analyses were performed using Stata V. 14 (StataCorp, College Station, TX, USA).

2.6. Ethics

The study was conducted in accordance with the principles of the

Table 1
Characteristics of the 429 men from the Danish fire services and Armed Forces, 2023–2024.

		Primary occupational exposure group				
	Total	Municipal	Governmental	Airport	Air force	Navy
Participants, n	429	208	59	50	98	14
Age (years), Mdn ($P_{5\%}$, $P_{95\%}$)	50 (26, 67)	50 (27, 64)	38 (22, 62)	52 (38, 67)	56 (28, 72)	49 (35, 58)
Education						
Short, n (%)	–	22 (12)	≤5	9 (20)	31 (35)	–
Medium, n (%)	–	125 (66)	28 (53)	29 (66)	44 (49)	–
Long, n (%)	–	42 (22)	22 (42)	6 (14)	14 (16)	–
Body Mass Index, Mdn ($P_{5\%}$, $P_{95\%}$)	26.8 (22.6, 33.6)	26.6 (22.8, 32.8)	27.4 (21.3, 33.7)	26.2 (22.7, 36.4)	27.3 (22.2, 33.7)	26.1 (24.3, 31.2)
Current tobacco use[a], n (%)	73 (19)	38 (21)	11 (21)	5 (12)	14 (16)	5 (36)
Diet[b]						
Meat, n (%)	–	144 (79)	39 (75)	34 (81)	65 (74)	–
Eggs, n (%)	–	48 (26)	13 (25)	16 (38)	24 (27)	–
Dairy products, n (%)	–	126 (69)	32 (62)	29 (69)	70 (79)	–
Fruit, n (%)	–	86 (47)	22 (44)	20 (48)	50 (56)	–
Fish and shellfish, n (%)	–	115 (64)	31 (62)	31 (78)	61 (69)	–
Tea drinking, n (%)	–	53 (30)	19 (38)	11 (28)	35 (40)	–
Takeaway, n (%)	–	75 (41)	26 (50)	16 (38)	24 (27)	–
Alcohol units per week, Mdn ($P_{5\%}$, $P_{95\%}$)	3 (0, 12)	3 (0, 13)	3 (0, 13)	4 (0, 12)	3 (0, 11)	2 (0, 6)
Vitamin and/or omega-3 oil supplement, n (%)	203 (52)	98 (52)	23 (43)	22 (50)	54 (61)	6 (43)
Exercise (weekly hours)[c], Mdn ($P_{5\%}$, $P_{95\%}$)	5 (1, 15)	5 (1, 17)	3 (0, 19)	5 (1, 12)	4 (1, 15)	7 (3, 12)
Blood donation ever, n (%)	152 (40)	77 (42)	13 (25)	17 (40)	38 (43)	7 (50)
Cholesterol-lowering medicine, n (%)	44 (11)	19 (10)	≤5	5 (10)	16 (16)	≤5
Comorbidity[d], n (%)	–	26 (14)	6 (11)	8 (18)	16 (18)	≤5
Occupational information						
Primary employment type (full-time), n (%)	348 (81)	143 (69)	43 (73)	50 (100)	98 (100)	14 (100)
Year of commencing service, Mdn ($P_{5\%}$, $P_{95\%}$)	1999 (1979, 2020)	1999 (1983, 2019)	2009 (1984, 2023)	1998 (1979, 2021)	1992 (1972, 2020)	1996 (1986, 2012)
Years of service, Mdn ($P_{5\%}$, $P_{95\%}$)	23 (3, 41)	23 (4, 40)	14 (1, 39)	24 (2, 42)	30 (2, 43)	27 (10, 37)
Years of foam usage, Mdn ($P_{5\%}$, $P_{95\%}$)	21 (2, 40)	23 (3, 39)	10 (1, 31)	21 (6, 37)	26 (4, 43)	11 (5, 17)
Functions:						
Instructor, n (%)	–	62 (30)	40 (68)	28 (56)	34 (35)	≥10
Storage management, n (%)	27 (7)	8 (4)	10 (19)	≤5	6 (7)	≤5
Emergency vehicle technician or mechanic, n (%)	31 (8)	14 (8)	6 (12)	≤5	6 (7)	≤5
Other job(s)[e], n (%)	182 (47)	95 (50)	29 (55)	10 (23)	43 (48)	5 (36)

