MARIJA MEIŠUTOVIČ-AKHTARIEVA

IMPACT OF THE USE OF NEW NICOTINE CONTAINING PRODUCTS ON INDOOR AIR QUALITY

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MARIJA MEIŠUTOVIČ-AKHTARIEVA

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Doctoral dissertation
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LIST OF ABBREVIATIONS

3-EP – 3-ethenylpyridine
3R4F – Kentucky Reference Cigarette
ACH – Air Change per Hour
CC – Conventional Cigarette
DEHS – Di-Ethyl-Hexyl-Sebacat
EC – Electronic Cigarette
ETS – Environmental Tobacco Smoke
HnB – Heat-not-Burn
HPHCs – Harmful and Potentially Harmful Constituents
HTP – Heated Tobacco Product
IAQ – Indoor Air Quality
IQOS – "I Quit Original Smoking" (acronym)
NCP – Nicotine Containing Product
NTPs – Novel Tobacco Products
NTV – Novel Tobacco Vapour
PAHs –Polycyclic Aromatic Hydrocarbons
PM – Particulate Matter
PM$_{1.0}$ – Mass concentration of particles with aerodynamic diameter lower than or equal to 1.0 μm
PM$_{2.5}$ – Mass concentration of particles with aerodynamic diameter lower than or equal to 2.5 μm
PM$_{4.0}$ – Mass concentration of particles with aerodynamic diameter lower than or equal to 4 μm
PM$_{10}$ – Mass concentration of particles with aerodynamic diameter lower than or equal to 10 μm
PMC– Particle Mass Concentration
PNC– Particle Number Concentration
PSD – Particle Size Distribution
RD – Ventilation Rate of Decay
SHS – Secondhand Smoke
SJR – Scimago Journal and Country Rank
SMPs – Submicron Particles
SVOCs – Semivolatile Organic Compounds
TSNAs – Tobacco-Specific Nitrosamines
VOCs – Volatile Organic Compounds

INTRODUCTION

Novel nicotine containing products have been intensively developed during the past decade, aiming to replace burning/pyrolysis of tobacco with less harmful delivery of nicotine to the adult smoker. Vaping products, e.g., electronic cigarettes (ECs) and more recently, heated tobacco products (HTPs), have emerged as the most recently introduced options. While both of these technologies aim to reduce the generation of carcinogenic compounds in the mainstream aerosol, the operating principle is quite different.

There is a limited amount of published data available on the impacts of the usage of these devices on the indoor air quality and subsequently, the so-called "second hand" exposure. Since no tobacco burning takes place, the primary source of pollution results from the exhaled breath of a product user. The exhaled breath composition differs largely from the mainstream aerosol: the majority of exhaled particles are water based, while most of the nicotine is being absorbed in the user's body [1, 2]. However, a number of studies have indicated that the impact of EC usage, and to a lesser extent, HTP usage, has a tangible impact on the indoor air quality and potentially, the health of bystanders.

Heating tobacco, instead of burning, can offer a potentially lower risk of delivering nicotine compared to the conventional cigarettes (CCs), because it creates a less complex aerosol than burned tobacco [3]. Moreover, the research on heat-not-burn (HnB) tobacco products (alternatively called "tobacco heating systems" (THSs) or novel tobacco products (NTBs)) has consistently demonstrated that harmful and potentially harmful constituents (HPHCs) are reduced or absent in the aerosol of heated tobacco.

Generally, the usage of HTP has been associated with lower or comparable indoor air pollutant concentrations compared to other conventional indoor sources or environments, in most cases distinguishable above the background, thus potentially being associated with health effects at prolonged exposure, as any other artificial air pollution source [4].

Aim of the Dissertation

The aim of the doctoral dissertation is to investigate the spatial and temporal dynamics of environmental aerosol generated by the novel nicotine containing products, its chemical composition and the impact on indoor air quality.

Objectives of the Dissertation

1. To review available data on the indoor air quality and compare the levels of pollutants in environments where novel tobacco products are used with general indoor environments (including micro-environments, such as transport cabins), paying specific attention to the environments, having combustion-based pollution sources present.
2. To perform physical modelling under controlled conditions for the assessment of temporal and spatial variations of exhaled aerosol from using HTP products, EC and CC in the chamber, based on particle matter (PM)

measurements supplemented by the averaged values of tobacco specific gaseous air pollutants with the aim to establish important factors on how to manage air quality in premises where such devices are used.

3. To simulate the variation of IAQ following the usage of HTP in the non-operating nightclub with varying number of users, and compare it with the typical IAQ in the nightclub under operation.

Scientific Novelty

While there is a rapid market transition to new nicotine containing products, the data on the environmental aerosol generated by these products is very sparse. For the first time, the quantitative indications were provided to compare possible hazards caused by HTP versus majority researched pollution sources (conventional cigarettes, water pipe, incense, mosquito coils, etc.).

No data was available on the time-resolved patterns of dispersion of aerosol in a room under varying environmental conditions. In this study, temporal and spatial variations of exhaled HTP, EC and CC aerosol in a chamber, based on a highly time resolved aerosol concentration and particle size distribution measurements, were compared.

The impact of HTP products on IAQ in real-life settings has not been investigated yet. The experiment in the real nightclub was conducted to supplement the results gained in the chamber.

Structure of the Dissertation

This doctoral thesis consists of the following chapters: acknowledgements, an introduction, literature review, materials and methods, results and discussion, general discussion and recommendations, conclusions, a list of references, a list of publications and appendices. The thesis is comprised of 170 pages, 31 figures, 6 tables and 8 appendices. The dissertation consists of material that has been previously published in articles, reproduced with the publisher's permission.

Publication of the Research Results

Three scientific articles based on the thesis research have been published in international journals registered in the *CA Web of Science* database. One research article has been published with Scimago Journal and Country Rank (SJR) impact indicator (Scopus). In addition, the experimental results were presented at three international conferences (see List of publications).

Practical Value of the Work

The methodology for the assessment of the impact of HTP on the indoor air quality has been developed during the research. The literature review showed that HTP has been associated with significantly lower pollution levels if compared to the majority of the conventional indoor pollution sources. However, the research results revealed that aerosols produced by EC and HTP lead to passive smoking. Several important factors, affecting aerosol particle number concentration as well as the concentration of chemical compounds, were determined in the chamber and real life environments. These factors can be practically used to create a healthier environment for the novel NCP users and bystanders.

It should be acknowledged that the empirical results of this dissertation were included in the WHO's latest air quality recommendations in 2019 [5].

Author's Contribution

The research was carried out during the literature review and in 3 consecutive experiments, resulting in four research articles.

In Article 1 (A Review of the Impacts of Tobacco Heating System on Indoor Air Quality versus Conventional Pollution Sources), the author planned and performed data collection and participated in the preparation of the manuscript for the publication of results.

In Article 2 (Impact of Exhaled Aerosol from the Usage of the Tobacco Heating System to Indoor Air Quality: A Chamber Study), the author helped with planning and conducted the experiments in a chamber, performed the data analysis and participated in preparing the manuscript for the publication of results.

In Article 3 (Impact of Using a Tobacco Heating System (THS) on Indoor Air Quality in a Nightclub), the author planned and conducted the experiments of simulating the variation of IAQ, following the usage of HTP in a non-operating nightclub with varying amount of users, and compared it with the typical IAQ in a nightclub under operation, performed data analysis and participated in the preparation of the manuscript for the publication of results.

In Article 4 (The Dynamics of Exhaled Aerosol Following the Usage of Heated Tobacco Product, Electronic Cigarette, and Conventional Cigarette), the author planned and conducted all the experiments of a new HTP product and compared it with a vaping product (EC) and a conventional cigarette (CC) in term of highly time resolved particle concentrations and PSD, performed data analysis and graphs. Based on the analysis of the experimental data, the author prepared the manuscript for the publication.

The published articles were prepared by the author under professional guidance of a supervisor and co-authors at Kaunas University of Technology.

1 LITERATURE REVIEW

1.1. Impact of Nicotine Containing Products on Indoor Air Quality

It has been revealed in different studies that indoor air can be several times more polluted than the outdoor air [6, 7]. Especially vulnerable groups are children and adults with asthma, who are at the high risk of poor indoor air quality [8, 9]. The daily time spent indoor among child and adult asthmatics is 66.7% and 83.3%, accordingly [10]. Most of the airborne indoor air contaminants, commonly found in indoor air, contain combustion byproducts (CO, PM, ETS), substances of natural origin (Radon (Rn), pet dander, mold), biological agents (mold), pesticides, lead and asbestos, ozone O_3 and various VOCs (from a variety of products and materials) [11]. It means that the identification of sources of airborne air pollutants in the indoor environment is important for the implementation of appropriate air quality control strategies. Due to the differences in functions and structures of buildings, it is useful, but very difficult to have a complete list of sources of air pollutants: gas stoves in kitchens, painting or coatings on walls, building construction materials, ceilings, and personal care products contain contaminants that can be harmful in the indoor environment [12].

Environmental tobacco smoke (ETS) is a mixture of particles that are emitted from the burning end of a cigarette, pipe or cigar and from the smoke exhaled by the smoker. Tobacco smoke can contain any of more than 4000 compounds, including carbon monoxide (CO) and formaldehyde (CH_2O) [13, 14]. Tobacco use is the leading cause of global preventable morbidity and mortality. In spite of much evidence on the adverse health effects of tobacco use, many people continue to smoke [15]. Over the past century, the number of worldwide smokers of tobacco products has increased to more than one and a half billion in the world [16]. A comparative risk assessment of the burden of disease and injury attributable to 67 risk factors in 21 worldwide regions concluded that tobacco smoking (including second-hand) and household air pollution were among the leading risk factors for global disease burden [17]. It is evident that the traditional smoking must be ceased or at least modified to reduce the risk to both smokers and bystanders [4].

It is important to distinct mainstream, secondhand and thirdhand smoke, giving appropriate evaluation of their impact on the indoor air quality (IAQ) and human well-being. Smoking tobacco product makes two kinds of tobacco smoke: mainstream smoke, which is inhaled by the smoker, and sidestream smoke from the smouldering tobacco product. Secondhand smoke is a mixture of sidestream smoke, and the mainstream smoke that is exhaled by the smokers. The mainstream smoke can be defined as a tobacco smoke that is exhaled by the smoking individuals. Among the substances found in such smoke, there is nicotine as well as a number of harmful, potentially carcinogenic chemicals. The inhaling of mainstream smoke increases the risk of lung cancer and can be an indirect cause for a higher probability of other types of cancer. Moreover, it contributes to other health problems, such as heart disease and lung disease [18].

Secondhand smoke (SHS) is the intricacy of smoke from the glowing end of a tobacco product and the smoke exhaled by the smokers. SHS has hundreds of toxic chemicals, and about 70 of them can cause cancer [19–22]. There is no risk-free value of SHS exposure; even short-term exposure has a potential to be detrimental to one's well-being, raising risks of heart disease, lung cancer and heart attacks [19, 20, 23]. The residents have a good chance of being exposed to secondhand smoking from family members or neighbours. SHS travels through buildings: doors, windows, hallways, ventilation system, outlets and gaps around pipes. The smoking cessation policy has been successful in defending those who do not smoke and is considered a unified methodology to defend one's well-being wholeheartedly [19, 20, 24].

Thirdhand smoke is a tobacco contamination that remains in the air and on all sorts of surfaces after stopping smoking. The term refers to the secondhand smoke gases and particles that attach themselves to and become embedded in the materials and objects, such as walls, carpet, household objects, blankets, furniture, various fabrics and toys. Thirdhand smoke is not the smoke itself, but rather the chemicals in the smoke, which adhere to the objects, of which there is a high possibility to be released back into the air or accumulate in the household dust.

Some chemicals in the thirdhand smoke do not occur due to the cigarette smoke: instead, they are a product of chemical transformation of tobacco smoke components that occurs in the environment. Thirdhand smoke can linger in indoor environments for a prolonged period of time, from several months to as long as several years. People may be exposed to thirdhand smoke when in contact with contaminated surfaces (smoke gets absorbed through the skin), by consuming contaminated food or breathing in dust as well as breathing in suspended thirdhand smoke components that remain in the air of the room [25].

In recent years, much attention has been focused on alternative nicotine-containing products to traditional tobacco-based cigarettes for reducing the risk of smoking, known to adversely affect nearly every organ of human body, result in diseases and reduce health status in general [26]. Heated tobacco products (HTPs) and electronic cigarettes (ECs) represent a potentially less harmful alternative to traditional tobacco products, such as conventional cigarettes (CCs), with growing popularity amongst the adult smokers worldwide [27]. The operation mechanism of CC, EC and HTP is different as well as temperature regime (see Fig. 1).

