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ANTTI KARJALAINEN

Exposure to particulate matter and organic chemicals, and controlling flour dust in the Finnish bakery industry

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ABSTRACT

The bakery industry is the largest food sub-industry in Finland, comprising international, and national enterprises, as well as local, traditional bakeries, confectioneries, in-store bakeries, and bake-off units. The purpose of this thesis was to investigate exposure to particulate matter and organic chemicals and the effectiveness of an intervention to control flour dust exposure in the Finnish bakery industry.

This thesis showed that personal exposure levels of dough makers (1.3–14.5 mg/m^3) and general bakers (4.5–22.7 mg/m^3) to inhalable dust exceeded the Finnish (8-hour) occupational exposure limit (OEL) of 2 mg/m^3 for flour dust. The $PM_{0.5}$ fraction was made up of 9–15% of the inhalable dust in the breathing zone of a dough maker. The real-time mass concentrations (PM_{10}) ranged from < 0.1 to 28.3 mg/m^3 at stationary locations. Several work tasks contributed to peak concentrations.

In the microscopic analysis, small, agglomerated flour dust particles, spherical particles, and soot agglomerates were found in the dust samples collected in the traditional bakery. Considering the PM_1 samples, carbon comprised 42–64% of the total PM_1 mass, and approximately 99% was organic carbon.

Regarding the intervention study, the mass concentrations of inhalable dust increased 24–55% in the breathing zone post-intervention in the traditional and industrial bakery. At most stationary locations, reductions (39–45%) in inhalable dust levels were obtained. The real-time, peak mass concentrations (PM_{10}) decreased only in the traditional bakery post-intervention.

The real-time number concentrations of particulate matter varied between 3.7×10^2 and 4.1×10^6 cm^{-3} at stationary locations in the traditional bakery, in-store bakery, and bake-off unit. Fine particles and nanoparticles contributed significantly to the number concentrations. The peak concentrations were detected when all the ovens were operated simultaneously in the facilities.

In the in-store bakery and bake-off unit, the TVOC concentration (31–214 $\mu g/m^3$) was lower than the target value of 300 $\mu g/m^3$ for industrial workplaces, and the concentrations of individual VOCs (1–81 $\mu g/m^3$) and short-chained carbonyls (< 1–59 $\mu g/m^3$) were lower than the Finnish OEL.

The findings of this thesis provided new knowledge on the number concentrations of particulate matter and concentrations of organic chemicals in the bakery industry. The study highlights the importance of personal protective equipment and local control measures to reduce exposure levels to particulate matter. Further research is required for planning interventions accompanied by technical control methods in bakeries.

Keywords: Bakery, exposure, fine particles, flour dust, indoor air, inhalable dust, nanoparticles, occupational, particulate matter, retail store

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Finally, I want to thank all the important people outside the academia for support, encouragement, and leisure activities.

Kuopio, September 2023
Antti Karjalainen

LIST OF ABBREVIATIONS

AM	arithmetic mean
BO	bronchiolitis obliterans
BZ	breathing zone
C	concentration
C_m	mass concentration
C_n	number concentration
CPC	condensation particle counter
D_{ae}	aerodynamic diameter
DNPH	2,4-dinitrophenylhydrazine
DRX	DustTrak DRX aerosol monitor 8533
EDS	energy-dispersive X-ray spectroscopy
EFSA	European Food Safety Authority
EIA	inhibition enzyme immunoassay
ELPI	electrical low-pressure impactor
EM	electron microscope
FEMA	The Flavor and Extract Manufacturers Association
FIOH	Finnish Institute of Occupational Health
FMPS	fast mobility particle sizer
GM	geometric mean
GMD	geometric mean diameter
GSD	geometric standard deviation
IBS	irritable bowel syndrome
IC	ion chromatography
ICP-MS	inductively coupled plasma mass spectrometry
ICRP	International Commission on Radiological Protection
IgE	immunoglobulin E
IgG	immunoglobulin G
IOM	Institute of Occupational Medicine
ISO	International Organization for Standardization
LC-MS/MS	liquid chromatography-tandem mass spectrometer
LEV	local exhaust ventilation

NCWS	non-coeliac wheat sensitivity
NIH	National Institutes of Health
NIOSH	National Institute for Occupational Safety and Health
NIST	National Institute of Standards and Technology
OEL	occupational exposure limit
OPC	optical particle counter
OSHA	Occupational Safety and Health Administration
PAS	personal air sampler
PM	particulate matter
PPE	personal protective equipment
P-Trak	P-Trak ultrafine particle counter 8525
PVC	polyvinyl chloride
RAST	radioallergosorbent test
RIA	rabbit IgG inhibition radioimmunoassay
RPE	respiratory protective equipment
S	stationary location
SCOEL	Scientific Committee on Occupational Exposure Limits
SD	standard deviation
SEM	scanning electron microscope
SMPS	scanning mobility particle sizer
TD-GC-MS	thermal desorption-gas chromatography-mass spectrometer
TEOM	tapered element oscillating microbalance
TVOC	total volatile organic compound
TWA	time-weighted average
VOC	volatile organic compound
WHO	World Health Organization
UFP	ultrafine particle

LIST OF ORIGINAL PUBLICATIONS

This thesis is based on data presented in the following articles, referred to by the Roman Numerals I-III.

I Karjalainen A, Leppänen M, Leskinen J, Torvela T, Pasanen P, Tissari J, Miettinen M. (2018). Concentrations and number size distribution of fine and nanoparticles in a traditional Finnish bakery. *Journal of Occupational and Environmental Hygiene,* 15(3): 194–203.

II Karjalainen A, Leppänen M, Ruokolainen J, Hyttinen M, Miettinen M, Säämänen A, Pasanen P. (2022). Controlling flour dust exposure by an intervention focused on working methods in Finnish bakeries: a case study in two bakeries. *International Journal of Occupational Safety and Ergonomics,* 28(3): 1948–1957.

III Karjalainen A, Väisänen A, Leppänen M, Ruokolainen J, Hyttinen M, Miettinen M, Säämänen A, Pasanen P. Exposure to particulate matter, volatile organic compounds, and carbonyls in an in-store bakery and a bake-off unit in Finland. Submitted manuscript.

The above publications have been included at the end of this thesis with their copyright holders' permission.

AUTHOR’S CONTRIBUTION

I) Indoor air measurements were conducted together with colleagues. Laboratory analyses were done by colleagues. Data analyses, interpreting the results, and writing of the paper were done by the author with the guidance of Senior Researcher Mirella Miettinen. The author received valuable editorial support from the co-authors.

II) The author planned the experiments with colleagues. Indoor air measurements were carried out by the author with the assistance of Early Stage Researcher Joonas Ruokolainen. The author was responsible for analysing the data. Writing the paper was conducted by the author with significant editorial input from the co-authors.

III) The author planned the experiments with the main supervisor. He conducted indoor air measurements with the assistance of Doctoral Researcher Antti Väisänen. Interpreting the results and writing of the paper was done by the author with the supportive co-operation of the co-authors.

Table of contents

1 Introduction

The bakery sector is highly diverse in Finland. Most bakeries are family-operated. Regarding the number of businesses and locations, the bakery sector is the largest sector within the food industry, employing approximately 7,000 full-time personnel in nearly 600 companies. In 2020, the majority (60%) of bakery industry companies employed fewer than five persons, whereas approximately 140 companies employed more than ten persons. A few large companies play a major role in the bakery industry, which is typical also in the other sectors of the food industry. Most of the bakery sector focuses on the production of fresh breads, bread rolls, and fresh pastries such as buns and cakes. (Hyrylä 2021)

Typically, the companies in the bakery industry are national, international, and regional enterprises, as well as local bakeries (small-scale, traditional bakeries), and confectioneries. Furthermore, many retail stores, including supermarkets and hypermarkets, have launched either an in-store bakery or a bake-off unit. (Hyrylä 2017) Many gas stations and kiosks also have a bake-off unit (Väyrynen 2016). The number of in-store bakeries and bake-off units is constantly increasing, and their selection of products has grown rapidly, which has led to the customers tending to purchase bakery products from various shops. The growing market share of in-store bakery products has also led to increased competition in the Finnish bakery industry. (Hyrylä 2015)

Exposure to flour dust occurs predominantly in bakeries and mills. The dustiest tasks are mixing, dough-making, bread-forming, and cleaning in the bakeries, whereas milling, packing, cleaning, and maintenance are associated with exposure in the mills. Furthermore, the following occupations also involve flour dust exposure: confectionary (weighing, mixing, production), pasta and pizza bakeries, animal feed plants, malt factories (drying, sieving, packing), and agriculture (milling, feeding). (Health Council of the Netherlands 2004, SCOEL 2008) Exposure to flour dust may have consequences for both the staff (e.g., various symptoms, absence from work and loss of income) and enterprise (expenses from absences due to sicknesses, occupational diseases, and losses of production outputs) (Säämänen et al. 2012).

The dust in bakeries comprises particles from cereal flours as well as various other ingredients, and several of these components are known to be sensitizers (Health Council of the Netherlands 2004, SCOEL 2008). Respiratory, dermal, and conjunctival reactions are associated with cereal flour dust among bakery workers (Health Council of the Netherlands 2004). Baker's asthma is one of the most common occupational asthmas (Houba et al. 1998a, Brant et al. 2007). Exposure to flour dust is also related to rhinitis, conjunctivitis, chronic bronchitis, and bronchial obstruction (Laurière et al. 2008, Fahim and El-Prince 2013). These symptoms may stem from sensitization of the worker, since the flour dust proteins are potential allergens, or caused by non-specific irritation (Health Council of the Netherlands 2004, SCOEL 2008).

According to the latest information provided by the Finnish Institute of Occupational Health (FIOH), in 2008–2016, the FIOH conducted over 240 flour dust measurements, of which 42% exceeded the Finnish occupational exposure limit (OEL) of 2 mg/m^3 (Ministry of Social Affairs and Health 2020) and 56% were greater than half of the OEL in the Finnish food industry (all occupations considered). Approximately 75% of the full-time bakery workers are exposed to flour dust concentrations that exceed the Finnish OEL of 2 mg/m^3 by at least 50%. Flour dust causes approximately 40 occupational diseases annually in Finland.

Nieuwenhuijsen et al. (1995a) showed that the time-weighted average (TWA) mass concentration (C_m) in bakeries is attributable to peak exposures associated with specific work activities. Meijster et al. (2008) found that > 75% of TWA exposure was directly related to peak exposures. These exposures are usually frequent in bakeries and may be linked to work-related adverse health effects (Health Council of the Netherlands 2004, SCOEL 2008). This warrants the need for controlling peak exposure levels to lower sensitization as well as both allergic and other symptoms. Therefore, intervention strategies are required for the work tasks that contribute to peak exposures. However, there are only four studies that focuse on the effectiveness of interventions in the bakery industry (Meijster et al. 2009, Baatjies et al. 2014, Hakala et al. 2016, Martinelli et al. 2020).

Furthermore, exposure to fine particles and nanoparticles in the bakery industry has been investigated in only two previous studies (Tissari et al. 2002, Tissari et al. 2005), which raises the need for further research. The two previous studies found that bakery air contained nanoparticles (< 100 nm), and most of the fine particles (< 2.5 μm) and nanoparticles were released into the bakery air from oven operations. Research has shown that smaller and lighter particles stay longer in the air and may penetrate the alveolar region of the lung (WHO 1999).

Workers are also exposed to various food flavor compounds during production processes. These volatile organic compounds (VOCs) are formed from, for example, yeast, milk, sugar, salt, and butter. (Cho and Peterson 2010). Furthermore, heating of fats and oils may produce carbonyls (Ho et al. 2006), which are also reactive volatile substances (Feng and Zhu et al. 2004). Research on the exposure levels of VOCs and carbonyls in the bakery industry is scarce. Only three studies (Tissari et al. 2002, Curwin et al. 2015, Chang et al. 2018) regarding the concentrations of VOCs and carbonyls in the bakery industry were found, highlighting the importance of further research.

This thesis aimed to examine the variation of mass concentrations, number concentrations, and number size distribution of particulate matter in the Finnish bakery industry. The morphology and composition of particles was also studied. Furthermore, concentrations of VOCs and carbonyls were investigated. In the present thesis, particulate matter covers total aerosol including particles from, for example, flours, spices, and oven operations in the facilities.

2 Literature review

2.1 Flour dust

2.1.1 Structure and components

Flour dust means finely ground particles of taxonomically related cereal grains of the subfamily *Festucoidea* and the tribes *Triticeae* and *Aveneae*, such as wheat (*Triticum sp.*), rye (*Secale cereale*), barley (*Hordeum sp.*) and oats (*Avenea sativa*), produced by milling or some other form of processing (Health Council of the Netherlands 2004, SCOEL 2008). The terms 'flour dust' and 'grain dust' need to be distinguished. Whereas 'flour dust' refers to particles regarding finely milled cereal or non-cereal grains, 'grain dust' comprises particles considering grain harvesting and handling, excluding milling. Grain dust may contain dry plant particles (non-grain plant matter), fungi (mainly from *Fusarium*, *Aspergillus*, *Cladosporium*, and *Alternaria genera*) with their mycotoxins, bacteria with their fragments (including endotoxins) and excretions (proteolytic enzymes), mites, insects, rodent excrements, sand, and residues of pesticides. (Stobnicka and Górny 2015) Due to its different health effects than flour dust, grain dust is excluded from this thesis.

Wheat is the main cereal grain used in the bakery industry. A wheat seed is composed of the endosperm (85%), husk (13%) and germ (2%). During the milling process, the endosperm is separated from the husk and germ and reduced to small particles. Wheat flour, made from the endosperm, contains starch and four groups of proteins: glutelins (glutenins), prolamins (gliadins), albumins, and globulins. The structure and texture of bread is predominantly determined by viscous complexes called gluten, which is formed by gliadins and glutenins. (Health Council of the Netherlands 2004, SCOEL 2008, Stobnicka and Górny 2015) The protein contents of whole wheat and wheat flour are nearly equal (12% vs. 11%). In airborne flour dust, the protein content is about 10%. (Tikkainen et al. 1996)

In the bakery industry, flour dust may contain several non-cereal components. These components include dough-improvers, such as a variety of enzymes (e.g., α-amylase, malt enzymes, cellulase, hemi-cellulase, xylanase), chemical ingredients (e.g., preservatives, bleaching agents, antioxidants), flavourings, spices, other additives (e.g., baker's yeast, egg powder, sugar), as well as contaminants (e.g., storage-related mites and microbes). Several of these components are sensitizers. (Health Council of the Netherlands, SCOEL 2008) In this thesis, flour dust is contained in the total aerosol, including both cereal and non-cereal components. In the context of occupational hygiene, an aerosol is defined as a system of particles suspended in a gaseous medium, usually air (Hinds 1999, WHO 1999). This thesis did not investigate the concentrations of allergens (e.g., wheat or rye allergens) or non-cereal components.

2.1.2 Aerosol behavior

The deposition and elimination of flour dust particles in the lungs follow the patterns of other similar types of solid aerosols. Particle deposition in the lungs is determined by the size, density, shape, aerodynamic properties of flour dust particles, and the volume of respiration. (Tikkainen et al. 1996)

The diameter is usually used to measure the of size of spherical particles. In occupational hygiene, the particle size is usually presented in terms of the spherical equivalent aerodynamic diameter (D_{ae}) (WHO 1999). This is defined as the diameter of a hypothetical sphere of density 1 g/cm^3 having the same terminal settling velocity when settling under gravity as the particle under consideration (Kulkarni et al. 2011).

The ICRP (International Commission on Radiological Protection) deposition model of particles, as described by Hinds (1999), characterizes the behavior of airborne particles in the respiratory system. This model focuses on how particles of different sizes and properties are deposited within the human respiratory tract based on their aerodynamic characteristics. Larger particles are more likely to be deposited in the upper airways, while smaller particles can penetrate deeper into the lungs. Inhaled particles may be exhaled or deposited in the various regions of the respiratory system. The most important deposition mechanisms are impaction, settling, and diffusion.

Regarding the deposition of particles in the head airways, the largest particles are removed by settling and impaction on nasal hairs and at bends in the airflow path. During mouth breathing, approximately 20% of 5 μm (D_{ae}) particles and 70% of 10 μm particles are deposited before the inhaled air reaches the larynx. In the tracheobronchial region, the dominant mechanisms are impaction for particles of > 3 μm and settling for particles of 0.5–3 μm. Generally, particles of > 10 μm do not reach the alveolar region, whereas a low number of 2–10 μm particles reach the alveolar region. Considering particles of 0.1–1 μm, the rate of alveolar deposition is approximately 10–20%. During mouth breathing, particles of about 3 μm have the highest deposition in the alveolar region, a rate of approximately 50%, whereas, during nose breathing, alveolar deposition is highest for particles of about 2.0 μm, with a rate of approximately 10–20%. (Hinds 1999)

Flour particles are cleared from the lungs by macrophages and the mucociliary system. However, in cases of heavy exposure, dust particles may penetrate to the interstitium due to the overloading of macrophages. (Stobnicka and Górny 2015) The individual characteristics of an exposed person also affect the kinetics of dust particles, such as age, pre-existing respiratory conditions, and exposure to other respiratory hazards (e.g., cigarette smoke) (Maynard and Kuempel 2005).

The terms inhalable (≤ 100 μm), thoracic (4–10 μm), and respirable (≤ 4 μm) are used for particles that may be hazardous when inhaled. The inhalable fraction refers to airborne particles inhaled through the nose and mouth. The thoracic fraction includes inhaled particles penetrating beyond the larynx, and the respirable fraction comprises particles penetrating to the unciliated airways. (Health Council of the Netherlands 2004) Eye and nose irritation are caused by particles with aerodynamic diameters of ≥ 10 μm, whereas particles of 5–10 μm may provoke asthmatic reactions, and particles of ≤ 5 μm may evoke an allergic alveolitis type of reaction (Stobnicka and Górny 2015).

2.1.3 Particle size range

Particle sizes of airborne flour dust have been reported in several studies. Lillienberg and Brisman (1994) showed that the aerodynamic diameters of flour dust have a bimodal distribution. They found that the smallest particles were around 5 μm and the larger ones approximately 15–30 μm. The largest particles are usually formed as agglomerates of smaller ones. (Roberge et al. 2012)

Tikkainen et al. (1996) estimated that more than 50% of the airborne flour dust particle mass has an aerodynamic diameter of over 15 μm, whereas in dusty areas, about 20wt% of these particles have the size of ≤ 4 μm. Stobnicka and Górny (2015) also suggested that > 50% of the airborne flour dust particle mass has an aerodynamic diameter of > 15 μm.

Burdorf et al. (1994) showed that the thoracic fraction (4–10 μm) contributed 39% to the total mass of inhalable dust, whereas the respirable fraction amounted to 19%. Lillienberg and Brisman (1994) reported that thoracic and respirable fractions were 26wt% and 9wt% of inhalable flour dust, respectively. Roberge et al. (2012) showed that an average mass percentage of 0.2–4.0 and 10–20 μm fractions made up 12% and 61% of inhalable dust, respectively.

Tissari et al. (2002, 2005) found that operating trolley ovens affected the number size distribution of particulate matter and resulted in ultrafine particles (an aerodynamic diameter of 100 nm or less) in the air of a bakery, whereas larger particles (> 5 μm) consisted mainly of flour dust.

2.1.4 Chemical composition and morphology of particles

There is a lack of studies regarding the chemical composition and morphology of particles in bakeries. Only two studies (Tissari et al. 2002, Ielpo et al. 2020) that analyzed the chemical composition of particles in bakeries were found. Tissari et al. (2002) also examined the morphology of particles.

Tissari et al. (2002) showed that in a traditional bakery, particles in the air comprised predominantly carbon compounds. The total carbon consisted mainly of organic carbon (88% on average) in total dust and PM_1 samples.

They found that the proportion of organic carbon increased when several work phases were ongoing. In a few samples collected in the middle of a working day, the proportion of organic carbon was 99%. In addition to carbon, 31 elements were determined from Teflon filters. The proportion of these elements was an average of < 1% in the filter samples. All the samples included Al, Mg, Na, and Ti. Furthermore, Ca, Cu, Si, and Zn were found in a few samples. The morphological analysis of particles was found to be challenging since the particulate matter in the bakery air consisted of agglomerated particles originating from flours, grease, and ovens (combustion). However, the results showed that there were nanosized particles in the bakery air.

Ielpo et al. (2020) found that in a traditional bakery, the carbonaceous fraction (organic carbon, elemental carbon, levoglucosan) in $PM_{2.5}$ samples was 35wt% when ovens were turned on and decreased to 17–22wt% when there was more intense activity of the ovens. In $PM_{2.5}$ samples, the mass fraction of water-soluble ions (Cl^-, NO_2^-, NO_3^-, SO_4^{2-}, $C_2O_4^{2-}$, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) varied between 42 and 60%.

2.1.5 Allergenic properties

The proteins in flour dust are potential allergens. Regarding wheat, proteins can be divided into the water/salt-soluble fraction (including albumins and globulins) and the water/salt-insoluble gluten (comprising gliadins and glutenins). The latter fraction represents about 80wt% of all wheat proteins. (Bittner et al. 2008) Water/salt-soluble albumin and globulin fractions of wheat flour have been shown to contain most of the allergens involved in bakers' allergies, whereas gliadins and glutenins are considered less allergenic (Tikkainen et al. 1996).

Various methods to measure concentrations of airborne wheat allergens have been introduced, for example, a human IgE inhibition RAST (radioallergosorbent test), rabbit IgG inhibition radioimmunoassay (RIA), human IgG4 inhibition enzyme immunoassay (EIA), and rabbit IgG inhibition EIA. These assays measure a wide range of water/salt-soluble wheat proteins that are the most relevant IgE-inducing allergens. (Bogdanovic et al. 2006b)

Considering biological activity, cereals belonging to the family *Poaceae* are the most important. Particles derived from wheat (*Triticum* sp.), barley (*Hordeum* sp.), rye (*Secale cereale*) and oats (*Avena sativa*) have high allergenic potency, whereas the flour dust coming from corn (*Zea mays*) is much less reactive instead. (Health Council of the Netherlands 2004, SCOEL 2008) Significant flour dust sensitizers also derive from non-cereal grains, such as soy (*Glycine hispida*), buckwheat (*Fagopyrum esculentum*), and peas (*Pisum sativum*) (Stobnicka and Górny 2015).

Wheat flour contains at least 40 allergens that can cause adverse health effects in exposed workers (Sander et al. 2001). The job category and bakery size are important determinants of elevated exposure to flour dust allergens (Baatjies et al. 2010). The major allergens of flours belong to the amylase/trypsin inhibitor family, which may contribute to natural defense against pests and pathogens (Geisslitz et al. 2021). In addition to cereal allergens, flour dust may also contain other allergic components like storage mites, fungi, enzymes, and spices. The allergenic components of spices and flavorings are proteins and chemicals, such as cinnamon compounds. Dough-improvers include allergens from enzymes. (Tikkainen et al. 1996)

Alpha-amylase is the most common starch-cleaving enzyme used as a flour additive (Brisman and Belin 1991). Cereal amylases should be distinguished from fungal amylases added as dough improvers. Wheat flour has been analyzed to contain cereal α-amylases 0.1–1.0 mg/g flour (Jauhiainen et al. 1993). Fungal α-amylase is usually derived from *Aspergillus oryzae* or *Aspergillus niger* (Stobnicka and Górny 2015), and it is often added to flours at mills, but may also be added at bakeries (Elms et al. 2001). It has been shown that occupational exposure to industrial fungal α-amylase increases the risk of respiratory allergy in bakery workers (Houba et al. 1997a). Studies in which α-amylase sensitization was examined using skin-prick tests and radioallergosorbent test (RAST) have suggested that approximately 10–15% of bakers are sensitized to α-amylase (Houba et al.

1996, Smith et al. 1997, Jeffrey et al. 1999). There are currently no specific OELs for exposure to flour antigens in Finland.

Both enzymatic techniques and immunoassays have been developed to analyze α-amylase. The enzymatic methods measure all active α-amylase, whereas immunoassays detect immunoreactive α-amylase. (Elms et al. 2001) The reported α-amylase levels might differ by a factor of two or more between different methods, which warrants the need for standardization since, thus far, no recommended methods for the quantification exist (Lillienberg et al. 2000).

Besides α-amylase, the following enzymes have been reported to cause sensitization in bakery workers: amyloglucosidase, cellulase, glucoamylase, hemicellulase, lipoxygenase, papain, pectinase, and xylanase (Houba et al. 1997a). Enzymes are used for reducing the viscosity of dough, giving the desired coloring and volume to bread, and lengthening shelf-life. The improver, which is typically only about 1wt% of the dough, usually has an enzyme content of 0.2–1wt%. (Vanhanen et al. 1996) No occupational exposure limit (OEL) values currently exist for enzymes in Finland.

2.1.6 Health effects

Different types of cereal flour dust may cause respiratory, dermal, or conjunctival reactions in workers. Symptoms observed in the respiratory tract and eyes are the primary health effects of flour dust exposure. (Health Council of the Netherlands 2004)

Baker's asthma is the most severe reaction evoked by flour dust exposure (Houba et al. 1998a, Brant et al. 2007). Occupational asthma stems from immunologic sensitization and subsequent allergic reactions to occupational-specific airborne allergens in the airways (Houba 1996). Variable airflow limitation and/or airway hyperresponsiveness because of causes and conditions related to a particular occupational environment (without stimuli encountered outside the workplace) are characteristics of occupational asthma. The prevalence of baker's asthma has been reported to be 5–17%. (Baatjies et al. 2009) However, in epidemiological studies, possible sources of bias have included the presence of atopic persons, a lack of knowledge of job history, and the existence of a healthy worker effect (symptomatically sensitized workers change to jobs with no or low exposure risks) (Health Council of the Netherlands 2004).

However, other clinical symptoms have also been reported, such as rhinitis (with frequent sneezing, nasal obstruction, and rhinorrhea) and conjunctivitis (with itching and inflamed, red eyes). Most of these symptoms are allergic in origin and preceded by sensitization of the worker. (Health Council of the Netherlands 2004, SCOEL 2008) Usually, baker's rhinitis can be considered a pre-stage of asthma; however, in some cases, asthma and rhinitis can develop simultaneously (Lehto et al. 2010). Symptoms develop after a latency period of months, years, or decades, and risk increases with increased exposure. Besides allergy, non-specific mucous membrane and respiratory irritation are also frequent and possibly more common than allergic symptoms among bakers. (Page et al. 2010) The following groups of workers have an increased risk of adverse health effects regarding exposure to flour dust: workers with flour sensitization because of constant exposure to low flour dust levels, workers with an atopic or allergic status, and workers with general respiratory symptoms or pre-existing asthma. (Stobnicka and Górny 2015)

A substantial proportion of work-related asthma and rhinitis has been shown to be of allergic origin, mediated by immunoglobulin E (IgE) antibodies to flour dust antigens. Approximately 5–28% of the workers in the bakery and flour milling industry are sensitized against flour dust allergens, whereas in the non-occupational exposed population, the prevalence of sensitization is estimated to be between 2 and 4%. Most studies on flour dust have been done on wheat. (Health Council of the Netherlands 2004) IgE immediate hypersensitivity reaction can be shown by, for example, skin-prick tests and a radioallergosorbent test (RAST) (Sandiford et al. 1994, Sandiford et al. 1995, García-Casado et al. 1996, Baur et al. 1998, Ehrlich and Prescott 2005). Regarding non-IgE-mediated respiratory symptoms, they are probably caused by non-specific irritation responses (Health Council of the Netherlands 2004).

Bakers also belong to the high-risk occupations for irritant contact dermatitis. In addition, occupational allergic contact dermatitis and protein contact dermatitis to flours, enzymes and mites occur, although they are rare compared to the number of exposed workers. (Tikkainen et al. 1996) Plenty of agents have been identified as potential dermal sensitizers and allergens, including cereal flour, dough-improvers like fungal α-amylase, cellulase and xylanase enzymes, cinnamon oil/cinnamic aldehyde, certain emulsifiers, baker's yeast, bleaching agents (benzoyl peroxide) and antioxidants (propyl gallate) (Stobnicka and Górny 2015).

Regarding sensitization and symptoms, the shape of exposure-response relationships is not well characterized (Jacobs et al. 2008). Houba et al. (1998b) suggested that work-related sensitization risk will be negligible when average exposure levels to inhalable dust are reduced to approximately 0.5 mg/m^3. However, concentrations < 1 mg/m^3 may trigger symptoms in already sensitized workers (SCOEL 2008). Brisman et al. (2000) found that the risk of asthma increased at inhalable flour dust concentrations of ≥ 3 mg/m^3, whereas the risk of rhinitis was increased at concentrations of ≥ 1 mg/m^3.

Peak exposures related to work activities have been frequently observed in bakeries. These exposures may contribute to work-related adverse health effects, meaning that prevention of peak exposures might lower specific sensitization, as well as allergic and other symptoms. However, there is a lack of research regarding the abundance and frequency of these peaks in the dose-response relationship. (Nieuwenhuijsen et al. 1995a, Meijster et al. 2008)

It has been shown that wheat antigens are the most reactive allergens, and the most significant allergens in wheat flour are related to the water/salt-soluble fraction (albumins and globulins) (Gómez et al. 1990, Sanchez-Monge 1992). The major water/salt-soluble proteins have a molecular mass of 12–15 kDa and are associated with baker's asthma (Sanchez-Monge et al. 1992). These allergens are present in the seeds of various cereals (including wheat, barley, rye, maize, millet, and rice) and belong to the α-amylase/trypsin inhibitor family (Geisslitz et al. 2022). That group of proteins comprises approximately 2–4% of the total wheat grain proteins (Geisslitz et al. 2021). A majority of the members of that family show IgE binding capacity *in vitro* and positive responses in skin prick tests (García-Casado et al. 1995).

Cross-reactivity of IgE antibodies to different cereal flours has been demonstrated, which can be explained by the fact that some allergens from different cereals are chemically and functionally closely related. The degree of cross-reactivity closely follows the taxonomic relationship of cereals in the following order of decreasing closeness: wheat, triticale, rye, barley, oat, rice, and corn. The cross-reactivity of specific IgE may indicate that persons who are sensitized to an allergen are likely to develop hypersensitivity to other components sharing similar or closely related allergens. (Sandiford et al. 1995, Houba et al. 1996)

Alpha-amylase/trypsin inhibitors play a role in immediate-type, IgE-induced respiratory and food allergies, and in irritable bowel syndrome (IBS) (El Hassouni et al. 2021). They have also been suggested to trigger the innate immune system, contributing to the development of celiac disease, and affecting about 1% of the Western population. Furthermore, α-amylase/trypsin inhibitors have been postulated to be associated with non-celiac wheat sensitivity (NCWS). An estimated prevalence of NCWS is 1–10%, being higher in women than men, and mainly based on self-diagnosis. (Geisslitz et al. 2021)

Aside from the α-amylase/trypsin inhibitor family, lipid transfer protein, peroxidase, thioredoxin, serine proteinase inhibitor, thaumatin-like protein, and some prolamins have been linked to baker's asthma. Furthermore, flour additives, such as fungal enzymes (mainly α-amylase) have also been associated with baker's asthma. (Salcedo et al. 2011)

2.1.7 Monitoring of particulate matter

European Standards has published standard BS EN 481:1993 which defines three categories for size-selective sampling: an inhalable fraction, a thoracic fraction, and a respirable fraction (see definitions in Section 2.1.2). Quantitative characterization of particulate matter in bakeries is usually based on air or settled dust sampling. Different filters (e.g., Teflon, PVC, glass) and samplers (e.g., IOM, Millipore cassette, PAS6) are used, which makes it difficult to interpret measurement results. (Stobnicka and Górny 2015) For example, Lillienberg and Brisman (1994) found that the IOM sampler collects almost twice as much flour dust as the conventional total dust sampler (Millipore cassette).