Mdn, Median. Medians and other percentiles are displayed as pseudo-percentiles based on five adjacent values.
[a] Includes cigarette, pipe and e-cigarette smoking and snuff use.
[b] For meat, eggs, dairy products and fruit, consumption is daily or almost daily (≥4 days per week) while intake of fish, tea and takeaway is weekly.
[c] Includes moderate and vigorous physical activity.
[d] Includes cancer, diabetes, inflammatory bowel disease, renal and liver disease, acute myocardial infarction and stroke.
[e] Includes jobs outside the fire services and Armed Forces from 2000 to 2024 categorized according to db07 sector.

Declaration of Helsinki. Study approval was obtained from the Capital Region's Committee on Health Research Ethics (H-23027326) on June 8, 2023. Further, the study was registered by the Knowledge Center on Data Protection Compliance under the records of processing regarding health science research projects within the Capital Region of Denmark (p-2023-14170) in accordance with regulations from the Danish Data Protection Agency. The privacy rights of all participants were observed and written informed consent was given prior to their participation in the study.

3. Results

A full overview of characteristics for the 429 participants in the study is shown in Table 1. The median age of participants was 50 years. Participants from the governmental fire services had the lowest median age and the highest percentage with long educations. Conversely, participants from the air force had the highest median age and the highest percentage with short educations. Median BMI was above the normal range in all groups. Dietary intake varied among the primary occupational exposure groups. Thus, a higher percentage of participants from airport fire services had a frequent intake of eggs, fish and shellfish. Notably, 40% of all participants had donated blood at some point.

Participants from airport fire services, the air force and the navy were exclusively full-time employees. For the 267 men from the municipal and governmental fire services, characteristics according to primary employment type is shown in Supplementary Table S3.

Correlations in serum concentrations of PFAS are shown in Supplementary Table S4. Concentrations of PFHxS, PFHpS and PFOS were strongly or very strongly correlated. Concentrations of PFAS with the longest carbon chains (PFOS, PFNA, PFDA and PFUnDA) were also strongly or very strongly correlated. Concentrations of PFOA were, however, not strongly correlated with the other PFAS.

The median serum concentrations of PFHxS, PFHpS and total PFOS (1.49, 0.28 and 6.46 ng/mL, respectively) were higher among participants from airport fire services compared to participants from other primary occupational exposure groups (Table 2). Age-adjusted geometric means for serum concentrations of PFHxS, PFHpS and total PFOS (1.42, 0.28 and 6.92 ng/mL) were also higher among airport fire service participants in comparison to reference measurements from ENFORCE (0.72, 0.14 and 5.86 ng/mL for PFHxS, PFHpS and total PFOS, respectively) (Fig. 2 and Table 3). In analyses restricted to participants commencing service from 2011 and onwards, the age-adjusted geometric means for serum PFAS were, however, closer to even among the primary occupational exposure groups (Supplementary Table S5). In analyses according to primary employment type, the medians and age-adjusted geometric means were lowest among the volunteers, intermediary among the part-time employees and highest among the full-time employees for almost all the measured PFAS (Supplementary Tables S6 and S7).

In our main regression analysis, serving in civilian airport fire services was associated with higher serum concentrations of PFHxS, PFHpS and PFOS (59.6%, 32.6% and 14.5% difference in adjusted analyses) compared to participants from municipal fire services (Table 4). Concentrations of PFNA and PFUnDA were also slightly higher among the civilian airport fire service participants. Conversely, serving in the air force was associated with lower serum concentrations of PFHxS, PFOS and PFNA (−16.3%, −16.2% and −12.8% difference in adjusted analyses). Finally, service in the navy seemed to be associated with a positive difference in PFDA and PFUnDA (15.5% and 35.0% difference in adjusted analyses).