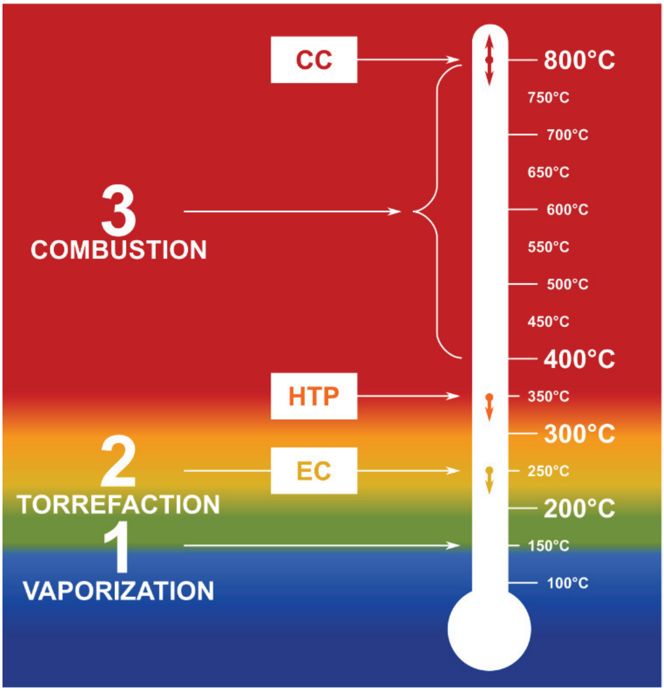


Figure 1. Temperature regimes of nicotine containing products [28]

HTP and EC are not yet thoroughly researched nicotine containing products on the market; however, in the latest years, these products have caught more attention from the scientific society.

1.1.1 Conventional Cigarettes (CC)

Conventional cigarettes (CCs) were the most commonly used tobacco product in most countries up to the recent decade. The current tobacco sector is becoming tremendously complex with the rapid emergence and popularity of alternative products [29]. The search for alternatives is motivated by numerous researches, showing that the usage of conventional cigarettes results in numerous hazardous pollutants that are emitted to the ambient air and cause adverse health effects to both mainstream smokers and bystanders [30]. The other reason for alternatives is that the use of tobacco cigarette is a highly addictive habit, making cessation a difficult and challenging task [31]. However, EC or HTP may act as a substitution for CC, potentially leading to less smoking or quitting at all.

A burning conventional cigarette is an example of a self-sustaining smoldering (flameless) combustion process, during which, the temperatures of tobacco at the tip of the cigarette can go up to 850 °C and higher when the air is drawn through the lit tip. The resulting smoke aerosol is a product of complex combustion, pyrolysis and pyrosynthesis processes that happen simultaneously with lower-temperature distillation and sublimation processes. The smoke aerosol that is produced during the combustion of this type is a complex and dynamic mixture composed of gases, liquid droplets and solid particles suspended in the air [32]. The energy created during the combustion process breaks down the tobacco components. The result of this process is a complex smoke aerosol (made up of various gases, liquid droplets as well as solid particles (soot) suspended in the air); it leaves a residue of ash (composed of inorganic compounds that were present in the tobacco material from the start). Cigarette smoke is a complex mixture, in which more than 8,400 chemicals have been identified [33]. A smaller number of these chemicals are considered by the public health authorities as most probably capable of causing smoking-related diseases, such as lung cancer, heart disease and emphysema. The majority of these HPHCs are generated at high levels during the processes that are taking place at temperatures, associated with tobacco combustion [34, 35].

1.1.2 Electronic Cigarettes (EC) vs. CC and HTP

The first widespread alternative products on the market were electronic cigarettes (ECs): battery-powered devices, electrically heating and vaporizing a liquid solution to produce an inhalable aerosol typically containing nicotine, flavourings and other compounds [36]. ECs are currently the most popular harm-reduction products, but many smokers do not find them sufficiently effective or satisfactory in fully substituting smoking [37]. However, only several researches on the impact of EC to IAQ that were performed using human volunteers in natural settings were reported in a systematic review [38].

EC liquids can contain varying amounts of nicotine, which makes it different from the HTP products, containing naturally occurring nicotine in the tobacco leaf. Liquids additionally contain glycerol and propylene glycol as the primary carriers of vaporized nicotine and may or may not contain flavourings [27]. EC utilizes heating at roughly 100–250 °C within a chamber to generate aerosol from liquid [39]. The vaporization of glycerol and propylene glycol is only possible in comparatively high temperatures; besides, glycerol can only be purified by distillation in a vacuum environment, as it begins to polymerize and decompose at temperatures lower than its boiling point of 290 °C under atmospheric pressure. In comparison, propylene glycol has one fewer hydroxyl group than glycerol (Fig. 2), and this is somewhat less dense and viscous than the triol, boiling at 189 °C under atmospheric pressure. It can be distilled without decomposition [40].

Heated EC vaping liquid containing nicotine (as opposed to nicotine in the tobacco matrix in HTPs) in an aerosol propellant matrix has known chemical composition [41]. Several studies have demonstrated that the ECs' exhaled aerosol is very volatile due to being composed of fast-evaporating liquid particles [42, 43]. The physical process of volatility is described in greater detail in sub-chapter 3.3.3.

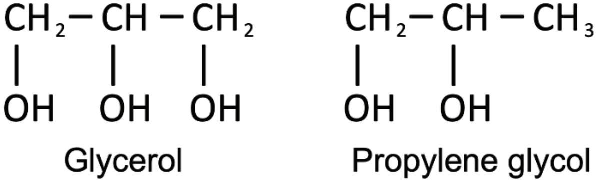


Figure 2. Chemical structure of glycerol and propylene glycol

At higher temperatures together with vaporization, the thermal degradation begin to occur, that is why EC liquids consist of chemicals requiring lower temperatures for vaporization. For example, HTP aerosol droplets are generated by the condensation of vaporized substances at higher temperatures. At the temperature range of 200–300 °C, mild pyrolysis begins to occur. It is as well known as torrefaction [44, 45]. Torrefaction is the production of gaseous substances (volatiles) from solids due to pyrolysis. The vaporization is a physical process when a liquid becomes a gas, while pyrolysis is a chemical process, when the decomposition of molecules at elevated temperatures occur [46]. However the HTP is heated to temperatures below the ignition temperature of the HTP tobacco material, the tobacco material undergoes processes such as drying and vaporization and close to the heater, thermal decomposition (torrefaction and low temperature pyrolysis), but no combustion (neither incomplete, nor complete).

Combustion is a more complex process, since it includes many more chemical reactions undergoing at different temperatures. During combustion, high-temperature pyrolysis forms smoke particles, in addition to substances transferred by vaporization and thermal degradation. The process of tobacco combustion in a cigarette is a fairly good example of a smouldering combustion, when the temperature can go up to 850 °C at the burning end of the cigarette [47, 48]; this happens when air is, for example, inhaled by a smoker through the burning tip. The temperature goes up because more oxygen is available, and the exothermic (heat-generating) characteristics of combustion reactions can be observed. These processes do not occur in EC and HTP.

The evaporation of relatively stable high-molecular weight species in the tobacco occurs mostly below 250 °C [49–51]. For example, the evaporation of glycerol, which is added to the tobacco as humectant and aerosol former with a boiling point of 290 °C occurs at temperatures between 120–250 °C [51], while the nicotine evaporates between 110 °C and 220 °C [52], even though the boiling point of nicotine is 247 °C (see Fig. 1).

Both EC and HTP mainstream aerosol has been well characterized and has been shown to contain a complex mixture of nicotine, propellent, flavourings and other trace compounds [3, 53, 54]. The HTP and EC aerosols as well contain fewer and significantly lower levels of toxicants compared to the CC smoke. The composition of aerosol may as well depend on the type of the device and heating regime, flavour additives, operational voltage and smoker puffing patterns [55–59]. The HTP does not produce any side-stream smoke emissions, thus the potential impacts to indoor air quality from the usage of HTP may come only from the exhaled aerosol [42, 43].

Despite positive reviews of ECs, there are studies demonstrating that the levels of airborne nicotine concentrations in the EC user's home are much higher than the in the home of people who smoke nicotine cigarettes. Ballbe et al. performed a study to single out the most prominent characteristics of passive exposure to nicotine from ECs' vapour and CCs' smoke at home among non-smokers under real-use conditions. The airborne markers in homes where CCs were used were statistically higher compared to other homes, where ECs were used (to be precise, 5.7 times higher). However, the concentrations of both biomarkers among non-smokers exposed to CCs' and ECs' vapour were not very different statistically (only 2 and 1.4 times higher, respectively) [60]. These findings serve to show that non-smokers that are passively exposed to ECs do absorb certain amounts of nicotine.

ECs are positioned on the market as a safer option for smokers; however, there are evidences showing that ECs' vapour can be harmful and lead to passive smoking.

1.1.3 Heated Tobacco Products (HTP) vs. CC and EC

Another viable approach to reduce the levels of hazardous substances associated with cigarette burning smoke is the generation nicotine-containing aerosol by heating tobacco in reduced temperatures as opposed to burning, thus entitling the process as "heat-not-burn" (HnB). The products are marked as Tobacco Heating System (THS) or Heated Tobacco Product (HTP). In this work, all products involving "heat-not-burn" process will be abbreviated as HTP.

The first commercial HnB, PremierTM (R. J. Reynolds, Winston-Salem, NC, USA), was introduced in 1988, followed by EclipseTM (R. J. Reynolds) and AccordTM (Philip Morris) in the 1990s, Heat BarTM (Philip Morris) in 2007 and HTPTM (Philip Morris International, Lausanne, Switzerland) in 2016. British American Tobacco has recently developed a THP comprising an electrical heating device, commercially known as GloTM, and consumable tobacco rods, commercially known as KentNeostiksTM, referred as THP1.0 [61]. Initially, such devices were referred to as Electrically Heated Cigarette Smoking System (EHCSS) [62]. In later developments, the term Tobacco Heating System (THS) has been established.

There is a big selection of HTP or other heat-not-burn products on the market already, though slightly differing in their working principle, e.g., IQOS from Philip Morris International, 3T from Vapour Tobacco Manufacturing, Glo from Britsh American Tobacco, Ploom Tech from Japan Tobacco, and others. Both the e-cigarettes and HTPs are gaining increasing popularity among the users [63].

The mainstream aerosol is produced by "distilling" nicotine and flavours at temperatures up to 350 °C via a heating blade inserted into a uniquely processed tobacco plug. Such electronically controlled heating prevents high temperature pyrolysis/combustion from occurring [64]. A schematic view of the HTP with components inserted in the Holder are provided in Figure 3.

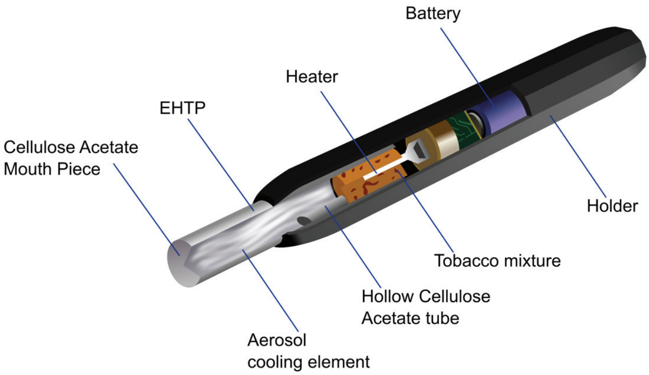


Figure 3. Schematic view of the HTP [65]

The investigations performed by governmental institutions confirmed the findings of tobacco companies and independent researchers. Mallock et al. [66] reported that the yields of the carbonyl compounds in the mainstream HTP (IQOS 2.2) aerosol were reduced by 80–96%, VOCs and SVOCs reduced by 97–99% compared to the reference cigarette 3R4F [53]. Having performed a systematic review of 31 publication on HnB, Simonavicius et al. [67] concluded that although limited by methodological heterogeneity, the findings were largely similar to the independent and industry-funded studies. Simonavicius et al. [67] reviewed thirty-one publications on HnB products and concluded that compared with CCs, HnB products delivered up to 83% of nicotine and reduced the levels of harmful and potentially harmful toxicants by at least 62% and particulate matter (PM) by at least 75%. However, most of studies were related to the mainstream aerosol. The data on the second-hand HnB aerosol is limited to several studies [68–73].

Although previous studies have reported many data on the mainstream aerosol, the data on the second hand HnB aerosol is still very limited.

Several authors have conducted studies in a controlled environment in order to distinguish HTP generated pollution and compare it to the background level [68, 69, 74]. The comparison between the background and HTP environmental aerosol samples generated by smoking machines in an environmentally controlled room showed a statistically significant increase in concentration of only five compounds (nicotine, acetaldehyde, toluene, benzene and solanesol) [69]. Such testing, however, did not take into account the transformation of mainstream aerosol in the human body. The comparisons were extended in other researches with smokers in an environmentally controlled room, simulating “Office”, “Residential” and “Hospitality” environments, and was compared with smoking a lit-end conventional cigarette (CC) [68]. The concentrations of most measured indoor air constituents during the use of HTP were similar to the background levels (with the exception of acetaldehyde and nicotine), and for the most analytes, were an order of magnitude lower than found in the assessments with the traditional cigarette. The data resulting from “residential” setting was compared with the other chamber-based studies, since the conditions were the most comparable to the reported elsewhere. Malysheva et al. [74] reported a 10% increase in carbon monoxide and formaldehyde air concentration above the background during the usage of HTP.