Regarding the particulate matter, air monitoring should take place during the whole working period (8-hour time-weighted average, TWA) when assessing occupational exposure levels, since concentrations vary considerably during working days (Health Council of the Netherlands 2004). In Finland and many other countries, the IOM samplers are typically used for measuring inhalable dust. They have a 50% cut-off D_{ae} of about 50 μm, which enables the collection of airborne inhalable dust in various environments, including bakeries. Considering respirable dust, for example, size-selective heads and cyclones have been used to measure exposure levels. (Tikkainen et al. 1996) Real-time monitoring using various devices is also commonly conducted to measure different size fractions of airborne particulate matter (Health Council of the Netherlands 2004). There are currently no validated methods for determining the amount of flour dust or allergens from biological samples.

Personal sampling is usually recommended since it considers the exposure of different individuals or task groups. Stationary sampling is also commonly used, but it usually results in lower concentrations than personal sampling and reflects the general area situations. The results are typically presented as arithmetic means (AM) with a range and standard deviation. (Lillienberg and Brisman 1994) As previously mentioned, short-term peak exposures are frequent and contribute significantly to TWA mass concentration in bakeries (Nieuwenhuijsen et al. 1995; Meijster et al. 2008).

Concerning nanoparticles (ultrafine particles, UFP), there are no generally agreed parameters for measuring nanoparticle concentrations in workplace air, nor is there an agreement on instruments to conduct these measurements (Leskinen et al. 2012). For example, number, mass, and surface area exposure concentrations have been suggested as metrics for exposure to nanoparticles (van Broekhuizen et al. 2012). Particle number concentration and number size distribution have been the most common metrics in studies (Kuhlbusch et al. 2011). Standards CSN EN 16966:2018 and CSN EN 17058:2018 provide guidelines for the assessment of exposure by inhalation of nano-objects and their aggregates and agglomerates in workplaces.

In workplaces, nanoparticles can be measured using, the following instruments, for example: condensation particle counter (CPC), scanning mobility particle sizer (SMPS), electrical low-pressure impactor (ELPI), and aerosol diffusion charger. However, relatively few of these instruments are readily applicable for routine exposure monitoring due to their lack of portability, difficulty of use, and high cost. (NIOSH 2009) Furthermore, the size, shape, and morphology can vary between different nanoparticles, which poses a significant challenge for measurement methods (Leskinen et al. 2012).

2.1.8 Mass concentrations of particulate matter

Table 1 presents mass concentrations obtained in previous studies in which personal and stationary measurements of particulate matter have been conducted for various particle size fractions and job titles in different types of bakeries. The job titles in which exposure to dust was the greatest in various papers were included in the table. For some job titles, arithmetic mean (AM), geometric mean (GM), standard deviation (SD), and geometric standard deviation (GSD) were calculated if they were not reported and if exposure information for repeated measurements was provided in the paper.

Most previous studies reported the mass concentrations (C_m) of inhalable dust in bakeries. A few studies reported the C_m of respirable and thoracic dust. In older studies predominantly, the C_m of total dust was measured. Bakers working in the dough-making area (weighing and mixing ingredients) usually had the most significant levels of dust exposure to both inhalable dust (0.1–37.7 mg/m^3) and total dust (0.9–86.0 mg/m^3). Regarding stationary samples, the C_m has varied at 0.1–19.0 mg/m^3 (inhalable dust) and < 0.1–16.5 mg/m^3 (total dust) in the dough-making area.

Considering all job titles, personal exposure has been < 0.1–318 mg/m^3 (inhalable dust), 0.2–2.3 mg/m^3 (respirable dust), 0.1–1.1 mg/m^3 (thoracic dust), and < 0.1–86.0 mg/m^3 (total dust), whereas at stationary locations, the C_m has varied at < 0.1–19.0 mg/m^3 (inhalable dust), < 0.1–0.8 mg/m^3 (respirable dust), and < 0.1–16.5 mg/m^3 (total dust). No studies on stationary measurements of thoracic dust in bakeries were found.

Some studies have conducted real-time monitoring of C_m for the particle size fractions of PM_1, $PM_{2.5}$, PM_4, and PM_{10} in bakeries. Personal exposure has been measured only for $PM_{2.5}$ and PM_{10} size fractions. In various work tasks, the C_m has been 0.2–1.5 mg/m^3 ($PM_{2.5}$) and 0.2–4.0 mg/m^3 (PM_{10}). Concerning stationary measurements, the C_m has varied at < 0.1–0.1 mg/m^3 (PM_1), < 0.1–0.7 mg/m^3 ($PM_{2.5}$), < 0.1–0.1 mg/m^3 (PM_4), and < 0.1–0.7 mg/m^3 (PM_{10}) in various job categories.

Table 1. Personal and stationary measurements of particulate matter in bakeries according to job title.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m³)	SD/GSD
Masalin et al. (1988)	NR	Personal	NR (total dust)	NR (1)	29	2	Bread baking	3.9/3.9 (3.3–4.5)	0.8/1.2
						3	Dough production	3.9/3.0 (1.7–7.8)	3.4/2.3
						2	Preparation of cake mixes	6.1/5.5 (3.6–8.5)	3.5/1.8
Musk et al. (1989)	NR	Personal	Casella or Millipore (total dust)	NR (1)	79	12	Cleaning staff/dough makers (bread baking)	NR/2.1 (< 0.1–16.8)	NR/NR
						10	Dough makers/mixers (confectionary/bakery)	NR/2.7 (0.6–14.1)	NR/NR
						2	Staff preparing ingredients (confectionary)	NR/11.0 (10.0–12.1)	NR/NR
Jauhiainen et al. (1993)	4–7	Personal, stationary	Three-piece cassettes (total dust)	NR (6)	44	7	Making of bread (BZ)	2.3/NR (1.5–3.4)	0.9/NR
						13	Making of dough (BZ)	4.6/NR (0.9–14.7)	3.6/NR
						7	Weighing (BZ)	4.2/NR (2.3–6.5)	1.6/NR
						5	Making of bread (S)	1.2/NR (0.4–4.2)	1.1/NR
						12	Making of dough (S)	4.3/NR (0.8–14.2)	4.5/NR
Bohadana et al. (1994)	3–6	Personal	Millipore (inhalable dust)	Industrial (1)	21	4	General baker	3.4/NR (0.7–8.7)	3.7/NR
							Special baker	41.3/NR (10.1–98.1)	39.5/NR
Burdorf et al. (1994)	NR	Personal	IOM sampler (inhalable dust)	Large (3), medium-size (7) Small (2)	129	62	Bread-formers	3.4/2.7 (0.6–14.2)	NR/2.0
						34	Dough makers	6.9/5.5 (1.2–16.9)	NR/2.1
						7	Mixed tasks	3.1/2.7 (1.2–4.9)	NR/1.7

Table 1. Continues.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m^3)	SD/GSD
Kolopp-Sarda et al. (1994)	NR	Personal	NR	Industrial (1)	10	10	Various tasks	4.9/NR (NR)	9.1/NR
Lillienberg and Brisman (1994)	5–8	Personal	IOM sampler (inhalable dust), Millipore (total dust)	NR (5)	58	6	Bread forming (inhalable dust)	8.0/NR (NR)	6.7/NR (NR)
						6	Dough mixing (inhalable dust)	7.5/NR (NR)	5.4/NR (NR)
						6	Bread forming (total dust)	4.3/NR (NR)	4.8/NR (NR)
						6	Dough mixing (total dust)	3.6/NR (NR)	2.5/NR (NR)
Nieuwenhuijsen et al. (1994)	Full-shift*	Personal	Casella (total dust)	Industrial (3)	352	9	Conf/dough brake	7.5/6.4 (2.9–15.3)	NR/1.8
						24	Dispense/mixing	9.0/5.0 (1.4–86.0)	NR/2.5
						32	Roll production	3.6/2.4 (0.4–21.1)	NR/2.5
Nieuwenhuijsen et al. (1995a)	NR	Personal	Casella (total dust)	Industrial (3)	128	9	Bread production	11.8/9.0 (2.2–25.0)	NR/2.3
						19	Dough brake	6.2/4.2 (0.9–21.1)	NR/2.5
						17	Roll production	18.1/8.8 (1.3–80.7)	NR/3.5
Nieuwenhuijsen et al. (1995b)	Full-shift*	Personal	Casella (total dust)	NR (3)	277	8	Confectionery (dough brake)	NR/6.1 (2.2–12.0)	NR/1.6
						37	Dispense/mixing	NR/3.8 (0.7–15.7)	NR/1.7
						30	Roll production	NR/1.4 (0.2–7.3)	NR/2.5

Table 1. Continues.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m^3)	SD/GSD
Houba et al. (1996)	6–8	Personal	PAS6 sampler (inhalable dust)	Large (2), small (19)	546	66	All-round staff (large bakeries)	NR/0.9 (0.1–26.8)	NR/NR
						105	Dough makers (large bakeries)	NR/3.0 (0.4–37.7)	NR/NR
						36	Bread baker (small bakeries)	NR/3.3 (1.2–8.8)	NR/NR
						57	Mixed baker (small bakeries)	NR/2.0 (0.3–14.2)	NR/NR
Vanhanen et al. (1996)	1–4	Personal, stationary	Millipore (total dust)	NR (4)	38	7	Dough making (BZ)	8.4/NR (3.0–18.8)	NR/NR
						10	Bread making (BZ)	3.2/NR (1.2–5.5)	NR/NR
						9	Dough making (S)	2.5/NR (0.7–8.4)	NR/NR
						11	Bread making (S)	1.1/NR (0.1–2.9)	NR/NR
Burstyn et al. (1997)	Full-shift*	Personal	7-hole sampler (inhalable dust)	Large (2), small (5)	88	88	Various tasks	21.4/NR (0.1–110)	NR/1.6–6.7
Houba et al. (1997b)	6–8	Personal	PAS6 sampler (inhalable dust)	Large (5) small (16)	571	124	All-round staff (large bakeries)	3.4/0.9–1.2 (1.3–7.5)	NR/ 1.2–2.0
						70	Dough makers (large bakeries)	3.5/0.9–4.5 (1.0–6.8)	NR/ 1.3-2.1
						36	Bread bakers (small bakeries)	3.8/3.3 (NR)	NR/1.6
						57	Mixed bakers (small bakeries)	2.7/2.0 (NR)	NR/1.9

Table 1. Continues.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m^3)	SD/GSD
Jeffrey et al. (1999)	8–10	Personal	7-hole sampler or Casella (inhalable dust)	NR (NR)	144	144	Various tasks	4.1/1.0–4.9 (0.1–23.7)	1.5–5.3/NR
Tissari et al. (2002)	NR	Personal, stationary	IOM sampler (inhalable dust), PM_1 Dekati impactor (PM_1), EPA-WINS impactor ($PM_{2.5}$ and PM_{10})	Traditional (1)	9	3	Dough maker (inhalable dust, BZ)	7.3/NR (4.1–10.2)	NR/NR
						6	General baker (inhalable dust, BZ)	2.7/NR (1.1–5.8)	NR/NR
					3	3	Various tasks (PM_1, S)	0.5/NR (0.2–0.8)	NR/NR
					10	10	Various tasks ($PM_{2.5}$, S)	0.5/NR (0.3–0.7)	NR/NR
					20	20	Various tasks (PM_{10}, S)	0.7/NR (0.5–0.7)	NR/NR
Elms et al. (2003)	NR	Personal	IOM sampler (inhalable dust)	Large, medium, small (19)	96	41	Various tasks (large bakeries)	NR/NR (< 0.1–36.8)	NR/NR
						28	Various tasks (medium bakeries)	NR/NR (< 0.1–25.9)	NR/NR
						27	Various tasks (small bakeries)	NR/NR (< 0.1–27.8)	NR/NR
de Pater (2003)	≥ 6	Personal	PAS6 sampler (inhalable dust)	Industrial (16), traditional (55)	348	186	Various tasks (industrial bakeries)	NR/1.0 (NR)	NR/3.8
						162	Various tasks (traditional bakeries)	NR/1.5 (NR)	NR/3.0

Table 1. Continues.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m^3)	SD/GSD
Bulat et al. (2004)	5–7	Personal, stationary	PAS6 sampler (inhalable dust)	Industrial (4), traditional (66)	410	22	Baker (industrial bakeries, BZ)	NR/1.1 (0.2–8.5)	NR/3.6
						29	Bread production (traditional bakeries, BZ)	NR/2.1 (0.3–13.3)	NR/2.4
						57	Pastry production (traditional bakeries, BZ)	NR/1.1 (0.2–8.7)	NR/2.6
						77	Dough making (traditional bakery, S)	NR/1.6 (0.1–9.0)	NR/2.2
						59	Worktable (traditional bakery, S)	NR/1.4 (0.2–6.6)	NR/2.3
Elms et al. (2005)	NR	Personal	IOM sampler (inhalable dust)	Large, medium, micro, small (55)	208	108	Baker/table/dough break	NR/3.3 (< 0.2–47.0)	3.4/NR
						59	Mixer/siever/weigher	NR/4.7 (< 0.2–30.6)	3.4/NR
Tissari et al. (2005)	NR	Stationary	EPA-WINS impactor (PM_{10})	Traditional (1)	16	16	Various tasks	0.3/NR (0.1–0.6)	NR/NR
Baatjies et al. (2007)	Full-shift*	Personal	PAS6 sampler (inhalable dust)	In-store (NR)	42	18	Baker	1.1/0.9 (0.4–2.2)	NR/1.8
							Confectioner	0.6/0.5 (0.2–1.2)	NR/1.8
Meijster et al. (2007)	4–10	Personal	PAS6 sampler (inhalable dust)	Industrial (20), traditional (65)	581	381	Various tasks (industrial bakeries)	NR/1.0 (0.2–292)	NR/3.5
						200	Various tasks (traditional bakeries)	NR/1.5 (0.2–318)	NR/2.7
Mounier-Geyssant et al. (2007)	5–11	Personal	Harvard Chempass sampler ($PM_{2.5}$ and PM_{10})	Craft, large (NR)	73	55	Baker and pastry apprentices ($PM_{2.5}$)	0.5/NR (0.2–1.5)	0.1–0.4/NR
						51	Baker and pastry apprentices (PM_{10})	0.7/NR (0.2–4.0)	0.1–0.8/NR

Table 1. Continues.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m³)	SD/GSD
Meijster et al. (2008)	NR	Personal	PAS6 sampler (inhalable dust)	Industrial, traditional (NR)	59	8	All baker	2.0/NR (NR)	NR/NR
						30	Breadbaker	4.5/NR (NR)	NR/NR
						16	Dough maker	1.8/NR (NR)	NR/NR
Page et al. (2009)	7	Personal	IOM sampler (inhalable dust)	NR (1)	102	8	Mixer	NR/4.0 (1.3–18.0)	NR/NR
						4	Moulder	NR/2.2 (0.1–7.3)	NR/NR
						5	Pan line	NR/3.4 (0.5–12.0)	NR/NR
						4	Sponge mixer	NR/25.2 (8.2–65.0)	NR/NR
Baatjies et al. (2010)	Full-shift*	Personal	PAS6 sampler (inhalable dust)	In-store (18)	211	112	Bread baker	1.8/1.3 (0.3–7.3)	NR/2.3
						38	Confectioner	0.9/0.7 (0.1–3.3)	NR/2.1
Page et al. (2010)	Full-shift*	Personal, stationary	IOM sampler (inhalable dust)	Large (1)	102	102	Various tasks (BZ, S)	NR/0.2–3.0 (ND–65.0)	NR/NR

Table 1. Continues.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m³)	SD/GSD
Roberge et al. (2012)	8	Stationary	Cassette (total dust), cassette + cyclone (respirable dust), IOM sampler (inhalable dust)	Tradition al (11)	144	66	Dough mixers (total dust)	6.0/3.0 (< 0.1–16.5)	5.0/6.0
							Dough mixers (inhalable dust)	8.6/4.9 (0.2–19)	6.9/4.2
							Dough mixers (respirable dust)	0.1/0.1 (< 0.1–0.5)	0.1/2.5
						78	Table (total dust)	2.5/1.1 (< 0.1–8.7)	2.5/6.2
							Table (inhalable dust)	3.7/2.2 (0.2–9.2)	2.8/3.7
							Table (respirable dust)	0.1/0.1 (< 0.1–0.3)	0.1/2.2
Baatjies et al. (2014)	Full-shift*	Personal	NR (inhalable dust)	In-store (15)	176	98	Bread baker (pre-intervention)	1.9/1.4 (0.3–7.3)	NR/2.2
						28	Confectioner (pre-intervention)	0.9/0.7 (0.3–3.3)	NR/2.0
					208	88	Bread baker (post-intervention)	1.0/0.7 (< 0.1–4.1)	NR/2.2
						57	Confectioner (post-intervention)	0.5/0.4 (< 0.1–1.7)	NR/2.1
Kirkeleit et al. (2017)	8	Personal	PAS6 sampler (inhalable dust), thoracic cyclone (thoracic dust)	Craft (2), industry (4), soft bread (1)	107	23	Craft baker (inhalable dust)	NR/2.3 (0.8–4.5)	NR/1.7
						21	Dough forming (inhalable dust)	NR/2.3 (0.4–5.2)	NR/2.0
						58	Dough making (inhalable dust)	NR/2.2 (0.3–9.9)	NR/2.0
						17	Craft baker (thoracic dust)	NR/0.3 (0.1–1.1)	NR/1.9
						10	Dough forming (thoracic dust)	NR/0.5 (0.2–0.7)	NR/1.5
						33	Dough making (thoracic dust)	NR/0.3 (0.1–1.1)	NR/2.0

Table 1. Continues.

Reference	Sampling time (h)	Type of sample	Instrument (type of dust)	Type of bakery (No.)	N	n	Job title	AM/GM (range) (mg/m^3)	SD/GSD
Chang et al. (2018)	5	Stationary	IOM sampler, Teflon filter, and MultiDust Foam disk, (inhalable and respirable dust)	In-store (5)	45	NR	Dough shaping and cake decorating (inhalable dust)	NR/NR (< 0.1–0.8)	NR/NR
						NR	Weighing, thieving, and mixing (inhalable dust)	NR/NR (0.1–0.8)	NR/NR
						NR	Weighing, thieving, and mixing (respirable dust)	NR/NR (ND–0.3)	NR/NR
Ielpo et al. (2020)	6	Stationary	Sequential air sampler ($PM_{2.5}$)	Traditional (1)	40	40	Various tasks	0.1/NR (0.1–0.3)	< 0.1–0.5/NR
Martinelli et al. (2020)	4	Personal, stationary	IOM sampler (inhalable dust)	Pre-intervention: craft (10), industrial (14) Post-intervention: craft and industrial (4)	105	29	Dough making (BZ, pre-intervention)	3.2/NR (0.1–14.1)	2.9/NR
						46	Dough forming (BZ, pre-intervention)	1.7/NR (0.2–12.1)	2.0/NR
						31	Dough making (S, pre-intervention)	3.0/NR (0.2–16.8)	3.2/NR
						40	Dough forming (S, pre-intervention)	1.5/NR (0.1–8.7)	2.0/NR
						28	Various tasks (S, post-intervention)	1.7/NR (0.2–5.6)	1.4/NR
Viegas et al. (2020)	3–8	Personal	IOM sampler (inhalable dust)	Traditional (2)	15	15	Various tasks	8.6/6.5 (1.3–18.3)	NR/2.4
Rumchev et al. (2021)	8	Personal, stationary	Cyclone (respirable dust), DustTrak 8530 (PM_1, $PM_{2.5}$, PM_4, PM_{10})	Industrial (1)	3	3	Various tasks (respirable dust, BZ)	0.9/NR (0.2–2.3)	1.2/NR
						1	Various tasks (PM_1, S)	< 0.1–0.1**	NR/NR
						1	Various tasks ($PM_{2.5}$, S)	< 0.1–0.1**	NR/NR
						1	Various tasks (PM_4, S)	< 0.1–0.1**	NR/NR
						1	Various tasks (PM_{10}, S)	< 0.1–0.2**	NR/NR

* = Exact duration of the full shift period was not reported.
** = Only median concentrations were reported.
AM = arithmetic mean, BZ = breathing zone, GM = geometric mean, GMD = geometric standard deviation, N = total number of samples in a bakery, n = number of samples in an occupational title, ND = not detected, No. = number of bakeries in a study, NR = not reported, S = stationary location, SD = standard deviation.

2.1.9 Number concentrations of particulate matter

Only two previous studies (Tissari et al. 2002, Viegas et al. 2018) were found on number concentrations (C_n) of particulate matter in bakeries. Tissari et al. (2002) obtained an average C_n of 6.0×10^4–2.5×10^5 cm^{-3} using an electrical low-pressure impactor (ELPI, size range: 6 nm – 10 μm) at stationary locations in a traditional bakery. Viegas et al. (2018) examined the personal exposure of workers to particle size fractions of 0.3–10 μm using a Lighthouse 3016/5016 handheld particle counter in 12 bakeries. In a production area, the C_n was 2.6×10^5–8.1×10^6 cm^{-3} ($PM_{0.3}$), 1.5×10^4–1.6×10^6 cm^{-3} ($PM_{0.5}$), 3.7×10^3–1.7×10^5 (PM_1), 1.9×10^3–6.2×10^4 cm^{-3} ($PM_{2.5}$), 4.4×10^2–1.8×10^4 cm^{-3} (PM_5), and 9.1×10^2–1.1×10^5 cm^{-3} (PM_{10}).

2.1.10 Controlling exposure to flour dust

Good working practices are known to reduce exposure to flour dust (Elms et al. 2005). These practices include measures regarding plant and equipment (engineering controls) and working practices (Patouchas et al. 2009). Establishing safe work practices (e.g., worker training programs and administrative controls) is also an important part of an effective occupational safety health program (Maynard and Kuempel 2005). Standard ISO 31000:2018 provides guidelines on managing risks in workplaces. Furthermore, the Finnish Occupational Safety and Health Act (738/2002) aims to improve the working environment and conditions.

Local exhaust ventilation (LEV) is one of the most studied exposure control technologies. The type of LEV (e.g., integrated into process or mobile), worker behavior and type of process (e.g., working below the LEV or at a distance) affect the effectiveness of LEV. Ineffective design and use (e.g., long distance from the source, weak airflow, poor maintenance) may reduce effectiveness of LEV. (Meijster et al. 2008)

Control measures regarding working practices include avoiding of spillages of flour, cleaning floors and surfaces, avoiding the use of compressed air for cleaning, loading ingredients into mixers carefully to avoid raising dust, and running mixers at a slow speed at the beginning, until wet and dry ingredients are combined. Furthermore, respiratory protective equipment (RPE) and personal protective equipment (PPE) (e.g., clothes, gloves, and goggles) should be worn if other control measures are not applicable or do not provide adequate control. (Patouchas et al. 2009)

Regarding preventive measures in bakeries, cooperation between an occupational hygienist and an occupational medicine specialist is needed (Patouchas et al. 2009). Lung function monitoring (spirometry, methacholine challenge test) and allergy tests (skin-prick tests, measurement of specific IgE antibodies) are part of the medical surveillance that aims to prevent adverse health outcomes of flour dust and other chemical agents (Stobnicka and Górny 2015).

There is a lack of experimental data and evaluation protocols on engineering control systems regarding nanoscale particles since aerosol control is mainly developed for mass-based exposures. However, nanoscale particles closely follow the movements of air and other gases and vapors if no significant thermal, electrostatic, or magnetic fields exist. Therefore, it is presumable that an engineering control system that is effective for gases and vapors would also be applicable for nanoscale particles. (Maynard and Kuempel 2005)

Considering intervention studies in bakeries, intervention measures have been focused on both technical control methods and work practices. Most studies have investigated the effectiveness of an intervention in the breathing zone only. Meijster et al. (2008) found that the reduction of exposure to inhalable flour dust was, in most cases > 50%, in Dutch bakeries. Meijster et al. (2009) examined changes in exposure over time and found a modest downward annual trend of −2% for flour dust in Dutch bakeries. Baatjies et al. (2014) obtained reductions of 23–67% in inhalable flour dust levels in South African supermarket bakeries. Hakala et al. (2016) reported an average reduction of 64% in inhalable flour dust concentrations in Finnish supermarket bakeries. Martinelli et al. (2020) showed that reductions in inhalable flour dust levels were between 16% and 70% in Italian bakeries.

In studies where flour was substituted with divider oil, there were clear reductions in flour dust exposure levels (Burstyn et al. 1997; Baatjies et al. 2014). Meijster et al. (2007) reported that control measures introduced while weighing ingredients, for example, limiting the use of bagged flour products and the enclosure of silos (when dumping flour), significantly decreased exposure levels. However, rather low reduction effect was observed when dusting flour was substituted with oil. That result contradicted previous studies (Burstyn et al. 1997; Baatjies et al. 2014) that showed a 30-fold decrease in exposure when dusting flour was replaced with oil. Meijster et al. (2008) suggested that the most effective control measures to reduce flour dust exposure were wet cleaning, no shaking of the cotton hose attached to the flour silo and no flour dusting. When sprinkling flour was substituted for oil, no significant reduction in exposure was found at the task level. Baatjies et al. (2014) found the best results in reducing flour dust levels when engineering controls and training were combined. Martinelli et al. (2020) observed significant reductions in flour dust levels when a sleeve on the end of a flour feeder's pipeline, a lid on the mixer tub, and an LEV system were installed.

2.2 Organic chemicals

2.2.1 Volatile organic compounds (VOCs)

Volatile organic compounds (VOCs) are defined as organic compounds boiling points ranging from 50–100 °C to 240–260 °C. Another criterion for classification of compound volatility is vapor pressure: VOCs are defined as compounds with a vapor pressure of 0.01 kPa or more at 20 °C, or as compounds having a corresponding volatility under particular conditions. (Tuomi and Vainiotalo 2016). VOCs are always present in both indoor and outdoor air where the most common VOCs are BTXS (benzene, toluene, xylenes, and styrene) and terpenes (α-pinene, limonene) (Sarigiannis et al. 2011).

Typical indoor emission sources of VOCs include, for example, paints and lacquers, cleaning supplies, organic solvents, cosmetic products, building materials and furnishings, office equipment such as photocopiers and printers, and materials including glues and adhesives. Levels of these chemicals are an average of 2–5-fold higher indoors than outdoors. Common symptoms associated with exposure to VOCs include eye irritation, nose and throat discomfort, headache, allergic skin reaction, nausea, fatigue, or dizziness. (Barro et al. 2009)

The concentration of total VOCs (TVOCs) is usually determined as toluene equivalents by considering compounds with a retention time between n-hexane (C_6) and n-hexadecane (C_{16}). TVOC concentration is frequently used as an indicator of indoor air quality. Regarding industrial environments, TVOC concentration is usually higher compared to non-industrial environments (e.g., homes, schools, daycare centers, and offices) because of the use of chemicals and factory processes that produce chemical substances. (Tuomi and Vainiotalo 2016)

In the food industry, workers may be exposed to a wide variety of flavoring substances in the form of solids, liquids, vapors, or liquid or vapor encapsulated within a particulate (Curwin et al. 2015). As far as bakeries are concerned, plenty of ingredients, including yeast, milk, sugar, salt, and butter, are used besides cereal flours, resulting in workers' exposure to various food flavor compounds during production processes (Cho and Peterson 2010). For example, regarding white bread, a wide array of volatile organic compounds (VOCs) has been found in the bread-making process, including alcohols, aldehydes, esters, ethers, ketones, acids, hydrocarbons, pyrazines, pyrrolines, furans, lactones, and sulfur compounds. These compounds may originate from the crumb, crust, or both. In the crumb, VOCs are formed by enzymatic reactions during dough kneading and the fermentation of dough sugars by yeasts and lactic acid bacteria. In the crust, VOCs stem from thermal reactions occurring during oven-baking, such as the Maillard reactions. (Pico et al. 2015)

The Flavor and Extract Manufacturers Association (FEMA) of the USA has identified 27 'high priority' flavoring substances that have the potential to pose respiratory hazards in workplaces (FEMA 2012). One of the most common VOCs observed in food and flavor manufacturing facilities is diacetyl, which has been associated with a severe lung disease, bronchiolitis obliterans (BO) (Day et al. 2011, Curwin et al. 2015, OSHA). BO has been identified in workers in the microwave popcorn industry and in flavoring and diacetyl manufacturing workers (Day et al. 2011).

Regarding bakery products, diacetyl is used as a natural and artificial flavoring ingredient and aroma carrier, imparting a buttery taste. In flavor formulations, diacetyl is a typical component in liquid solutions but can also be added to powders. (Curwin et al. 2015) Recently, some facilities have replaced diacetyl with alpha-diketone substitutes, such as 2,3-heptanedione, 2,3-hexanedione, and 2,3-pentanedione. According to preliminary data, these compounds might also pose health risks for workers because of their structural similarities with diacetyl. (OSHA)

Table 2 shows concentrations of VOCs considering personal and stationary measurements in the previous studies in bakeries. Compounds with ≥ C_6 were included in the table. For some compounds, AM, GM, SD, and GSD were calculated if they were not reported and if exposure information of repeated measurements was provided in the paper.

Table 2. Personal and stationary measurements of VOCs in bakeries.