In Table 5, our regression analysis according to primary employment type is shown. Compared to volunteers, part-time service was associated with a positive trend difference in serum concentrations of almost all the measured PFAS with differences appearing more pronounced for full-time service.

In the expanded regression model, primary occupational exposure group and employment type, age, blood donations, body mass index and intake of eggs were the most influential factors in relation to serum PFAS. Results for PFHxS, total PFOS and PFOA are shown in Supplementary Table S8. However, adding adjustment for body mass index and intake of eggs to our regression analyses did not change results substantially (Supplementary Table S9).

4. Discussion

In this cross-sectional study of serum PFAS among men from the Danish fire services and Armed Forces, findings signified occupational exposure of civilian airport firefighters. While serum concentrations of PFAS among most participants were at level with recent age-adjusted measurements from the general population in Denmark, civilian airport firefighters had slightly higher concentrations of especially PFHxS, PFHpS and PFOS in serum (Hull et al., 2023). Regression analyses also confirmed positive associations for these PFAS among civilian airport firefighters in comparison to municipal firefighters.

Airport fire services specialize in mitigation of aviation emergencies with requirements for response times and equipment dictated by a risk of mass casualties. Fires may involve large quantities of aviation fuels and similar highly flammable liquids prompting widespread use of AFFF in relation to both civilian and military airports in the past (Nilsson et al., 2020; Miljøstyrelsen, 2016; Xu et al., 2020; Forsvarsministeriet, 2021). Analyses of AFFF used in airports and resulting contaminations have documented the presence of a range of non-polymeric PFAS in previous generations of firefighting foams with a predominance in detection of PFOS, PFOA and PFHxS (NIRAS, 2021; Miljøstyrelsen, 2016; Interstate Technology Regulatory Council, 2023). In Denmark, foams containing PFOS were banned in 2006 with a transition period

Table 2
PFAS serum concentrations (ng/mL) among the 429 men from the Danish fire services and Armed Forces, 2023–2024.

		Primary occupational exposure group				
	Total	Municipal	Governmental	Airport	Air force	Navy
	(n = 429)	(n = 208)	(n = 59)	(n = 50)	(n = 98)	(n = 14)
PFAS	Mdn ($P_{5\%}$, $P_{95\%}$)	Mdn ($P_{5\%}$, $P_{95\%}$)	Mdn ($P_{5\%}$, $P_{95\%}$)	Mdn ($P_{5\%}$, $P_{95\%}$)	Mdn ($P_{5\%}$, $P_{95\%}$)	Mdn ($P_{5\%}$, $P_{95\%}$)
PFHxS	0.76 (0.36, 2.03)	0.74 (0.37, 1.70)	0.66 (0.27, 1.43)	1.49 (0.49, 3.99)	0.74 (0.36, 1.41)	0.81 (0.48, 1.38)
PFHpS	0.19 (0.07, 0.41)	0.18 (0.06, 0.39)	0.17 (0.06, 0.39)	0.28 (0.08, 0.68)	0.19 (0.08, 0.37)	0.19 (0.11, 0.35)
PFOA	1.02 (0.44, 1.91)	1.03 (0.45, 2.11)	0.89 (0.39, 1.91)	1.11 (0.41, 1.92)	0.95 (0.46, 1.81)	0.89 (0.47, 1.82)
nPFOS	3.96 (1.41, 10.19)	4.00 (1.36, 10.03)	3.32 (1.34, 9.99)	5.12 (1.80, 14.70)	3.91 (1.61, 8.75)	4.48 (2.20, 8.00)
Total PFOS	5.09 (1.91, 12.36)	5.08 (1.77, 12.42)	4.41 (1.67, 11.92)	6.46 (2.35, 18.31)	5.17 (2.09, 11.22)	5.68 (2.76, 9.91)
PFNA	0.43 (0.20, 0.90)	0.45 (0.18, 0.96)	0.39 (0.19, 1.03)	0.54 (0.23, 0.95)	0.41 (0.23, 0.77)	0.48 (0.28, 0.86)
PFDA	0.15 (0.08, 0.34)	0.16 (0.07, 0.36)	0.13 (0.07, 0.34)	0.16 (0.07, 0.31)	0.15 (0.09, 0.29)	0.18 (0.12, 0.33)
PFUnDA	0.08 (0.02, 0.20)	0.08 (0.01, 0.21)	0.07 (0.01, 0.25)	0.09 (0.03, 0.22)	0.08 (0.03, 0.17)	0.10 (0.06, 0.18)