The HTP has shown to deliver nicotine to the aerosol at levels higher than ECs but lower than CCs, when tested using Health Canada Intense puffing regime [31]. At the same time, significantly lower concentrations of harmful and potentially harmful constituents (HPHCs) [3, 53] and tobacco-specific nitrosamines (TSNAs, [54]) were found in the aerosol of HTP (as compared to the mainstream smoke of reference cigarette 3R4F). The reduction of constituents in the mainstream aerosol of HTS (IQOS) in comparison to a reference cigarette 3R4F was more than 90%: carbon monoxide (> 97.00%), volatile organic compounds (VOCs), aromatic amines, hydrogen cyanide, phenol and polycyclic aromatic hydrocarbons (PAHs) (> 98%) [15]. The mainstream aerosol of another type of HnB - THP1.0 has as well shown to have an overall average reduction of 97.5% for the abbreviated list of HPHCs of smoke, specified by the US Food and Drug Administration Tobacco Products Scientific Advisory Committee for reporting in cigarette smoke (excluding nicotine) in comparison to the CC [75].

Ruprecht et al. [71] investigated the environmental pollution from HTP, EC and conventional cigarettes in a sitting room in a flat owned by habitual smokers, furnished with typical home appliances, where the use of nicotine containing products was performed in a controlled manner. Only background subtracted data was reported, thus allowing the data to be used only in a comparative analysis of controlled environments. Polycyclic aromatic hydrocarbons (PAHs) were mostly non-detectable in the particulate emission of organic matter from these devices. Metal concentrations were similar to the background levels and lower during the HTP usage, compared to both ECs and CCs. The concentrations of aldehydes in the environmental tobacco aerosol of HTP were higher in comparison to the EC aerosol; however, the levels were substantially lower compared to CCs. Polycyclic aromatic hydrocarbons (PAHs) were reported to be mostly non-detectable in the particulate emission of organic matter from HTP. Metal concentrations were similar to the background levels and lower during the HTP usage, compared to both ECs and CCs [71].

Comparing all studies together, it can be concluded that the concentrations of the most investigated indoor air constituents during the use of HTP products in a controlled environment are similar to the background levels (with the exception of aldehydes) and the order of magnitude lower than in the CC environmental aerosol [68, 71–73].

1.2 Impact of Heated Tobacco Products on Indoor Air Quality vs. Conventional Pollution Sources

The data in sub-chapter 1.1. shows that the use of new nicotine containing products leads to significantly lower levels of ambient air pollutants in various indoor conditions in comparison to a traditional cigarette. This leads to a question whether it is possible to distinguish such air pollution from the process of smoking as well as environments that are containing other pollution sources, commonly encountered in households or other micro-environments worldwide. Therefore, the sub-chapter 1.2. aims to review the available data on the indoor air quality and analyse the levels of pollutants in such environments, where HTP is used in comparison to the general indoor environments. In this sub-chapter, a specific attention was paid to the environments having combustion based pollution sources present, such as candles, incense or mosquito coils.

The parameters of simulated indoor environments (chambers) [68, 71, 76–83] as well as "real-life" indoor environments [84–107] were analysed in this sub-chapter. The "real life" included studies reporting on the indoor concentration of pollutants in two major categories, i.e., residential and public. The latter one was comprised of a wide span of environments, including schools, kinder-gardens, offices, coffee shops, libraries, copy centres, pharmacies, newspaper stands, gymnasiums, hairdressers, restaurants, supermarkets as well as various modes of motorized transport. The "controlled" included studies performed in environmentally controlled chambers as well as chamber-type environments, conducted not exactly in a completely controlled manner, but having the aim of investigating environmental pollution generated by a particular source as well as studies that are representing pollution levels in various non-smoking indoor environments. Detailed information on sampling conditions and analytical methods that are employed in the research studies is summarized in tables provided in Appendix 1 and Appendix 2.

The potential health effects of the previously reported concentration levels may be discussed through the proximity of the values to the guideline threshold values. Interestingly, the guideline values vary broadly among different regions and organisations as presented in Table 1, making such comparison rather complicated. Guideline values for some pollutants (acetaldehyde, toluene) differ by an order of magnitude. Only occupational exposure limit values for nicotine are defined in Europe (EU, [108]) and the USA (OSHA, [109]). No guidelines for indoor particulate matter are available; thus, ambient air annual limit values are presented instead. The median concentration of formaldehyde in chambers exceeded WHO guideline (100 µg/m^3) value in case of CC smoking [68, 71] as well as burning incense [76]. Smoking of CC as well resulted in exceeding the EU exposure limit for acetaldehyde (200 µg/m^3) [68, 71] and the US inhalation RfC for benzene (30 µg/m^3) [68]. The concentrations of toluene did not exceed any guideline value.

Table 1. Indoor air quality guideline values [4]

Pollutant	**WHO guideline value, µg/m^3** [110]	**Proposed EU exposure limits, µg/m^3**[111]	**The US EPA inhalation RfC^a, µg/m^3**	**Lithuanian hygiene standards, µg/m^3**
Acetaldehyde	NA	200	9 [112]	10 [113]
Formaldehyde	100 short-term (30-minute) guideline value	30 (NOAEL)^b	NA	100 short-term 10 daily rate [113]

Benzene	NA[c]	NA[c]	3 [114]	1500 short-term100 daily rate [113]
Toluene	NA	300	5000 [115]	600 [113]
Nicotine	NA	500[d] [108]	500[d] [109]	500 [116]
No guidelines for indoor particulate matter are available. Ambient air annual limit values are presented.				
$PM_{2.5}$	10 [110]	25 [117]	15 [118]	40 [113]

[a] **Reference Concentration (RfC)** is an estimate (with uncertainty spanning, perhaps, the order of magnitude) of a continuous inhalation exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime (US EPA, IRIS).

[b] NOAEL is no observed adverse effect level that is not equivalent to the exposure limit but used in deriving it.

[c] Benzene is a genotoxic carcinogen in humans, and no safe level of exposure can be recommended.

[d] occupational exposure limit value, 8 hour average.

The selection of analytes for the previously presented literature review included the most commonly investigated indoor air pollutants, having sufficient number of references reporting their levels. Among gaseous pollutants, nicotine and acetaldehyde are among the most commonly associated with the HTP usage [68], while combustion byproducts CO, NO and NO_X, are not detected [65]. In fact, 62 trace compounds were qualitatively identified in the exhaled HTP aerosol [120]. Aldehydes, nitrogenated species and aromatic species are listed among these trace compounds [121]. The concentrations of aldehydes (formaldehyde, acetaldehyde), monoaromatic hydrocarbons (benzene, toluene), nicotine and particulate matter ($PM_{2.5}$ or respirable fraction) were compared in case of chamber studies, while in the real-life studies, only the concentrations of formaldehyde, acetaldehyde, benzene and toluene were considered. Formaldehyde and benzene are among the most researched pollutants known for their hazardousness to health and often found indoors in the concentrations of health concern [122]. Formaldehyde, acetaldehyde and nicotine are common indicators of nicotine containing product aerosol, as is toluene [20]. The critical appraisal of the setting and implementation of indoor exposure limits in the EU has identified formaldehyde and benzene as high priority chemicals, while acetaldehyde and toluene were identified as the chemicals of second priority based on the risk assessment [111]. The inhalable PM is a well-documented pollutant that is associated with smoking and having health effects, including respiratory and cardiovascular morbidity, such as aggravation of asthma, respiratory symptoms and an increase in hospital admissions, mortality from cardiovascular and respiratory diseases and lung cancer [123].

The concentration data of combustion based pollution sources from chamber studies have been obtained in a variety of chambers, having various volume and air change rate values. Such data was normalised for the chamber volume and air change rate in order to match conditions suggested by the CEN/TS 16516 standard (CEN standards), i.e., chamber volume of 30 m^3 and air change rate of 0.5 ach. At the same time, the data has not been normalised for a number of test products that were used, assuming that the realistic scenarios were chosen by the investigators.

The statistical significance of differences in HTP vs. other groups of pollution sources was tested by the independent-samples t test, setting confidence interval percentage to 95%. Such test was conducted only for cases where 3 and more data points were available and indicated by the reported p-value.

In data processing for literature review, a variety of descriptive statistics estimates have been reported, including parametric ones (mean, standard deviation) and non-parametric ones (median, interquartile range) as well as min-max range. Such various estimates pose a challenge for intercomparing them due to a different nature of the statistical estimate. All these estimates were plotted on the same graph but visually denoting the used statistics (Fig. 5, 6, 7, 8, 9, 11). A logarithmic concentration scale was applied as the data spans over several orders of magnitude. The scientific articles served as a primary source of references, reporting indoor air quality in settings that are described below. Web of Science search engine (Clarivate Analytics, Philadelphia, USA) was employed for searching specific keywords. Afterwards, a cross-reference check was performed to search for additional studies.

1.2.1 Carbonyl Compounds

Volatile organic compounds with carbonyl group, aldehydes and ketones, are found in tobacco product smoke. Formaldehyde, acetaldehyde, acrolein, crotonaldehyde and acetone are usually found in higher concentrations than other carbonyl compounds. Moreover, some of these compounds, e.g., formaldehyde, acrolein and crotonaldehyde, are toxic and carcinogenic compounds. Acetaldehyde and acetone are respiratory irritants. The design features of cigarettes, including ventilation properties, filters and paper porosity, have an impact on the levels of toxic and carcinogenic chemicals in the inhaled smoke [20].

It is important to notice that carbonyl compounds undergo oxidation processes (Fig. 4) in the human body and become carboxylic compounds with their own spectrum of harmful and toxic properties.

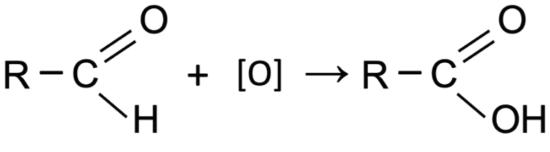


Figure 4. Scheme of oxidation of carbonylic compounds

Formaldehyde is among the most abundant pollutants, having reported occurrences in a broad range of indoor environments. There are plenty of various ways in which humans can be exposed to formaldehyde, and smoking is not the only one. Other ways include contact with insulating materials, particle board or plywood furniture, water based paints, fabrics, household cleaning agents, disinfectants, pesticide formulations, paper products and adhesives, containing formaldehyde used for plastic surfaces, parquet, carpets, and other building materials, containing urea-formaldehyde resins [111].

Other carbonyl compound that is reviewed in real life indoor non-smoking environment and chamber studies is acetaldehyde. Acetaldehyde is a frequent ingredient in manufacturing perfume, polyester resins and basic dyes; besides, it may be used as a fruit and fish preservative as well as a flavouring agent and a denaturant for alcohol. Moreover, its use extends to fuel, gelatine hardening of gelatine and solvent in the rubber, tanning and paper industries. Acetaldehyde is an intermediate product of higher plant respiration and is a result of incomplete wood combustion in fireplaces and woodstoves, coffee roasting, burning of tobacco and vehicle exhaust fumes [4].

Residential fireplaces and wood stoves are the two most releasing sources indoors [124]. The presence of humans and their habits might appear as another important factor, influencing acetaldehyde levels, as it is an abundant component of human exhaled breath, which is a metabolic product of sugars and ethanol [125].

Chamber Studies

Generally, the most recent reported levels of carbonyls for HTP were lower than most of other sources, except for several cases of EC. The usage of HTP in chambers resulted in the average levels of formaldehyde at 15.3 μg/m^3 [68] and 13.3 μg/m^3 [71] as well as 29.3 μg/m^3 and 17.2 μg/m^3 for acetaldehyde, respectively (Fig. 5).

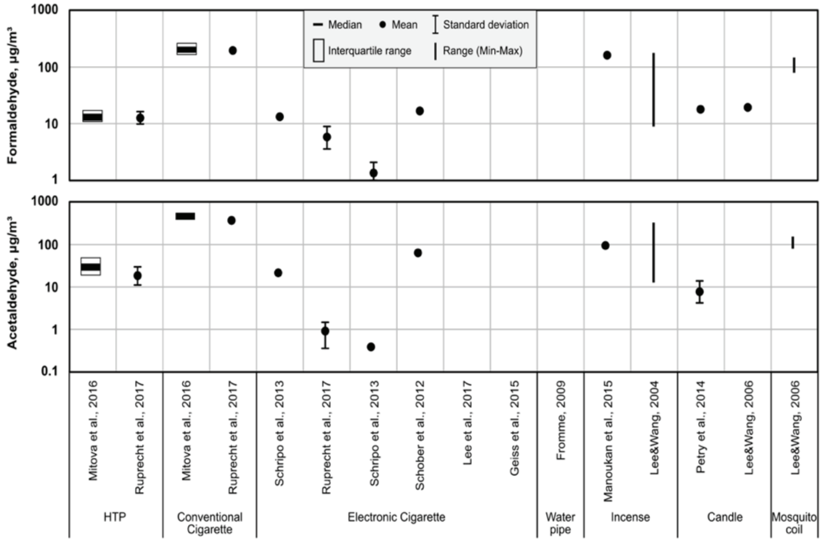


Figure 5. Concentration of airborne formaldehyde and acetaldehyde as reported in chambers with active pollution sources [4]

Such levels are an order of magnitude lower ($p < 0.05$) compared to the smoking of CC in chambers, e.g., 205.9 μg/m^3 and 197.1 μg/m^3 for formaldehyde as well as 528.1 μg/m^3 and 349.9 μg/m^3 for acetaldehyde. In other research, aldehydes were higher in comparison with the EC environmental aerosol; however, the levels were substantially lower compared to CCs [71]. Interestingly, Schripp et al. [79] reported CC generated concentrations of aldehydes by an order of magnitude lower than Mitova et al. [68] and Ruprecht et al. [71], but the amount of smoked CC is not clearly presented.