Reference	Sampling time (h)	Type of sample	Instrument	Type of bakery (No.)	Compound	n	AM/GM (Range) (µg/m³)	SD/GSD
Tissari et al. (2002)[a]	NR	Stationary	Multisorbent, DNPH-silica	Traditional (1)	2-Hexanone	4	9/9 (7–11)	3/1
					3-Methyl-2-pentanone	4	19/19 (19–19)	NA
					3,3-Dimethylpentane	4	141/126 (62–197)	70/2
					Benzaldehyde	4	55/55 (55–55)	NA
					Decanal	4	40/15 (2–101)	53/7
					Dimethylhexane	4	176/175 (159–192)	23/1
					Eucalyptol	4	16/15 (9–25)	8/2
					Heptanal	4	25/10 (1–52)	26/8
					Hexanal	4	248/248 (248–248)	NA
					Nonanal	4	173/76 (7–309)	153/8
					Octanol	4	42/41 (31–53)	16/1
					Toluene	4	6/5 (3–8)	4/1
					Trimethylpentane	4	206/189 (110–305)	98/2
					TVOC	52	647/287 (26–1265)	631/6
Curwin et al. (2015)[b]	Full-shift*	Personal, stationary	DNPH-silica	NR (1)**	Benzaldehyde	73 (BZ) 105 (S)	NR/3–9 (NR) NR/3–81 (NR)	NR/6–10 NR/7–10
Chang et al. (2018)[c]	5	Stationary	Silica gel tube, XAD-2 tube	In-store (5)	2,3-Heptanedione	20	4/NR (ND–20)	5/NR
					2,3-Hexanedione	20	3/NR (ND–8)	1/NR
					TVOC	24	2334/NR (150–8657)***	1982/NR

[a] = Samples were collected in an oven area, a doughnut frying area, and a confectionary.
[b] = Samples were collected for the following processes: Control (S), handling (BZ, S), and production (BZ, S).
[c] = Samples were collected for a few processes (dough shaping and cake decorating, oven and baking, weighing, thieving, and mixing) and in a few areas (office, reception, stairs, and warehouse).
* = Exact duration of the full-shift period was not reported. The DNPH tubes were changed approximately every 3 h.
** = The study included ten food-manufacturing facilities (including one bakery). The results comprise all these facilities since the concentrations were not presented for each facility separately.
*** = Concentration is presented in ppb.
AM = arithmetic mean, BZ = breathing zone, GM = geometric mean, GSD = geometric standard deviation, n = number of samples regarding a compound, NA = not applicable since compound was detected only in one sample, ND = not detected, No. = number of bakeries in a study, NR = not reported, S = stationary location, SD = standard deviation.

In the two previous studies (Tissari et al. 2002, Chang et al. 2018), the TVOC concentrations were 7–336 ppb (the concentrations were converted from µg/m^3 into ppb using the equation presented by Boguski (2022) and 150–8660 ppb, respectively. However, the sampling methods were different between these studies. Tissari et al. (2002) determined the TVOC concentrations using a toluene-equivalent method, whereas Chang et al. (2002) measured TVOCs using a direct reading instrument, a ppbRAE 3000 detector, which was calibrated for isobutylene.

2.2.2 Carbonyls

Carbonyls is a common term for aldehydes and ketones that are reactive volatile substances (Feng and Zhu et al. 2004). They can be either formed in the atmosphere from atmospheric oxidation of hydrocarbons or directly emitted from sources. A major emission source for carbonyls is incomplete combustion of carbonaceous material. (Ho et al. 2006)

Regarding indoor carbonyls, the possible sources include construction materials, furniture, cooking, smoking, and painting (Weng et al. 2010). Major carbonyl compounds in occupational and residential indoor environments are aldehydes, mainly formaldehyde and acetaldehyde. Besides the above-mentioned sources, aldehydes can also be released from indoor ozone reactions with unsaturated VOCs. Indoor aldehyde concentrations are 2–13-fold higher than outdoor ones. (Barro et al. 2009)

Carbonyls are of concern to the public because of their adverse health effects (Feng and Zhu 2004). For example, formaldehyde, acetaldehyde, benzaldehyde, and acrolein are suspected carcinogens and mutagens. They may also cause odor problems. (Barro et al. 2009) Furthermore, formaldehyde and acetaldehyde are also potent sensory irritants (Feng and Zhu et al. 2004).

Considering the food industry, a few cooking activities, such as combusting fuel and heating oil, may emit carbonyls. Various cooking fuels (e.g., natural gas, liquefied petroleum gas, kerosene, and coal) produce formaldehyde and acetaldehyde. (Ho et al. 2006) Aldehydes and ketones may also originate from lipid oxidation (Pico et al. 2015). Partial conversion of fats and oils to volatile chain scission products is caused by heating fats and oils in the presence of air (Ho et al. 2006).

Moreover, the degradation of the flour amino acids via the Ehrlich pathway can form some aldehydes inside the yeast cell (Pico et al. 2015). Besides formaldehyde and acetaldehyde, acrolein and nonanal are also common carbonyls stemming from cooking operations in which oils are used (Ho et al. 2006).

Table 3 presents the carbonyl concentrations obtained in personal and stationary measurements in previous studies in bakeries. The short-chained carbonyls with < C_6 were included in the table. For some compounds, AM, GM, SD, and GSD were calculated if they were not reported and if exposure information of repeat measurements was provided in the paper.

In the previous studies, acetaldehyde was the most dominant compound in the bakeries. The acetaldehyde concentrations have varied between ND (not detected) and 1653 $\mu g/m^3$ in the samples collected in various locations.

Table 3. Personal and stationary measurements of short-chained carbonyls in bakeries.

Reference	Sampling time (h)	Type of sample	Instrument	Type of bakery (No.)	Compound	n	AM/GM (Range) (µg/m³)	SD/GSD
Tissari et al. (2002)[a]	NR	Stationary	Multisorbent, DNPH-silica	Traditional (1)	2-Butanone	4	8/8 (8–8)	NA/NA
					2-Pentanone	4	5/4 (4–5)	1/1
					Acetaldehyde	4	821/565 (229–1653)	710/3
					Acrolein + acetone	4	155/150 (116–204)	46/1
					Buturaldehyde	4	9/6 (2–16)	10/4
					Formaldehyde	4	42/23 (7–91)	42/4
					Propanal	4	10/6 (1–19)	9/5
					Valeraldehyde	4	22/22 (22–22)	NA/NA
Curwin et al. (2015)[b]	Full-shift*	Personal, stationary	DNPH-silica	NR (1)**	Acetaldehyde	73 (BZ)	NR/55–78 (NR)	NR/5–7
						105 (S)	NR/43–81 (NR)	NR/4–11
					Furfural	73 (BZ)	NR/4–6 (NR)	NR/7–9
						105 (S)	NR/6–8 (NR)	NR/10–11
					Isovaleraldehyde	73 (BZ)	NR/6–12 (NR)	NR/7–13
						105 (S)	NR/5–15 (NR)	NR/4–12
					Propionaldehyde	73 (BZ)	NR/4–4 (NR)	NR/5–6
						105 (S)	NR/3–5 (NR)	NR/3–11
Chang et al. (2018)[c]	5	Stationary	Silica gel tube, XAD-2 tube	In-store (5)	2,3-Pentanedione	20	1/NR (ND–3)	1/< 1
					Acetaldehyde	20	190/NR (ND–830)	260/NR
					Acetoin	20	2/NR (ND–75)	2/NR
					Diacetyl	20	1/NR (ND–3)	1/NR
					Furfural	20	4/NR (ND–66)	15/NR

[a] = Samples were collected in an oven area, a doughnut frying area, and a confectionary.
[b] = Samples were collected for the following processes: Control (S), handling (BZ, S), and production (BZ, S).
[c] = Samples were collected for a few processes (dough shaping and cake decorating, oven and baking, weighing, thieving, and mixing) and in a few areas (office, reception, stairs, and warehouse).
* = Exact duration of the full-shift period was not reported. The DNPH tubes were changed approximately every 3 h.
** = The study included ten food-manufacturing facilities (including one bakery). The results comprise all these facilities since the concentrations were not presented for each facility separately.
AM = arithmetic mean, BZ = breathing zone, GM = geometric mean, GSD = geometric standard deviation, n = number of samples regarding a compound, NA = not applicable since compound was detected only in one sample, ND = not detected, No. = number of bakeries in a study, NR = not reported, S = stationary location, SD = standard deviation.

3 Aims of the study

The main objectives of this study were to examine (1) concentrations, number size distribution, chemical composition, and morphology of particulate matter, (2) the effectiveness of an intervention to control flour dust exposure, and (3) concentrations of organic chemicals in the Finnish bakery industry. The research includes three studies, and their detailed aims are listed below.

Study I: (i) To study the variation of concentrations (mass, number) and number size distribution of particulate matter, (ii) to investigate the chemical composition and morphology of particles, and (iii) to determine exposure of a dough maker to the $PM_{0.5}$ fraction of inhalable dust in a traditional bakery.

Study II: (i) To examine the effectiveness of the intervention focused on working methods to control mass concentrations of flour dust, and (ii) to study the effect of the intervention on the proportions of various size fractions of the PM_{15}in an industrial and a traditional bakery.

Study III: (i) To investigate the mass and number concentrations, and number size distribution of particulate matter, and (ii) to determine the concentrations of volatile organic compounds (VOCs) and short-chained carbonyls in an in-store bakery and a bake-off unit.

4 Materials and methods

4.1 Study facilities

The study was conducted in different types of Finnish baking facilities, including two traditional bakeries (Papers I–II), an industrial bakery (Paper II), an in-store bakery (Paper III), and an in-store bake-off unit (Paper III). Table 4 presents the floor areas and average daily workforces of the facilities.

Table 4. Floor area and average daily workforce in the facilities.

Study	Study facility	Floor area (m^2)	Daily workforce (workers)
Paper I	Traditional bakery 1	450	10
Paper II	Traditional bakery 2	130	8
Paper II	Industrial bakery	3,500	15
Paper III	In-store bakery	120	3
Paper III	In-store bake-off unit	100	2

The traditional bakeries, as well as the in-store bakery and bake-off unit produced a wide variety of products, for example, bagels, baguettes, bread rolls, breads, buns, pasties, pastries, and pies. In the industrial bakery, the main products were filled and non-filled shaped buns, but cakes and gateaux were also produced.

Regarding traditional bakery 1 (Paper I), the facility included a main production and packaging unit and an outlet store. Traditional bakery 2 (Paper II) had two floors, including a main production unit upstairs, and a confectionary unit downstairs. A packaging unit and an outlet store also existed upstairs besides the main production unit. The industrial bakery (Paper II) comprised three units: a bun-baking (including two production lines), confectionary, and packaging unit. The in-store bakery and bake-off unit (Paper III) were located in a hypermarket and supermarket, respectively. In the bake-off-unit, partially baked and frozen products (parbaked products) were oven-baked inside the store.

There was one dough maker in traditional bakery 1 and the industrial bakery. These workers also occasionally participated in other work tasks besides dough-making (weighing and mixing ingredients). Two dough makers worked in traditional bakery 2, one on a night shift and another on a day shift. They also contributed to other work tasks. In the in-store bakery, all workers participated in dough-making and other work tasks. This thesis uses the term ‘general baker’ for workers who actively contributed to various tasks (including dough-making). The term ‘dough maker’ is used for workers who predominantly focused on dough-making. Considering the in-store bake-off unit, two persons worked in the facility, one in the morning and early afternoon, and another in the late afternoon and evening.

In each facility, the duration of the workers’ (concerning all job titles) working days was eight hours on average. The workers usually brushed the floor and tables each day at the end of the work shifts in most of the facilities. Regarding personal protective equipment (PPE), the workers wore working clothes and caps in all facilities, but respiratory protective equipment (RPE), goggles or gloves were not worn.

Mechanical ventilation existed in all facilities, except in traditional bakery 2, which had natural ventilation. Considering traditional bakery 1, there were hoods above the ovens and the doughnut baking spot, and the devices were connected to a local dust control system with flexible hoses. In the industrial bakery, the bakery machines were connected to the ventilation system. In the in-store bakery and bake-off unit, local exhaust ventilation (LEV) was connected to trolley ovens.

4.2 Indoor air measurements

The indoor air measurements were conducted on three consecutive working days in each facility, except in the industrial bakery (Paper II). Because of schedule reasons of the industrial bakery, a pre-intervention study included two consecutive measurement days, and then a day two weeks later; the post-intervention study included three consecutive measurement days (see Section 4.2.2). In traditional bakery 1 (Paper I), two separate measurement campaigns (both including three measurement days) were performed. Considering the in-store bake-off unit (Paper III), the measurements were conducted in the morning and early afternoon on each measurement day.

The mass concentrations (C_m) (Papers I–III), number concentrations (C_n) (Papers I and III), and number size distribution ($dN/dlogD_p$) (Papers I and III) of particulate matter were examined, as well as the chemical composition and morphology of particles (Paper I), and concentrations (C) of organic chemicals (VOCs and carbonyls) (Paper III).

4.2.1 Particulate matter

The measurements of particulate matter were performed in the breathing zone (BZ) and at stationary locations (S). The sampling locations of each facility are described in Table 5.

Table 5. Sampling locations for the measurements of particulate matter in the facilities.

Study	Study facility	Breathing zone (BZ)	Stationary location (S)
Paper I	Traditional bakery 1	Dough maker (BZ1)	Baking area (S1), oven area (S2), flour depository (S3)
Paper II	Traditional bakery 2	General baker (BZ2)	Beside a dough divider (S4)*, beside a dough roller (S5)*
Paper II	Industrial bakery	Dough maker (BZ3), line worker (BZ4)	Beside a production line (S6), working area of the dough maker (S7)
Paper III	In-store bakery	General baker (BZ5)	Beside a flour mixer tub and flour feeder (S8), beside a baking table and showcase (S9), near a baking table and trolley ovens (S10)
Paper III	In-store bake-off unit	NA	Beside a table near trolley ovens (S11), Beside a showcase (S12)

* = Near a baking table

NA = Not applicable (personal samples were not collected at the request of a supermarket manager)

Both gravimetric sampling and real-time monitoring were used in the measurements. Figure 1 illustrates the layouts of the facilities and stationary sampling locations. Table 6 shows a list of instruments used in the facilities.

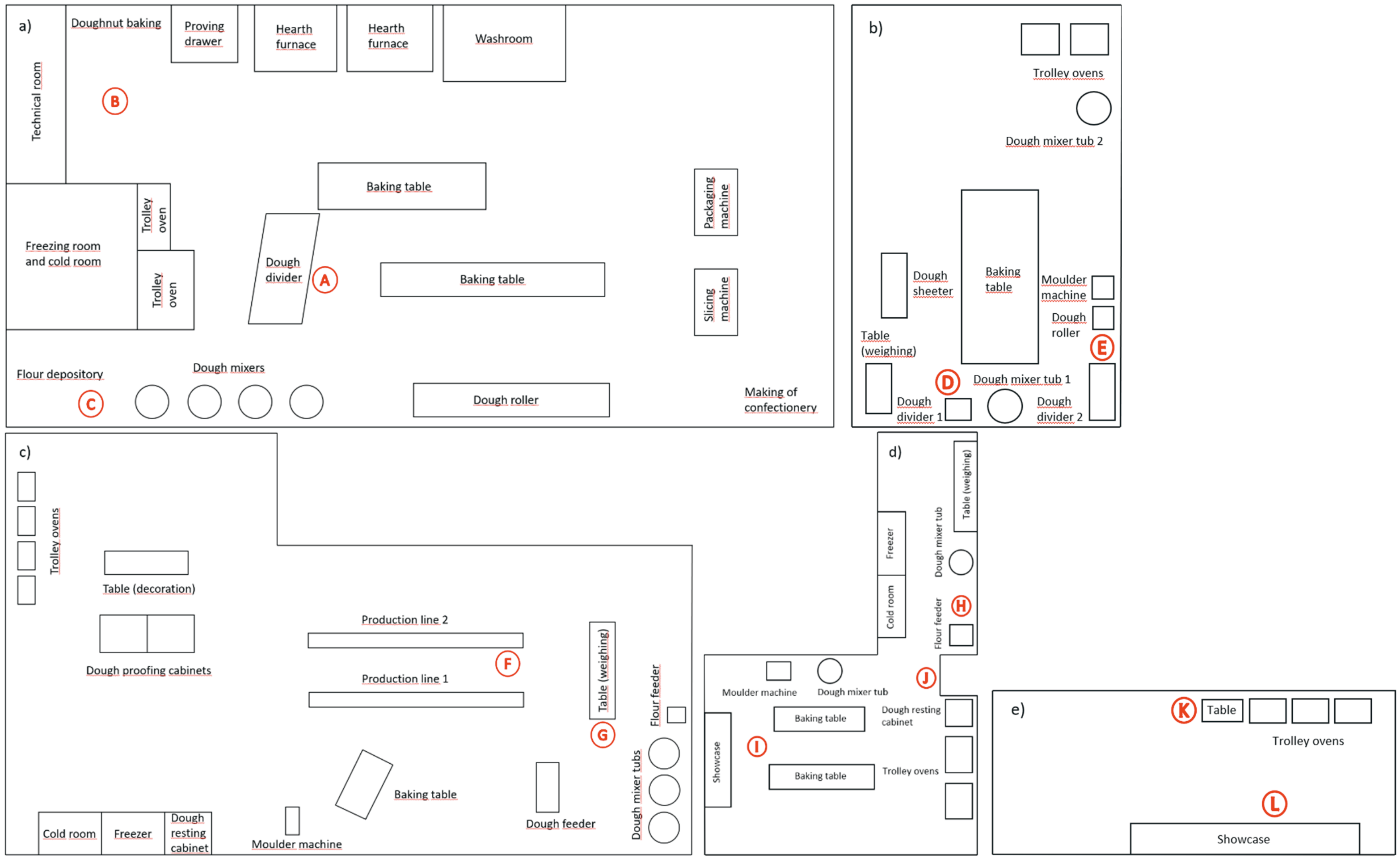
a)
Technical room
Doughnut baking
B
Proving drawer
Hearth furnace
Hearth furnace
Washroom
Freezing room and cold room
Trolley oven
Trolley oven
Dough divider
A
Baking table
Baking table
Packaging machine
Slicing machine
Flour depository
C
Dough mixers
Dough roller
Making of confectionery
b)
Trolley ovens
Dough mixer tub 2
Dough sheeter
Baking table
Moulder machine
Dough roller
E
Table (weighing)
D
Dough mixer tub 1
Dough divider 1
Dough divider 2
c)
Trolley ovens
Table (decoration)
Dough proofing cabinets
Production line 2
Production line 1
F
Table (weighing)
G
Flour feeder
Dough mixer tubs
Baking table
Dough feeder
Moulder machine
Cold room
Freezer
Dough resting cabinet
d)
Freezer
Cold room
Dough mixer tub
Table (weighing)
H
Flour feeder
J
Moulder machine
Dough mixer tub
Baking table
Dough resting cabinet
Showcase
I
Baking table
Trolley ovens
e)
K
Table
Trolley ovens
L
Showcase

Figure 1. Baking facility of the (a) traditional bakery 1, (b) traditional bakery 2, (c) industrial bakery (bun-baking unit), (d) in-store bakery, and (e) in-store bake-off unit, and stationary sampling locations: A = baking area (S1), B = oven area (S2), C = flour depository (S3), D = beside a dough divider (S4), E = beside a dough roller (S5), F = beside a production line (S6), G = working area of the dough maker (S7), H = beside a flour mixer tub and flour feeder (S8), I = beside a baking table and showcase (S9), J = near a baking table and trolley ovens (S10), K = beside a table near trolley ovens (S11), L = beside a showcase (S12).

Table 6. Instruments used for measuring particulate matter in the facilities.

Instrument	Parameter	Unit	Size range (µm)	Flow rate (L/min)	Study	Sampling locations
DustTrak DRX 8533[a] (TSI Inc.)	C_m	mg/m^3	0.5–15	2.0	I, II	S1, S5, S7
IOM sampler[b] (SKC Inc.)	C_m	mg/m^3	< 100	2.0	I–III	BZ1–5, S1, S4–7, S8, S9, S11, S12
OPS 3330[a] (TSI Inc.)	C_m	mg/m^3	0.3–10	2.0	III	S10, S11
PM_1 impactor[b] (Dekati Ltd.)	C_m	mg/m^3	< 1	10	I	S1, S2
TEOM 1405[a] (Thermo Scientific)	C_m	mg/m^3	< 100	2.0	I	S1
CPC 3022A[a] (TSI Inc.)	C_n	cm^{-3}	0.007–3	1.5	III	S10
CPC 3775[a] (TSI Inc.)	C_n	cm^{-3}	0.004–3	1.5	I	S1, S2
CPC 3776[a] (TSI Inc.)	C_n	cm^{-3}	0.0025–3	1.5	I	S1, S3
P-Trak 8525[a] (TSI Inc.)	C_n	cm^{-3}	0.02–1	0.7	III	S11
FMPS 3091[a] (TSI Inc.)	$dN/dlogD_p$	cm^{-3}	0.006–0.5	10	I, III	S1, S2 S10
SMPS[a] (TSI Inc.)	$dN/dlogD_p$	cm^{-3}	0.01–0.7	0.3	I	S1
EM[b]*	morphology	NA	NA	0.3–0.4	I	BZ1, S1

[a] = Real-time monitoring
[b] = Gravimetric sampling
* = Flow rate of an aspiration sampler was 0.3 L/min in campaign 1 and 0.4 L/min in measurement campaign 2.
BZ = breathing zone, C_m = mass concentration, C_n = number concentration, EM = electron microscope, NA = not applicable S = stationary location.

Full-shift dust samples (C_m of inhalable dust, aerodynamic diameter D_{ae} < 100 μm) were collected on nitrocellulose filters (0.8 μm AAWP, Merck KGaA, Germany) using Institute of Occupational Medicine (IOM) samplers (SKC Inc., USA) and sampling pumps (SKC AirChek Sampler, Model 224-PCXR4, SKC Inc., USA). Sampling was conducted at BZ and S (Papers I–III) (Table 6). In traditional bakery 2 (Paper II), the measurements were only performed during the morning shift. The IOM samplers consisted of stainless-steel cassettes and plastic housing bodies. The pumps were calibrated using a mini-BUCK Calibrator Model M-5 (A.P. Buck Inc., USA). The sampling height at S was approximately 1.4 m.

In traditional bakery 1 (Paper I), the IOM samples were collected during measurement campaign 2. A customized pre-cyclone with a cut-point of approximately 0.5 μm was also used with the IOM sampler to determine the C_m of $PM_{0.5}$. At BZ1 and S1, two parallel IOM samplers were used, one with the pre-cyclone installed. Furthermore, PM_1 samples were collected at S1 and S2 on 47 mm Teflon filters (Teflo R2PJ047, Pall Life Sciences, USA) and quartz fiber filters (Tissuquartz 2500QAT-UP, PALL Life Sciences) in two parallel lines during measurement campaign 1. Particles of > 1 μm were cut off using a pre-impactor (Dekati Ltd., Finland).

Real-time C_m was measured using a DustTrak DRX aerosol monitor 8533 (DRX) (TSI Inc., USA) (Papers I and II), a tapered element oscillating microbalance (TEOM 1405, Thermo Scientific, USA) (Paper I), and an optical particle sizer (OPS) 3330 (TSI Inc., USA) (Paper III) at S (Table 6). Monitoring was conducted at 10-s (DRX and TEOM in Paper I, OPS in Paper III) and 30-s (DRX in Paper II) intervals. The devices were positioned on a table (DRX and OPS) and on the floor (TEOM). Considering the DRX and OPS, the sampling height was approximately 0.8 m.

The DRX measured the following particulate matters simultaneously: PM_1 (D_{ae} < 1 μm), $PM_{2.5}$ (D_{ae} < 2.5 μm), PM_4 (D_{ae} < 4 μm), PM_{10} (D_{ae} < 10 μm), and PM_{15} (D_{ae} < 15 μm) size fractions. During measurements, a gravimetric sample was collected simultaneously on a nitrocellulose filter (37 mm AAWP, Merck KGaA, Germany) placed on the filter body inside the DRX. The OPS measured the following size fractions simultaneously: 0.3–0.5 μm, 0.5–1 μm, 1–3 µm, 3–5 µm, 5–8 µm, 8–10 µm, and PM_{10} (D_{ae} < 10 µm). Regarding the TEOM, the pre-cyclone was attached to the device to measure real-time C_m of nanoparticles (D_{ae} < 100 nm).

The filters (nitrocellulose, Teflon, and quartz fiber) were weighed using a Mettler-Toledo MT5 microbalance (Mettler-Toledo, LLC, USA) before and after sampling in an acclimatization room where the filters were stabilized for approximately 24 hours (relative humidity 30%, 21 °C). Static charges on the filters were eliminated using a Statickmaster 2U500 Alpha Ionizer (StaticTek, USA). The pre-sampling and post-sampling weights of the filters were measured after 24 hours of conditioning in the acclimatization room. For the DRX, a correction factor for C_m was calculated as a quotient of the filter sample C_m and an average C_m of the PM_{15} obtained from the device.

Real-time C_n was monitored using condensation particle counters (CPC, TSI Inc., USA) and a P-Trak ultrafine particle counter 8525 (TSI Inc., USA) at S (Table 6). Concerning the CPCs, the following models were used: CPC 3775 (Paper I), CPC 3776 (Paper I), and CPC 3022A (Paper III). Number size distribution (dN/dlogD_p) was measured using a fast mobility particle sizer (FMPS 3091, TSI Inc., USA) (Paper I and III) and a scanning mobility particle sizer (SMPS, TSI Inc., USA) (Paper I) at S (Table 6). The SMPS system included a pre-impactor, charger (Kr-85), platform (Model 3080), differential mobility analyzer (Model 3081), and CPC 3776. The FMPS and SMPS were positioned on the floor in the facilities. The CPC 3775 and CPC 3776 were placed above the FMPS and SMPS in traditional bakery 1 (Paper I), respectively. In the in-store bakery (Paper III), the CPC 3022A was placed above the FMPS. Monitoring was conducted at 1 s (Paper I) and 10 s intervals (Paper III).

Alcohol-based CPCs, including butanol as a working fluid, were used. Alcohol vapors were eliminated from the air using plastic hoses connected to the CPCs' exhaust outlets. No exhaust outlet existed in the in-store bake-off unit (Paper III), and it was not possible to insert a plastic hose into the LEV connected to the trolley ovens, so the CPC was not used in that facility. Furthermore, the FMPS was not used in the bake-off unit because of space issues. In the bake-off unit, the P-Trak was used at S11 (Table 6) instead to measure the C_n. The device was positioned on a table, and the sampling height was approximately 0.8 m.

In traditional bakery 1 (Paper I), the DRX, TEOM, CPCs, FMPS, and SMPS operated constantly for three consecutive days. In traditional bakery 2 (Paper II), the DRX constantly measured for three and two consecutive days during the pre- and post-intervention study, respectively. In the industrial bakery (Paper II), the DRX operated constantly for two and three consecutive days during the pre- and post-intervention study, respectively. There was a technical problem with the device on one day in both traditional bakery 2 and the industrial bakery, so the monitoring time varied between two and three days in the bakeries. In the in-store bakery (Paper III), the OPS monitored constantly for one day since the device had a technical problem on measurement days 2 and 3. The CPC and FMPS operated constantly for three consecutive days. Considering the in-store bake-off unit (Paper III), the OPS was used for approximately six hours on each measurement day. The P-Trak was used on two measurement days with a daily monitoring time of four and six hours.

Electron microscopy (EM) samples (Paper I) were collected on porous carbon films (S147-4400 Holey Carbon Film 400 Mesh Cu, Agar Scientific, USA) using an aspiration sampler designed and built by Lyyränen et al. (2009) at BZ1 and S1 (Table 6). The pre-impactor of the SMPS was attached to the aspiration sampler to cut off particles with $D_{ae} > 0.6$ µm.

4.2.2 Intervention study

Paper II examined the effectiveness of intervention strategies focused on working methods to control mass concentrations and peak exposures of flour dust. Two bakeries were selected for the study considering the size of the bakery and the number of workers: an industrial bakery and a traditional bakery. The degree of automation was high in the industrial bakery; however, the pre-intervention working methods in the dough-making process were similar in both bakeries. The C_m was measured near the dough-making area (Tables 5 and 6) pre- and post-intervention in both bakeries.

Work activities associated with peak exposures and various other work activities that required improvement to control dust levels in the bakeries were identified during the pre-intervention study. The intervention strategies to control flour dust exposure were planned and implemented using checklists developed by Säämänen et al. (2012) for dust reduction in various working activities. Relevant strategies for the bakeries were selected from the checklists with the bakery managers.

In both bakeries, the workers aimed to follow the following intervention strategies to control flour dust exposure after the pre-intervention study: Ingredients added to bowls/dough mixers at as low a height as possible, empty bags handled with care to avoid dusting, bags emptied holding their mouth near to the bottom of the bowls/tubs, and empty bags flattened outside the work area. The following strategies were applied only in the industrial bakery: floor and surfaces cleaned immediately when weighing ingredients, and floor and surfaces cleaned immediately when handling bags. One strategy the workers aimed to follow only in the traditional bakery: flour thrown from as low a height as possible onto the baking table. The bakery managers introduced the checklists and intervention measures to the workers and trained them to follow the new working methods.

Both bakeries were visited once to check and reinforce intervention adherence before the post-intervention measurements. The interventions were implemented for approximately 6 months in the industrial bakery and 3.5 months in the traditional bakery. Because of schedule reasons in the bakeries, different follow-up times were selected. The follow-up times were relatively short since the intervention strategies were assumed to be readily adopted. Therefore, the effect of the intervention was expected to be seen in a few months in both bakeries.

4.2.3 Organic chemicals

The measurements of organic chemicals (VOCs and carbonyls) were conducted at BZ and S (Paper III). The sampling locations are described in Table 7.

Table 7. Sampling locations in the breathing zone (BZ) and at stationary locations (S) for the measurements of organic chemicals in the facilities.

Sampling location	In-store bakery		In-store bake-off unit	
	VOCs	Carbonyls	VOCs	Carbonyls
General baker (BZ5)	x	–	–	–
Beside a flour mixer tub and flour feeder (S8)	x	x	–	–
Beside a baking table and showcase (S9)	x	–	–	–
Beside a table near trolley ovens (S11)	–	–	x	x
Beside a showcase (S12)	–	–	x	–

The VOC samples were collected and analyzed according to the ISO-6:2021 standard. Tenax TA adsorption tubes (Sigma Aldrich Corporation, USA), which included 200 mg sorbent, and SKC AirChek 3000 pumps (SKC Inc., USA) with a calibrated air flow of 200 ml/min were used for the sampling. The pumps were calibrated using the mini-BUCK Calibrator Model M-5 (A.P. Buck Inc., USA). The sampling was conducted at BZ5 and S8–9 in the in-store bakery, and at S11–12 in the bake-off unit. Ten samples were collected in the bakery, including four at BZ and six at S. Two background samples (B1) were collected in a café next to the bakery. In the bake-off unit, six samples were collected. Personal samples were excluded at the request of the supermarket manager. Two background samples (B2) were collected in the store beside the bake-off unit. In both facilities, the sampling time was one hour, and the sampling was conducted at a height of 1.4 m for each sample.

Short-chained (< C_6) carbonyls were collected and analyzed according to the ISO 16000-3:2011 standard. Waters Sep-Pak 2,4-dinitrophenylhydrazine (DNPH) Silica cartridges (Waters Corp., USA), which contained 350 g of sorbent, and a Sartorius 16692 diaphragm vacuum pump (Sartorius AG, Germany) with a calibrated air flow of 1.5 L/min were used for sampling. The pump was calibrated using the mini-BUCK Calibrator Model M-5 (A.P. Buck Inc., USA). One full-shift sample was collected at a height of 1.4 m in the in-store bakery (S8) and bake-off unit (S11).