PFAS, per- and polyfluoralkyl substances; Mdn, Median, nPFOS, linear PFOS.
Medians and other percentiles are displayed as pseudo percentiles based on five adjacent values.

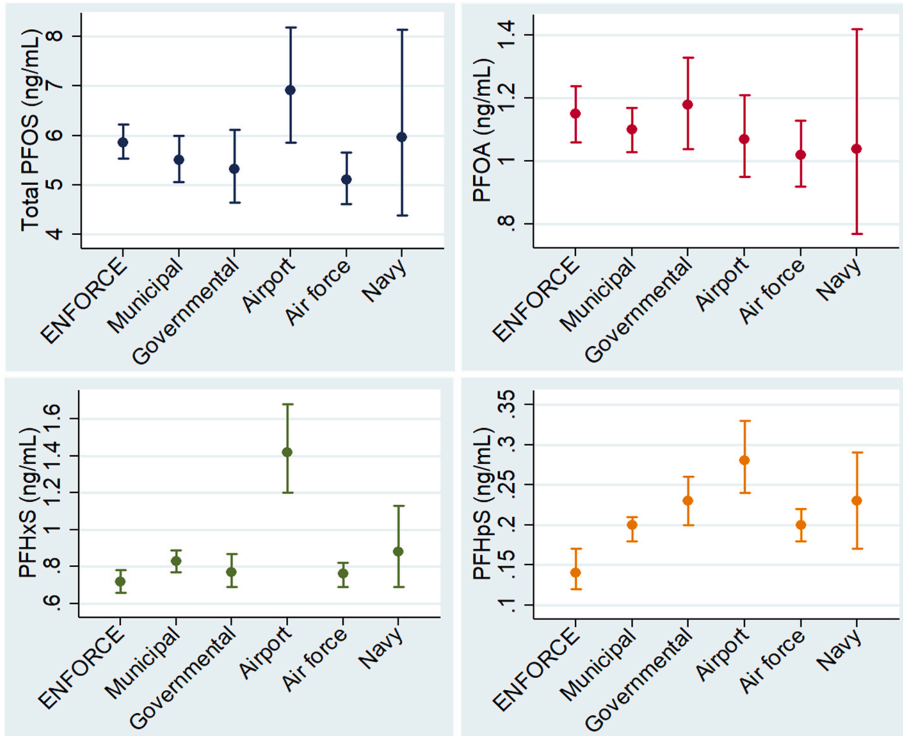

Fig. 2. Age-adjusted geometric means for serum concentrations (ng/mL) of total PFOS, PFOA, PFHxS and PFHpS among men from the Danish fire services and Armed Forces (n = 429, 2023–2024) compared to national measurements from the ENFORCE study (n = 496 men, 2021).

Table 3
Age-adjusted geometric means for PFAS serum concentrations (ng/mL) among men (n = 429, 2023–2024) from the Danish fire services and Armed Forces compared to national measurements from the ENFORCE study (n = 496 men, 2021).