The reported data for EC varies broadly and was found to be not statistically different from the HTP ($p > 0.05$). Schober et al. [79] determined similar levels of formaldehyde to HTP (16.0 μg/m^3), while acetaldehyde levels were 2–3 times higher. Schripp et al. [79] and Ruprecht et al. [71] reported substantially lower EC generated concentrations of aldehydes. Such variation of the airborne levels of carbonyls is predetermined by the large variation in carbonyl concentrations generated by the ECs, as determined by multiple studies. The emission of formaldehyde (μg/10 puffs) from 13 brands of Japanese e-cigarettes varied from 0.7 ± 0.8 to 34 ± 35 and from 0.2 ± 0.1 to 26 ± 28 for acetaldehyde [126]. The content of formaldehyde ranged from 2.0 to 56.1 mg and acetaldehyde from 1.1 to 13.6 mg per one EC (150 puffs), as determined in 12 brands of ECs distributed within Poland [127]. Similarly, high variation of aldehyde emission from 10 various ECs of the same brand was reported by Uchiyama et al. [128]. Formaldehyde, acetaldehyde, acrolein and other aldehydes are known to form during the heating of mixtures of glycerin and propylene glycol, the most common solvent formulation for EC liquids [129]. The emission of aldehydes from ECs are very much dependant on the temperature of heating coil [31, 41, 81, 130] as well as the composition of the EC liquids [131]. EC solutions may contain numerous compounds in addition to the vendor listed propylene glycol, glycerol and nicotine, resulting in a very different aerosol composition [132].

To summarize, the burning of incense resulted in a high mean concentration of formaldehyde (158 μg/m^3) and acetaldehyde (94.8 μg/m^3) [76], almost reaching the CC generated levels 205.9 μg/m^3 [68] and 197.1 μg/m^3 [71]. Candle burning produced formaldehyde at the same level ($p > 0.05$), while the concentration of acetaldehyde was lower than in case of HTP.

Real Life Indoor Non-smoking Environment

In residential settings, a broad range of concentrations of formaldehyde was reported, ranging from 5.3 μg/m^3 [84] to 33 μg/m^3 [86]. In the majority of residential environments, the concentration of formaldehyde was higher compared to the levels of formaldehyde in simulated indoor environments where HTP was used (22.4 μg/m^3 [68] and 13.3 μg/m^3 [71]); however, the overall difference among the mean of reviewed concentration did not appear to be statistically significant ($p > 0.05$).

In schools, the concentrations of formaldehyde varied from 14.1 μg/ m^3 [96] to 17.8 μg/m^3 [98] and were at the same level as the HTP generated in the "office" conditions (14.0 μg/m^3 [68]). The office environments have been reported to contain a relatively high concentration of formaldehyde (52.3 µg/m^3 [101]); the same has been noticed in shopping malls 30–120 µg/m^3 [102] or transportation micro-environments, such as new cars 80.0 µg/m^3 [99]. This may be attributed to the release of formaldehyde from plastic materials that are abundant in these environments. Generally, the levels of formaldehyde in public environments were statistically significantly ($p < 0.05$) higher than HTP generated in the "office" conditions (14.0 µg/m^3 [68]) (Fig. 6).

The highest median concentration of acetaldehyde 23.5 µg/m^3 among all environments was reported by Bari et al. [85]; it was measured in winter and summer time in residencies in Canada. Hult et al. [86] found the mean concentration of 18.6 µg/m^3 at the air exchange rate of 0.5 h^{-1} at the US residences; Derbez et al. [133] reported the concentration of 16.5 µg/m^3 in 6 newly build energy efficient homes in France. Such concentrations were statistically significantly higher ($p < 0.05$) compared to those obtained during the use of HTP in simulated residential setting (7.4 µg/m^3 [68]) (Fig. 6).

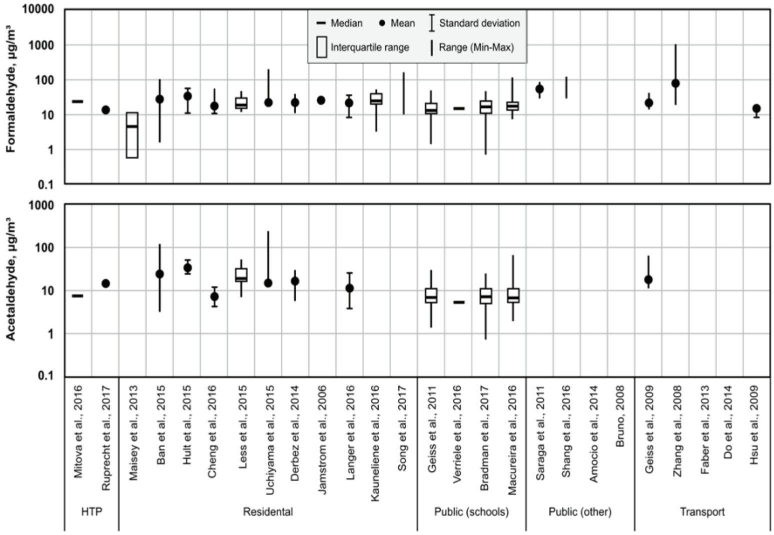


Figure 6. Concentration of airborne formaldehyde and acetaldehyde as reported in various indoor environments [4]

In schools, the concentrations of acetaldehyde were at the same order of magnitude (5.2 µg/m^3 [97], 7.7 µg/m^3 [100]) as HTP generated in a simulated public environment (9.4 µg/m^3 [68]). The transportation micro-environments (cabin air of used private cars) were found to contain 15.6 µg/m^3 of acetaldehyde [104] (Fig. 6).

1.2.2 Monoaromatic Hydrocarbons

Aromatic compounds have their unique cyclic structure and form separate class among other organic compounds. The first member of homologous series of aromatic compounds is benzene (C_6H_6); the second is toluene ($C_6H_5–CH_3$). These two monoaromatic compounds are usually found in tobacco product smoke.

Benzene is found in the air from coal and oil burning, gasoline service stations and motor vehicle exhaust. Benzene is as well used in the manufacture of detergents, explosives, pharmaceuticals and dyestuffs. Tobacco smoke contains benzene and accounts for nearly half of the national exposure [114].

The major use of toluene is as an additive to gasoline to improve octane ratings. Toluene is used to produce benzene and as a solvent in paints, coatings, synthetic fragrances, adhesives, inks, cleaning agents, in the production of polymers used to make polyamide, plastic bottles, polyurethanes, for pharmaceuticals, dyes, cosmetic nail products and the synthesis of organic chemicals. The highest concentrations of toluene usually occur in indoor air from the use of common household products (paints, paint thinners, adhesives, synthetic fragrances and nail polish) and a cigarette smoke [134].

Chamber Studies

While monoaromatic hydrocarbons are some of the most researched indoor pollutants that are related to the combustion processes, very few studies on nicotine containing products reported benzene concentrations (Fig. 7). The usage of HTP has resulted in 0.93 μg/m^3 [68], while the use of EC resulted in lower levels of benzene (0.17 μg/m^3 [80]). The water pipe smoking appeared to be a strong emitter of benzene, producing levels at 17.0 μg/m^3 [83].

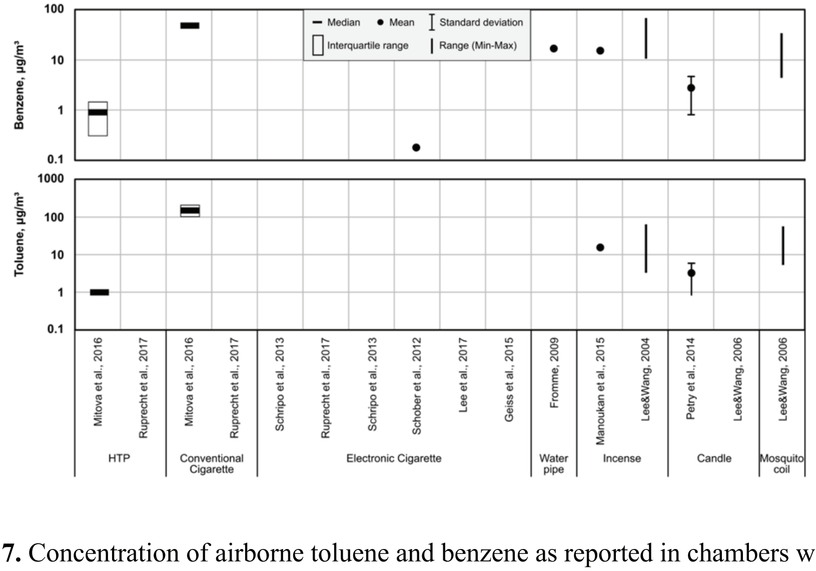


Figure 7. Concentration of airborne toluene and benzene as reported in chambers with active pollution sources [4]

As a non-nicotine containing product, the candle burning as well resulted in higher concentrations of benzene and toluene (2.7 μg/m^3 and 3.1 μg/m^3, respectively

[78]). Toluene levels were below the background for HTP, while CC smoking has resulted in 151.1 µg/m^3 [68]. In case of incense and mosquito coil burning, toluene concentrations were at a similar level, ranging from 4.6 to 53.6 µg/m^3 [77] and from 2.9 µg/m^3 to 60.0 µg/m^3 [135], respectively.

Real Life Indoor Non-smoking Environment

The concentrations of benzene in the residential environment ranged from 0.8 µg/m^3 [94] to 3 µg/m^3 [91], in schools from 1.1 µg/m^3 [97] to 2.6 µg/m^3 [96] and were statistically insignificantly ($p > 0.05$) higher than the HTP generated (0.57 µg/m^3 [68], 2.7 µg/m^3 [71]). In other public environments, the concentrations were higher than the HTP generated by an order of magnitude (15.3 µg/m^3 [101]) in the office, 14.8 µg/m^3 in a drugstore [103]. In new cars, the average benzene concentration was 11.8 µg/m^3, but it increased after 20k km to 38.5 µg/m^3, as attributed to the fuel leakage [105]. The highest benzene levels were observed in the parked new cars [99], reaching a concentration level of 270.0 µg/m^3 (Fig. 8).

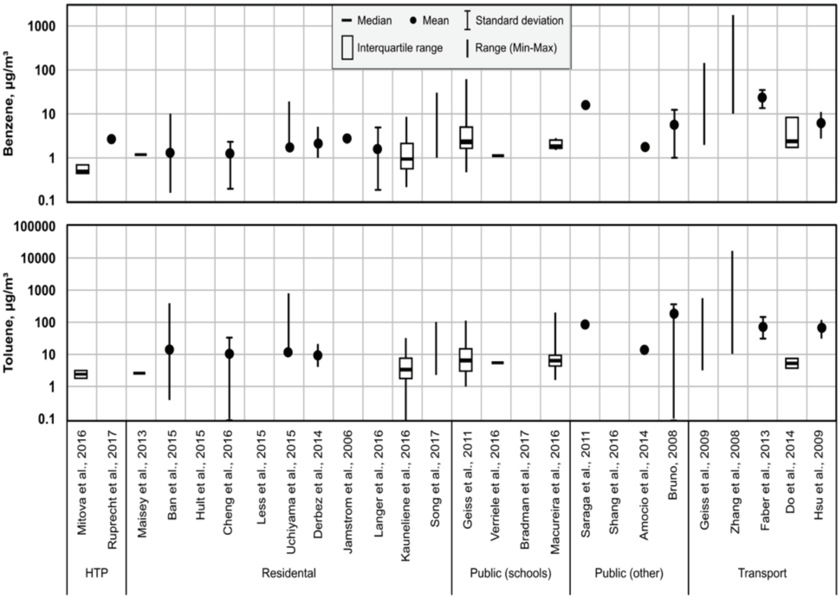


Figure 8. Concentration of airborne benzene and toluene as reported in various indoor environments [4]

HTP generated median toluene concentration in simulated “residential” conditions was 0.57 µg/m^3 in the “office” and in the “hospitality” conditions, 0.25 µg/m^3 [68], while in the residences, the concentrations varied in the range from 2.62 µg/m^3 [84] to 14.1 µg/m^3 [85] and was statistically significantly higher ($p < 0.05$). In schools, it varied from 5.5 µg/m^3 [97] to 7.1 µg/m^3 [96]. Bruno et al. [103] reported toluene concentration as high as 589.0 µg/m^3 in a newspaper kiosk, 303.4 µg/m^3 in a drugstore, 243.0 μg/m^3 in a photocopy shop. High mean toluene concentration (90.2 μg/m^3) was as well measured in the office [101]. Transportation micro-environments as well stand out as a site for high exposure of toluene. In used cars, a mean concentration of 98.8 μg/m^3 was reported [104], while in the new vehicles, it was as high as in Zhang et al. [99].