4.3 Analytical methods

Concentrations of organic carbon (OC) and elemental carbon (EC) were determined from quartz fiber filters (S1, n = 2) using a thermal-optical carbon analyzer (Sunset Laboratory, USA) (Paper I). The analyses were conducted according to the National Institute for Occupational Safety and Health (NIOSH) method 5040. Other 31 elements (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Sb, Se, Sr, Th, Ti, Tl, U, V, and Zn) were analyzed from the Teflon filter (S1, n = 1) using inductively coupled plasma mass spectrometry (ICP-MS) according to the ISO 17294-2:2003 standard. Six water-soluble ions (Br^-, Cl^-, NO_3^-, SO_4^{2-}, F^- and PO_4^{3-}) were determined from the Teflon filter (S2, n = 1) using ion chromatography (IC) according to the ISO 10304-1:2007 standard. Furthermore, the 31 above-mentioned elements were also determined from two IOM filters (BZ1 with and without pre-cyclone) using ICP-MS according to the ISO 17294-2:2003 standard.

The morphology and chemical composition of the particles were examined from the porous carbon films (S1) using a scanning electron microscope (SEM, Sigma HD VP, Carl Zeiss NST, UK) connected to two energy-dispersive X-ray spectroscopy detectors (EDS, Thermo Scientific, USA). SEM imaging was conducted at an accelerating voltage of 2 kV using SE2 and InLens detectors. An accelerating voltage of 10 kV was used in the EDS analyses. Considering the PM_1 samples, the OC and EC and 31 above-mentioned elements were not analyzed from the S2 samples. The water-soluble ions were not determined from the S1 samples.

A thermal desorption-gas chromatography-mass spectrometer (TD-GC-MS) was used to analyze the VOC samples. The TD-GC-MS consisted of a TD-100 thermal desorber (Markes International Ltd., UK), 7890A gas chromatograph (Agilent Technologies Inc., USA), and 5975C mass selective detector (Agilent Technologies Inc., USA) (Paper III). The device included an HP-5MS UI (60 m x 0.25 mm x 0.25 µm) column. The Tenax TA adsorption tubes were treated with +280 °C helium for 10 min to extract the collected compounds. Before analyzing the samples, blank samples and autotune were run to ensure the absence of contamination.

The compounds were identified using an MSD ChemStation program (version F.01.00.1903, Agilent Technologies Inc., USA) that included an MS library, NIST 20 (National Institute of Standards and Technology, USA). Standards were prepared for both VOC sample sets (all samples from the in-store bakery and bake-off unit) using a standard solution (HC 48 Component Indoor Air Standard, Supelco Inc., USA). The standard solution comprised 48 compounds with a concentration of 50 ng/µL dissolved in methanol. The standards were analyzed together with the VOC samples, and standard curves were constructed. Concerning quantification, the toluene-equivalent method was used.

The DNPH derivatives of short-chained carbonyl compounds were analyzed using a liquid chromatography-tandem mass spectrometer (LC-MS/MS) system that consisted of Nexera X2 LC-30AD pump, Nexera X2 SIL-30AC autosampler, DGU-20A5R degassing unit, CTO-20AC column oven, LCMS-8040 triple quadrupole mass spectrometer (all manufactured by Shimadzu Corp., Japan), and Kinetex® reversed-phase C18 column (1.7 µm x 100 mm x 3 mm internal) (Phenomenex Inc., USA) (Paper III). Water and acetonitrile were used as eluents. Four-point standard curves were constructed for the following compounds to quantify the collected compounds selectively: 2-butanone, acetaldehyde, acetone, acrolein, butyraldehyde, crotonaldehyde, formaldehyde, methacrolein, propionaldehyde, and valeraldehyde. A Carbonyl-DNPH Mix 1 (Sigma Aldrich Corporation, USA), which included the DNPH derivatives of the above-mentioned compounds, was used as reference material. LabSolutions Insight software (Shimadzu Corp., Japan) was used to identify the individual compounds by their retention times and ions.

4.4 Data analysis

Arithmetic means (AM) for the C_m of the IOM (Papers I–III) and PM_1 (Paper I) samples (averaged over the repeat full-shift samples), and for the CPC (Papers I and III), DRX (Papers I–III), OPS (Paper III), P-Trak (Paper III), and TEOM (Paper I) results (averaged over the complete dataset comprising the C_m and C_n of the daily working hours) were calculated. Geometric means (GM) and geometric standard deviations (GSD) were calculated for the IOM, CPC, DRX, OPS, and P-Trak results. For the CPCs (traditional bakery 1, in-store bakery), DRX (traditional bakery 1 and 2, industrial bakery), OPS (traditional bakery 1, in-store bakery), and TEOM (traditional bakery 1), an AM was also calculated over the complete dataset including the daily C_m and C_n outside working hours (average background concentration). Regarding FMPS (traditional bakery 1 and in-store bakery), an average number size distribution ($dN/dlogD_p$) was determined for various work phases (averaging periods were 10–60 min).

The mass percentages of carbon were calculated for the gravimetric PM_1 samples (S1) (Tables 5 and 6) in Paper I. The mass percentages and C_m of the other 31 elements (excluding water-soluble ions) were determined for the PM_1 (S1) and IOM (BZ1 with and without pre-cyclone) samples. Moreover, the mass percentages and C_m of the six water-soluble ions were also calculated for the PM_1 (S2) samples.

The effectiveness of the intervention was assessed by comparing the AM of the pre- and post-intervention C_m of the IOM samples and DRX results in Paper II. Post-intervention percentages were calculated from the average C_m results (either an increase or reduction in the average C_m). The average pre- and post-intervention proportions of various size fractions of the PM_{15} were calculated by dividing the C_m of each size fraction by the C_m of the PM_{15} at each time point and averaging over the complete dataset. In Paper II, peak exposures measured using the DRX were also examined pre- and post-intervention. Peak exposure was defined as C_m > 2 mg/m^3, and the duration of the C_m peaks was computed by considering the peaks of > 2 mg/m^3 of the PM_{15} size fraction during each measurement day.

In Paper III, the concentration (C) of the individual VOCs was calculated as toluene equivalents for each sampling location (Table 5). The C of TVOC for each sampling location was computed by summing the concentrations of compounds with retention times between n-hexane (C_6) and n-hexadecane (C_{16}). In this study, all the compounds with the retention time between C_6 and C_{16} (including aldehydes) were classified as VOCs. Average concentrations were calculated for the compounds of each sampling location, and then the compounds were sequenced alphabetically. Based on the concentrations, the ten most dominant compounds detected in the sample sets of each sampling location were considered when compiling the VOC results. Furthermore, the compounds with a prevalence of ≥ 50% in the sampling locations were considered for the compilation. Eventually, the compounds were compiled into one table (see Section 5.2.1). Concerning short-chained (< C_6) carbonyls, which were collected using the DNPH Silica cartridges, concentrations (C) were calculated using the compounds' own response factors.

5 Results

5.1 Particulate matter

5.1.1 Mass concentrations: Gravimetric sampling (Papers I–III)

Table 8 shows the results of the full-shift inhalable dust (IOM) samples collected at BZ and S. In the bakeries, the C_m was 0.3–15.1 mg/m^3 in the breathing zone (BZ) and 0.1–3.0 mg/m^3 at stationary locations (S), whereas in the in-store bake-off unit where flours were not used, the C_m was 0.1 mg/m^3. Considering the $PM_{0.5}$ particles collected using the IOM sampler with the pre-cyclone in traditional bakery 1, the C_m made up of 9–15wt% and 4–8wt% of inhalable dust at BZ1 and S1, respectively.

Table 8. Mass concentrations (C_m) of the IOM samples collected in the breathing zone (BZ) and at stationary locations (S).

Sample	C_m (mg/m^3)				
	n	**AM**	**GM**	**GSD**	**Range**
Traditional bakery 1[a]					
BZ1	3	9.5	8.9	1.5	6.8–14.5
BZ1 pre-cyclone	2	0.8	0.8	1.5	0.6–1.1
S1	3	2.5	2.5	1.1	2.3–2.6
S1 pre-cyclone	3	0.2	0.1	1.4	0.1–0.2
Traditional bakery 2[b]					
BZ2	3	11.6	11.3	1.3	9.4–15.1
S4	3	2.6	2.6	1.2	2.1–3.0
S5	3	0.8	0.7	1.4	0.6–1.1
Industrial bakery[b]					
BZ3	3	1.6	1.6	1.2	1.3–2.0
BZ4	2	0.4	0.4	1.5	0.3–0.5
S6	3	0.4	0.4	1.4	0.3–0.5
S7	3	0.5	0.5	1.7	0.3–0.7
In-store bakery					
BZ5	3	5.4	5.3	1.2	4.5–5.9
S8	3	0.4	0.3	1.6	0.2–0.5
S9	3	0.2	0.2	2.8	0.1–0.4
In-store bake-off unit					
S11	3	0.1	0.1	1.6	0.1–0.1
S12	3	0.1	0.1	1.2	0.1–0.1

[a] = Measurements were conducted during measurement campaign 2.
[b] = Pre-intervention C_m is presented.
AM = arithmetic mean, BZ1 = dough maker, BZ2 = general baker, BZ3 = dough maker, BZ4 = line worker, BZ5 = general baker, GM = geometric mean, GSD = geometric standard deviation, IOM = Institute of Occupational Medicine, n = number of samples, S1 = baking area, S2 = oven area, S3 = flour depository, S4 = beside a dough divider (near a baking table), S5 = beside a dough roller (near a baking table), S6 = beside a production line, S7 = working area of the dough maker, S8 = beside a flour mixer tub and flour feeder, S9 = beside a baking table and showcase, S10 = beside a baking table and trolley ovens, S11 = beside a table near trolley ovens, S12 = beside a showcase.

In traditional bakery 1, three PM_1 samples were collected on Teflon filters during measurement campaign 1. The C_m was 0.1 and 0.2 mg/m^3 at S1 and 0.2 mg/m^3 at S2.

5.1.2 Mass concentrations: Real-time monitoring (Papers I–III)

The real-time C_m of PM_{10} size fraction measured using the DRX and OPS at stationary locations (S) is presented in Table 9. The C_m varied at < 0.1–28.3 and < 0.1–0.1 mg/m^3 in the bakeries and in-store bake-off unit, respectively. For all facilities, the average C_m (arithmetic mean) was low, and ranged from < 0.1 to 1.0 mg/m^3. The average C_m was 40 μg/m^3 in the in-store bakery and 30 μg/m^3 in the in-store bake-off unit. In the bakeries, the average background C_m measured outside working hours was < 0.1 mg/m^3. The background C_m was not measured in the in-store bake-off unit.

Table 9. Real-time mass concentration (C_m) of PM_{10} size fraction measured using the DRX and OPS at stationary locations (S) during working hours.

Location	C_m (mg/m^3)			
	AM	GM	GSD	Range
Traditional bakery 1[a]				
S1 (DRX)	0.4	1.4	1.3	< 0.1–2.3
Traditional bakery 2[b]				
S5 night shift (DRX)	1.0	0.3	4.9	< 0.1–28.3
S5 morning shift (DRX)[c]	0.3	0.1	3.3	< 0.1–25.0
Industrial bakery[b]				
S7 (DRX)[c]	0.3	0.4	2.5	< 0.1–8.1
In-store bakery				
S10 (OPS)	< 0.1	< 0.1	4.2	< 0.1–2.5
In-store bake-off unit				
S11 (OPS)	< 0.1	< 0.1	1.5	< 0.1–0.1

[a] = Measurements were conducted during measurement campaign 2.
[b] = Pre-intervention C_m is presented.
[c] = Measurements were performed on two working days.
AM = arithmetic mean, DRX = DustTrak DRX aerosol monitor 8533, GM = geometric mean, GSD = geometric standard deviation, OPS = optical particle sizer 3330, PM_{10} = size fraction with aerodynamic diameter D_{ae} < 10 μm, S1 = baking area, S5 = beside

a dough roller (near a baking table), S7 = working area of the dough maker, S10 = beside a baking table and trolley ovens, S11 = beside a table near trolley ovens.

Figure 2 illustrates the real-time mass concentration (C_m) time series of the PM_{10} size fraction measured using the DRX and OPS at stationary locations (S) during working hours and all measurement days when the monitoring was conducted in each facility. In the traditional bakeries (Figures 2a–c) and industrial bakery (Figure 2d), the C_m fluctuated significantly during the measurement days, and several peak concentrations were detected when dusty work phases were ongoing. The daily peak C_m in each stationary location was 1.1–2.3 mg/m^3 (S1, Figure 2a), 7.0–25.0 mg/m^3 (S5, morning shift, Figure 2b), 14.8–28.3 mg/m^3 (S5, night shift, Figure 2c), and 4.3–8.1 mg/m^3 (S7, Figure 2d). However, only a few C_m peaks ranging between 0.5 and 2.5 mg/m^3 (S10) were observed in the in-store bakery (Figure 2e). In the in-store bake-off unit (Figure 2f), the C_m remained predominantly below 0.1 mg/m^3.

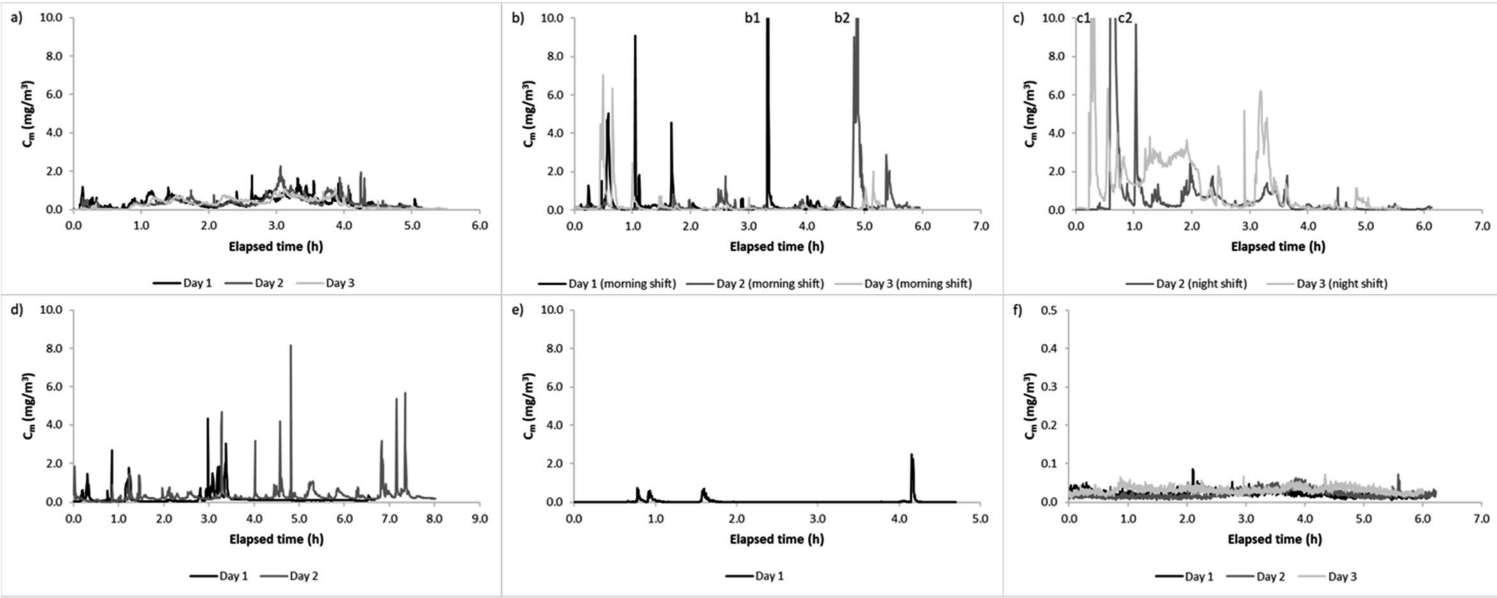

Figure 2. Real-time mass concentration (C_m) time series of the PM_{10} size fraction measured using the DRX during working hours at (a) S1 (traditional bakery 1, measurement campaign 2), (b) S5 (traditional bakery 2, pre-intervention, morning shift), (c) S5 (traditional bakery 2, pre-intervention, night shift), (d) S7 (industrial bakery, pre-intervention), and using the OPS at (e) S10 (in-store bakery) and (f) S11 (in-store bake-off unit). Note: Peak C_m is 25.0 mg/m^3 (b1), 15.2 mg/m^3 (b2), 14.8 mg/m^3 (c1), and 28.3 mg/m^3 (c2). DRX = DustTrak DRX aerosol monitor 8533, OPS = optical particle sizer 3330, PM_{10} = size fraction with aerodynamic diameter D_{ae} < 10 μm, S1 = baking area, S5 = beside a dough roller (near a baking table), S7 = working area of the dough maker, S10 = beside a baking table and trolley ovens, S11 = beside a table near trolley ovens.

The TEOM was used on three measurement days at S1 in traditional bakery 1, two days with the pre-cyclone and one day without the pre-cyclone during measurement campaign 2. The average C_m was 0.2 mg/m^3 when the pre-cyclone was applied and 0.5 mg/m^3 when the pre-cyclone was not used. At the beginning of dough-making and when several work phases were ongoing, the real-time C_m was notably greater compared to the DRX; however, during oven operations, the difference was slighter. Furthermore, the difference between the real-time C_m measured using the TEOM with and without the pre-cyclone leveled when small particles stemming from the oven operations were dominant in the bakery air.

5.1.3 Chemical composition and morphology of particles (Paper I)

Large flour dust particles, comprising predominantly phosphorus (P), potassium (K), nitrogen (N), sulfur (S), carbon(C), and oxygen (O), were found particularly in the BZ samples in the SEM images and EDS analyses (see Figure of the SEM images in Appendix 1, Paper 1). Small, agglomerated flour dust particles, spherical particles, and soot agglomerates were also observed in the samples. The agglomerated flour dust particles contained mainly carbon and small amounts of silicon (Si) or sulfur, and also included nanosized particles. The spherical particles consisted of, for example, nitrogen, silicon, phosphorus, sulfur, potassium, sodium (Na), calcium (Ca), and chlorine (Cl).

Considering the 31 elements (excluding carbon and water-soluble ions) analyzed using the ICP-MS, the C_m was < 0.1–0.7 µg/m^3 at S1 (PM_1), < 0.1–27.0 µg/m^3 at BZ1 without the pre-cyclone (inhalable dust), and < 0.1–3.7 µg/m^3 at BZ1 with the pre-cyclone ($PM_{0.5}$ particles). Carbon, of which at least 99% was organic according to an analysis conducted using the thermal-optical carbon analyzer, made up 42–64% of the total mass of the PM_1 samples at S1. The other 31 elements made up 1.1% of the total PM_1 mass at S1, and the main elements were natrium (0.7 µg/m^3), potassium (0.6 µg/m^3), and bismuth (0.3 µg/m^3). In the IOM samples collected at BZ1, the proportion of the 31 elements was 0.4% in inhalable dust and 0.5% in $PM_{0.5}$ particles. The main elements were potassium (27.0 µg/m^3), calcium (12.5

μg/m^3), and sodium (12.0 μg/m^3) in inhalable dust, and calcium (3.7 μg/m^3), aluminum (Al) (0.4 μg/m^3), and magnesium (Mg) (0.3 μg/m^3) in $PM_{0.5}$ particles.

The C_m of the water-soluble ions, determined from the PM_1 filter samples collected at S2 and analyzed using the IC, were 0.5 μg/m^3 (Cl^-), 0.4 μg/m^3 (NO_3^-), 2.3 μg/m^3 (SO_4^{2-}), and 1.6 μg/m^3 (PO_4^{3-}). These ions comprised 2.2% of the total mass of the PM_1 samples. In the analysis, Br^- and F^- were not detected.

5.1.4 Intervention study (Paper II)

Table 10 presents the pre- and post-intervention results of the full-shift inhalable dust (IOM) samples and real-time monitoring conducted using the DRX in traditional bakery 2 and the industrial bakery. The IOM samples were collected in the breathing zone (BZ) and at stationary locations (S), and the real-time measurements were performed at S.

Table 10. Average pre- and post-intervention mass concentrations (C_m) measured using the IOM samplers and DRX in the breathing zone (BZ) and at stationary locations (S), and percentual differences in the average C_m.

Sample/size fraction	C_m (mg/m^3), AM (range)		D (%)
	Pre-intervention	**Post-intervention**	
Traditional bakery 2			
IOM[a]			
BZ2	11.6 (9.4–15.1)	14.4 (8.2–22.7)	+24
S4	2.6 (2.1–3.0)	1.6 (0.9–2.1)	–39
S5	0.8 (0.6–1.1)	1.2 (1.0–1.4)	+54
DRX (S5)			
PM_1 night shift	0.7 (0.0–12.5)*	1.2 (0.0–9.8)*	+73
PM_1 morning shift	0.1 (0.0–10.1)	0.4 (0.0–6.8)*	+217
$PM_{2.5}$ night shift	0.7 (0.0–12.5)*	1.2 (0.0–9.8)*	+73
$PM_{2.5}$ morning shift	0.1 (0.0–10.1)	0.4 (0.0–6.8)*	+216
PM_4 night shift	0.7 (0.0–12.7)*	1.2 (0.0–9.9)*	+71
PM_4 morning shift	0.1 (0.0–10.9)	0.5 (0.0–6.8)*	+208
PM_{10} night shift	1.0 (0.0–28.3)*	1.3 (0.0–15.0)*	+34
PM_{10} morning shift	0.3 (0.0–25.0)	0.6 (0.0–14.5)*	+130
PM_{15} night shift	1.4 (0.0–56.9)*	1.7 (0.0–36.7)*	+22
PM_{15} morning shift	0.4 (0.0–39.2)	0.8 (0.0–20.9)*	+93
Industrial bakery			
IOM[b]			
BZ3	1.6 (1.3–2.0)	2.1 (2.7–3.7)*	+28
BZ4	0.4 (0.3–0.5)*	0.7 (0.6–0.7)*	+55
S6	0.4 (0.3–0.5)	0.2 (0.2–0.3)	–45
S7	0.5 (0.3–0.7)	0.3 (0.2–0.5)	–40
DRX (S7)			
PM_1	0.2 (0.0–5.0)*	0.1 (0.0–12.1)	–22
$PM_{2.5}$	0.2 (0.0–5.0)*	0.1 (0.0–12.1)	–21
PM_4	0.2 (0.0–5.1)*	0.1 (0.0–12.1)	–22
PM_{10}	0.3 (0.0–8.1)*	0.2 (0.0–17.5)	–25
PM_{15}	0.4 (0.0–18.7)*	0.3 (0.0–33.1)	–20

[a] = Three samples at BZ and S were collected post-intervention.
[b] = Two samples at BZ and three samples at S were collected post-intervention.
* = Measurements were conducted on two working days.
AM = arithmetic mean, BZ2 = general baker, BZ3 = dough maker, BZ4 = line worker, D = difference, DRX = DustTrak DRX aerosol monitor 8533, GM = geometric mean, GSD = geometric standard deviation, IOM = Institute of Occupational Medicine, PM_1 = size fraction with D_{ae} < 1 μm, $PM_{2.5}$ = size fraction with aerodynamic diameter D_{ae}

< 2.5 μm, PM_4 = size fraction with D_{ae} < 4 μm, PM_{10} = size fraction with D_{ae} < 10 μm, PM_{15} = size fraction with D_{ae} < 15 μm, S4 = beside a dough divider (near a baking table), S5 = beside a dough roller (near a baking table), S6 = beside a production line, S7 = working area of the dough maker.

In traditional bakery 2, the average C_m (arithmetic mean) of inhalable dust decreased 39% at S4, whereas the average C_m increased 24% and 54% at BZ2 and S5, respectively. No reductions in the average C_m were achieved in the DRX results. Regarding the night shifts, the average C_m increased 22–73% in the PM_1, $PM_{2.5}$, PM_4, PM_{10} and PM_{15} size fractions. Considering the morning shifts, the average C_m increased 93–217% in the above-mentioned size fractions. In the industrial bakery, the average C_m of inhalable dust decreased 45% and 40% at S6 and S7, respectively, but increased 28% and 55% at BZ3 and BZ4, respectively. Concerning the DRX results, the average C_m decreased 20–25% in the above-mentioned size fractions.

Several peak exposures were measured in both bakeries during measurement days. The peak C_m of the PM_{10} size fraction is presented as an example in Figures 2b–d and 3a–c. In traditional bakery 2, the number of C_m peaks was, on average, greater during morning and night shifts post-intervention. The daily peak C_m was 7.0–25.0 mg/m^3 (morning shift) and 14.8–28.3 mg/m^3 (night shift) pre-intervention (Figures 2b–d), and 5.2–14.5 mg/m^3 (morning shift) and 6.9–15.0 mg/m^3 (night shift) post-intervention (Figures 3a–c). Regarding the duration of the C_m peaks (duration of peaks > 2 mg/m^3 for the PM_{10} size fraction is presented as an example hereafter), the shortest peaks were 30 s during both morning and night shifts pre- and post-intervention. The longest peaks were 7 min (morning shift) and 55 min (night shift) pre-intervention, and 17 min (morning shift) and 1 h 3 min (night shift) post-intervention.

In the industrial bakery, the number of C_m peaks was, on average, the same pre- and post-intervention. The daily peak C_m was 4.3–8.1 mg/m^3 pre-intervention (Figure 2d) and 11.1–17.5 mg/m^3 post-intervention (Figure 3c).

The shortest C_m peaks were 30 s pre- and post-intervention, whereas the longest peaks were 1 min pre- and post-intervention.

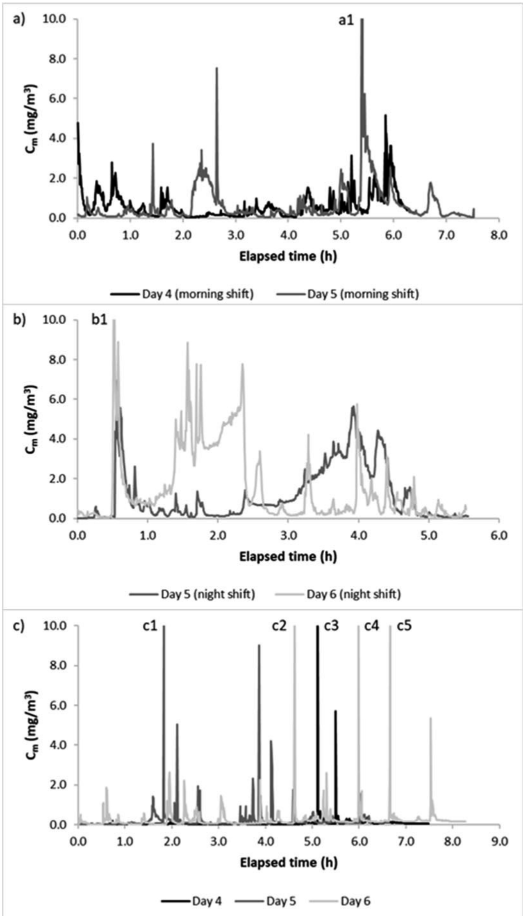

Figure 3. Real-time post-intervention mass concentration (C_m) time series of the PM_{10} size fraction measured using the DRX during working hours at (a) S5 (traditional bakery 2, morning shift), (b) S5 (traditional bakery 2, night shift), and (c) S7 (industrial bakery). Note: Peak C_m is 14.5 mg/m^3 (a1), 15.0 mg/m^3 (b1), 11.1 mg/m^3 (c1), 12.0 mg/m^3 (c2), 13.7 mg/m^3 (c3), 12.8 mg/m^3 (c4), and 17.5 mg/m^3 (c5). DRX = DustTrak DRX aerosol monitor 8533, PM_{10} = size fraction with aerodynamic diameter D_{ae} < 10 μm, S5 = beside a dough roller (near a baking table), S7 = working area of the dough maker.

Table 11 shows that the PM_{15} consisted mainly of the PM_1 (D_{ae} < 1 μm), $PM_{4–10}$ (4 μm < D_{ae} < 10 μm), and $PM_{10–15}$ (10 μm < D_{ae} < 15 μm) size fractions in both bakeries pre- and post-intervention. The PM_1 fraction clearly dominated, and the average proportions of the PM_{15} were 43% (traditional bakery 2, morning shift), 69% (traditional bakery 2, night shift), and 61% (industrial bakery) pre-intervention, whereas the corresponding proportions were 61%, 76%, and 58% post-intervention. The real-time measurements showed that when C_m of the PM_{15} was < 1 mg/m^3, the PM_1 fraction dominated, whereas when C_m of the PM_{15} was > 1 mg/m^3, the proportion of the $PM_{10–15}$ fraction was usually greater.

Table 11. Average pre- and post-intervention proportions (%) of the real-time mass concentrations (C_m) of the PM_{15} for PM_1 (aerodynamic diameter D_{ae} < 1 μm), $PM_{1–2.5}$ (1 μm < D_{ae} < 2.5 μm), $PM_{2.5–4}$ (2.5 μm < D_{ae} < 4 μm), $PM_{4–10}$ (4 μm < D_{ae} < 10 μm), and $PM_{10–15}$ (10 μm < D_{ae} < 15 μm) size fractions.

Study facility	**AM (%), pre-/post-intervention**				
	PM_1	**$PM_{1–2.5}$**	**$PM_{2.5–4}$**	**$PM_{4–10}$**	**$PM_{10–15}$**
Traditional bakery 2, morning shift	43/61	1/1	2/1	23/18	31/19
Traditional bakery 2, night shift	69/76	1/0	2/1	15/9	15/14
Industrial bakery	61/58	0/0	0/1	14/14	24/27

Note: The C_m was measured using the DRX at S5 (traditional bakery 2) and S7 (industrial bakery). AM = arithmetic mean, DRX = DustTrak DRX aerosol monitor 8533, PM = particulate matter, PM_{15} = size fraction with D_{ae} < 15 μm, S5 = beside a dough roller, S7 = working area of the dough maker.

Adherence to the intervention was controlled by a walk-through survey prior to the post-intervention measurements in the bakeries. After the pre-intervention measurements the implementation of the interventions strategies was left to the responsibility of the managers and employees. The walk-through survey showed that all the control measures were implemented frequently in traditional bakery 2. In the industrial bakery, the intervention strategies related to cleaning were not implemented; however, the other control measures were followed frequently, and the floor and surfaces were cleaned each day at the end of the work shifts.

5.1.5 Number concentrations (Papers I and III)

The real-time C_n measured using the CPCs and P-Trak at stationary locations (S) is presented in Table 12. The C_n was 1.0×10^3–4.1×10^6 cm^{-3} in the bakeries and 2.2×10^3–1.5×10^5 cm^{-3} in the in-store bake-off unit, respectively. Considering all facilities, the average C_n (arithmetic mean) varied between 1.3×10^4–3.3×10^5 cm^{-3}. In traditional bakery 1 and the in-store bakery, the average background C_n was approximately 1.0×10^3 and 8.5×10^3 cm^{-3}, respectively. The background C_n was not measured in the in-store bake-off unit.