		Primary occupational exposure group				
		Municipal	Governmental	Airport	Air force	Navy
	ENFORCE	(n = 208)	(n = 59)	(n = 50)	(n = 98)	(n = 14)
PFAS	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)	GM (95% CI)
PFHxS	0.72 (0.66, 0.78)	0.83 (0.77, 0.89)	0.77 (0.69, 0.87)	1.42 (1.20, 1.68)	0.76 (0.69, 0.82)	0.88 (0.69, 1.13)
PFHpS	0.14 (0.12, 0.17)	0.20 (0.18, 0.21)	0.23 (0.20, 0.26)	0.28 (0.24, 0.33)	0.20 (0.18, 0.22)	0.23 (0.17, 0.29)
PFOA	1.15 (1.06, 1.24)	1.10 (1.03, 1.17)	1.18 (1.04, 1.33)	1.07 (0.95, 1.21)	1.02 (0.92, 1.13)	1.04 (0.77, 1.42)
Total PFOS	5.86 (5.53, 6.22)	5.51 (5.05, 6.00)	5.32 (4.64, 6.11)	6.92 (5.85, 8.19)	5.11 (4.62, 5.66)	5.97 (4.38, 8.14)
PFNA	0.55 (0.50, 0.60)	0.48 (0.44, 0.51)	0.48 (0.42, 0.54)	0.53 (0.47, 0.59)	0.44 (0.40, 0.47)	0.54 (0.41, 0.71)
PFDA	0.19 (0.16, 0.22)	0.17 (0.16, 0.18)	0.16 (0.14, 0.19)	0.16 (0.14, 0.18)	0.16 (0.15, 0.17)	0.20 (0.16, 0.26)
PFUnDA	0.11 (0.09, 0.13)	0.08 (0.07, 0.11)	0.08 (0.06, 0.12)	0.09 (0.07, 0.11)	0.08 (0.08, 0.09)	0.11 (0.08, 0.16)

PFAS, per- and polyfluoralkyl substances; GM, geometric mean; CI, confidence interval.

allowing for stockpiled use until 2011 (NIRAS, 2021). In the last decades, production of AFFF has been adjusted favoring the use of PFHxS, short-chain perfluoroalkyl acids and fluorotelomers with increasing competition from PFAS free alternatives (NIRAS, 2021; Interstate Technology Regulatory Council, 2023). In a large recent study of airport firefighters (n = 799) from 27 airports across Australia, serum concentrations of PFHxS, PFHpS and PFOS were elevated among participants commencing service prior to 2005 (Nilsson et al., 2020, 2022a, 2022b). At this point, use of Lightwater AFFF from 3M had been fully replaced by a fluorotelomer based solution (Nilsson et al., 2022a). Median concentrations were 14 ng/mL for PFOS, 6.5 ng/mL for PFHxS and 0.85 ng/mL for PFHpS for serum sampled in 2018–2019 (Nilsson et al., 2022a). In an earlier and smaller study among a subset of participants (n = 149, 2013), medians were 66 ng/mL for PFOS and 25 ng/mL for PFHxS with no measurement of PFHpS (Rotander et al., 2015). While our more recent measurements may appear less striking, peak concentrations among our civilian airport firefighters may also have been an order of magnitude higher during their actual use of AFFF. In our study, the civilian airport firefighters were almost exclusively from a single metropolitan airport ceasing their use of AFFF from 3M in 2008. Since then, PFAS free foam (Solberg Re-Healing RF3x6 foam) has been used. In our analyses restricted to participants commencing service from 2011 and onwards, civilian airport firefighters no longer had markedly higher serum PFAS. Though contaminated soil and surface water in relation to training grounds and fire stations may have contributed some exposure in the years trailing the end of the AFFF era, our findings do not indicate substantial ongoing occupational exposure to PFAS among airport firefighters (Koordinerende arbejdsgruppe ved Dragør Kommune TKoKLAS, 2024). Regarding other potential sources of PFAS, the turnout gear applied was coated with PFOA until manufacturers in Denmark gradually transitioned to treating textiles with PFHxA instead from 2020 to 2023 (Kruse, 2021). We found no indications of higher exposure to either of these compounds.

In a smaller American study (n = 47) from Ohio from 2019, civilian airport firefighters also had higher concentrations of PFHxS and PFOS in serum compared to both suburban firefighters and measurements from

Table 4
Multiple linear regression of associations between primary occupational exposure group and PFAS serum concentrations (ng/mL) among 429 men from the Danish fire services and Armed Forces, 2023–2024.