1.2.3 Nicotine and Other Markers of Environmental Tobacco Smoke

Nicotine is naturally present in the leaves of tobacco plants and can evaporate from tobacco at temperatures below 300 °C [34, 50].

Nicotine is used in all new tobacco products, since it is a central nervous system stimulant that acts as a mediator for the release of several neurotransmitters, including acetylcholine, beta-endorphin, dopamine, norepinephrine, serotonin and adrenocorticotropic hormone (ACTH) [136]. When a body is exposed to nicotine, the individual experiences the release of adrenaline. This surge of adrenaline stimulates the body. When humans, mammals and most of other types of animals are exposed to nicotine, it increases their heart rate, heart muscle oxygen consumption rate and heart stroke volume. Consuming nicotine is as well linked to raised alertness, euphoria and a sensation of being relaxed. People who regularly consume nicotine and then suddenly stop experience withdrawal symptoms, which may include: anxiety, cravings, irritability, etc. The American Heart Association says that nicotine that is consumed from smoking tobacco is one of the hardest substances to quit [137]. Nicotine as well acts as one of the main markers of the environmental tobacco smoke.

The markers of environmental tobacco smoke: (RSP, UVPM, FPM, solanesol, nicotine, 3-ethenylpyridine), toluene, carbon monoxide, propylene glycol, glycerol, and triacetin, were below the limit of detection or the limit of quantification in simulated non-smoking and smoking environments after using of electrically-heated tobacco product referred to as the Novel Tobacco Vapour (NTV) product, while the concentrations of ammonia, carbonyls and total volatile organic compounds were found at the same levels in the chamber without NTV use [72].

Chamber Studies

HTP generated a concentration of 10.4 μg/m^3, which is again an order of magnitude lower compared to CC (168.0 μg/m^3[68]). EC generated lower nicotine levels, although similarly to aldehydes, it demonstrated a high variation from 0.08 μg/m^3 [82], 0.2–0.6 μg/m^3 [138] to 3.1 μg/m^3 [80]. Smoking of water pipe resulted in a twice higher concentration of nicotine (20.5 μg/m^3[83]) (Fig. 9).

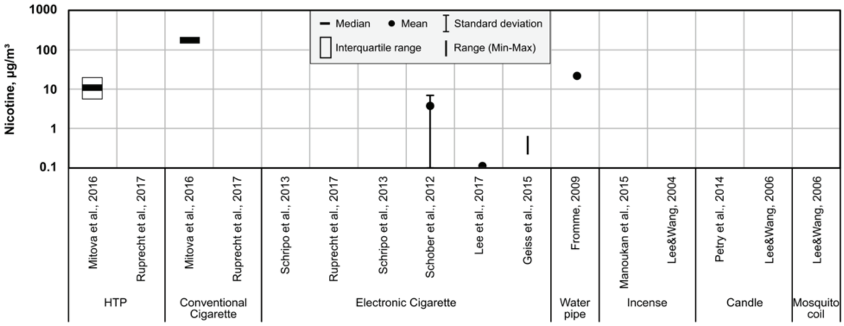


Figure 9. Concentration of airborne nicotine as reported in chambers with active pollution sources [4]

1.2.4 Particulate Matter

Among pollutants, the particulate matter is frequently associated with the usage of thermal nicotine containing devices. The exhaled aerosol usually contains sub micrometer/ultrafine (mostly liquid) particles, having mode at around 100 nm. Due to a highly volatile nature, these particles are further subjected to transformations, such as nucleation/condensation/coagulation and evaporation. The prevailing mechanism depends on the type of device (EC vs. HTP) and environmental conditions, such as operating air conditioner [139] and ventilation intensity [73]. Meanwhile, the particle size is the most important property of particles, because it affects the behaviour, transport and control technologies of the particles [12].

In various research projects concerning aerosol inhalation into the human respiratory tract, the analysis of aerosol particle/droplet size distribution is a useful way of estimating the doses of exposure: the method is indirect but fairly accurate. When solid water–insoluble particles settle on the lung surface, the dose exposure is calculated by checking the particulate matter shape and their size-determined chemical characteristics. When it comes to the aerosols containing liquid droplets, the surrounding gas phase usually results in a higher chemical deposition in contrast to the suspended particulate matter alone.

This can be explained by the fact that the diffusion of gases happens in a shorter time period than droplets; thus, the active compounds may penetrate into the alveolar region more efficiently if they are sufficiently volatile. This is why the aerosol size distribution, as the only factor, is not sufficient for evaluating the dose exposure in human respiratory system in cases of liquid-/droplet-based aerosols. Another method to assess how much aerosol is delivered to the lungs is to classify the droplets into the classes of larger sizes. For this purpose, coarse particulate matter was defined as PM_{10}: this refers to the mass concentration of suspended particulate matter that is lower than or equal to 10 µm. The subcategories, for example, $PM_{4.0}$, which stands for a respirable aerosol, are covered as well. $PM_{4.0}$ denotes a size below, which the minimum of 50% of the mass concentration is able to reach the alveolar region. In other words, from the

50% of particulate matter that goes through the trachea, it is just a small fraction settles in the alveolar area. Both PM_{10} and $PM_{4.0}$ are frequently used to estimate the doses that penetrates to the upper part of the human respiratory tract and stay there when analysing aerosols, in which the particles of large diameter are found, for example, some of those are used for the therapeutic drug deliver [140].

There are other categories that are used to assess the mass concentrations of aerosols that get into the alveolar region even deeper. For instance, $PM_{2.5}$ is used in a case of a fine particulate mass concentration fraction, where 85% and, in many cases, a higher number of suspended objects are able to penetrate into the alveolar area. $PM_{1.0}$ corresponds to the fine particulate matter that is equal to or smaller than 1 μm; in this case, at least 95% of the suspended objects are able to penetrate into the alveolar area.

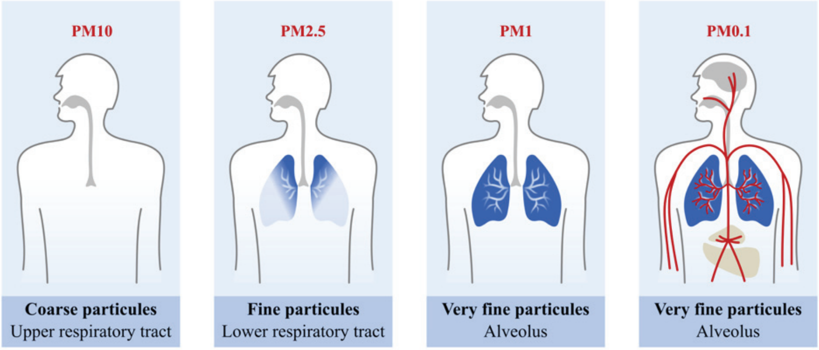


Figure 10. Particulate matter penetration in the human body [141]

Lastly, PM0.1 represents the ultrafine particles, as well called nanoparticles/nano-droplets; 100% of them are able to penetrate into the alveolar region. Because of the simple definition of particulate matter as well as to the fact that it is possible to achieve gravimetric measurements directly, the state regulatory institutions generally recommend calculating PMx gravimetrically in order to have a clue about the dose exposure in the case of pollutants or pharmaceutical aerosols [110].

In order to characterize the particle deposition in human airways, it is essential to evaluate the impact that the particulate air pollution has on the health. This task is rather complex due to the fact that the deposition on lungs is complicated, and its main influencing factors remain unclear to date. The particles that were less than 2 μm in diameter only entered the lungs deeply in the areas where the airflow rate was low, but larger particles, those of 3 μm, were more likely to settle in this region, because of the combination of gravitational sedimentation and inertia. Meanwhile, the larger particles are mainly deposited in the proximal bifurcations as a result of inertia. It has been discovered that the particle size and inhalation rate were among the determining factors of the site, amount and mechanism (inertial impaction or gravitational sedimentation) of deposition, and the accumulation mostly occurred on the inner walls of the tubes that were subjected to a higher axial flow rate and stronger inward secondary velocity [142].

Submicronic particles (SMPs) were four-times higher during smoking CCs than those released by ECs and HTP and remained high for at least 1 h, while SMPs values returned immediately similar to the background for ECs and HTP [70]. Sometimes, the submicron particles form on the nanoparticles, in this particular case, the latter act as the hubs of vapour condensation when present in the aerosol-formation (nucleation) zone. The nucleation zone is the area, in which the threshold vapour supersaturation is achieved; thus, the phase transition (from vapour to aerosol) may occur [143].

The concentration levels of particle number concentrations following the usage of HTP vary broadly in the range from 1E+4 to 1E+7 #/cm^3, while they directly depend on the proximity of the user to a bystander as well as the number of users, such as 10 to 30 [71, 144]. Particulate matter mass concentrations were reported to vary between 14 to 21 µg/m^3 during the exhalation of the HTP smoke [145].

Chamber Studies

The use of HTP resulted in the lowest levels of fine particulate matter among the sources investigated with the reported concentration of $PM_{2.5}$ of 32.0 µg/m^3 [71]. Mitova et al.[68] found even lower levels of fine PM as represented by the respirable suspended particle concentration, which fell below the limit of quantification (< 14.7 µg/m^3) (Fig. 11).

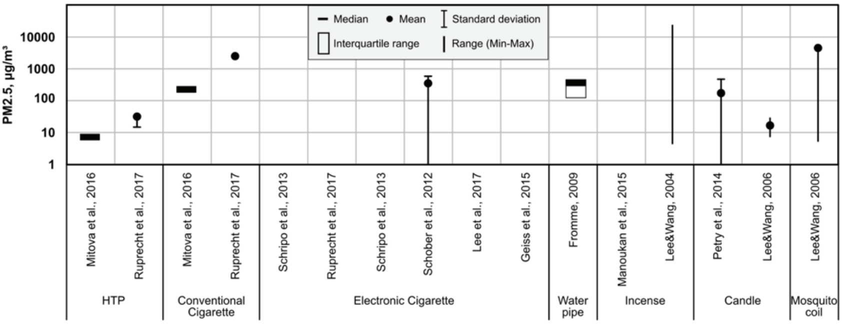


Figure 11. Concentration of airborne $PM_{2.5}$ as reported in chambers with active pollution sources [4]

At the same time, the smoking of CC resulted in 268 µg/m^3 RSP median concentration, thus again exceeding that of HTP by an order of magnitude. The burning of mosquito coil [135] resulted in $PM_{2.5}$ mean concentration of 4324 µg/m^3, what is almost twice higher than in case of CC smoking (2464 µg/m^3) [71]. The concentration of $PM_{2.5}$ from the candle burning varied from actually the same level (31.6 µg/m^3) [135] as HTP generated to more than five times higher mean concentration (168.0 µg/m^3) [78]. It must be considered that the reported level of fine particulate matter may be affected by the measurement method. The environmental aerosol that was formed from the exhaled vapours of new nicotine products is composed primarily of volatile liquid particles (water vapour, propylene glycol).

Thus, the standard gravimetric methods may report lower concentration than those measuring the real time aerosol particles. At the same time, the evaporation as well occurs in the environment, thus potentially minimizing the health risk associated with the liquid particles.

1.2.5 Other Combustion-Related Pollutants

A group of 93 pollutants has been established by the US FDA in tobacco products and tobacco smoke entitled harmful and potentially harmful constituents (HPHCs), including carcinogens, respiratory, cardiovascular, reproductive or developmental toxicants and additives [146]. Jaccard et al. [3] reported a mean reduction of about 90% that has been observed on average across a broad range of HPHCs measured in the HTP mainstream aerosol, compared with the levels of HPHCs of CC. The aerosol produced by experimental tobacco plug blends in the HTP was shown to contain significantly lower concentrations of harmful and potentially harmful constituents (HPHCs) than found in the mainstream smoke of reference cigarette 3R4F [3, 53]. Most importantly, the reduced formation of HPHCs indicated to lead to a reduced exposure when used ad libitum in a short-term clinical study that was conducted with adult smokers in a controlled environment [64].

The use of HTP was reported not to increase the indoor concentrations of carbon monoxide and nitrogen oxides [68], which are listed as WHO priority pollutants. This is expected, since no high temperature combustion/pyrolysis takes place in the HTP. However, the combustion-related sources have been indicated to result in significant increases of the combustion oxides. In 2004, Lee and Wang [77] concluded that incense burning is an important indoor air pollution source for CO. It was the major gas pollutant, resulting from the smoldering effect of mosquito coils [135].

Polycyclic aromatic hydrocarbons (PAHs) form another indicator of incomplete combustion, reported as mostly non-detectable in the HTP smoke [71]. At the same time, the concentration of putative carcinogenic PAHs in indoor air increased by 20% during the vaping of EC [80]. The pyrolysis-related smoking is undoubtedly a strong source, e.g., seven carcinogenic PAHs were found to be a factor that was 2.6 higher during the smoking session of water pipe compared to the control day [83].

Metal concentrations were not significantly higher compared to the background levels during the use of HTP. At the same time, ECs were reported to emit higher amounts of trace metals [71]. Moreover, the emission factors of Cr, Ni, Ag from ECs and La were even higher than from the CCs.