Table 12. Real-time number concentration (C_n) of particulate matter measured using the CPCs and P-Trak at stationary locations (S) during working hours.

Location	C_n (cm^{-3})			
	AM	GM	GSD	Range
Traditional bakery 1				
S1 (CPC 3775)[a]	4.0×10^4	3.1×10^4	2.3	1.5×10^3–2.2×10^5
S1 (CPC 3776)[b]	1.1×10^5	6.9×10^4	3.5	3.7×10^2–5.6×10^5
S2 (CPC 3775)[a]	3.3×10^5	1.7×10^5	3.0	1.0×10^3–4.1×10^6
S3 (CPC 3776)[b]	1.0×10^5	8.7×10^4	1.7	2.5×10^4–5.5×10^5
In-store bakery				
S10 (CPC 3022A)	1.3×10^4	1.2×10^4	1.7	3.5×10^3–9.9×10^4
In-store bake-off unit				
S11 (P-Trak 8525)	2.6×10^4	1.8×10^4	2.3	2.2×10^3–1.5×10^5

[a] = Measurements were conducted during measurement campaign 1

[b] = Measurements were conducted during measurement campaign 1 and 2

AM = arithmetic mean, CPC = condensation particle counter, GM = geometric mean, GSD = geometric standard deviation, P-Trak = P-Trak ultrafine particle counter, S1 = baking area, S2 = oven area, S3 = flour depository, S10 = beside a baking table and trolley ovens, S11 = beside a table near trolley ovens.

Figure 4 depicts the real-time number concentration (C_n) time series measured using the CPCs and P-Trak at stationary locations (S) during working hours and all measurement days when the monitoring was performed in traditional bakery 1 (Figure 4a,b), the in-store bakery (Figure 4c), and the in-store bake-off unit (Figure 4d). The C_n varied widely during the measurement days in all facilities. The highest C_n peaks in each stationary location were 5.6×10^5 cm^{-3} (S1), 4.1×10^6 cm^{-3} (S2), 5.5×10^5 cm^{-3} (S3), 9.9×10^4 cm^{-3} (S10), and 1.5×10^5 cm^{-3} (S11).

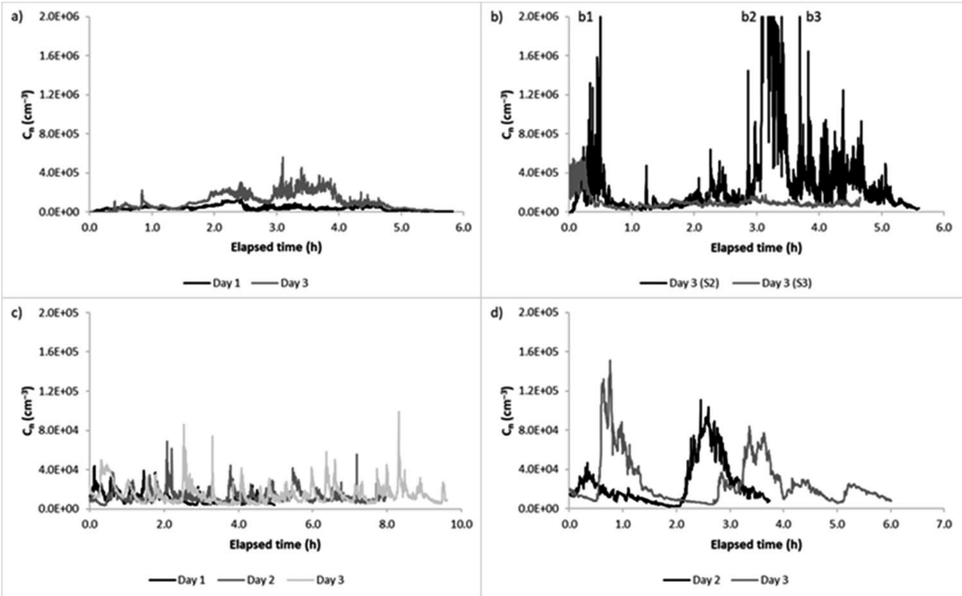

Figure 4. Real-time number concentration (C_n) time series measured using the CPC during working hours at (a) S1 (traditional bakery 1, CPC 3775 on day 1 in measurement campaign 1, CPC 3776 on day 3 in measurement campaign 2), (b) S2/S3 (traditional bakery 1, CPC 3775 at S2 on day 3 in measurement campaign 1, CPC 3776 at S3 on day 3 in measurement campaign 2), (c) S10 (in-store bakery, CPC 3022A each day), and using the P-Trak at (d) S11 (in-store bake-off unit). Note: Peak C_n is 2.6×10^6 cm^{-3} (b1), 4.1×10^6 cm^{-3} (b2), and 2.5×10^6 cm^{-3} (b3). CPC = condensation particle counter, P-Trak = P-Trak ultrafine particle counter, S1 = baking area, S2 = oven area, S3 = flour depository, S10 = beside a baking table and trolley ovens, S11 = beside a table near trolley ovens.

5.1.6 Number size distribution (Papers I and III)

Figure 5 shows examples of the average number size distribution of particulate matter in various work phases measured at stationary locations (S) using the SMPS and FMPS in traditional bakery 1 and the in-store bakery, respectively. Averaging periods were 10–60 min.

The average number size distribution of fine particles and nanoparticles fluctuated greatly in both bakeries. In traditional bakery 1, nanoparticles with a geometric mean diameter (GMD) of 10 nm and a geometric standard deviation (GSD) of 1.5 were formed after the trolley ovens were turned on. That work phase (number size distribution was measured using the FMPS at S2 during measurement campaign 2) was excluded from Figure 5 because of the very low average C_n (< 1.0×10^3 cm^{-3}) compared to other work phases (the number size distribution was measured using the SMPS at S1 and S2 during measurement campaign 1). During shaping of doughs on a baking table, the GMD of particles was 110 nm (GSD 2.2) at S1, and when the workers operated the trolley ovens, the GMD decreased to 60 nm (GSD 1.9) at S1 (Figure 5a). Doughnut baking resulted in nanoparticles with a GMD of 130 nm (GSD 1.6) at S2, and when all ovens were in operation, and doughnut baking was ongoing, the GMD of particles was 90 nm (GSD 1.8) at S2 (Figure 5a).

In the in-store bakery, operating trolley ovens induced nanoparticles with a GMD of 34 nm (GSD 1.2), whereas shaping of doughs resulted in particles with a GMD of 52 nm (GSD 1.8) at S10 (Figure 5b). When dough mixers were running, the GMD of particles was 52 nm (GSD 1.3), and cleaning (after oven and baking activities had ended) also resulted in particles with a GMD of 52 nm (GSD 1.1) at S10 (Figure 5b).

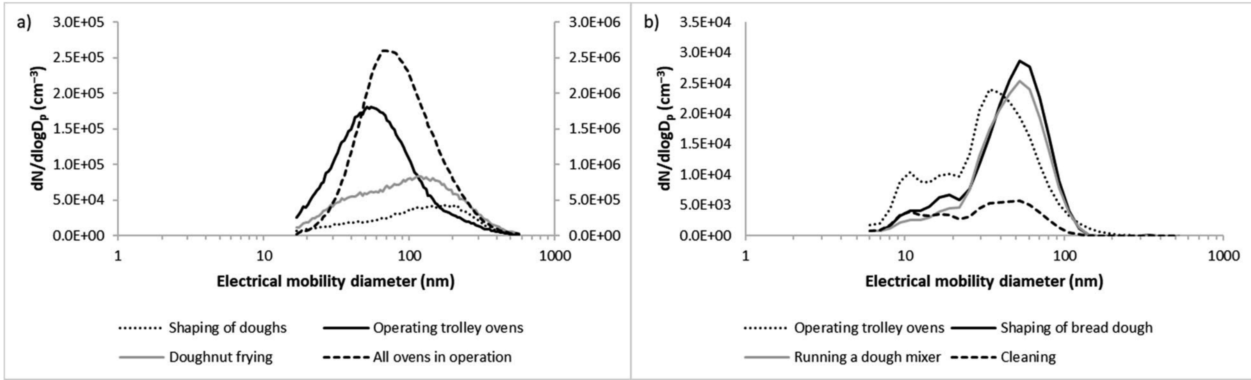

Figure 5. Average number size distribution (dN/dlogD$_p$) of particulate matter in various work phases measured using the SMPS at (a) S1 and S2 (traditional bakery 1, days 1 and 3 in measurement campaign 1), and using the FMPS at (b) S10 (in-store bakery, day 1). Note: FMPS = fast mobility particle sizer 3091, S1 = baking area, S2 = oven area, S10 = beside a baking table and trolley ovens, SMPS = scanning mobility particle sizer. The work phases in (a) are as follows: shaping of doughs (day 1, S1, left vertical axis), operating trolley ovens (day 1, S1, left vertical axis), doughnut frying (day 3, S2, left vertical axis), and all ovens in operation (day 3, S2, right vertical axis).

5.2 Organic chemicals

5.2.1 Concentrations of VOCs and short-chained carbonyls (Paper III)

The concentrations (C) of the VOCs (the retention time between C_6 and C_{16}) and short-chained carbonyls collected in various sampling locations in the in-store bakery and bake-off unit are presented in Table 13. Regarding the VOCs that were not among the ten most dominant compounds, their C was included in the table (marked with *) in cases when these compounds were the most dominant in some other sampling locations. Moreover, when some VOCs had a prevalence of < 50wt%, their C was included in the table (marked with **) if these compounds were the most dominant in some other sampling locations.

In the bakery, the C of TVOCs was 57–169 $\mu g/m^3$ at BZ5 and 31–70 $\mu g/m^3$ at S8/S9. Regarding individual VOCs, the C was 1–20 $\mu g/m^3$ at BZ5 and 1–23 $\mu g/m^3$ at S8/S9. The most dominant compounds detected at BZ5, S8, and S9 were d-limonene (8–16 $\mu g/m^3$), nonanal (6–7 $\mu g/m^3$), and decamethylcyclopentasiloxane (the abbreviation D5 is used hereafter) (8–23 $\mu g/m^3$), respectively. The average C of these compounds accounted for 14–28wt% of the average TVOC concentrations. For carbonyls, the C varied between < 1 and 45 $\mu g/m^3$ at S8. Acetaldehyde (45 $\mu g/m^3$), 2-butanone (20 $\mu g/m^3$), and acetone (17 $\mu g/m^3$) were the most dominant compounds.

In the in-store bake-off unit, the measurements were conducted only at stationary locations, and the C of TVOCs and individual VOCs was 82–214 and 2–81 $\mu g/m^3$ at S11/S12, respectively. The most dominant compounds detected at these locations were 1-butoxy-2-propanol (14–81 $\mu g/m^3$) and d-limonene (7–18 $\mu g/m^3$), respectively. Their average C made up 13–28wt% of the TVOC concentrations. For carbonyls, the C ranged from < 1 to 59 $\mu g/m^3$. Acetaldehyde (59 $\mu g/m^3$), butyraldehyde (20 $\mu g/m^3$), and formaldehyde (16 $\mu g/m^3$) were detected at the highest concentrations.

The average C of VOCs was at the same level in the background samples and other samples in both facilities. The C of the TVOCs was, on average, slightly greater in the in-store bake-off unit compared to the in-store bakery. For carbonyls, the C was consistent in both facilities.

Table 13. Concentrations (C) of the organic chemicals (VOCs and short-chained carbonyls) in the samples collected in various locations (B = background sample, BZ = breathing zone, S = stationary location) in an in-store bakery (B1, BZ5, S8, S9) and a bake-off unit (B2, S11, S12).

Compound	C ($\mu g/m^3$), AM (range)			
	B1/B2	**BZ5**	**S8/S9**	**S11/S12**
VOCs				
1-Butanol	ND/ND	8 (6–12)	5 (3–7)/ 3 (2–3)	ND/ND
1-Butoxy-2-propanol	ND/ 2 (2–2)*	ND	ND/ND	38 (14–81)/ 9 (4–19)
2-Etylhexanol	2 (1–2)*/ 4 (3–5)	2 (2–2)*	2 (2–2)**/ 2 (2–2)*	3 (3–3)*/ 3 (3–3)
2-Furantmethanol	ND/ND	11 (4–20)	ND/ND	ND/ND
2-Methyl-2-propanol	4 (3–4)/ 1 (1–1)*	3 (2–4)*	ND/ 2 (1–2)*	7 (5–8)/ 8 (2–17)
3-Carene	4 (3–5)/ 1 (1–1)*	2 (2–2)*	1 (1–1)**/ 2 (2–2)*	4 (3–4)*/ 3 (3–3)*
3-Methyl-1-butanol	ND/ 2 (2–2)*	4 (2–9)	1 (1–2)*/ 2 (2–2)**	4 (3–6)*/ 5 (3–6)
Acetoin	ND/ND	2 (2–2)**	ND/ND	4 (3–5)*/ 4 (2–6)
Allyl isothiocyanate	ND/ND	3 (2–7)	ND/ 3 (2–4)	ND/ND
Alpha-pinene	7 (5–8)/ 3 (3–4)	3 (3–4)	2 (2–3)/ 3 (3–4)	5 (4–6)*/ 4 (3–5)
Benzene	3 (2–3)/ 2 (2–2)	ND	ND/ND	ND/ND
D5	5 (3–6)/ 8 (3–12)	7 (4–11)	5 (3–8)/ 16 (8–23)	9 (5–11)/ 10 (4–14)
Decanal	2 (2–2)*/ 2 (2–2)	3 (1–4)*	4 (4–4)/ 2 (2–2)*	7 (5–8)/ 4 (3–5)*
D-Limonene	22 (18–27)/ 12 (10–14)	13 (8–16)	6 (5–8)/ 14 (12–17)	13 (8–19)/ 13 (7–18)
Ethyl acetate	ND/ 3 (3–3)	3 (2–4)*	2 (1–2)/ 3 (2–4)	3 (3–4)*/ 3 (3–3)*
Eucalyptol	ND/ 2 (2–2)*	2 (2–3)*	2 (2–2)**/ ND	10 (10–11)/ 3 (2–4)*
Hexanal	2 (2–3)/	3 (2–3)*	2 (2–2)/	4 (4–5)*/

	ND		2 (2–2)	4 (3–4)*
Nonanal	4 (4–4)/ 3 (3–3)	6 (5–9)	6 (6–7)/ 5 (4–5)	12 (10–14)/ 9 (8–11)
Octyl acrylate	ND/ND	3 (2–6)*	2 (1–2)/ ND	ND/ND
TVOCs	54 (45–62)/ 58 (45–72)	92 (57–169)	38 (31–42)/ 58 (46–70)	137 (119–214)/ 103 (82–124)
Carbonyls				
2-Butanone	NA/NA	NA	20/NA	10/NA
Acetaldehyde	NA/NA	NA	45/NA	59/NA
Acetone	NA/NA	NA	17/NA	3/NA
Acrolein	NA/NA	NA	< 1/NA	1/NA
Butyraldehyde	NA/NA	NA	7/NA	20/NA
Crotonaldehyde	NA/NA	NA	< 1/NA	1/NA
Formaldehyde	NA/NA	NA	10/NA	16/NA
Methacrolein	NA/NA	NA	< 1/NA	< 1/NA
Propionaldehyde	NA/NA	NA	3/NA	6/NA
Valeraldehyde	NA/NA	NA	1/NA	4/NA

* = not among the ten most dominant compounds in the sampling location.
** = prevalence < 50wt% in the sampling location.
AM = arithmetic mean, B1 = in a café next to the bakery, B2 = in the store beside the bake-off unit, BZ5 = general baker, D5 = decamethylcyclopentasiloxane, NA = not available, ND = not detected, S8 = beside a dough mixer tub and flour silo, S9 = beside a baking table and trolley ovens, S11 = beside a table and trolley ovens, S12 = beside a showcase, TVOCs = total volatile organic compounds, VOC = volatile organic compound.

6 Discussion

6.1 Mass concentrations of particulate matter

In Finland, the 8-hour occupational exposure limit (OEL) for inhalable flour dust is 2 mg/m^3 (Finnish Ministry of Social Affairs and Health 2020). Regarding bakeries, in this study, the C_m of inhalable dust in the breathing zone (BZ) was 340–725% (traditional bakery 1), 408–1133% (traditional bakery 2, pre- and post-intervention), 15–185% (industrial bakery, pre- and post-intervention), and 224–294% (in-store bakery) of the Finnish OEL. At the stationary locations (S), the concentrations were 115–130%, 28–148%, 8–35%, and 3–23% of the OEL respectively. In the in-store bake-off unit where flours were not used, the C_m of organic dust at S was 1–2% of the Finnish OEL (8-hour) of 5 mg/m^3 (Finnish Ministry of Social Affairs and Health 2020). The results showed that the average C_m was highest in the traditional bakeries at both BZ and S, which may be explained by the greater degree of craft work there compared to the other bakeries.

Considering various job titles, the C_m of inhalable dust was 1.3–14.5 mg/m^3 (dough makers: BZ1, BZ3 pre-intervention), 4.5–22.7 mg/m^3 (general bakers: BZ2 pre- and post-intervention, BZ5), and 0.3–0.5 mg/m^3 (line worker: BZ4). In previous studies, the C_m of inhalable dust was 0.1–65.0 mg/m^3, 0.1–26.8 mg/m^3, and 0.5–12.0 mg/m^3 for dough makers, all-round stuff/general bakers/mixed tasks, and line workers, respectively (see Section 2.1.8, Table 1).

At the stationary locations, the C_m of inhalable dust was 0.1–3.0 mg/m^3 in the bakeries (S1, S4–9), and 0.1 mg/m^3 in each sample collected in the in-store bake-off unit (S11, S12). In other studies, the exposure levels obtained in area sampling varied between < 0.1 and 19.0 mg/m^3 (see Section 2.1.8, Table 1).

In the previous Finnish studies in which exposure to inhalable flour dust has been investigated, the C_m was 0.9–4.2 mg/m^3 at stationary samples in a traditional bakery (Tissari et al. 2002, 2005). These concentrations align with those obtained in the current study at the stationary locations (0.1–3.0 mg/m^3 at S1 and S4–9). In their intervention study, Hakala et al. (2016) measured C_m of inhalable flour dust in Finnish in-store bakeries; however, only percentage changes in the C_m post-intervention were reported without information on the exposure levels.

Regarding the $PM_{0.5}$ particles, collected using the IOM sampler with the pre-cyclone in traditional bakery 1, the C_m accounted for 9–15% and 4–8% of inhalable dust at BZ1 and S1, respectively. The dough maker (BZ1) also participated in other work tasks in addition to dough making, which led to exposure to both small and large particles. The C_m of the PM_1 particles (0.1–0.2 mg/m^3) determined gravimetrically at S1 and S2 were slightly lower than the levels (0.2–0.8 mg/m^3) measured by Tissari et al. (2002) using a Dekati PM_1 impactor.

The real-time C_m of the PM_{10} size fraction measured at stationary locations (S1, S5, S7, S10, S11) was < 0.1–28.3 mg/m^3 in the bakeries and < 0.1–0.1 mg/m^3 in the in-store bake-off unit. The average C_m was highest in the traditional bakeries, which agrees with the results obtained using the IOM samplers. In the in-store bakery, the average C_m was < 0.1 mg/m^3, although the stationary site (S10) was located near a baking table. This result is likely related to low-dust flours being used while shaping doughs on the baking table. Low-dust flours are less-dusty flours developed for bakeries to reduce exposure to dust.

In previous Finnish studies (Tissari et al. 2002, 2005), the C_m of PM_{10} size fraction, measured using an EPA-WINS impactor in a traditional bakery, was 0.1–1.3 mg/m^3. Regarding other studies, Mounier-Geyssant et al. (2007) obtained personal exposure levels of 0.2–4.0 mg/m^3 to PM_{10} size fraction using a Harvard Chempass Sampler in traditional and industrial bakeries, whereas Rumchev et al. (2021) measured area PM_{10} levels of 0.1–6.2 mg/m^3 using a DustTrak II Aerosol Monitor 8530 in industrial bakeries. Previous studies on exposure to PM_{10} particles in in-store bakeries or bake-off units were not found. In a study conducted by Zaatari and Siegel (2014) in retail stores, the average C_m of PM_{10} size fraction was 20 µg/m^3, which is in accordance with the results (30–40 μg/m^3) obtained in the in-store bakery (S10) and bake-off unit (S11). There are no limit values for PM_{10} concentrations in industrial workplaces in Finland.

The real-time C_m (PM_{10}) varied widely in the traditional bakeries (Figures 2a-c) and industrial bakery (Figure 2d) during the measurement days, and several peak concentrations were observed. In traditional bakery 1, the daily peak C_m (1.1–2.3 mg/m^3) arose from weighing flour and adding flour from sacks to dough mixers, whereas in traditional bakery 2, the daily peak C_m (7.0–28.3 mg/m^3 pre-intervention, 5.2–15.0 mg/m^3 post-intervention) stemmed from the production of breads (during night shifts), bread rolls, buns, and sourdough. In the industrial bakery, the daily peak C_m (4.3–8.1 mg/m^3) was attributed to weighing ingredients, adding flour from sacks to dough mixers, running the dough mixers, and dumping flour from a flour feeder. In the in-store bakery (Figure 2e), only a few C_m peaks (0.5–2.5 mg/m^3) were observed, which might be related to the low-dust flours that possibly contributed to the number of peaks. The peak C_m was related to dumping flour from a flour feeder into dough mixers, operating the dough mixers, and cleaning. The findings in the traditional bakeries, industrial bakery, and in-store bakery are in accordance with those of previous studies (Meijster et al. 2008, Roberge et al. 2012) that showed high C_m peaks during dough-making (weighing and mixing ingredients). In the in-store bake-off unit (Figure 2f), the C_m was very low, predominantly below 0.1 mg/m^3, and the particles stemmed mainly from the background dust, such as the floor, surfaces, or clothes.

Considering the TEOM, the average C_m at S1 was 0.2 mg/m^3 with the pre-cyclone and 0.5 mg/m^3 without the pre-cyclone in traditional bakery 1. The average C_m without the pre-cyclone is consistent with that (0.4 mg/m^3) measured using the DRX at S1. However, the real-time C_m measured using the TEOM was greater compared to the DRX at the beginning of dough-making and when several work phases were ongoing, which can be explained by the broader particle size range (< 100 μm) of the TEOM. During oven operations, the difference in the real-time C_m between the TEOM and

DRX was slighter, and the difference between the C_m measured using the TEOM with and without the pre-cyclone leveled.

6.2 Chemical composition and morphology of the particles

Small, agglomerated flour dust particles, spherical particles, and soot agglomerates were observed in the samples collected in traditional bakery 1. The agglomerated particles included nanosized particles. This finding is consistent with Tissari et al. (2002), who also found nanosized particles in SEM images in a previous Finnish study.

Regarding the PM_1 samples collected at S1, carbon comprised 42–64% of the total PM_1 mass, and approximately 99% of that was organic carbon. In addition to carbon, the rest of the particles most likely comprised mainly hydrogen, nitrogen, and oxygen, since these compounds are the major ingredients of carbohydrates and proteins. Tissari et al. (2002) found that an average of 88wt% of total carbon consisted of organic carbon in total dust and PM_1 samples. The proportion of organic carbon increased during a working day and was 99wt% at its highest when several work phases were ongoing. Ielpo et al. (2020) reported that the carbonaceous fraction (organic carbon, elemental carbon, levoglucosan) accounted for 17–35% of the total $PM_{2.5}$ mass. The proportion of the carbonaceous fraction was at its highest when the ovens were turned on and decreased during the operation of the ovens.

In the current study, the mass proportion of the 31 elements (excluding carbon and water-soluble ions) was 1.1% at S1 (PM_1), 0.4% at BZ1 (inhalable dust), and 0.5% at BZ1 ($PM_{0.5}$, collected with the pre-cyclone). Considering BZ1, the proportions align with the result obtained by Tissari et al. (2002), who showed that the mass proportion of the same 31 elements was < 1% in the total dust and PM_1 samples. The water-soluble ions (Cl^-, NO_3^-, SO_4^{2-}, PO_4^{3-}) made up 2.2% of the total PM_1 mass at S2. This finding is contrary to that of Ielpo et al. (2020), who found that the mass proportion of water-soluble ions (Cl^-, NO_2^-, NO_3^-, SO_4^{2-}, $C_2O_4^{2-}$, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+})

varied at 42–60% in the $PM_{2.5}$ samples. This discrepancy could be attributed to the facts that $PM_{2,5}$ samples were investigated instead of PM_1 samples, and the combustion processes were different since a wood oven was used in the previous study.

6.3 Intervention

In traditional bakery 2, a 39% reduction in the average C_m of inhalable dust was achieved at S4, whereas the average C_m increased 54% at S5. The C_m reduction might be explained by either random variation in the exposure levels or the intervention measures implemented at S4, where ingredients were weighed and mixed. Intervention strategies were also targeted at S5; however, an increase in the average C_m was observed. At BZ2 (general baker), the average C_m of inhalable dust increased 24%. The C_m increases at S5 and BZ2 may be attributable to the greater workload of the workers during post-intervention measurements and lack of adherence to intervention strategies related to cleaning (the floor and surfaces were not cleaned immediately when weighing ingredients and handling bags). This highlights the employers' need to motivate all the workers to commit to each intervention strategy. Post-intervention, the average C_m exceeded the Finnish OEL of 2 mg/m^3 for inhalable flour dust at BZ2 (14.4 mg/m^3) but was lower than the OEL at S4 (1.6 mg/m^3) and S5 (1.2 mg/m^3).

In the industrial bakery, the average C_m of inhalable dust decreased by 45% at S6 and 40% at S7 post-intervention, and the exposure levels were lower than the Finnish OEL. However, the average C_m of inhalable dust increased by 28% at BZ3 (dough maker) and 55% at BZ4 (line worker). Post-intervention, the average C_m exceeded the Finnish OEL at BZ3 (2.1 mg/m^3) but was lower than the OEL at BZ4 (0.7 mg/m^3). The line workers spent no time in the dough-making area, which explains the lower exposure levels at BZ4 compared to BZ3. The C_m reduction at the stationary locations and the C_m increase in the breathing zone may possibly be related to the facts discussed above.

The real-time measurements showed that in traditional bakery 2, no reduction in the average C_m was achieved at S5, which is consistent with the result obtained using the IOM sampler. Concerning all size fractions, the average C_m increased 93–217% (morning shift) and 22–73% (night shift) at S5 post-intervention. The average C_m was greater in the night shifts pre- and post-intervention, which might be related to dustier work tasks during bread-making or poorer adherence to the intervention strategies. In the industrial bakery, the average C_m decreased 20–25% (all size fractions included) at S7 post-intervention, whereas a reduction of 40% in average C_m was observed at S7 using the IOM sampler. The C_m increase at S5 and the C_m reduction at S7 could also be attributed to the facts discussed earlier.

In previous intervention studies, measurements were conducted only in the breathing zone, and intervention strategies focused on both technical control methods and work practices were investigated. Meijster et al. (2009) found a modest downward annual trend of −2% for flour dust in Dutch bakeries. Baatjies et al. (2014) showed reductions of 23–67% in inhalable flour dust levels in South African supermarket bakeries. Hakala et al. (2016) achieved an average reduction of 64% in inhalable flour dust levels in Finnish supermarket bakeries. Martinelli et al. (2020) reported reductions of 16–70% in inhalable flour dust levels in Italian bakeries.

Regarding the C_m peaks of the PM_{10} size fraction, in traditional bakery 2, the number of peaks was, on average, greater, but the highest C_m peaks were lower during morning and night shifts post-intervention. The maximum duration of the peaks was slightly longer post-intervention, and the difference between the longest pre- and post-intervention peaks was 10 min (morning shift) and 8 min (night shift). In the industrial bakery, the number of C_m peaks was, on average, the same pre- and post-intervention, but the highest C_m peaks were greater post-intervention. Furthermore, there was no difference between the duration of the longest peaks pre- and post-intervention. The peaks were vastly shorter compared to traditional bakery 2, which can be explained by the higher degree of automation in baking activities and lower exposure levels.

Concerning the proportions of the size fractions of the PM_{15}, the PM_1 fraction clearly dominated in both bakeries pre- and post-intervention. In traditional bakery 2, the average proportion of the PM_1 particles of the PM_{15} was 43–69% pre-intervention and 61–76% post-intervention, whereas in the industrial bakery, the average proportion was 61% pre-intervention and 58% post-intervention. The average C_m increased in traditional bakery 2 but decreased in the industrial bakery post-intervention, which explains the differences between the pre- and post-intervention proportions. The higher average mass proportion of PM_1 particles is likely related to the fact that smaller and lighter particles tend to stay longer in the bakery air compared to the larger particles.

Tikkainen et al. (1996) and Stobnicka and Górny (2015) suggested that > 50% of the airborne flour dust particle mass has an aerodynamic diameter of > 15 µm. Burdorf et al. (1994) and Lillienberg and Brisman (1994) showed that the thoracic fraction (4–10 µm) and respirable fraction (≤ 4 µm) accounted for 26–39% and 9–19% of inhalable dust, respectively. Roberge et al. (2012) reported that an average mass percentage of particles of 0.2–4.0 and 10–20 µm contributed 12 and 61% of inhalable dust, respectively. The particle size range of the DRX used in the present study is 0.5–15 µm, which explains the differences in the mass percentages between various studies. Furthermore, the baking areas were not cleaned during the measurements, which was also attributed to the high proportions of PM_1 particles in both bakeries.

6.4 Number concentrations of particulate matter

The real-time C_n of particulate matter measured at stationary locations (S1–3, S10, S11) was 3.7×10^2–4.1×10^6 cm^{-3} (traditional bakery 1), 3.5×10^3–9.9×10^4 cm^{-3} (in-store bakery), and 2.2×10^3–1.5×10^5 cm^{-3} (in-store bake-off unit). Only one previous study (Tissari et al. 2002) was found on the number concentrations of nanosized particles in a bakery. In traditional bakery 1, the average C_n (4.0×10^4–3.3×10^5 cm^{-3}) was greater compared to the in-store bakery (1.3×10^4 cm^{-3}) and bake-off unit (2.6×10^4 cm^{-3}) and aligns with that (6.0×10^4–2.5×10^5 cm^{-3}) obtained by Tissari et al. (2002) using an electrical low-pressure impactor (ELPI) in a traditional bakery. The greater average C_n and peak C_n in traditional bakery 1 may be explained by the fact that both hearth ovens and trolley ovens existed in the facility, which resulted in high concentrations of particles in the bakery air.