	Primary occupational exposure group				
	Municipal	Governmental	Airport	Air force	Navy
PFAS		% diff (95% CI)	% diff (95% CI)	% diff (95% CI)	% diff (95% CI)
PFHxS					
Crude	ref	**−15.7 (-26.9, -2.8)**	**76.8 (51.9, 105.8)**	−5.9 (−16.3, 6.0)	3.6 (−20.6, 35.2)
Adjusted[a]	ref	−12.4 (−24.5, 1.7)	**59.6 (36.6, 86.5)**	**−16.3 (-26.0, -5.3)**	−3.0 (−24.1, 24.0)
PFHpS					
Crude	ref	−2.2 (−17.0, 15.2)	**50.7 (26.5, 79.5)**	7.8 (−6.0, 23.5)	10.1 (−19.0, 49.7)
Adjusted	ref	13.4 (−3.8, 33.6)	**32.6 (11.8, 57.4)**	−4.7 (−16.8, 9.2)	9.9 (−16.1, 43.9)
PFOA					
Crude	ref	−6.1 (−18.1, 7.6)	2.3 (−11.6, 18.4)	−2.7 (−13.1, 9.0)	−8.5 (−29.1, 18.2)
Adjusted	ref	−2.8 (−16.5, 13.2)	−5.2 (−19.2, 11.1)	−9.9 (−20.6, 2.3)	−10.6 (−30.4, 14.9)
nPFOS					
Crude	ref	−15.5 (−28.5, 0.0)	**33.5 (11.7, 59.7)**	−4.6 (−17.0, 9.6)	6.6 (−22.1, 45.8)
Adjusted	ref	−7.7 (−23.1, 10.7)	17.8 (−2.6, 42.5)	**−17.1 (-28.7, -3.6)**	1.0 (−25.1, 36.3)
Total PFOS					
Crude	ref	−15.1 (−28.1, 0.4)	**31.4 (10.0, 57.0)**	−3.0 (−15.5, 11.5)	4.7 (−23.4, 43.1)
Adjusted	ref	−6.3 (−21.6, 12.1)	14.5 (−5.0, 38.0)	**−16.2 (-27.7, -2.8)**	−0.2 (−25.7, 33.9)
PFNA					
Crude	ref	−10.7 (−21.8, 1.9)	**15.4 (0.2, 32.9)**	−5.5 (−15.4, 5.5)	9.7 (−14.4, 40.5)
Adjusted	ref	−4.7 (−17.5, 10.2)	8.8 (−6.5, 26.6)	**−12.8 (-22.7, -1.7)**	4.9 (−17.3, 33.2)
PFDA					
Crude	ref	−11.3 (−22.0, 0.9)	−3.4 (−15.8, 10.9)	−4.6 (−14.3, 6.1)	17.7 (−7.6, 49.8)
Adjusted	ref	−9.1 (−21.4, 5.1)	−5.7 (−18.9, 9.8)	−9.4 (−19.6, 2.2)	15.5 (−9.0, 46.7)
PFUnDA					
Crude	ref	−12.0 (−28.4, 8.1)	10.6 (−11.3, 37.9)	3.4 (−12.8, 22.8)	31.6 (−10.5, 93.7)
Adjusted	ref	−0.3 (−20.6, 25.2)	**14.6 (9.7, 45.3)**	−1.9 (−18.7, 18.6)	35.0 (−7.2, 96.4)

PFAS, per- and polyfluoralkyl substances; diff, difference; CI, confidence interval; nPFOS, linear PFOS.
Statistically significant associations marked in bold.
[a] Adjusted for age, years of service, education, blood donations and primary employment type. Participants providing no questionnaire information were not included in the adjusted analyses (n = 40).

the US general population (NHANES data)(Leary et al., 2020). Further, a study from Finland documented changes in especially PFHxS and PFNA in serum among firefighters (n = 8) from the Oulu Airport during a three-session ARFF training programme using AFFF in 2010 (Laitinen et al., 2014).

Table 5
Multiple linear regression of associations between primary employment type and PFAS serum concentrations (ng/mL) among men (n = 267) from the municipal and governmental fire services in Denmark, 2023–2024.