Ruprecht et al. [71] investigated the concentrations of particle-phase alkanes, organic acids and levoglucosan from the use of HTP and provided a comparison with previously reported values from the use of EC and CC [147]. Several n-alkanes and organic acids after HTP use were found at elevated concentrations (two fold) indoors compared to the outdoors. The concentrations of heptacosane, nonacosane, hentriacontane, titriacontane, linoleic and eicosanoic acids and levoclucosan were significantly higher in HTP generated aerosol, while hexatriacontane, octatriacontane, dodecanoic and tetradecaoic acid concentrations were significantly higher in EC, although they were well below the levels associated with the CC environmental smoke. The information on the health risks associated with the previously mentioned organic compounds is very limited, and none of them are included in the US FDA list of tobacco aerosol HPHCs [146]. Moreover, the comparison of the HPHC levels, resulting from the conventional household non-nicotine combustion sources, is not possible, since the list of compounds that was reported from these studies is much shorter.

1.3 Tobacco Product Generated Aerosol Dynamics

The aerosol that is generated by various tobacco products are solid and/or liquid particles that are suspended in the carrier gas, which in most cases, is the air. When the particulate phase is made up of liquid droplets, in many cases, the aerosol is formed via the process of nucleation and condensation, followed by the coagulation and break-up [148].

The aerosols have been studied for a long time; however, most discoveries in the dynamics of aerosols are relatively recent, because lately, a more advanced technological base has become available. According to these studies, the aerosol dynamics depends on a number of aspects, such as volatility, accumulation etc.

1.3.1 Nucleation and Accumulation

Recently, a considerable amount of scientific literature has been revolving around the topic of dynamics of aerosol that is generated by new nicotine containing products as well as nucleation process [149]. Modern methods, such as molecular dynamics simulation, enables scientists to test different parameters and evaluate the nucleation and accumulation processes that are taking part in the aerosols that are formed by different sources, including nicotine containing products. It allows scientists to evaluate their theories and empirical results more thoroughly.

Aerosol particulate matter from HTP or EC liquid is not formed from the condensation of the byproducts of combustion like in CC. HTP or EC aerosol droplets are generated when solvent (propylene glycol) in case of EC or tobacco additive (glycerol) in case of HTP is vaporized. When vaporized solvent reaches supersaturation and condense on cooling, nuclei are formed, onto which more solvent, water, nicotine or other particles can condense to form aerosol droplets during the accumulation process [150].

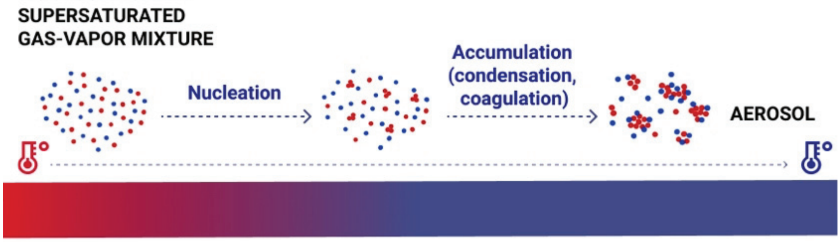


Figure 12. Nucleation and accumulation processes of nicotine containing products (adapted from [151])

When aerosols spread indoors or outdoors, a variety of mechanisms influence the changes in their physicochemical characteristics. The modifications take place due to the following factors: hygroscopic properties, evaporation/condensation rates, transport mechanics as well as coagulation/aggregation [12]. When the vapour pressure reaches the right balance with the liquid phase, the seeds come to a specific and relatively stable size. In a very short period of time, from milliseconds to seconds, each suspended object moves randomly (which is called Brownian motion), but the speed is lower than that observed in the gas phase. This random movement causes the particulate matter to collide and merge: coagulation happens in the case of droplets, whereas in a case of solid particles, the aggregation is observed. This means that the average aerosol particle size gets larger, and at the same time, their number concentration diminishes. When the time period is longer, from several minutes to several hours, the chemical reactions or reevaporation/condensation take place [149]. In essence, nicotine containing products emit gases or vapours, which in turn interact via chemical reactions or homogeneous and heterogeneous nucleation (Fig. 12).

It was demonstrated that the aerosol produced as a result of using HTP, consists of liquid droplets only. The droplets are formed once a homogeneous nucleation process at a relatively low operating temperature takes place. Meanwhile, CC contain a significant number of solid carbon-based nanoparticles, which are a result of the combustion [152]. However, in systems like EC, nucleation takes place in a very narrow zone [143]. Since the aerosol formation rate is an exponential function of vapour concentration and temperature, the performance of valid calculations is rather complicated as well as the accurately predicting conditions of aerosol formation in the EC. Defining the location of the nucleation zone is challenging as well. It may happen that some of the nanoparticles do not pass through the nucleation zone; alternatively, there may be not enough vapour to condense on all nanoparticles. If the latter is the case, some of the nanoparticles would not grow to submicron size [143]. These scientific challenges are dealt with the help of molecular dynamics simulation.

There are three main theories that are used to describe the nucleation processes: Classical Nucleation Theory (CNT), Molecular Dynamics (MD) and Kinetic Model (KM). These three theories were used for the simulation of nucleation of propylene glycol from vapour state at 70 °C as shown in Figure 13 [153].

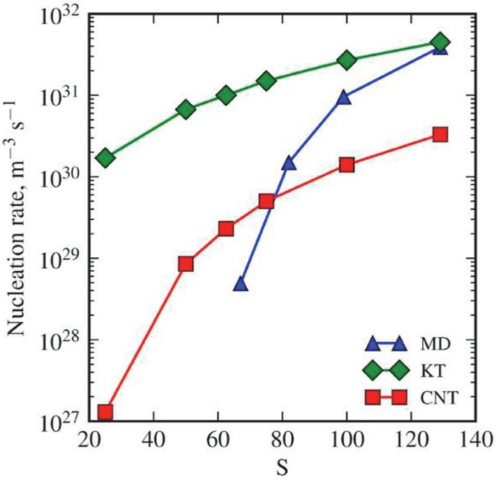


Figure 13. Nucleation rate isotherms at 70 °C, computed using molecular dynamics simulations (MD), kinetic theory (KT) and classical nucleation theory (CNT) [153]

Propylene glycol is the main solvent that is used in the EC liquids. Molecular dynamics simulation results have shown that propylene glycol nucleation events occur at about 70 °C. The simulation results have as well revealed that CNT underestimates the nucleation rate at high supersaturations. KT assumes no free energy barrier to nucleation, thus this theory has a better agreement with the MD. Molceular dynamic simulation has shown that MD predict over a factor of two larger clusters and 50% higher free energy barriers than the CNT. Thus MD theory is more feasible to describe propylene glycol droplet nucleation rate from the vapour phase than other two theories [153]. However, most of the researchers in their articles refer to CNT to describe the nucleation rate. In case of this doctoral thesis, CNT will be used as well when there are references to the nucleation processes.

1.3.2 Gravitational Deposition

The deposition on interior surfaces is considered an essential mechanism for removing tobacco smoke particles. Particle deposition depends on a variety of characteristics, such as particle size, ventilation rate, air circulation, surface properties and temperature differences between the air and surfaces [154, 155]. There are several ways of aerosol interaction with the surface: diffusional deposition, impaction and gravitational deposition (Fig. 14.).

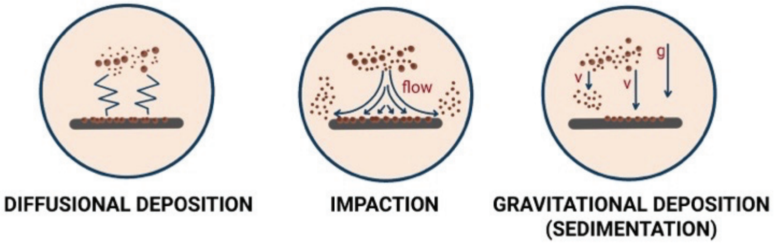


Figure 14. Types of aerosol interaction with the surface (adapted from [156])

The diffusion is the main driving force in particle transportation when it comes to particles smaller than 0.1 m in diameter with standard density. As for the larger ones are concerned, the gravitational deposition is the main factor that impacts their movement. The particles that are below 0.1 m in diameter normally settle onto surfaces via diffusional deposition; however, if coagulation takes place, larger particles will be formed. The particles over 1 m in size frequently settle onto a surface because of the gravitation. A particle size of interest is 0.525 m; this is due to the fact that this size is the one when the net displacement taking place due to the diffusion is the same as the one caused by the gravitational settling. When particles are about this size, neither diffusive nor gravitational forces dominate; this is why they are relatively steady (in comparison to the particles of other diameter). This may be the reason why a lot of particles that are smaller than 0.5 m often float in the air [12]. For example, cigarette smoke contains particles ranging from less than 0.01 m to larger than 1 m.

1.3.3 Volatility

Volatility is a quality that describes how easily a substance volatilizes. In the surrounding medium and the test tube, any organic chemical substance is divided between its own aqueous and gaseous phases based on the Henry's Law. Volatility does not contain a specific numerical meaning, but it is often described by the introduction of the net pressure or boiling point (for liquids). A higher vapour pressure indicates higher volatility, while the maximum boiling point indicates low volatility [157].

Volatility depends on two main contributing factors, i.e., intermolecular forces and molecular weight. That is why it is important to know the chemical structures of smoke or vapour components in order to predict their dynamics in chamber or real-life premises. Like water, organic components in the tobacco are vaporized when the tobacco is heated either by an external source or the exothermic (heat-generating) processes [51]. Intermolecular forces between organic compounds are usually weaker than between the water molecules. This is the reason why organic compounds tend to be more volatile than water.

The awareness of the effects of volatilisation on a particle size and compositions is fundamental to the understanding of the effects of human well-being and the surrounding indoor environment. Recently, the particles that are mainly consisting of volatile compounds, such as tobacco smoke originating from the EC and HTP, have attracted the concern of the scientific community. Therefore, the effects of volatilisation on particle volume dispersion and particle size distribution are considered important; however, the measuring of PSD in tobacco smoke generated by the HTP and EC is difficult because of the liquid nature and high coagulation rate of aerosols [143, 158–160]. In case of EC high volatility of the exhaled particles, containing up to 76% of water, it causes rapid evaporation to nucleation mode; thus, possibly, they cannot be measured efficiently by various particle detection techniques [68].

The key difference in volatility between EC, HTP and CC aerosol can be explained by their different physical and chemical properties. Sub-chapter 1.2.4. presents the evidence that cigarette smoke contains particulate matter associated with many low-volatile compounds, whereas HTP aerosol has fewer low-volatile compounds and no solid particles. Due to the particles in the lead, consisting of propylene glycol and/or glycerol, the volatility of these particles was higher than in CC tobacco smoke [161].

It has been found that the composition of exhaled EC aerosol was greater than 99.9% of water and glycerin (about 75% water, 25% glycerol) [162]. The main constituents of the EC aerosol are polar compounds that include water, glycerin and propylene glycol. Propylene glycol has higher volatility (b.p. = 188 °C) than glycerol (b.p. = 290 °C). Besides, glycerol and propylene glycol, which were originally in the e-liquids, other compounds like benzaldehyde, diphenyl ether, diethyl phthalate and naphthalene were detected in the EC filters [163].

The negative correlation between the particle volatility and the volumetric ratio of vegetable glycerin in the e-liquid was revealed. It has been found that the addition of flavouring chemicals into the e-liquids significantly decreased the volatility of EC generated aerosols as well [164]. In another study, the same e-liquids were analysed, i.e., propylene glycol (PG) in a concentration of 100% and glycerol or "vegetable glycerin" in a concentration of 99.7%. There was no nicotine in these two liquids. PG is more than 100 times more volatile than vegetable glycerin. It is worth noting that secondhand exposures to $PM_{2.5}$ from EC aerosols are likely to be short-lived for most scenarios, and a major portion of the EC aerosol (88%; CI 77–99%) evaporates within a few minutes. The volatility and the rate of evaporation of pure compounds are mainly controlled by their vapour pressure [165]. Feng et al. [166] have found that EC droplets, which are more hygroscopic than the CC aerosol, grew in size faster when exposed to the humid environment of the lung. Moreover, the volatility at three different temperatures (37, 100, 170 °C) was examined, and it has been discovered that the particle number decreases by about 50% between the low and high temperatures, suggesting rapid evaporation [167].

The aerosols emitted from ECs contain a large number of volatile droplets [168]. When the EC aerosols went through the thermodilution system (from 37 °C to 300 °C), it was observed that the mode of the mainstream EC aerosol decreased by about 24 nm, and the final mode that was noted was about 10 nm; meanwhile, the particle concentration decreased by approximately 30%. These findings correlate with the results of the study conducted by Pacitto et al. [169] on the thermodilution of the HTP aerosol and Pratte et al. [170] where the thermodenuder technique was used: a residual peak was visible at about 0.01 m.

HTP generated aerosols as well contain glycerol and propylene glycol, but since the working principle of HTP differs from the EC, the concentrations of these components are much lower in HTP aerosols. Glycerin and propylene glycol are used as the humectants in the HTP HeatSticks [171]. The HTP enables various ingredients to be added to the tobacco powders as carriers, because of that, aerosol dynamics of different HTP brands may differ significantly. In a case of EC, the ingredients must be uniformly dispersed within the liquid, and the list of possible constituents is shorter.