The average C_n and peak C_n measured using the P-Trak at S11 (in-store bake-off unit) were higher than those obtained using the CPC at S10 (in-store bakery). This result might be attributed to the greater number of trolley ovens in the bake-off unit. Furthermore, the detection ranges of the CPC and P-Trak were different, which could also explain the difference between the results. The ventilation rates might also have differed in the facilities; however, this study did not examine the ventilation.

In all the facilities, the C_n fluctuated greatly during the measurement days (Figure 4a–d). Tissari et al. (2002) also demonstrated a wide variation in the real-time C_n in a traditional bakery. In the current study, a rapid increase in the C_n at S2 and S3 was observed when the trolley ovens were turned on at the beginning of measurement day 3 in traditional bakery 1 (Figure 4b). This finding indicated that the ovens released particles into the bakery air, which was also reported by Tissari et al. (2002). Ielpo et al. (2020) found that fine particles formed from wood burning in a wood oven in a traditional bakery.

In traditional bakery 1, on measurement day 3 (measurement campaign 2), when the DRX and CPC were used at S1, both the C_m (Figure 2a) and C_n (Figure 4a) were occasionally high between the time points of 3.0 and 4.0 hrs. During that time, all the ovens were operating simultaneously, and several other work phases were ongoing at the same time. The highest C_m and C_n peaks were detected between the above-mentioned time points. This result may be explained by the fact that both nanoparticles (arising out from oven operations mainly) and larger particles (stemming predominantly from flour dust) were in the bakery air at the same time. The C_n was, on average, higher in the oven area (S2, Figure 4b) compared to the baking area (S1, Figure 4a), indicating that the oven operations resulted in the highest C_n peaks. Tissari et al. (2002) suggested that the inner surfaces of the ovens might release particles into the bakery air since in their study, opening the doors of the oven led to an increase in C_n, even though no bakery products were in the ovens.

In the in-store bakery, on measurement day 1, when the OPS and CPC were used at S10, simultaneous C_m (Figure 2e) and C_n (Figure 4c) peaks were detected between the time points of 0.8 and 1.8 hrs. This result is likely related to the facts discussed above. In the in-store bake-off unit, the C_m and C_n time series were not consistent. Since flours were not used, predominantly small and light particles stemming from the oven operations existed in the air, which resulted in high C_n at several time points, although the C_m remained very low. The results show that the high C_n stems from oven operations instead of flour dust, and fine particles and nanoparticles significantly affect the C_n.

6.5 Number size distribution of particulate matter

The average number size distribution of particulate matter measured at S1 (traditional bakery 1), S2 (traditional bakery 1), and S10 (in-store bakery) also varied widely (Figure 5). In various work phases, the GMD of particles was 10–130 nm (GSD 1.5–2.2) and 34–52 nm (GSD 1.1–1.8) in traditional bakery 1 and the in-store bakery, respectively. Tissari et al. (2002, 2005) also showed that the number size distribution measured using the ELPI fluctuated greatly in a traditional bakery. No other previous studies were found on number size distribution ($dN/dlogD_p$) of nanosized particles in bakeries.

In traditional bakery 1, the smallest particles (GMD 10 nm, GSD 1.5) were formed at S2 when the trolley ovens were turned on. When the trolley ovens were operating, the GMD of particles at S1 was 60 nm (GSD 1.9). In the in-store bakery, operating the trolley ovens resulted in particles with a GMD of 34 nm (GSD 1.2) at S10. Tissari et al. (2002, 2005) found that the particle size was < 40 nm when operating ovens. They also showed that particles of approximately 300 nm originating from grease were formed during doughnut frying. In the present study, doughnut frying induced particles with a GMD of 130 nm (GSD 1.6) at S2. When doughs were shaped on the baking table, the GMD of particles was 110 nm (GSD 2.2) and 52 nm (GSD 1.8) at S1 and S10, respectively.

6.6 Concentrations of VOCs

In the in-store bakery and bake-off unit, the C of the TVOCs and individual VOCs was low. The C of the TVOCs was 57–169 µg/m^3 (BZ5, in-store bakery), 31–70 µg/m^3 (S8/S9, in-store bakery), and 82–214 µg/m^3 (S11/S12, in-store bake-off unit). Tuomi and Vainiotalo (2016) suggested a target value of 300 µg/m^3 for TVOCs in industrial workplaces. The C of TVOCs was lower than the target value in both facilities. A TVOC concentration can be applied as an indicator of indoor air quality; however, it is irrelevant in terms of toxicity and health since it disregards the adverse effects of individual VOCs.

Only two previous studies (Tissari et al. 2002, Chang et al. 2018) on the C of TVOCs in bakeries were found. Tissari et al. (2002) obtained TVOC concentrations of 26–1265 µg/m^3 at stationary locations in a traditional bakery. These concentrations are, on average, greater compared to those detected in the current study. Chang et al. (2018) measured TVOC concentrations of 150–8657 ppb at stationary locations in in-store bakeries. Using the equation presented by Boguski (2022), in the current study, the C of the TVOCs was 8–45 and 22–57 ppb in the in-store bakery and bake-off unit, respectively. The TVOC concentrations detected in the present study are notably lower than those of Chang et al. (2018), which could be explained by the different sampling methods. In the previous study, the TVOCs were measured using a direct reading instrument, a ppbRAE 3000 detector, which was calibrated for isobutylene.

The average C of the TVOCs was greater in the in-store bake-off unit compared to the in-store bakery. This result might be attributed to the greater number of trolley ovens in the bake-off unit since there was only one worker during the measurements, and no flavors were used. Previous studies (Cho et al. 2010, Petisca et al. 2014, Pico et al. 2015) have shown that the thermal processing of bakery products is related to VOC emissions.

The C of the individual VOCs was 1–20 μg/m^3 (BZ5), 1–23 μg/m^3 (S8/S9), and 2–81 μg/m^3 (S11/S12). The average C of VOCs was vastly lower than the Finnish OELs (8 h) for VOCs; however, an OEL is not set for some VOCs detected in this study. The most dominant VOCs were d-limonene (BZ5, S12), nonanal (S8), D5 (S9), and 1-butoxy-2-propanol (S11). Possible sources for these compounds were building and construction materials and personal care products (NIH). The same compounds were also observed in the background samples, and the C was at the same level compared to other sampling locations.

Considering the other VOCs, the following compounds were not detected in the background samples: 1-butanol, 2-furantmethanol (furfural), acetoin, allyl isothiocyanate, and octyl acrylate. Plausible sources for 1-butanol and octyl acrylate were building and construction materials (NIH). According to previous research, furfural is formed during heating by the Maillard reaction and sugar pyrolysis (Petisca et al. 2014), acetoin is a buttermilk flavoring (Day et al. 2011), and allyl isothiocyanate is possibly related to the packaging materials of products (EFSA 2010). Cleaning and personal care products might also be possible sources, at least for acetoin and furfural.

Two previous studies (Tissari et al. 2002, Chang et al. 2018) reported C for a few VOCs observed in the current study. Tissari et al. (2002) measured VOC concentrations (calculated as toluene equivalents) of 2–309 μg/m^3 for eucalyptol, hexanal, nonanal, and decanal at stationary locations in a traditional bakery. These concentrations are, on average, greater compared to those (2–14 μg/m^3) of the present study. Acetoin is another compound detected in both the current and previous (Chang et al. 2018) study. The average C of acetoin at BZ5 and S11/S12 was 2–4 μg/m^3, which aligns with that of 2 μg/m^3 (calculated as toluene equivalents) obtained by Chang et al. (2018). 2,3-heptanedione and 2,3-hexanedione, which may pose respiratory hazards (Day et al. 2011, Curwin et al. 2015, OSHA), were not observed in the present study.

6.7 Concentrations of short-chained carbonyls

The C of short-chained carbonyls was also low in the in-store bakery (< 1–45 μg/m^3 at S8) and bake-off unit (< 1–59 μg/m^3 at S11). These exposure levels were remarkably lower than the Finnish OELs (8 h), but no OEL is set for some carbonyls observed in this study. The most dominant compounds were 2-butanone, acetaldehyde, acetone, butyraldehyde, and formaldehyde, possibly originating from building and construction materials, cleaning products, coatings, paints, and personal care products. The current study did not detect diacetyl and 2,3-pentanedione, which may be associated with respiratory hazards (Day et al. 2011, Curwin et al. 2015, OSHA).

Three previous studies (Tissari et al. 2002, Curwin et al. 2015, Chang et al. 2018) on short-chained carbonyls in bakeries were found. They reported C for some carbonyls that were detected in the present study. Regarding 2-butanone, acetaldehyde, acetone, acrolein, butyraldehyde, formaldehyde, and valeraldehyde, Tissari et al. (2002) measured concentrations of 2–1653 μg/m^3 at stationary locations in a traditional bakery. These concentrations are, on average, significantly higher than those (< 1–59 μg/m^3) detected in the current study. However, the C of butyraldehyde (7–20 μg/m^3) measured at S8 and S11 corresponds with that (2–16 μg/m^3) obtained in the previous study. Acetaldehyde was also detected in the current study and previous studies (Curwin et al. 2015, Chang et al. 2018). Curwin et al. (2015) obtained geometric mean (GM) concentrations of 43–81 μg/m^3 for acetaldehyde at stationary locations in ten food-manufacturing facilities (including one bakery). The GM concentration in the present study was 52 μg/m^3, which aligns with that of the previous study. Chang et al. (2018) reported an average C of 190 μg/m^3 for acetaldehyde at stationary locations in in-store bakeries. That concentration is vastly greater than the levels (45–59 μg/m^3) measured in the present study.

7 Conclusions

The results of this thesis showed that the personal exposure levels of dough makers and general bakers to inhalable dust were high and exceeded the Finnish (8-hour) occupational exposure limit (OEL) of 2 mg/m^3 for flour dust. In the traditional bakeries, the mass concentrations of inhalable dust also exceeded the OEL at one stationary location. However, considering all the facilities, the mass concentrations were vastly lower than the OEL at most of the stationary locations. The $PM_{0.5}$ fraction accounted for 9–15% of the inhalable dust in the breathing zone of a dough maker.

Regarding the real-time mass concentrations (PM_{10}), several peak concentrations were observed in the traditional bakeries and industrial bakery, whereas in the in-store bakeries where low-dust-flours were used, only a few peak exposures were measured. The mass concentrations were very low in the in-store bake-off unit, where flours were not used. The work tasks that affected the peak concentrations included weighing of ingredients (e.g., flours, spices), adding flour from sacks to dough mixers, and operating dough mixers.

The microscopic analysis showed small, agglomerated flour dust particles, spherical particles, and soot agglomerates in the dust samples collected in the traditional bakery. Nanosized particles were found in particle agglomerates. The PM_1 samples comprised mainly carbon and very small amounts of other elements and water-soluble ions. The total carbon consisted nearly entirely of organic carbon.

In the intervention study, no reductions in the exposure levels to inhalable flour dust were achieved in the breathing zone post-intervention in the traditional and industrial bakery. However, the mass concentrations of inhalable dust decreased at most of the stationary locations. Considering real-time measurements, the peak mass concentrations (PM_{10}) decreased in the traditional bakery post-intervention, but no reductions in the peak levels were achieved in the industrial bakery. In both bakeries, the PM_{15} size fraction comprised predominantly PM_1 particles and large particles (aerodynamic diameter of > 10 μm) pre- and post-intervention.

The real-time number concentrations were greater in the traditional bakery compared to the in-store bakery and bake-off unit. The results showed that the ovens had a remarkable effect on the number concentrations since turning on the trolley ovens increased the concentrations rapidly in the traditional bakery. Several peak number concentrations were detected when all the ovens were operating simultaneously in the facilities. High number concentrations were detected in the bake-off unit, although the mass concentrations were very low. That result indicated that high number concentrations arise from oven operations instead of flour dust, and fine particles and nanoparticles predominantly contributed to high number concentrations. Nanoparticles with a geometric mean diameter of 10 nm were detected when the trolley ovens were turned on, whereas operating the trolley ovens resulted in nanoparticles with a geometric mean diameter of 30–60 nm.

The concentrations of TVOCs, VOCs, and carbonyls were low in the in-store bakery and bake-off unit. The TVOC concentrations were lower than the target value of 300 μg/m^3 for industrial workplaces, whereas the concentrations of the individual VOCs and carbonyls were lower than the Finnish OELs. The most dominant VOCs and carbonyls possibly originated from building and construction materials and personal care products. A few VOCs were possibly related to work in the facilities and plausible sources were flavorings, baking products in the trolley ovens, and packaging materials.

Based on the results of this thesis, local control measures are required to reduce exposure levels to inhalable dust in the breathing zone and to peak mass concentrations of particulate matter in bakeries. Further studies are needed to plan more rigorous interventions supplemented by technical control methods in bakeries. The results also highlight the need for further research on the number concentrations of particulate matter and concentrations of organic chemicals in various types of bakeries.

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Appendices

ARTICLES

ARTICLE I

Karjalainen A, Leppänen M, Leskinen J, Torvela T, Pasanen P, Tissari J, Miettinen M. (2018). Concentrations and number size distribution of fine and nanoparticles in a traditional Finnish bakery. *Journal of Occupational and Environmental Hygiene,* 15(3): 194–203.

ARTICLE II

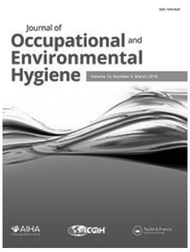
Karjalainen A, Leppänen M, Ruokolainen J, Hyttinen M, Miettinen M, Säämänen A, Pasanen P. (2022). Controlling flour dust exposure by an intervention focused on working methods in Finnish bakeries: a case study in two bakeries. *International Journal of Occupational Safety and Ergonomics,* 28(3): 1948–1957.

ARTICLE III

Karjalainen A, Väisänen A, Leppänen M, Ruokolainen J, Hyttinen M, Miettinen M, Säämänen A, Pasanen P. Exposure to particulate matter, volatile organic compounds, and carbonyls in an in-store bakery and a bake-off unit in Finland. Submitted manuscript.

ARTICLE I

Karjalainen A, Leppänen M, Leskinen J, Torvela T, Pasanen P, Tissari J, Miettinen M. (2018). Concentrations and number size distribution of fine and nanoparticles in a traditional Finnish bakery. *Journal of Occupational and Environmental Hygiene,* 15(3): 194–203.



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Concentrations and number size distribution of fine and nanoparticles in a traditional Finnish bakery

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ABSTRACT

In bakeries, high concentrations of flour dust can exist and ovens release particles into the air as well. Particle concentrations (mass, number) and number size distribution may vary considerably but the variation is not commonly studied. Furthermore, the role of the smallest size fractions is rarely considered in the exposure assessment due to their small mass. The objectives of this work were to find out how concentrations and number size distribution of fine and nanoparticles vary in a traditional Finnish bakery and to determine the exposure of a dough maker to the nanoparticle fraction of the inhalable dust.

Two measurement campaigns were carried out in a traditional, small-scale bakery. Sampling was performed at the breathing zone of the dough maker and three stationary locations: baking area, oven area, and flour depository. Both real-time measurements and conventional gravimetric sampling were conducted. Nanoparticle fraction of the inhalable dust was determined using an IOM sampler with a customized precyclone.

Number concentration of fine and nanoparticles, and mass concentrations of both the inhalable dust and nanoparticles were high. The nanoparticle fraction was 9–15% of the inhalable dust at the breathing zone of the dough maker. Different sources, such as ovens and doughnut baking affected the number size distribution.

Flour dust contained nanoparticles but most of the fine and nanoparticles were released into the air from the oven operations. However, nanoparticles are not a primary concern in bakeries compared to health effects linked to the large flour particles such as flour-induced sensitization or asthma and development of occupational rhinitis.

KEYWORDS
Baker; exposure; flour dust; occupational

Introduction

The bakery industry in Finland is diverse and the largest subfood-industry in terms of locations and number of jobs. Over 700 businesses located all over Finland employ approximately 8,000 workers. A majority of the bakeries are family businesses with less than ten employees.[1] In Finland, 15,000–20,000 workers perform work activities that include handling of flour dust. In 2008–2011, 40% of flour dust measurements exceeded the Finnish occupational exposure limit (OEL), 2 mg/m^3.[2]

Flour dust usually contains several noncereal components such as enzymes, preservatives, spices, and other possible sensitizers.[3] The major health effects of flour dust are occupational asthma, conjunctivitis, rhinitis, and dermal reactions.[4] Baker's asthma is one of the most common occupational asthmas.[5–7] Exposure to flour dust can also lead to chronic bronchitis and bronchial obstruction.[8,9]

High concentration periods related to specific work activities in bakeries may be decisive in causing the adverse health effects. The prevention of these periods could decrease allergic and respiratory symptoms.[10] However, data on the variation of concentrations (mass, number) in bakeries is insufficient. In addition, thus far, fine and nanoparticles in bakeries have been rarely studied, partly due to the lack of appropriate instruments but mainly because of their low mass concentration. However, the smallest size fractions may play a role, for example, in allergic alveolitis type of reactions.[4] To the best of our knowledge, there are only two studies where nanoparticles have been measured in bakeries.[11,12]

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In this study, the variation of concentrations (mass, number) and number size distribution of fine and nanoparticles was studied in a traditional Finnish bakery. In addition, the exposure of a dough maker to the nanoparticle fraction of the inhalable dust was determined.

Methods

Bakery

The study was carried out in a traditional, small-scale bakery in Finland. The bakery produced a wide variety of products including breads, rolls, bagels, baguettes, rice pasties, pies, cakes, gateaux, buns, doughnuts, pastries, and biscuits. The average workforce was 10 employees; most of them had individual job tasks and the degree of automation was low. The floor area and volume of the bakery were approximately 450 m^2 and 1,700 m^3, respectively. There was mechanical ventilation (F7 filters) in the building. F7 is a filter for fine dust (according to the European standard for air filters EN779:2012) with an average efficiency (Em, %) for 0.4-µm particles of $80 \leq Em < 90$.[13] All the ovens were fueled by oil. Above the ovens and doughnut baking spot were hoods that were equipped with metal grease filters (G2). G2 is a filter for coarse dust with an average arrestance (Am, %) for synthetic dust of $65 \leq Am < 80$.[13] Local dust control system in a bakery was a tubular bag filter unit (CT-34-54-1600 L, Clevertek, Finland) with a blower (CT-5, 5–6000, 5.5 kW). It consisted of 54 tubular filter bags that were cleaned automatically by compressed air. Total filter area was 34 m^2 and capacity 6000 m^3/hr. Devices, for example dough mixers and dough roller (Figure 1), were connected to the system with flexible hoses. However, local dust controls were in most of the areas used infrequently. The workers brushed the floor and tables each day during and at the end of a work shift.

In the bakery, two separate measurement campaigns were conducted in October 2014 and March 2016, both on three consecutive days during a week. The work shift started at around 2:00 a.m. and finished at around 7:30 a.m. Every work shift began with dough making. Trolley ovens were turned on at the beginning of each day, followed by hearth ovens 30 min later. Baking of the products started at around 2:50 a.m. and operating of the trolley ovens 10 min later. Doughnut baking also began at 3:00 a.m. and continued until 7:00 a.m. Operating the hearth ovens started at approximately 4:00 a.m. and continued to 6:00 a.m. In addition to dough making (weighting and mixing ingredients), the dough maker participated in other tasks such as dough forming, placing products into hearth ovens, and cleaning.

Particle measurements

Measurements were performed at the breathing zone (BZ) of the dough maker and at three stationary (S) locations: A) baking area, B) oven area, and C) flour depository (Figure 1) (S-A, S-B, and S-C hereafter). The sampling height at the stationary locations varied between 1.0 and 1.4 m. Mass concentrations (C_m), number concentrations (C_n), number size distribution (dn/dlogD_p), and particle morphology and composition were studied.

Real-time C_m was measured with a tapered element oscillating microbalance (TEOM; Thermo Scientific, USA) and a DustTrak DRX (TSI, USA). A customized precyclone with a cut point of approximately 0.5 µm was attached to the TEOM when real-time C_m of nanoparticles was measured. The DustTrak DRX is a light-scattering laser photometer that measures simultaneously PM1 ($D_{ae} < 1$ µm; D_{ae} is an aerodynamic diameter), PM2.5 ($D_{ae} < 2.5$ µm), PM4 ($D_{ae} < 4.0$ µm), PM10 ($D_{ae} < 10$ µm), and total particulate mass (TPM) ($D_{ae} < 15$ µm) size fractions.[14] The results were corrected with the correction factor obtained from the weighed

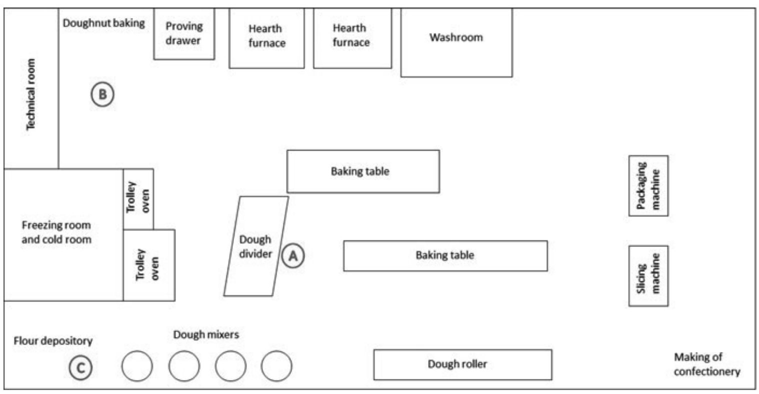

Figure 1. Baking facility and stationary sampling locations (A = baking area, B = oven area, C = flour depository).

Table 1. A list of instruments and the corresponding parameter (C_n = number concentration, C_m = mass concentration, dn/dlogD_p = number size distribution) used in the measurement campaigns 1 and 2.

Instrument	Parameter	Unit	Size range (μm)	Flow rate (dm^3/min)	Sampling time[a] (hr)	Campaign	Sampling location
CPC 3775	C_n	cm^{-3}	0.004–3	1.5	—	1	S-A, S-B
CPC 3776	C_n	cm^{-3}	0.0025–3	1.5	—	1, 2	S-C, S-A
SMPS	dn/dlogD_p	cm^{-3}	0.01–0.70	0.3	—	1	S-A
FMPS	dn/dlogD_p	cm^{-3}	0.056–0.56	10	—	1	S-A
TEOM 1405	C_m	mg/m^3	< 100	2.0	—	2	S-A
DustTrak DRX 8533	C_m	mg/m^3	0.1–15	3.0	—	2	S-A
IOM sampler	C_m	mg/m^3	< 100	2.0	5	2	BZ, S-A
PM1	C_m	mg/m^3	< 1	10	5	1	S-A
EM[b]	morphology	—	—	0.3, 0.4	—	1, 2	BZ, S-A

Notes: BZ = breathing zone; S-A, S-B, S-C = stationary location A, B, and C, respectively; – = not applicable.
[a]Sampling time is reported for the gravimetric samples.
[b]Sampling flow rate of was 0.3 dm^3/min in campaign 1 and 0.4 dm^3/min in campaign 2.

filter. The C_m of TPM and PM1 are reported in this study.

IOM (SKC, USA) samples were collected on nitrocellulose-filters (0.8 μm AAWP; Merck Millipore, Germany) to determine C_m of the 1) inhalable dust (D_{ae} < 100 μm) and 2) nanoparticles (in this study, $D_{ae} < 0.5$ μm) at the BZ of the dough maker and at the S-A. At both the BZ and the S-A, two parallel IOM samplers of which one had the precyclone installed, were used. In addition, PM1 samples were collected on 47-mm Teflon (Teflo R2PJ047; Pall Life Sciences, USA) and quartz fiber filters (Tissuquartz 2500QAT-UP; PALL Life Sciences) in two parallel lines. A pre-impactor (Dekati Ltd., Finland) was used to cut off particles larger than 1 μm.

Real-time C_n was measured with condensation particle counters (CPC; TSI, USA) at S-A, S-B, and S-C. Number size distribution was measured with a scanning mobility particle sizer (SMPS; TSI, USA) and a fast mobility particle sizer (FMPS; TSI, USA). The SMPS system consisted of a pre-impactor, charger (Kr-85), platform (Model 3080), differential mobility analyzer (Model 3081), and CPC 3776.

Electron microscopy (EM) samples were collected on porous carbon films (S147-4400 Holey Carbon Film 400 Mesh Cu, Agar Scientific, USA) with an aspiration sampler[15] at the BZ of the dough maker and at the S-A. In the measurement campaign 2, a pre-impactor of the SMPS was attached to the aspiration sampler to cut off large particles.

Instruments used in the measurement campaigns, corresponding parameters, and sampling location of each instrument are listed in Table 1.

Analytical methods

Concentrations of organic and elemental carbon were determined from quartz fiber filters using a thermal-optical carbon analyzer (Sunset Laboratory, USA). The analyses were performed according to National Institute for Occupational Safety and Health (NIOSH) method 5040.[16] In addition, 31 elements were determined from Teflon and IOM filters using inductively coupled plasma mass spectrometry (ICP-MS) and ion chromatography (IC) according to ISO 17294-2 and ISO 10304-1 standards.[17,18]

The morphology and composition of the particles were studied using a scanning electron microscope (SEM; Sigma HD VP, Carl Zeiss NST, UK) connected with two energy-dispersive X-ray spectroscopy detectors (EDS; Thermo Scientific, USA). SEM imaging was conducted at an accelerating voltage of 2 kV using SE2 and InLens detectors. An accelerating voltage of 10 kV was used in the EDS analyses.

Results

Number concentrations and number size distribution

The C_n of fine and nanoparticles measured with the CPC 3775 or the CPC 3776 at the S-A followed approximately the same trend during the work shift in both measurement campaigns (Figure 2a and b). Before the work shift,

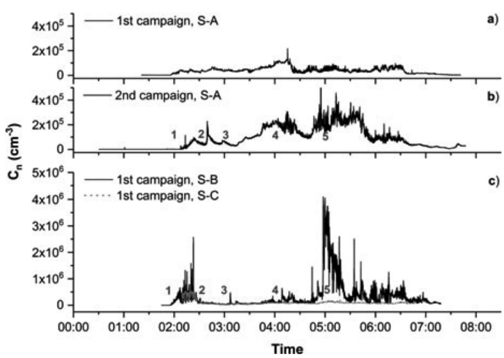

Figure 2. Particle number concentration (C_n) at the S-A (a, b), S-B and S-C (c), measured with the CPC 3775 or CPC 3776. Numbers in the figures indicate different phases of the work shift: 1 = start of the work shift, trolley ovens on; 2 = hearth ovens on; 3 = operating the trolley ovens begins; 4 = operating the hearth ovens begins; 5 = all ovens operating simultaneously.

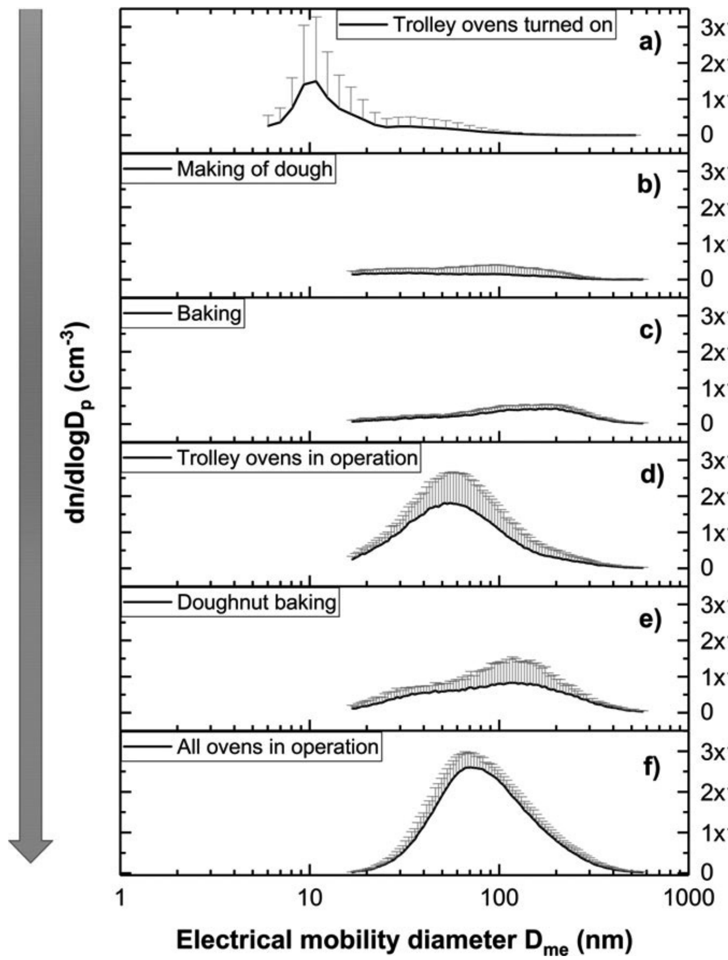

Figure 3. Average number size distribution (dn/dlogD_p) with standard deviation (grey lines) measured at the S-A (b-d) and S-B (a, e, and f). The number size distributions were measured with the SMPS, except (a) that was measured with the FMPS. Averaging periods were 10–60 min. The arrow in the left illustrates the chronological order in which the events started during the work shift.

the background C_n was low, below $1.0*10^3$ cm^{-3}. At the S-A, the C_n was approximately $5.6*10^5$ cm^{-3} at its highest during the work shift. In the first measurement campaign (Figure 2a), the C_n was lower during the latter part of the work shift due to opened window and door at 4:20 a.m.

Instantly after the ovens were turned on, particles were released into the bakery air. This can be seen from the C_n measured at the S-B and S-C (Figure 2c). At the S-B, the C_n was high, approximately $4.1*10^6$ cm^{-3}, when all the ovens were operated simultaneously (Figure 2c) at 5:00 a.m. At the S-C, the C_n was lower and ranged between $2.5*10^4$ cm^{-3} and $5.5*10^5$ cm^{-3} during the work shift.

Number size distributions were measured during the first measurement campaign with the SMPS and FMPS. The number size distribution of fine and nanoparticles fluctuated greatly (Figure 3). The formation of very small particles after turning the trolley ovens on can be seen from Figure 3a that shows the average number size distribution measured at the S-B using the FMPS. During dough making, the C_n at the S-A was low (Figure 2a and b) and the average number size distribution wide (Figure 3b). At the beginning of baking, a geometric mean diameter (GMD) of the particles was approximately 110 nm, and a geometric standard deviation (GSD) 2.2 at the S-A (Figure 3c). When operating the ovens became more frequent, the GMD at the S-A decreased to approximately 60 nm (GSD 1.9) (Figure 3d). At the beginning of doughnut baking, the average number size distribution at the S-B was bimodal (Figure 3e). When all the ovens were in operation and doughnut baking ongoing, the GMD of the particles at the S-B was approximately 90 nm (GSD 1.8) (Figure 3f).