	Primary employment type		
	Volunteer (n = 24)	**Part-time** (n = 57)	**Full-time** (n = 186)
PFAS		% diff (95% CI)	% diff (95% CI)
PFHxS			
Crude	ref	20.7 (−3.7, 51.3)	**43.3 (17.2, 75.2)**
Adjusted[a]	ref	13.0 (−9.6, 41.1)	**39.0 (14.9, 68.2)**
PFHpS			
Crude	ref	20.1 (−8.6, 57.9)	**32.7 (4.0, 69.3)**
Adjusted	ref	8.4 (−16.0, 39.9)	**25.6 (1.0, 56.2)**
PFOA			
Crude	ref	6.4 (−14.6, 32.7)	19.5 (−1.8, 45.5)
Adjusted	ref	−2.4 (−22.3, 22.6)	14.3 (−6.0, 38.9)
nPFOS			
Crude	ref	24.3 (−5.6, 63.8)	**57.7 (23.4, 101.6)**
Adjusted	ref	16.2 (−13.2, 55.6)	**51.2 (17.8, 94.0)**
Total PFOS			
Crude	ref	24.6 (−5.1, 63.5)	**55.6 (22.1, 98.3)**
Adjusted	ref	14.7 (−13.4, 52.1)	**48.5 (16.7, 88.9)**
PFNA			
Crude	ref	20.4 (−3.9, 50.8)	**44.6 (18.2, 76.7)**
Adjusted	ref	23.1 (−2.9, 56.2)	**42.4 (16.2, 74.5)**
PFDA			
Crude	ref	14.2 (−8.2, 43.0)	**27.5 (4.3, 55.8)**
Adjusted	ref	17.9 (−7.4, 50.3)	**25.1 (1.7, 54.0)**
PFUnDA			
Crude	ref	**57.1 (7.8, 129.0)**	**50.4 (7.5, 110.5)**
Adjusted	ref	**63.1 (9.1, 143.8)**	**53.1 (8.5, 115.9)**

PFAS, per- and polyfluoralkyl substances; diff, difference; CI, confidence interval; nPFOS, linear PFOS.
Statistically significant associations marked in bold.
[a] Adjusted for age, years of service, education and blood donations. Participants providing no questionnaire information were not included in the adjusted analyses (n = 25).

We found no evidence of higher serum PFAS among participants from the air force. If anything, serving in the air force was associated with lower serum concentrations of PFHxS, PFOS and PFNA. At present, the RDAF uses PFAS free foam (Solberg Re-Healing RF3x6 foam) (Forsvarsministeriet, 2021). AFFF containing PFOS was, however, used on the firetrucks at their air bases until 2014 with storage continuing as late as 2022 (Forsvarsministeriet, 2021; Rigsrevisionen, 2023). In a previous study of men (n = 1060) from the United States Air Force, employment in fire protection was the strongest service-related predictor of elevated serum PFHxS, PFOA and PFOS (Purdue et al., 2023). The number of men (n = 5) serving in fire protection was, however, rather limited (Purdue et al., 2023).

Airports are categorized in order to dimension their ARFF responses based on characteristics such as daily departure and landing frequencies, maximum aircraft lengths and fuselage widths, and all-cargo operations (International Civil Aviation Organization, 2014). Airport categories determine requirements for emergency vehicles and types and amounts of extinguishing agents used (International Civil Aviation Organization, 2014). Thus, differences in airport sizes and categories may explain some of the differences in exposure to PFAS observed between civilian and air force firefighters in this study. Job assignments may also differ among civilians and military employees.

Participants serving in the navy were relatively few in our study and their observed slightly positive associations for serum PFDA and PFUnDA were most likely random findings. The concentrations of these PFAS were quite low with narrow ranges among all groups.