Paccito et al. [169] has shown that the particle concentration in HTP smoke was less than 1E+08 #/cm^3, but their size distribution was found about 0.1 m. However, it has been shown that as the temperature rises, the particle size distribution drops roughly to 0.02 m (300 °C), and the volatility of particles increases. The other researchers used fatty acids as low polarity substances and applied in aerosol fixation in order to reduce the particle volatility, which is covered by fatty acids on the surface of the particles. It has been found that the solubility of the fixation agent was an important factor in the fixation. The researchers have shown that it is possible to lower the volatility, and therefore, the spread of HTP aerosols by adding fixation agents [172].

Other experiments illustrate the higher volatility of HTP as well as EC aerosols and their rather different physicochemical properties when compared to the CC smoke [149]. During the experiment data analysis, a standard mathematical approach was applied to the smoke mass concentration profile of cigarettes, when the rates of decay appeared to be between 0.80 and 0.83 h^{-1}. This exceeds the applied ventilation rate of 0.5 h^{-1}; in this case, the wall losses or evaporation were not considered. In other words, the smoke mass concentration goes down by 95% after 3.75 h, in contrast to 6 h, when only the room ventilation rate was considered. In the case of HTP aerosol, the researchers were only able to estimate the ventilation rate of decay (RD), which was equal to the aerosol lifetime of less than 1 min. This can be explained by the rapid evaporation of aerosol droplets, which takes place when they are released into a closed environment: as a result, very few data points are generated. When it comes to EC aerosol, enough data points were obtained, and a reliable curve fit could be achieved. It turned out that the calculated RD vary in the range of 145–155 h^{-1}, this range is substantially larger than the one of room ventilation alone (0.5 h^{-1}). These results lead to the conclusion that EC and HTP aerosols dissipate about 225 times faster than a cigarette.

1.4 Sub-Chapter Summary

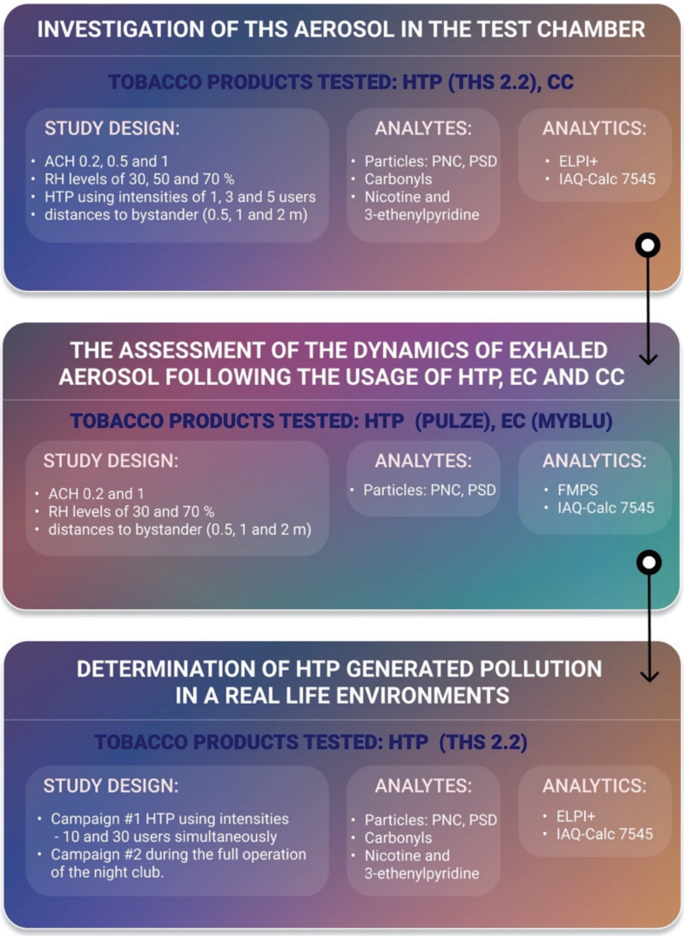
Generally, the data presented in sub-chapter 1.1. indicates that both EC and HTP usage results in lower adverse impact on indoor air, compared to CC, water pipe as well as many other indoor pollution sources, such as incense burning and mosquito coils. This is further supported by the outcomes of several studies on other combustion based tobacco products, such as cigars and pipes. Most importantly, the composition and qualitative and quantitative characteristics of EC and HTP environmental aerosols differ considerably from those of environmental tobacco smoke. The summary of recent studies on the impacts of HTPs, ECs and CCs on the indoor air quality that is presented before indicates that while the usage of HTP and EC devices results in a much lower deterioration of indoor air quality compared to the traditional combustible tobacco products, the issue of the second hand smoking remains. The exhaled aerosol, when using HTP and EC, is a very dynamic system that is affected by multiple factors.

The references that are analysed in sub-chapter 1.2. show that the usage of HTP is a relatively weak source of pollution based on carbonyl, mono-aromatic hydrocarbon, nicotine and fine particulate data, compared to the traditional cigarettes ($p < 0.05$) and water pipes, although statistically insignificantly higher emitter than EC ($p > 0.05$). Among the non-nicotine sources, higher levels of aldehydes, VOCs and $PM_{2.5}$ were generated by incense and mosquito coil, although at the same levels for candle, in case of formaldehyde ($p > 0.05$). This implies that the indoor combustion of incense and especially mosquito coils, if used in a similar temporal pattern, may provide a higher exposure to pollutants than standing near a user of novel nicotine containing products, including the EC and HTP.

The purpose of 1.3. sub-chapter was to give a summarized overview of the aerosol dynamics, physicochemical characteristics and associated applications. When EC and HTP are used, the process results in the emission of submicron liquid droplets made up from extremely volatile ingredients. Depending on the ventilation parameters, the volume of enclosed environment and the usage intensity, there may be certain variations in the aerodynamic diameter of the liquid droplets. Such droplets are dispelled within seconds; thus, the exposure to ultra-fine particles is restricted to a very short period of time and only to persons who are in close proximity to the HTP users. This is in contrast with the PM of environmental tobacco smoke, which is a lot more stable and lingers for a long time in enclosed environments, leading to substantial build-up of indoor pollution, especially in those cases when the smoking rates are significantly elevated and the ventilation levels are low. In addition, the particle nucleation and accumulation take place, since particle concentration during the exhalation rises above $10E+6\ cm^{-1}$. The aerosol dynamics and gravitational deposition depend on a number of factors, such as the type of aerosol generation device (EC or HTP), aerosol particles parameters, for instance, volatility, as well as environmental conditions, including operating air conditioner, distance of transport and ventilation intensity.

2 METHODS AND MEASUREMENTS

The main information on three separately conducted experiments is summarized in Fig. 15. A detailed description of each experiment is provided further in sub-chapters 2.1., 2.2. and 2.3.



INVESTIGATION OF THS AEROSOL IN THE TEST CHAMBER

TOBACCO PRODUCTS TESTED: HTP (THS 2.2), CC

STUDY DESIGN:
- ACH 0.2, 0.5 and 1
- RH levels of 30, 50 and 70 %
- HTP using intensities of 1, 3 and 5 users
- distances to bystander (0.5, 1 and 2 m)

ANALYTES:
- Particles: PNC, PSD
- Carbonyls
- Nicotine and 3-ethenylpyridine

ANALYTICS:
- ELPI+
- IAQ-Calc 7545

THE ASSESSMENT OF THE DYNAMICS OF EXHALED AEROSOL FOLLOWING THE USAGE OF HTP, EC AND CC

TOBACCO PRODUCTS TESTED: HTP (PULZE), EC (MYBLU)

STUDY DESIGN:
- ACH 0.2 and 1
- RH levels of 30 and 70 %
- distances to bystander (0.5, 1 and 2 m)

ANALYTES:
- Particles: PNC, PSD

ANALYTICS:
- FMPS
- IAQ-Calc 7545

DETERMINATION OF HTP GENERATED POLLUTION IN A REAL LIFE ENVIRONMENTS

TOBACCO PRODUCTS TESTED: HTP (THS 2.2)

STUDY DESIGN:
- Campaign #1 HTP using intensities - 10 and 30 users simultaneously
- Campaign #2 during the full operation of the night club.

ANALYTES:
- Particles: PNC, PSD
- Carbonyls
- Nicotine and 3-ethenylpyridine

ANALYTICS:
- ELPI+
- IAQ-Calc 7545

Figure 15. Key parameters of the conducted experiments

2.1 Investigation of HTP Aerosol in the Test Chamber

The previously mentioned studies are mostly aimed at the quantification of levels (average values) of aerosol particles or gaseous compounds in indoor air during the usage of HnB devices. At the same time, no data are yet available on the time-resolved patterns of dispersion of aerosol in a room under varying environmental conditions. The aim of this study was to assess temporal and spatial variations of exhaled HTP aerosol in a chamber, based on a highly time resolved aerosol concentration and particle size distribution measurements, supplemented by the averaged values of tobacco specific gaseous air pollutants. This data was regressed against the environmental conditions, such as ventilation rate, relative humidity, HTP usage intensity and distance of the user to a bystander, indicating important factors that are affecting the potential exposure.

2.1.1 Indoor Chamber Description

A test chamber (the floor area of 13 m^2 and a volume of 35.8 m^3), representing a standard room, was adapted for the purpose to evaluate the levels of various gaseous pollutants, aerosol particle concentration and size distribution at the location of potential exposure of an occupant in a room. The walls, floor and ceiling of the chamber were fabricated by using conventional construction materials, such as painted dry-wall, PVC lining and a panel ceiling. The supply and exhaust airflow via in-ceiling air diffusers were controlled by using an air handling unit (GOLD 04, Swegon AB, Sweden). The supply air temperature during the experiments was set to + 22 ± 2 °C. The relative humidity (RH) was regulated in the range of 30 to 70% using air humidification system (ES4, NORDMANN Engineering AG, Basel, Switzerland). The cleaning of chamber before each experimental day was done by water based floor mopping.

The supply air was conditioned in a heat exchanger and treated with three-step filtration: pre-filter of class F7, activated carbon bed (removing outdoor VOCs and ozone) and HEPA13 final filter. The four-way mixing ventilation was chosen for this study, as it is commonly used in residential buildings. Such setup has been tested to provide sufficient dispersion of aerosol, generated in a room [173]. Relative humidity (RH) has been controlled by humidifier (ES4, Nordmann Engineering AG, Switzerland) in the range from 30 to 70%, while the temperature was maintained within a range from 21 to 23 °C. One multi-nozzle air supply diffuser of 0.5 × 0.5 m with plenum box was used for the in-ceiling air supply. The air change rate inside the chamber was checked before and after each day experiments via the duct air velocity measurements (Innova 1221, Lumasense, Denmark). Additionally, the air change rate was verified by using CO_2 tracer gas decay method (ASTM, 2000) by CO_2 meter (7545 IAQ-CALC, TSI Inc., the USA) before and after the measurement campaign. The effectiveness of purging of the chamber between runs was additionally tested by using metal oxide VOC sensors (iAQ-2000, ams AG, Austria).

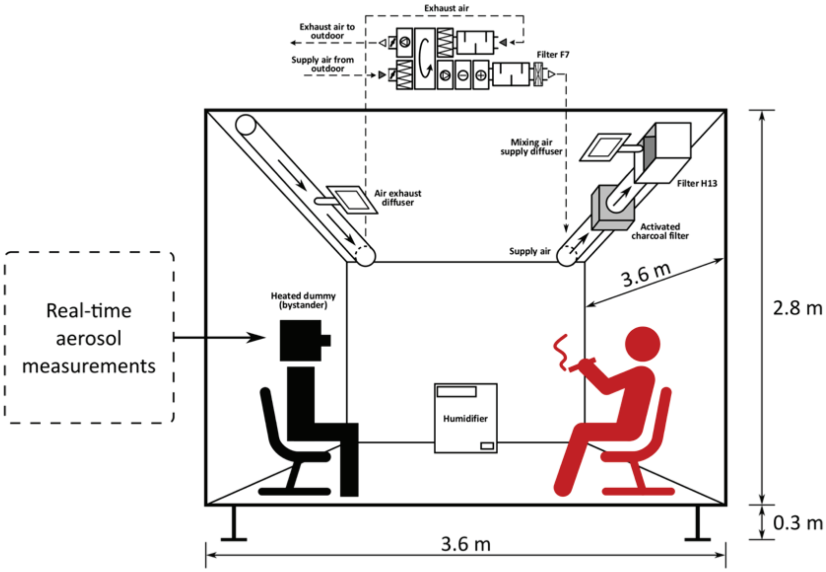


Figure 16. Experimental chamber [73]

Bystander

A "dummy" of a rectangular geometry was installed in the chamber to simulate a seated person with the inclusion of "legs". The surface area of the "dummy" was equal to 1.7 m^2 covered with a textile fabric. The surface temperature of the dummy was maintained in the range of + 31 °C to + 34 °C, similar to the human body surface temperature. The dummy was seated on a chair near a wall. The aerosol samples were drawn through a copper inlet tube and divided to sampling instruments, which were positioned immediately outside of the chamber in order to minimize the particle losses due to the diffusion and evaporation processes.