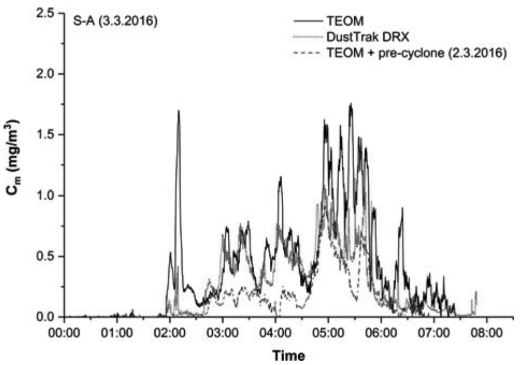

Figure 4. Real-time mass concentration (C_m) time series (5 min sliding average) measured at the S-A using the TEOM and the DustTrak DRX (3.3.2016), and the real-time C_m time series of nanoparticles measured using the TEOM with the precyclone (2.3.2016).

Mass concentrations

Real-time C_m was measured at the S-A during the second measurement campaign. During the work shift, the C_m varied remarkably, whereas background C_m was low before the work shift (Figure 4). The TPM ($D_{ae} < 15$ µm) measured with the DustTrak DRX was clearly lower than the total C_m measured with the TEOM at the beginning of the dough making (before 2:30 a.m.) but later the difference was slighter. Obviously, the C_m of nanoparticles (TEOM + precyclone) was much lower than the total C_m measured with the TEOM but the difference decreased at 4:30 a.m. to 5:30 a.m., when smaller particles released from the oven operations were more dominant.

The C_m of the inhalable dust that was determined in the measurement campaign 2 using the IOM sampler was 6.8–14.5 mg/m^3 at the BZ and 2.3–2.6 mg/m^3 at the S-A (Table 2). The nanoparticle fraction, collected using the IOM sampler with the precyclone, accounted for 9–15% and 4–8% of the inhalable dust at the BZ and the S-A, respectively. The C_m of the nanoparticles calculated from the data of TEOM with the precyclone agreed quite well with the C_m determined using the IOM with the precyclone. In addition, the PM1 concentrations determined in the measurement campaign 1 from the Teflon filters were close to the C_m measured in the campaign 2 using the IOM sampler with the precyclone. Instead, the PM1 concentrations calculated from the DustTrak DRX data were constantly higher and constituted 85–93% of the TPM.

Morphology and composition of the particles

Large flour dust particles existed in the bakery air, especially in the BZ samples (Figure 5a). These particles consisted mainly of phosphorus (P), potassium (K), nitrogen (N), sulphur (S), carbon (C), and oxygen (O). Smaller flour dust particles that resemble agglomerates were also found (Figure 5b–e). These agglomerates consisted mainly of carbon and small amounts of silicon (Si) or sulphur, and contained nanosized particles as well (Figure 5e). In addition, spherical particles containing, for example, nitrogen, silicon, phosphorus, sulphur, potassium, sodium (Na), calcium (Ca), and chlorine (Cl) were present (Figure 5b–d). Furthermore, soot agglomerates were occasionally observed in the samples (Figure 5f).

Carbon, determined with the thermal-optical carbon analyzer from the gravimetric PM1 samples, made up 42–64% of the total mass of the PM1 particles. At least 99% of the carbon was organic. The elements analyzed with the ICP-MS from the PM1 filter samples collected at the S-A composed of 1.1% of the total PM1 mass. In the IOM-samples collected from the BZ, the fraction of these elements was 0.4% of the inhalable dust, and 0.5% of the nanoparticles. In the inhalable dust, the main elements were potassium (27.0 µg/m^3), calcium (12.5 µg/m^3), and sodium (12.0 µg/m^3) (Table 3). In the nanoparticle fraction, potassium and sodium were not detected. Concentrations of water soluble ions, determined from

Table 2. Mass concentration (C_m) of the inhalable dust, nanoparticles, and PM1 at the BZ and at the S-A or S-B, determined with the IOM samplers and gravimetric PM1 sampling. In addition, arithmetic means of the total C_m and the C_m of the nanoparticles calculated from the TEOM data, and the C_m of total particulate matter (TPM) and PM1 calculated from the DustTrak DRX data for the same time period as the gravimetric IOM sampling took place, are presented.

	C_m (mg/m^3)								
	IOM		IOM + precyclone		PM1[a]	TEOM	TEOM + precyclone	DustTrak TPM	DustTrak PM1
Day	*BZ*	*S-A*	*BZ*	*S-A*	*S-A/S-B*	*S-A*	*S-A*	*S-A*	*S-A*
1	14.49	2.57	NA	0.10	0.09	NA	0.17	0.42	0.36
2	7.19	2.30	1.07	0.16	0.20	NA	0.19	0.41	0.36
3	6.81	2.49	0.62	0.20	0.21[b]	0.50	NA	0.32	0.30

NA = not available.
[a]gravimetric PM1 sampling was performed in the measurement campaign 1.
[b]sample collected from the point S-B.

Table 3. Mass concentrations (µg/m^3) of the elements determined with the ICP-MS from the PM1 Teflon filter samples collected at the S-A in the measurement campaign 1, and from the IOM filter samples collected at the BZ without (inhalable dust) and with the precyclone (nanoparticles) in the measurement campaign 2. Elements with measured mass (µg abs.) lower than the lowest observable quantity (LOQ) in all three samples are not listed.

Element	Al	B	Ba	Bi	Ca	Co	Cr	Cu	Fe	K	Mg	Mn	Mo	Na	Ni	Pb	Se	Sr	Ti	V	Zn	$C_{m,tot}$[b]
LOQ[a] *(µg abs.)*	*0.025*	*0.005*	*0.005*	*0.005*	*0.25*	*0.005*	*0.005*	*0.005*	*0.075*	*1*	*0.25*	*0.025*	*0.005*	*1*	*0.005*	*0.005*	*0.005*	*0.005*	*0.025*	*0.005*	*0.025*	—
S-A (PM1)	0.025	0.180	0	0.333	0.106	0	0	0.008	0.061	0.600	0	0	0.018	0.732	0.009	0.034	0.047	0	0	0.007	0	200
BZ (inhal. dust)	1.323	0.634	0.200	0	12.5	0.072	0.136	0.099	0.766	27.0	4.70	0.111	0.013	12.0	0.577	0	0.021	0.040	0.166	0.010	0.334	14490
BZ (nanop.)	0.386	0.286	0.012	0	3.74	0	0.084	0.076	0.258	0	0.315	0	0.013	0	0.013	0.010	0.041	0.022	0.098	0.007	0.132	1071

Notes: — = not applicable; 0 = measured mass (µg abs.) lower than LOQ.
[a]lowest observable quantity.
[b]total gravimetric mass concentration determined from the filter.

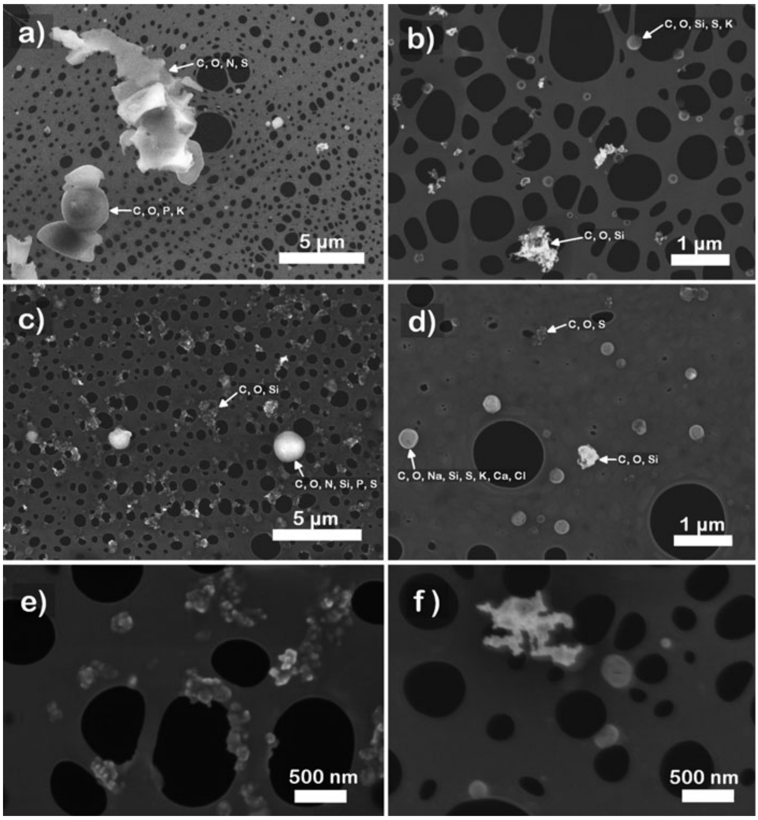

Figure 5. SEM images of the particles in the bakery air. Samples are collected at (a) the BZ without the pre-impactor, (b) the BZ with the pre-impactor, (c) the S-A without the pre-impactor, (d) the S-A with the pre-impactor. In (e), a higher magnification image of nanosized flour dust particles; and in (f), a soot agglomerate is presented. The results of the EDS analyses are inserted in figures a–d.

the PM1 filter samples collected at the S-A, were 0.5, 0.4, 2.3, and 1.6 µg/m^3 for Cl^-, NO_3^-, SO_4^{2-}, and PO_4^{3-}, respectively. The ions constituted 2.2% of the PM1 mass collected on the filter.

Discussion

To the best of our knowledge, this is the first study in which the smallest fractions of airborne particles found in a bakery are characterized and the proportion of nanoparticles of the inhalable dust is determined. In addition, the dough maker's personal exposure to nanoparticles was measured, which has not been done in previous studies. Furthermore, this study indicated that flour dust contains also nanosized particles.

The C_n and C_m fluctuated a lot in the bakery air that is in accordance with the previous studies in which real-time measurements have been performed.[7,11,12] The rapid increase in the C_n near (S-B) or behind (S-C) the trolley ovens instantly after the ovens were turned on was a clear evidence that the ovens released particles into the bakery air. The maximum temporary C_n ($4.1*10^6$ cm^{-3} at S-B) was significantly higher than in the previous study ($5.0*10^5$ cm^{-3}) in which the C_n was measured at the middle of the baking facility using an ELPI (Electrical Low Pressure Impactor; Dekati, Finland).[11] Time series of the C_n and C_m were different at the S-A at the beginning of the work shift. There was a high peak in the C_m (5 min sliding average 1.7 mg/m^3) at approximately 2:10 a.m. but not in the C_n. The peak in the C_m arose out from the weighting of flours and adding flours from sacks to the dough mixers that resulted in large particles into the air. Meijster et al.[19] and Roberge and Cloutier[7] have measured high C_m peaks at the beginning of dough mixing as well. At approximately 5:00 a.m., when all the ovens were operated simultaneously, both the C_n ($2*10^5$–$5*10^5$ cm^{-3}) and C_m (1.2–1.5 mg/m^3) were high at the S-A.

Number size distribution of the fine and nanoparticles varied remarkably, consistently with the previous studies.[11,12] Different sources, such as ovens, dough making, and doughnut baking affected the number size distribution. Very small particles were released into the bakery air from the ovens that shifted the number size distribution of the particles toward smaller sizes when operating the ovens became more frequent. Tissari et al.[11] have found that particle size reached the peak of 300 nm during doughnut baking, and, meanwhile, particle number size distribution changed to bimodal, which was also seen in this study at the beginning of doughnut baking. In other countries, number size distribution has been rarely measured with direct reading instruments. Roberge and Coultier[7] have measured number size distribution with a Grimm 1.108 optical particle counter and an eight-stage cascade impactor. A mode of smaller than 1-µm particles was observed by both methods.

In Finland, an 8-hr occupational exposure limit (OEL) for inhalable flour dust is 2 mg/m^3.[20] In this study, the C_m of the inhalable dust clearly exceeded the OEL, being 340–725% and 115–130% of the OEL at the BZ and S-A, respectively. Dusty tasks the dough maker was involved in besides dough making were, for example, rolling of dough and baking of rice pasties (including throwing rye flour to the baking table and dough), which may explain the high exposure level of the dough maker. Burstyn et al.[21] have reported that time spent pouring or dusting flours is among the main factors increasing inhalable dust exposure. However, the OEL was exceeded at the S-A as well. The C_m at the S-A was consistent with or slightly higher than the results (0.9–2.3 mg/m^3) of Tissari et al.[11,12] who determined the total C_m using US-EPA method.[22] In other Finnish studies that have covered dough making, the C_m of the total dust has been 0.9–18.8 mg/m^3 at the BZ and 0.7–14.2 mg/m^3 at the stationary locations.[23–25] In other countries, the C_m of the inhalable dust during dough making (including BZ and stationary samples) has ranged from 0.2 to 65 mg/m^3.[7,26–34] The C_m of the inhalable dust is typically larger than the C_m of the total dust because IOM-sampler is more effective for larger particles than closed cassette method.[7]

The nanoparticle fraction of the inhalable dust was 9–15% at the BZ and 4–8% at the S-A. The difference in the percentages at the BZ and S-A supports the presence of the nanosized flour dust particles at the BZ. The C_m of nanoparticles at the BZ was 0.6–1.1 mg/m^3, which is relatively high, 30–55% of the OEL of the inhalable dust. The C_m of the PM1 determined gravimetrically at the S-A, 0.1–0.2 mg/m^3, was slightly lower than the values (0.2–0.6 mg/m^3) measured by Tissari et al.[11] This is probably due to rapid fluctuation of the C_m and longer sampling times in the present study.

Carbon comprised 42–64% of the PM1 mass, and it was nearly entirely organic. The rest of the particles were probably composed mainly of hydrogen, oxygen, and nitrogen, which are the major ingredients of carbohydrates and proteins, in addition to carbon. Tissari et al.[11] have also observed that particulate carbon in the bakery was mainly organic but elemental carbon was detected too, especially in the samples taken near the ovens during heating. The elements determined with the ICP-MS analysis comprised of only 1.1% and the ions determined with the IC analysis 2.2% of the PM1, which was in accordance with the results of the previous study.[11] In the BZ samples, the fraction of the elements determined with the ICP-MS analysis was 0.4% of the inhalable dust and 0.5% of the nanoparticles. Distribution of the elements followed the composition of the ingredients used in the bakery. The SEM imaging proved that flour dust included also nanosized particles. In addition, spherical particles that originate probably at least partly from outdoor sources[35] were present in the bakery air.

Real-time measurements performed in this study revealed that high C_m period occurred for example at the beginning of the dough making when large flour particles were released into the air. Both the C_m and C_n were high when all the ovens were operated simultaneously and plenty of nanoparticles were released into the air. The primary health effects linked to the large flour particles are flour-induced sensitization and development of occupational rhinitis.[34] The smallest particles can reach the alveolar region and may play a role, for example, in allergic alveolitis type of reactions.[4,36]

Conclusions

This study reported that flour dust contained nanosized particles that made up 9–15% of the inhalable dust at the breathing zone of the dough maker. However, most of the fine and nanoparticles were released into the bakery air from the oven operations. Concentrations of fine and nanoparticles were the highest when all the ovens were operated simultaneously.

Acknowledgments

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ARTICLE II

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Controlling flour dust exposure by an intervention focused on working methods in Finnish bakeries: a case study in two bakeries

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ABSTRACT

Objectives. The purpose of this study was to examine the effectiveness of intervention strategies to control mass concentrations and peak exposures of flour dust in two Finnish bakeries. The effect of the intervention on the proportion of various particle size fractions of the total particulate matter was also investigated. *Methods.* Mass concentrations of flour dust were measured during three working days in a pre-intervention and post-intervention study in both an industrial and a traditional bakery. Gravimetric sampling and real-time measurements were performed. Relevant intervention strategies focused on working methods were planned in collaboration with the managers of the bakeries. *Results.* The average mass concentration of inhalable flour dust reduced in most of the stationary locations post intervention. The reductions in exposure levels were between 39 and 45%. However, the exposure levels increased 28–55% in the breathing zone. Real-time measurements showed reductions in the peak mass concentrations in the traditional bakery post intervention. In both bakeries, the total particulate matter size fraction consisted predominantly of particles with an aerodynamic diameter lower than 1 μm and greater than 10 μm. *Conclusion.* Further studies are needed to plan more effective intervention measures supplemented by technical control methods in both bakeries.

KEYWORDS
baker; fine particles; indoor air quality; mass concentration; occupational

1. Introduction

In Finland, the bakery sector includes small, medium-sized and large bakeries, as well as confectionary shops. The bakery industry is the largest food sub-industry in Finland, comprising nearly 700 companies and employing approximately 8000 full-time workers [1]. Of these workers, approximately 5000 are exposed to flour dust [2].

The dust in bakeries contains particles from cereal flours and various other ingredients, such as enzymes (e.g., fungal α-amylase), chemical ingredients (e.g., preservatives), flavourings, spices and other additives (e.g., baker's yeast, sugar) as well as contaminants, e.g., storage-related mites and microbes. Several of these components are sensitizers [3,4].

Cereal flour dust may cause respiratory, dermal and conjunctival reactions among bakery workers [3]. The most severe health outcome is baker's asthma, one of the most common occupational asthmas [5,6]. Exposure to flour dust can also lead to rhinitis, conjunctivitis, chronic bronchitis and bronchial obstruction [7,8]. These symptoms may be either allergic in origin, stemming from sensitization of the worker as the proteins of flour dust are potential allergens, or caused by non-specific irritation [3,4].

According to the latest information provided by the Finnish Institute of Occupational Health (FIOH), in 2008–2016, the FIOH conducted over 240 flour dust measurements, of which 42% exceeded the Finnish occupational exposure limit (OEL) of 2 mg/m^3 and 56% were greater than half of the OEL in the Finnish food industry. In Finland, flour dust causes approximately 40 occupational diseases annually [2]. Since exposure levels often exceed the OEL, interventions to reduce occupational flour dust exposure are urgently needed.

Nieuwenhuijsen et al. [9] showed that the time-weighted average (TWA) mass concentration (C_m) in bakeries is significantly determined by peak exposures associated with specific work activities. Meijster et al. [10] found that $> 75\%$ of TWA exposure was directly associated with peak exposures. These exposures are known to be frequent in bakeries and they may contribute to work-related adverse health effects [3,4]. This suggests that controlling peak exposure levels might play an important role in lowering sensitization as well as both allergic and other symptoms. Therefore, intervention strategies are needed for the work tasks that are linked to peak exposures.

Research on the effectiveness of interventions in bakeries is scarce, and especially studies focused on controlling peak exposures are lacking. To the best of our knowledge, there are only three studies focusing on the effectiveness of interventions in the bakery industry [11–13]. Two of these studies [12,13] were conducted in supermarket bakeries, whereas Meijster et al. [11] included traditional and industrial bakeries, as well as flour mills and ingredient producers, in their study.

This study presents the follow-up exposure results of a small-scale intervention focused on the working methods of bakery workers. The aim of the present study was to investigate the effectiveness of the intervention to control C_m and peak exposures of flour dust in Finnish industrial and traditional bakeries. The effect of the intervention on the proportions of various size fractions of the total particulate matter (TPM) was also examined.

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2. Methods

2.1. Study design

The study was conducted in two Finnish bakeries: an industrial, large-size bakery and a traditional, small-size bakery. They were randomly selected for the study considering the size of the bakery and the number of workers.

2.2. Industrial bakery

The industrial bakery, built in 2017, had a daily workforce of 15 workers on average. The floor area was approximately 3500 m^2, and the average production output was 20–25 doughs (147 kg/dough) and 400–600 products per day. Only wheat flours were used in the products. The bakery facilities were divided into three units consisting of a bun-baking unit (production of filled and non-filled shaped buns), a confectionary unit (production of cakes and gateaux) and a packaging unit. There were six to nine line workers on two production lines (cutting the dough batches, filling and forming the buns, placing the buns on baking sheets) and one dough maker in the bun-baking unit. The line workers also took the buns to a dough resting cabinet, decorated the buns and put them into trolley ovens.

The confectionary and packaging units each had approximately three workers. The dough maker of the bun-baking unit occasionally participated in the tasks of the production lines in addition to dough-making (weighing and mixing ingredients, using a dough feeder and dough divider). Mechanical ventilation existed in the building and the bakery machines were connected to the ventilation system.

Every working day began with dough-making between 05:30 and 06:00, and the working days ended between 12:30 and 14:00. The production lines, tables and floors were brushed each day, usually at the end of the work shifts. With regard to personal protective equipment (PPE), the workers used working clothes and caps, but not respiratory protective equipment (RPE), goggles or gloves.

2.3. Traditional bakery

The traditional bakery, built in 1960, had an average daily workforce of eight workers. The floor area was approximately 130 m^2, and the average production output was 40–50 doughs (40 kg/dough) per day and 2500–3000 products per day. Wheat, rye and barley flours were used in the products. The bakery had two floors, including a main production unit (production of bagels, breads, bread rolls, buns, doughnuts, pasties, pastries, pies and pizzas) upstairs and a confectionary unit (production of cakes and gateaux) downstairs. A packaging unit and an outlet store also existed upstairs besides the main production unit.

Upstairs, four workers baked the products (e.g., weighing ingredients and mixing them with dough mixer tubs, using the bakery machines, shaping, filling and decorating products on a baking table, putting the products into trolley ovens). One person packed the products and two persons worked in shifts in the outlet store, while one person worked downstairs in the confectionary unit. All of the products were baked in trolley ovens in the main production unit upstairs. The building had natural ventilation; however, local exhaust fans were connected to a doughnut fryer and trolley ovens.

The starting time of a working day ranged between 23:00 and 24:00. First, the bakery manager started dough-making for various breads and worked alone with bread baking for about 5–6 h. The other workers arrived between 05:00 and 06:00, when they started baking the other products. The duration of the working days of the bakery manager and other workers were 10–11 and 6–8 h, respectively. The workers brushed the floor and tables each day, usually at the end of the work shifts. They used working clothes and caps, but not RPE, goggles or gloves.

2.4. Particle measurements

The C_m of flour dust was measured during three normal working days in the bakeries in a pre-intervention and post-intervention study (see later). The measurements were conducted on three consecutive days, except in the industrial bakery where the pre-intervention study included two consecutive measurement days and then a day 2 weeks later due to schedule reasons in the bakery.

Full-shift dust samples were collected on nitrocellulose filters (0.8 µm AAWP; Merck KGaA, Germany) using Institute of Occupational Medicine (IOM) samplers (SKC Inc., USA) and sampling pumps (SKC AirChek Sampler, Model 224-PCXR4; SKC Inc., USA) with a calibrated air flow of 2 L/min. The calibration was done using a mini-BUCK Calibrator Model M-5 (A. P. Buck Inc., USA). The IOM samplers consisted of stainless steel cassettes and plastic housing bodies. Sampling was conducted to determine the C_m of inhalable dust (aerodynamic diameter [D_{ae}] $<$ 100 µm) in the breathing zone and stationary samples.

In the industrial bakery, the IOM samples were collected in the bun-baking unit in the breathing zone of the dough maker (BZ1) and a randomly selected line worker (BZ2), and at two stationary locations: beside a production line (S1) and beside a table where the dough maker weighed the ingredients (S2) (Figure 1). In the traditional bakery, sampling was conducted in the main production unit in the breathing zone of a general baker (BZ3) who had several work tasks, and at two stationary locations in the vicinity of a baking table beside a dough divider (S3) and beside a dough roller (S4) (Figure 2). The sampling height at the stationary locations was approximately 1.4 m. The filters placed inside the sampling cassettes were weighed using a microbalance (Mettler-Toledo MT5; Mettler-Toledo, LLC, USA) prior to and after the sampling in an acclimatization room where they were stabilized for at least 24 h (relative humidity 40%, 20 °C). A Statickmaster 2U500 Alpha Ionizer (StaticTek, USA) was used to eliminate static charges of the filters.

Real-time monitoring of C_m was performed with a DustTrak DRX Aerosol Monitor 8533 (DRX) (TSI Inc., USA) at stationary locations S2 and S4 with 30-s intervals. The DRX measures simultaneously the following particulate matters: PM1 (D_{ae} $<$ 1 µm), PM2.5 (D_{ae} $<$ 2.5 µm), PM4 (D_{ae} $<$ 4 µm), PM10 (D_{ae} $<$ 10 µm) and TPM (D_{ae} $<$ 15 µm) size fractions. A simultaneous gravimetric sample was collected on a nitrocellulose filter (37 mm AAWP; Merck KGaA, Germany) placed on the filter body inside the DRX, and the filter was changed for each measurement day. The pre-sampling and post-sampling weights of the filters were measured after 24 h of conditioning in the acclimatization room. The C_m for each measurement day was corrected with a correction factor calculated as a quotient of the filter sample C_m and an average C_m of the TPM obtained from the device. The DRX was positioned on a table,

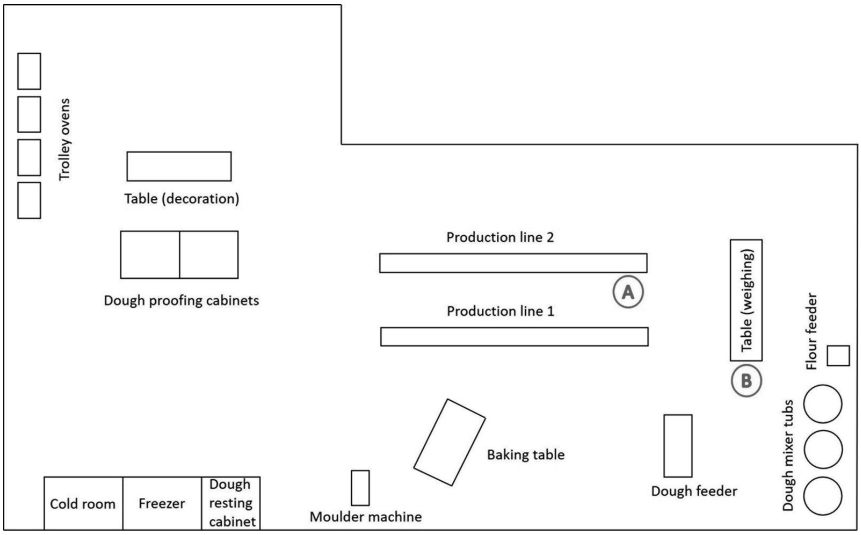

Figure 1. Baking facility of the industrial bakery (bun-baking unit) and stationary sampling locations (A = S1; B = S2). Note: Both production lines included a dough roller machine that was connected to the dough feeder. S1 = beside a production line; S2 = working area of the dough maker.

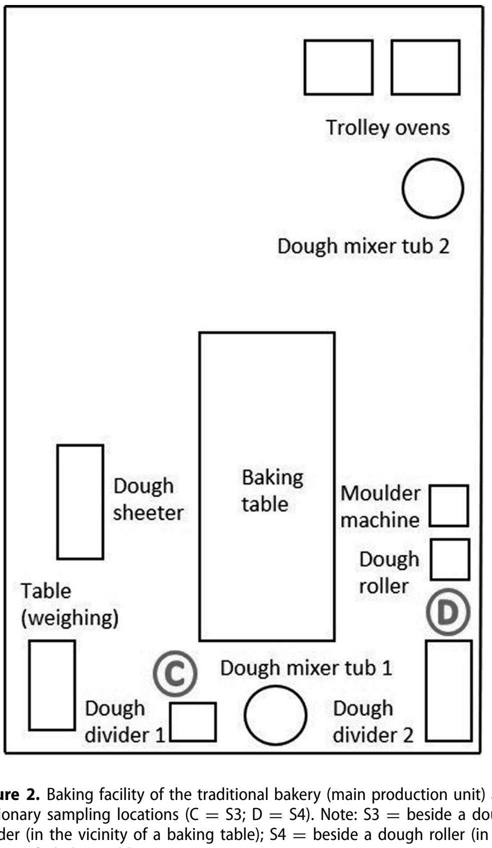

Figure 2. Baking facility of the traditional bakery (main production unit) and stationary sampling locations (C = S3; D = S4). Note: S3 = beside a dough divider (in the vicinity of a baking table); S4 = beside a dough roller (in the vicinity of a baking table).

the sampling height was approximately 0.8 m in both bakeries and the daily duration of the monitoring varied between 6 and 8 h.

2.5. Intervention

Work activities associated with peak exposures in the bakeries were evaluated during the pre-intervention study. In addition, various other working activities that required improvement to control dust levels in the bakeries were identified. Based on the information obtained from the pre-intervention study, intervention strategies focusing on working methods were planned and implemented, with the aim to reduce flour dust exposure. With regard to the intervention measures, only working methods were investigated in this study, because these control measures might provide a cost-effective strategy to reduce flour dust exposure in bakeries compared to technical control methods (e.g., local exhaust ventilation, a new ventilation system, etc.). Furthermore, these control measures can be applied in both industrial, large-scale bakeries and traditional, small-scale bakeries.

Two different-sized bakeries were selected for this study to examine whether there are differences in the effectiveness of and adherence to the intervention strategies between the industrial and traditional bakery. In the industrial bakery, the degree of automation was high; however, considering the dough-making process, the pre-intervention working methods were similar between the bakeries. The C_m was measured in proximity to the dough-making area in both bakeries.

The intervention strategies were planned using checklists developed by Säämänen et al. [14] for dust reduction in various working activities. Relevant strategies for the bakeries were selected from the checklists in collaboration with the managers, who introduced the checklists and intervention measures to the workers and trained them to follow the new working methods (Table 1). In the industrial bakery, the checklists were mounted on the bakery walls, whereas in the traditional bakery, the lists were available on a table in the vicinity of the workers. In both bakeries, all of the workers aimed to follow the new working methods.

Prior to the post-intervention measurements, both bakeries were visited once to check and reinforce intervention

Table 1. Intervention strategies the bakeries aimed to follow to control flour dust exposure after the pre-intervention study.

Working method	Industrial bakery	Traditional bakery
1. Ingredients added at as low a height as possible to bowls/dough mixers	x	x
2. Empty bags handled with care to avoid dusting	x	x
3. Floor and surfaces cleaned immediately when weighing ingredients	x	–
4. Floor and surfaces cleaned immediately when handling the bags	x	–
5. Bags emptied holding their mouth near to the bottom of the bowls/tubs	x	x
6. Empty bags flattened outside the work area	x	x
7. Flour thrown from as low a height as possible onto the baking table	–	x

adherence. Information on the adoption of the intervention strategies was obtained from a walk-through survey in the bakeries. The interventions in the industrial and traditional bakery were implemented for approximately 6 and 3.5 months, respectively. Different follow-up times were selected due to schedule reasons in the bakeries. The follow-up times were < 1 year as the intervention strategies were expected to be adopted in a relatively short time frame, and so the effect of the intervention was assumed to be seen in a few months in both bakeries. At the end of the implementation periods, post-intervention measurements were conducted to examine the overall effectiveness of the intervention strategies in the bakeries.

2.6. Data analysis

The effectiveness of the intervention was assessed by calculating arithmetic means for the pre-intervention and post-intervention C_m of the IOM samples (averaged over the repeat full-shift samples pre and post intervention) and DRX results (averaged over the complete dataset pre and post intervention). Post-intervention percentages were calculated from the average C_m results.