Among participants from the municipal and governmental fire services, differences in serum PFAS according to primary employment type can be interpreted as indication of an occupational exposure gradient. There are, however, distinct differences in socioeconomic status, geographical distribution, health behaviour and disease incidence among these groups and residual confounding may explain at least some of the gradient (Petersen et al., 2018; Petersen, 2018). Serum PFAS among the full-time employees was at level with reference measurements from the ENFORCE study. Ultimately, environmental exposure to PFAS is extremely common with diet as the main source (European Food Safety Authority, 2020).

The main strength of the study was the inclusion of participants from different occupational firefighting groups with different employment types, durations and times. Further, workplaces in all five regions of Denmark were represented in the study. Finally, participants provided extensive information on potential sources of PFAS exposure both within and beyond the working environment.

While recruitment efforts were extensive and flexible with numerous information meetings both during and after working hours at workplaces across the country, the overall participation rate (28%) was somewhat lower than expected. Firefighters assumed to be more exposed following intense use of foam may have been more likely to participate causing a potential bias from selection. Further, capturing the full occupational history of firefighters may be quite complicated with many potentially overlapping employments. Our approach of assigning a primary occupational exposure group and employment type was rather crude. Thus, 58% of participants belonged to more than one of the occupational exposure groups and 37% of participants had served with more than one employment type. Potential misclassification of occupational exposure was equal across all the groups in the study and, therefore, non-differential.

Our targeted analyses of PFAS were highly sensitive. Considering the vast number of PFAS, the targeted approach was, however, inexhaustive. No fluorotelomers were measured. Fluorotelomers may, however, act as precursors and ultimately degrade into the measured perfluoroalkyl acids (e.g. 6:2-fluorotelomer sulfonic acid can degrade into PFHxA, PFPeA and PFBA, while 8:2-fluorotelomer sulfonic acid can degrade into PFOA) (NIRAS, 2021). We also prioritized measurements of both short- and long-chained PFAS. Although the toxicokinetic documentation for short-chained PFAS remains limited, these compounds appear to have much shorter half-lives for excretion (44 days for PFBS, 230 days for PFPeS, 32 days for PFHxA, and 62 days for PFHpA) and their measured concentrations or their lack of such, therefore, reflect only more recent exposures (The Danish Environmental Protection Agency et al., 2015; Xu et al., 2020; U.S. Environmental Protection Agency, 2023). Ultimately, assessment of occupational exposure to PFAS remains challenging as manufacturers of products such as firefighting foams and turnout gear face limited requirements for declaring specific contents.

Serum concentrations of legacy PFAS have declined over the last decades in Denmark (Hull et al., 2023). Our reference measurements from the ENFORCE study were based on samples from 2021 analyzed in a different laboratory. Applying these older measurements from an older study population with no information on aspects such as blood donation in comparisons, we may underestimate the current differences between serum PFAS among the firefighters and the general population in Denmark.

5. Conclusion

Slightly higher concentrations of especially PFHxS, PFHpS and PFOS in serum among civilian airport firefighters compared to the general population most likely reflected remnants of past occupational exposure to firefighting foam. The use of firefighting foam containing PFAS was discontinued by the civilian airport fire services more than a decade prior to the study. Thus, our findings emphasize the importance of protection through robust regulation and substitution.

CRediT authorship contribution statement

Kajsa Ugelvig Petersen: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Dorthe Furstrand Lauritzen:** Writing – review & editing, Validation, Software, Investigation, Formal analysis, Data curation, Conceptualization. **Regitze Sølling Wils:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Anne Thoustrup Saber:** Writing – review & editing, Methodology, Conceptualization. **Ulla Vogel:** Writing – review & editing, Methodology, Conceptualization. **Niels Erik Ebbehøj:** Writing – review & editing, Methodology, Conceptualization. **Johnni Hansen:** Writing – review & editing, Methodology, Conceptualization. **Julie Elbæk Pedersen:** Writing – review & editing, Methodology, Conceptualization. **Tina Kold Jensen:** Writing – review & editing, Methodology, Conceptualization. **Maria Helena Guerra Andersen:** Writing – review & editing, Methodology, Investigation, Conceptualization.

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Conflicts of interest

The authors declare no conflicts of interests.

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Appendix A. Supplementary data

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