Regarding the real-time monitoring, peak exposure was defined as $C_m > 2$ mg/m^3 in this study. Duration of the C_m peaks was computed by considering the peaks of > 2 mg/m^3 of the TPM size fraction during each measurement day. Furthermore, the pre-intervention and post-intervention proportion of various size fractions of the TPM was calculated by dividing the C_m of each size fraction by the C_m of the TPM at each time point and averaging over the complete dataset.

3. Results

3.1. Inhalable dust

The pre-intervention and post-intervention results of the full-shift inhalable dust (IOM) samples collected from the breathing zone of the workers and stationary locations are presented in Table 2. In the industrial bakery, the average C_m reduced 45 and 40% at S1 and S2 post intervention, respectively. In the breathing zone, no reduction in the exposure levels was achieved post intervention, and in fact, the average C_m increased 28% (BZ1) and 55% (BZ2).

In the traditional bakery, the average C_m at S3 reduced 39% post intervention. The results showed no reduction in BZ3 and at S4 where, instead, the average C_m increased 24 and 54%, respectively.

3.2. Real-time monitoring

Table 3 presents the real-time, full-shift C_m values measured with the DRX at S2 (industrial bakery) and S4 (traditional bakery) pre and post intervention. In the industrial bakery, the average C_m reduced 22, 21, 22, 25 and 20% in the PM1, PM2.5, PM4, PM10 and TPM size fractions, respectively. No reductions in the average C_m were achieved in the traditional bakery. Regarding the night shifts, the average C_m increased 73% (PM1), 73% (PM2.5), 71% (PM4), 34% (PM10) and 22% (TPM). Concerning the morning shifts, the average C_m increased 217% (PM1), 216% (PM2.5), 208% (PM4), 130% (PM10) and 93% (TPM). The concentrations were higher during the night shifts compared to the morning shifts pre and post intervention.

During the working days, the C_m fluctuated greatly, and several peak exposures (the peak C_m of the TPM size fraction is presented as an example hereafter) were measured in the bakeries (Figures 3 and 4). In the industrial bakery, the C_m was at its highest when all the workers were working at the same time. The highest C_m peaks were approximately 18.7 and 33.1 mg/m^3 pre and post intervention, respectively. During measurement day 4, fewer peaks were measured because the DRX was positioned about 2 m away from S2 for approximately 5 h due to space issues. For the rest of the day, the DRX was placed at S2. The number of peaks varied between the measurement days depending on the production output and workload of the workers. The highest C_m peaks were lower pre intervention than post intervention. The duration of the single peaks (duration of peaks > 2 mg/m^3 for the TPM size fraction is presented as an example hereafter) were between 0.5 and 3 min pre and post intervention.

In the traditional bakery, the number of C_m peaks was clearly greater during both night and morning shifts post intervention. However, the highest C_m peaks were lower post intervention than pre intervention during both shifts. The peak C_m was 39.2 mg/m^3 (morning shift) and 56.9 mg/m^3 (night shift) pre intervention, and 20.9 mg/m^3 (morning shift) and 36.7 mg/m^3 (night shift) post intervention. The workload of the workers and production output differed from day to day, which explains the variation in the number of peaks between the measurement days. The durations of the shortest single peaks were approximately 30 s during both shifts pre and post intervention, whereas the longest peaks were approximately 10 min (morning shift) and 11 min (night shift) pre intervention, and 18 min (morning shift) and 1 h 4 min (night shift) post intervention.

In both bakeries, the TPM consisted mainly of the PM1 ($D_{ae} < 1$ μm), PM4–10 (4 μm $< D_{ae} <$ 10 μm) and PM10–15 (10 μm $< D_{ae} <$ 15 μm) fractions pre and post intervention. Figures 5 and 6 illustrate that the proportions of the PM1, PM4–10 and PM10–15 fractions of the TPM varied greatly during the working days. In both bakeries, the PM1 and PM10–15 fractions clearly dominated, and most of the time, the proportion of the PM1 fraction was significantly greater compared to the other fractions. In the industrial bakery, the PM1 fraction

Table 2. Mass concentration (C_m) of the full-shift IOM samples collected in the breathing zone and stationary locations.

	C_m (mg/m^3)							
	Pre intervention				Post intervention			
Sample	AM	GM	GSD	Range	AM	GM	GSD	Range
Industrial bakery								
BZ1	1.6	1.6	1.2	1.3–2.0	2.1	3.1	1.3	2.7–3.7[a]
BZ2	0.4	0.4	1.5	0.3–0.5[a]	0.7	0.6	1.1	0.6–0.7[a]
S1	0.4	0.4	1.4	0.3–0.5	0.2	0.2	1.5	0.2–0.3
S2	0.5	0.5	1.7	0.3–0.7	0.3	0.3	1.8	0.2–0.5
Traditional bakery								
BZ3	11.6	11.3	1.3	9.4–15.1	14.4	13.1	1.7	8.2–22.7
S3	2.6	2.6	1.2	2.1–3.0	1.6	1.5	1.6	0.9–2.1
S4	0.8	0.7	1.4	0.6–1.1	1.2	1.2	1.2	1.0–1.4

[a]Two measurements conducted.
Note: Pre-intervention and post-intervention studies included three measurement days. AM = arithmetic mean; BZ1 = dough maker; BZ2 = line worker; BZ3 = general baker; GM = geometric mean; GSD = geometric standard deviation; IOM = Institute of Occupational Medicine; S1 = beside a production line; S2 = working area of the dough maker; S3 = beside a dough divider (in the vicinity of a baking table); S4 = beside a dough roller (in the vicinity of a baking table).

Table 3. Real-time, full-shift mass concentration (C_m) measured with the DRX in stationary locations S2 and S4.

	C_m (mg/m^3)							
	Pre intervention				Post intervention			
Size fraction	AM	GM	GSD	Range	AM	GM	GSD	Range
Industrial bakery								
PM1	0.2	0.3	2.4	0.0–5.0[a]	0.1	0.1	2.6	0.0–12.1
PM2.5	0.2	0.3	2.4	0.0–5.0[a]	0.1	0.1	1.7	0.0–12.1
PM4	0.2	0.3	2.4	0.0–5.1[a]	0.1	0.1	2.6	0.0–12.1
PM10	0.3	0.4	2.5	0.0–8.1[a]	0.2	0.1	1.7	0.0–17.5
TPM	0.4	0.2	2.8	0.0–18.7[a]	0.3	0.1	2.6	0.0–33.1
Traditional bakery								
PM1 night shift	0.7	0.2	5.1	0.0–12.5[a]	1.2	0.5	4.5	0.0–9.8
PM1 morning shift	0.1	0.1	3.4	0.0–10.1	0.4	0.2	2.9	0.0–6.8[a]
PM2.5 night shift	0.7	0.2	5.0	0.0–12.5[a]	1.2	0.5	4.5	0.0–9.8
PM2.5 morning shift	0.1	0.1	3.3	0.0–10.1	0.4	0.6	2.9	0.0–6.8[a]
PM4 night shift	0.7	0.2	4.8	0.0–12.7[a]	1.2	0.5	4.5	0.0–9.9
PM4 morning shift	0.1	0.1	3.3	0.0–10.9	0.5	0.6	2.9	0.0–6.8[a]
PM10 night shift	1.0	0.3	4.9	0.0–28.3[a]	1.3	1.5	4.3	0.0–15.0
PM10 morning shift	0.3	0.1	3.3	0.0–25.0	0.6	0.3	2.9	0.0–14.5[a]
TPM night shift	1.4	0.4	5.1	0.0–56.9[a]	1.7	0.7	4.2	0.0–36.7
TPM morning shift	0.4	0.3	3.4	0.0–39.2	0.8	0.4	3.0	0.0–20.9[a]

[a]Measurements conducted on two working days.
Note: Pre-intervention and post-intervention studies included three measurement days. AM = arithmetic mean; D_{ae} = aerodynamic diameter; DRX = DustTrak DRX Aerosol Monitor 8533; GM = geometric mean; GSD = geometric standard deviation; PM = particulate matter; PM1 = size fraction with $D_{ae} < 1$ µm; PM2.5 = size fraction with $D_{ae} < 2.5$ µm; PM4 = size fraction with $D_{ae} < 4$ µm; PM10 = size fraction with $D_{ae} < 10$ µm; S2 = working area of the dough maker; S4 = beside a dough roller (in the vicinity of a baking table); TPM = total particulate matter.

made up an average of 62 and 58% of the TPM pre and post intervention, respectively. Generally, when C_m was < 1 mg/m^3 the PM1 fraction dominated, whereas during $C_m > 1$ mg/m^3 the proportion of the PM10–15 fraction was usually greater. In the traditional bakery, the PM1 fraction accounted for, on average, 64 and 72% in the night shifts pre and post intervention, respectively. Regarding the morning shifts, the proportions were 44 and 60% pre and post intervention, respectively.

In the industrial bakery, the time series of the measurement days were quite similar, except for measurement day 1 (Figure 5a) between 11:00 and 14:00 when the proportion of the PM1 fraction ranged from 80 to 100%. Regarding the night shifts of measurement day 2 (Figure 6b) and day 5 (Figure 6d) in the traditional bakery, the time series of the proportions were also quite similar. In the morning shifts, the proportion of the PM1 fraction was, on average, greater compared to the other fractions on measurement day 5 than on measurement day 2.

3.3. Adherence to the intervention

Implementation of the intervention strategies was left to the responsibility of the managers and employees. The walk-through survey prior to the post-intervention measurements in the bakeries showed that all of the control measures were implemented frequently by the workers, except the working methods related to cleaning. The floor and surfaces were not cleaned immediately when weighing ingredients and handling the bags. However, cleaning was conducted each day at the end of the work shifts in both bakeries.

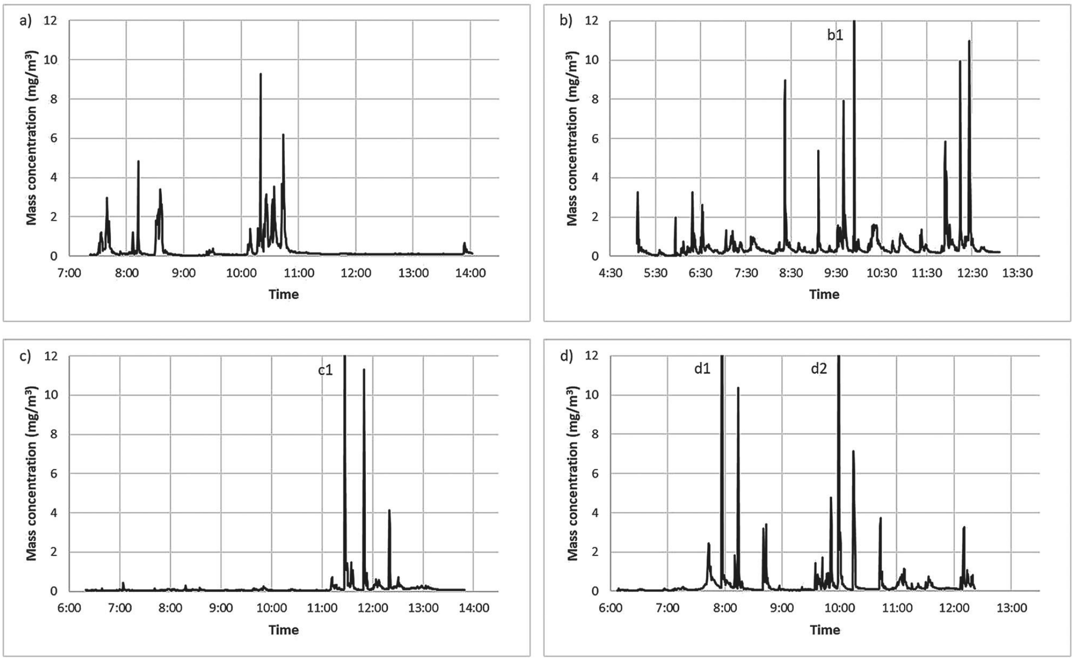

Figure 3. Real-time mass concentration (C_m) time series (24-h scale and 30-s sliding average) of the TPM size fraction measured at S2 in the industrial bakery using the DRX: (a), (b) pre-intervention C_m for measurement days 1–2; (c), (d) post-intervention C_m for measurement days 4–5. Note: Peak C_m is 18.7 mg/m^3 (b1), 27.2 mg/m^3 (c1), 20.0 mg/m^3 (d1) and 19.1 mg/m^3 (d2). DRX = DustTrak DRX Aerosol Monitor 8533; S2 = working area of the dough maker; TPM = total particulate matter (aerodynamic diameter [D_{ae}] < 15 μm).

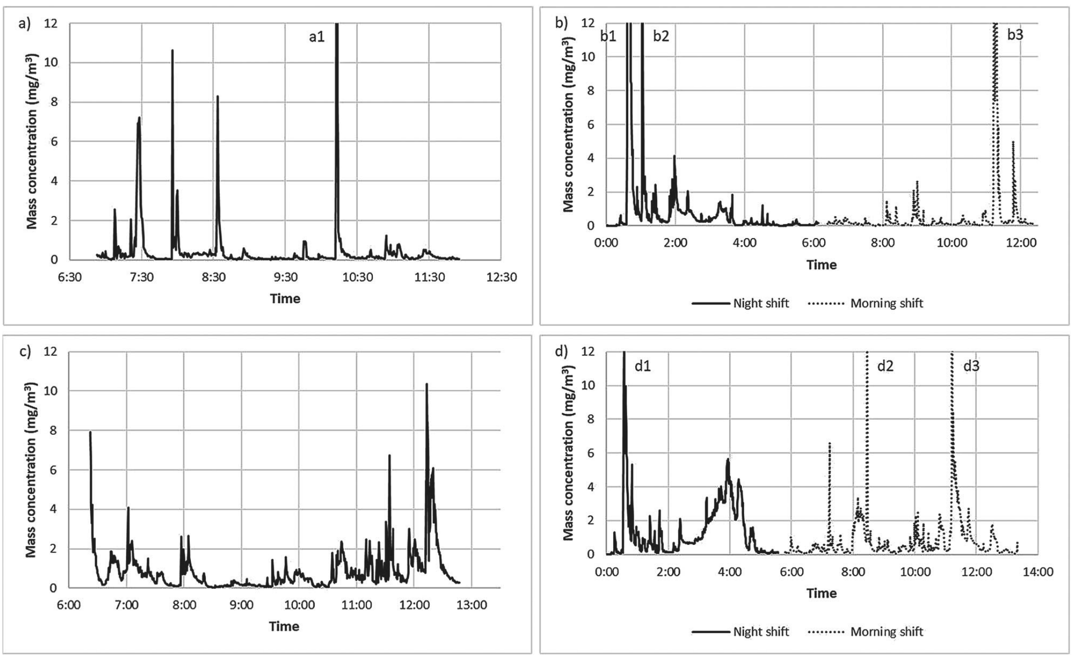

Figure 4. Real-time mass concentration (C_m) time series (24-h scale and 30-s sliding average) of the TPM size fraction measured at S4 in the traditional bakery using the DRX: (a), (b) pre-intervention C_m for measurement days 1–2; (c), (d) post-intervention C_m for measurement days 4–5. Note: C_m time series of (a), (c) morning shift and (b), (d) both night and morning shifts (separated by different lines). Peak C_m is 38.6 mg/m^3 (a1), 56.9 mg/m^3 (b1), 18.4 mg/m^3 (b2), 39.2 mg/m^3 (b3), 14.5 mg/m^3 (d1), 15.1 mg/m^3 (d2) and 20.9 mg/m^3 (d3). DRX = DustTrak DRX Aerosol Monitor 8533; S4 = beside a dough roller (in the vicinity of a baking table); TPM = total particulate matter (aerodynamic diameter [D_{ae}] < 15 μm).

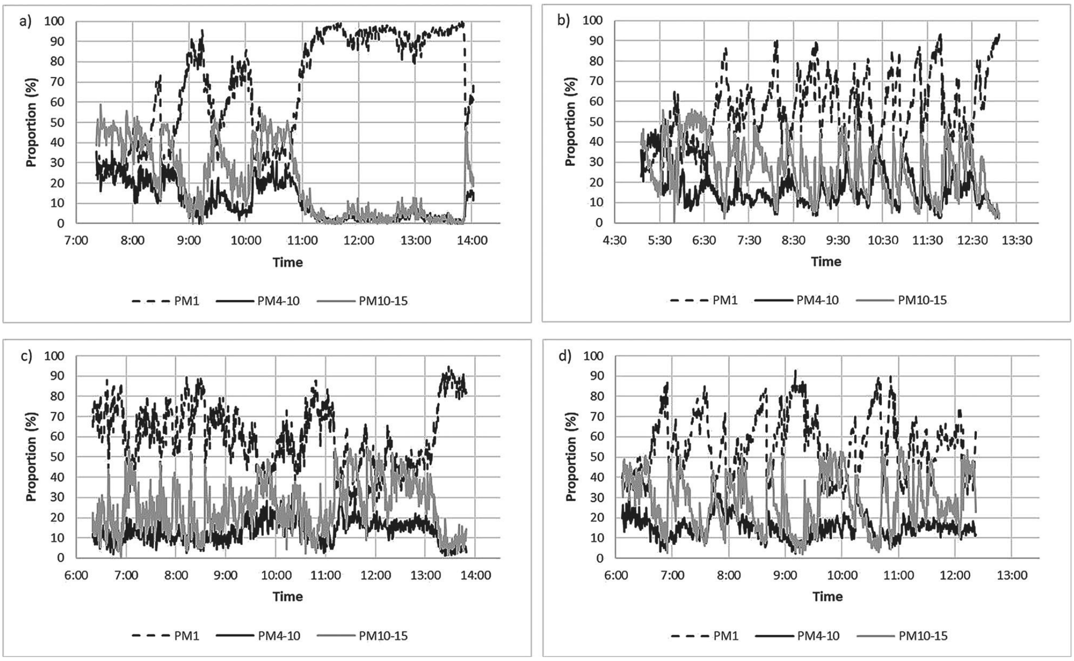

Figure 5. Real-time mass concentration (C_m) time series (24-h scale and 30-s sliding average) of the proportions of TPM in the industrial bakery presented for the PM1 (D_{ae} < 1 µm), PM4–10 (4 µm < D_{ae} < 10 µm) and PM10–15 (10 µm < D_{ae} < 15 µm) size fractions: (a), (b) pre intervention for measurement days 1–2; (c), (d) post intervention for measurement days 4–5. Note: C_m measured at S2 using the DRX. DRX = DustTrak DRX Aerosol Monitor 8533; PM = particulate matter; S2 = working area of the dough maker; TPM = total particulate matter (aerodynamic diameter [D_{ae}] < 15 µm).

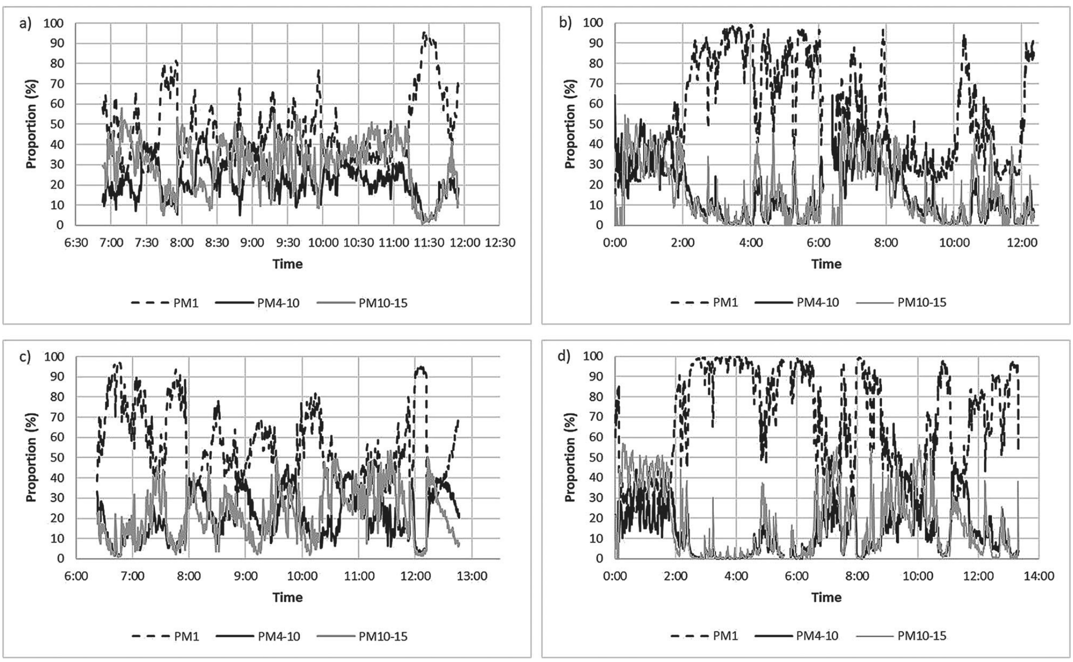

Figure 6. Real-time mass concentration (C_m) time series (24-h scale and 30-s sliding average) of the proportions of TPM in the traditional bakery presented for the PM1 (D_{ae} < 1 µm), PM4–10 (4 µm < D_{ae} < 10 µm) and PM10–15 (10 µm < D_{ae} < 15 µm) size fractions: (a), (b) pre intervention for measurement days 1–2; (c), (d) post intervention for measurement days 4–5. Note: C_m measured at S4 using the DRX. C_m time series of (a), (c) morning shift and (b), (d) both night and morning shifts (morning shift starts approximately at (b) 06:25 and (d) 05:50). DRX = DustTrak DRX Aerosol Monitor 8533; PM = particulate matter; S4 = beside a dough roller (in the vicinity of a baking table); TPM = total particulate matter (aerodynamic diameter [D_{ae}] < 15 µm).

4. Discussion

In the industrial bakery, the average C_m reduced 45 and 40% in stationary samples S1 and S2 post intervention, respectively. This finding may be explained by the intervention strategies or by random variation in the exposure levels between the measurement days. Post intervention, the average C_m was lower than the Finnish OEL of 2 mg/m^3 [15]. However, in the breathing zone samples, the average C_m increased 28 and 55% in BZ1 (dough maker) and BZ2 (line worker), respectively. A possible explanation for this may be that the workload of the workers and production output might have been greater during the post-intervention measurements, which could have contributed to the high exposure levels. Post intervention, the average C_m exceeded the OEL in BZ1 but was lower than the OEL in BZ2. The C_m of the dough maker was greater compared to the line worker, because dough-making included the dustiest work tasks in a relatively large area. The line workers spent no time in the dough-processing area, and so they had less contact with dust than the dough maker.

In the traditional bakery, a 39% reduction in the average C_m was achieved in stationary sample S3 post intervention, whereas the average C_m increased 54% at S4. S3 was located beside the area where the ingredients were weighed and mixed, and intervention measures were implemented in these work phases, which may explain the differences in the results. However, the C_m reduction might be also attributed to random variation in the exposure levels. S4 was located closer to the baking table, and even though one of the intervention strategies was to throw flour from as low a height as possible onto the table, no reduction in the average C_m was observed. Regarding the breathing zone, the average C_m increased 24%, which may be related to the facts discussed earlier (greater workload of the workers, production output, lack of adherence to some intervention strategies). Post intervention, the average C_m exceeded the OEL in BZ3 but was lower than the OEL at the stationary locations.

This study showed that the follow-up time (< 1 year) was sufficient for the workers to adapt to most of the intervention strategies in both bakeries. The workers employed the control measures frequently, but the new working methods related to cleaning were not applied. A possible reason for inadequate cleaning was the busy schedule in both bakeries. Due to the relatively short follow-up periods, the current study could not monitor flour dust levels in the longer term. Both bakeries should pay attention to the maintenance of control measures. Moreover, the bakeries should place emphasis on training and supervision of new workers regarding the control measures.

Previous intervention studies have examined the effectiveness of an intervention in the breathing zone only and included intervention measures focused on both technical control methods and work practices. Baatjies et al. [12] obtained reductions of 23–67% in inhalable flour dust levels in South African supermarket bakeries. Meijster et al. [11] investigated changes in exposure over time and found a modest downward annual trend of −2% for flour dust in Dutch bakeries. Regarding the previous Finnish intervention study [13], an average reduction of 64% in inhalable flour dust levels was achieved in supermarket bakeries.

In studies where flour was substituted with divider oil, clear reductions in flour dust exposure levels were achieved [12,16,17]. A study by Meijster et al. [18] showed that control measures introduced during the weighing of ingredients, e.g., limiting the use of bagged flour products and the enclosure of silos (when dumping flour), significantly decreased the exposure levels. A rather low reduction level was observed when dusting flour was substituted with oil, which is contradictory to what was found by Burstyn et al. [16,17] and Baatjies et al. [12]. Meijster et al. [10] suggested that the most effective control measures to reduce flour dust exposure in bakeries were wet cleaning, no shaking of the cotton hose attached to the flour silo and no flour dusting. Baatjies et al. [12] found that the best results in reducing flour dust levels were observed when combining engineering controls and training.

The C_m of the dough maker (1.3–3.7 mg/m^3) in BZ1 are lower than the levels (6.8–14.5 mg/m^3) reported by Karjalainen et al. [19]. However, the previous study was conducted in a traditional bakery. In other countries, personal exposure levels of inhalable dust during dough-making have varied between 0.1 and 65.0 mg/m^3 [20–28].

Regarding the C_m of the line worker (0.3–0.7 mg/m^3) in BZ2, the results are lower than the levels (0.5–12.0 mg/m^3) reported in previous studies [23,26]. The C_m of the general baker (8.2–22.7 mg/m^3) in BZ3 are, on average, greater compared to the all-round staff (0.1–26.8 mg/m^3) in the previous studies, which were, however, conducted in industrial bakeries [22,24,29].

Considering the C_m of inhalable dust in the stationary samples (0.2–3.0 mg/m^3), the results agree quite well with those (1.6–1.9 and 0.9–4.2 mg/m^3) obtained by Brisman et al. [23] and Tissari et al. [30], respectively, but are, on average, lower than the levels reported by Bulat et al. [24] (0.1–9.0 mg/m^3) and Roberge et al. [31] (0.2–19.0 mg/m^3).

The real-time measurements showed that the average C_m reduced 20–25% (all size fractions included) at S2 in the industrial bakery post intervention. These reductions are in line with those obtained by the IOM samplers. In the traditional bakery, no reductions in the average C_m at S4 were observed, which was also seen in the IOM samples. Regarding the night and morning shifts, the average C_m increased 22–73 and 93–217% (all size fractions included) at S4 post intervention, respectively. As discussed earlier, the workload of the workers and production output varied daily, and there was a lack of adherence to some intervention strategies, which explains the variation in the average C_m and number of peak exposures between the measurement days in both bakeries.

In the industrial bakery, the peak C_m arose from weighing ingredients, adding flour from sacks to the dough mixers, running the dough mixers and dumping flour from the silo. Previous studies [10,19,31] also measured high C_m peaks during dough-making. In the traditional bakery, the peak C_m stemmed from the production of breads (night shifts), bread rolls, buns and sourdough. Roberge et al. [31] also measured high C_m peaks at the baking table.

Regarding the duration of the C_m peaks, there were no differences in the industrial bakery pre and post intervention. However, the highest peak concentrations were greater during the post-intervention measurements, which may be explained by the facts discussed earlier. In the traditional bakery, the maximum C_m peak duration was several minutes longer post intervention (18 min) than pre intervention (10 min) during the morning shift, which is also likely attributed to the facts discussed earlier. Considering the maximum C_m peak duration during the night shift, there was a significant difference between the pre-intervention (11 min) and post-intervention (1 h 4 min) measurements. A possible explanation for this might be that the dough divider, which was located beside S4, was taken into use during the night shifts post intervention, causing long C_m peaks. However, the highest C_m peaks were lower during both the night and morning shifts post intervention. This result is possibly related to the intervention measures or random variation in the exposure levels. The average C_m was greater in the night shifts pre and post intervention, which might be attributed to dustier work tasks during bread baking or poorer adherence to the intervention strategies.

In both bakeries, the proportion of the PM1 fraction of the TPM was predominantly greater compared to the other fractions on each measurement day. This finding is related to low concentrations. When the C_m was $< 1\ mg/m^3$, the PM1 fraction generally dominated. During dusty work phases when the C_m was $> 1\ mg/m^3$, the proportion of larger particles (PM10–15) was usually greater.

Karjalainen et al. [19] reported that the PM1 fraction constituted 85–93% of the TPM in a traditional Finnish bakery. Roberge et al. [31] found that particles of 0.23–4.00 µm had an average mass percentage of 12%, whereas the average mass percentage was 61% for particles of 10–20 µm, obtained by the GRIMM PAS 1.108. Stobnicka and Górny [32] suggested that $>$ 50% of the airborne flour dust particle mass has $D_{ae} > 15$ µm. These discrepancies could be attributed to different sampling methods, as the DRX used in the current study is designed to measure particles with $D_{ae} < 15$ µm. Furthermore, smaller and lighter particles (PM1) stay longer in the bakery air compared to the larger particles, which tend to settle on the floor faster by gravity, resulting in a higher average proportion of PM1 particles in the bakeries.

In the industrial bakery, a huge difference between the PM1 fraction and the other fractions existed between 11:00 and 14:00. The increase in PM1 concentration after 11:00 is possibly related to the cleaning of the baking areas. The DRX was located further away from the areas being cleaned, which could explain the high concentration of the small particles. Moreover, after cleaning, small particles stay longer in the bakery air compared to the larger particles. The differences were also seen at the end of the work shifts during measurement days 2 and 4 in the industrial bakery and during each measurement day in the traditional bakery, which is also attributed to cleaning.

5. Conclusions

The results of this study show that reductions in the exposure levels to inhalable flour dust were not observed in the breathing zone post intervention, although the workers employed several intervention strategies frequently. However, the exposure levels reduced in most of the stationary locations post intervention. The peak exposure levels also reduced in the traditional bakery post intervention, but reductions in the peak concentrations were not achieved in the industrial bakery. In both bakeries, the TPM consisted predominantly of PM1 particles and large particles with $D_{ae} > 10$ µm pre and post intervention. More rigorous interventions supplemented by technical control methods are required for both bakeries to reduce flour dust exposure.

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Disclosure statement

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ARTICLE III

Karjalainen A, Väisänen A, Leppänen M, Ruokolainen J, Hyttinen M, Miettinen M, Säämänen A, Pasanen P. Exposure to particulate matter, volatile organic compounds, and carbonyls in an in-store bakery and a bake-off unit in Finland. Submitted manuscript.

ANTTI KARJALAINEN

The bakery sector is highly diverse and the largest food sub-industry in Finland. This thesis examined exposure to particulate matter and organic chemicals and the effectiveness of an intervention to control flour dust exposure in the Finnish bakery industry. The results showed that personal protective equipment, local control measures and more rigorous interventions are required to reduce workers' exposure levels to particulate matter.

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