2.1.2 Experimental Design and Analytics in the Chamber

The experiment aimed to research the quantitative effects of environmental variables, including ventilation intensity (V) as air changes per hour (0.2, 0.5 or 1 h^{-1}), HTP use intensity (I) as the number of parallel users (1, 3 or 5), relative humidity (RH, 30, 50 or 70%) and the distance to the bystander (D, 0.5, 1 or 2 m) onto pollutant concentration variations in a chamber. The ventilation intensity and humidity parameters were chosen as recommended comfort conditions (V 0.5; RH 50) as well as conditions outside the humidity comfort range (RH 30; RH 70). The ventilation intensity of 0.2 h^{-1} was chosen as typical for the indoor environments without mechanical ventilation, while 1 h^{-1} to represent intensively ventilated residential environment [174, 175]. The experimental plan with variables and responses are provided in supplementary material (A3). In total, 30 experimental runs were carried out with HTP and three with CC.

Particles

The real time size-segregated particle concentration was determined by using the electrical low pressure impactor (ELPI+, Dekati Ltd., Finland) at a flow rate of 10 l/min. ELPI+ divides aerosol particles to 15 fractions (from 0.006 μm to 10.0 μm). This instrument utilises the cascade impaction principle and has a direct-reading capability. The real-time concentrations of aerosol samples were registered at 1 s temporal resolution. Additionally, for the process control, a scanning mobility particle sizer (SMPS) (3910, TSI Inc., the USA) classified the aerosol particles across 13 size bins from 10 to 420 nm, based on the electrical mobility diameter during 60 s, or was used as a counter for a single size bin at 1 s resolution with the flow rate of 1 l/min. SMPS measures the particles, using electrical classification by the radial differential mobility analyser and optical counting, using isopropanol-based condensation particle counter; the resolution in terms of concentration is 1 $\#/cm^3$.

The verification of particle size distribution (PSD) as measured by the particle instruments was conducted by generating an aerosol of Polystyrene Latex (PSL) particles of 0.3 μm and 1.0 μm in the chamber, using an atomizer (Collison Nebulizer, CH Technologies, the USA), Inc. and passing the flow through the aerosol neutralizer (3054, TSI Inc, the USA).

Carbonyls

The concentration of formaldehyde and acetaldehyde was estimated following the ASTM D5197-03 procedure. This method involves drawing air (pump Universal PCXR8, SKC Inc., the USA) through a cartridge, containing silica gel coated with 2,4-dinitrophenylhydrazine (DNPH) reagent (SKC Inc.). The DNPH derivatives were analysed for parent aldehydes by utilizing high performance liquid chromatography (HPLC).

Nicotine and 3-Ethenylpyridine

The method was based on ISO 18145 procedure, i.e., the collection of nicotine by adsorption on a sorbent resin (XAD®-4, 7 x 70 mm size, 2 sections, 40/80 mg sorbent, 20/40 mesh, SKC Inc.), extraction of nicotine from the sorbent resin and determination by gas chromatography-mass spectrometry (GC-MS-QP2010 Ultra, Shimadzu Corp., Japan). The standard procedure was modified by using mass spectrometry detection instead of nitrogen selective detection.

Microclimate parameters include carbon dioxide (CO_2) and carbon monoxide (CO). The values of carbon dioxide and carbon monoxide together with temperature and relative humidity levels were continuously recorded by appropriate sensors (7545 IAQ-CALC, TSI Inc., the USA). The instrument was used as a low-drift NDIR sensor for CO_2 (range from 0 to 5000 ppm, accuracy ± 3.0% of reading or ± 50 ppm, resolution 1 ppm) and electrochemical sensor for CO (range 0 to 500 ppm, accuracy ± 3.0% of reading or ± 3 ppm, resolution 0.1 ppm). The meters were factory calibrated before each measurement campaign and additionally inter-compared every third day of the experiment.

2.1.3 Experimental Procedure in the Chamber

The experiment was realized as a D-optimal design, available via experiment planning package Modde 10 (MKS Umetrics, Sweden), supplemented with additional experiments, aiming to compare or validate the main experimental runs. The experiment plan included 20% of replicates. The experimental setup is visualised in Fig. 17.

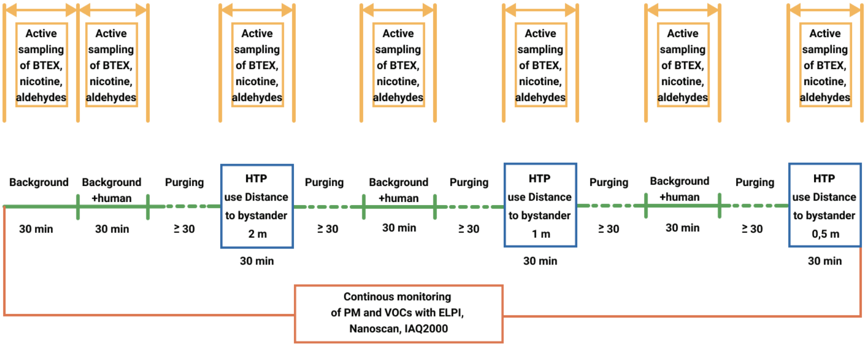


Figure 17. Experimental setup [73]

The real-time particle number concentration (PNC) and particle size distribution (PSD) that were obtained from the aerosol instruments were further analysed for maximum and mean concentration values during the entire experimental run and after the active period of the usage as well as recalculated to particle mass concentration for the $PM_{2.5}$ fraction (particles having aerodynamic diameter of less than 2.5 µm). Additionally, off-line carbonyls (acetaldehyde and formaldehyde), off-line nicotine and 3-ethenylpyridine and real-time CO_2 concentration were entered as responses to the model.

The concentration of pollutants before the experiment was monitored initially for 30 min. to assure low background concentration adequately. A background conditions with human presence but no product use were monitored afterwards, i.e., after 30 min; under the intensity of three HTP users, the background conditions were evaluated with three human subjects; 5 HTP users, background conditions with five HTP users; the experimental chamber was purged at least for 30 min. after each measurement session; ACH of 8 h^{-1} and portable air cleaners (KC-A60, Sharp Corporation; MC70L, Daikin; Therapy Air, Zepter International, Ciuzas et al. (2016)) were used to reach the background levels for both the aerosol particles and VOCs (PNC – 300–500 #/cm^3 or VOCs –500–600 ppm of CO_2 equivalents).

A human subject entered the chamber and used the HTP according to the ad libitum regime, until the full consumption of a single stick (12 puffs on average). The aerosol concentration was monitored for sufficient time to reach a lower asymptote of the exponential decay.

The experiment was performed in the identical sequence every day, only changing relative humidity, air change rate as well as the HTP use intensity.

The pollution generated by the smoking of a conventional cigarette (CC, Marlboro Gold) was investigated for the comparison with HTP-generated pollution. The experiment was run at 50% RH, 0.5 ACH, at 2 m distance to the bystander and 1 cigarette smoking intensity, representing the “medium” exposure conditions. In total, three experimental runs were carried out. The evaluation of background conditions, sampling duration and analyte measurement were identical as in the case of HTP experiments.

A volunteer checklist was issued before the experiments to ensure proper clothing and hygiene products, thus minimizing the background pollution. Each study participant was thoroughly inspected, and the potential risk was assessed by the research personnel before the experiment.

The data processing and analysis was carried out, according to the following procedures that are listed below.

Temporal variation of particle number concentration and particle size distribution. The measured distributions were based on the particle number concentration (unit particles/cm^3 or #/cm^3). The PSD was based on the number density (concentration) distribution function, which represented particle concentration normalized to the particle size bin where it was measured, i.e., the number of particles per volume of air with size between Dp and dDp. This is usually expressed mathematically as follows: Dp = dN/dlogDp (#/cm^3).

Regression modelling of factors affecting dispersion of particles. The results were fitted to a regression model to obtain the effects of air changes per hour, distance between the bystander and a user and the intensity of HTP use (number of human subjects using HTP). Multiple linear regression (interaction) model was employed in order to obtain linear functions, relating experimental variables and responses. This process was used to determine the significant process factors that are affecting the variations of pollutant concentrations between runs. The obtained models were assessed for their coefficient of determination (R^2), predictive squared correlation coefficient (Q^2), model validity and reproducibility.

The normalisation of data with respect to chamber size. The measured concentrations were normalised, aiming at the adequate comparison of measured concentrations and the results from other studies. The following conditions were suggested by CEN/TS 16516 standard [176], i.e., chamber volume of 30 m^3 and air change rate of 0.5 as well as the number of test products used in each study, according to the following formula: $C_{normal} = C_{measured} \text{ x } (V \text{ x } ACH)/(V_{st} \text{ x } ACH_{st})$.

2.2 Dynamics of Exhaled Aerosol Following the Usage of HTP, EC and CC

This study has researched an exhaled aerosol from the usage of a new HTP product and compared it with a vaping product (EC) and a conventional cigarette (CC) in terms of highly time resolved particle concentrations and size distributions. The important factors that are affecting the aerosol dynamics in a room were researched and estimated quantitatively.

2.2.1 Used Products

The following nicotine containing products were investigated in the research, i.e., a heated tobacco product “Pulze” and an e-cigarette “myblu”, both manufactured by Imperial Brands, PLC (the UK) (Fig. 18).

“Pulze” is a battery-powered heated tobacco device, which heats up a refined tobacco rod (branded as “iD Stick”) to a pre-set temperature: either 315 °C in eco mode (referred to as HTP_Eco in the experimental plan) or 345 °C in standard mode (HTP_Standard). Non-flavoured iD sticks were used. The tobacco rod component of the iD stick consists primarily of reconstituted tobacco and glycerin (the principal humectant ingredient that is used for aerosol formation in the heated tobacco product). The aerosol is produced by heating and subsequent distillation process where the active ingredients from tobacco are trapped in liquid mainstream aerosol, filtered within several stages of filtration within the ID Stick and thus delivered to the user.

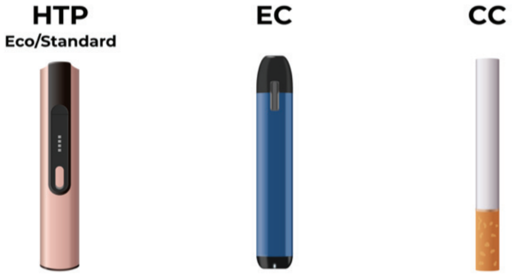


Figure 18. Used products [27]

The data on conventional cigarette has been taken from the previous study, comparing several types of ECs, and the CC Martuzevicius et al. [177], which was performed in the same setting by using Marlboro Gold (Philip Morris International, Inc), having tar, nicotine and carbon monoxide yields of 8 mg, 0.6 mg and 9 mg, respectively.

2.2.2 Measurement Methods

This experiment was conducted in the same chamber as described in sub-chapter 2.1.1. (Fig. 16). The particle number concentration (PNC) and particle size distribution (PSD) was measured by the Fast Mobility Particle Sizer (FMPS) spectrometer (model 3091, TSI Inc., USA) in the size range from 5.6 to 560 nm in 32 channels with 1 sec^{-1} time resolution. The measured distributions were based on the particle number concentration (unit particles cm^{-3}) measurement. Before the start of each run, a background concentration of particulate matter was recorded (3 min. before the start). The background concentration at the beginning of each experiment was aimed to be achieved as low as possible and ranged between 300 and 5000 particles cm^{-3}.

Relative humidity and temperature were measured continuously (IAQ-Calc 7545; TSI Inc., the USA) at a resolution of 30 sec. The same instrument was used for the verification of ventilation rate regimes based on the decay of CO_2 concentration.

2.2.3 The Experimental Design and Analytics

The experiment has been designed as a full factorial multiple linear regression interaction model, aided by an experiment planning software (Modde 10.1, Umetrics Inc, Sweden). Full experimental worksheet is presented in Appendix 7 (supplementary material). The experiment factors (controlled variables) were set to Product Type (qualitative variable, having values of HTP_eco, HTP_standard and EC), Ventilation Rate (0.2 h^{-1} representing minimum ventilation and 1 h^{-1} representing an intense ventilation regime), Relative Humidity (30%, representing dry air, and 70%, representing humid air) and Distance from a user to a dummy (0.5, 1.0 and 2.0 m.). Additionally, three male volunteers (aged between 20 to 30 years) have been included in the study to represent the user variable (marked as V1, V2 and V3). Such design resulted in 108 runs in total. No specific requirements were set to the volunteers that were participating in the study, except that they were regular smokers or/and experienced e-cigarette users. Three male volunteers within 20–30 years of age have signed for the study, acknowledging their consent to participate in a written form. The volunteers were regular smokers of traditional cigarettes and electronic cigarettes, but have not used the particular HTP product. No other user related data has been collected, thus treating volunteers as an independent variable that brings random variation in the results.

The responses to the model that were derived from the real time particle number concentration and size distribution measurements are presented in Table 2. All responses (except ratios) have been log-transformed in order to correct for skewness in distribution. The model data was fitted into multiple linear regression interaction model. After initial runs, the insignificant term (interaction mostly) terms were manually removed, aiming to balance the goodness of the model based on the estimate of model fit (R^2) and the estimate of the future prediction precision (Q^2).

Table 2. Parameters that derived from the real time measurements of particle size resolved number concentrations

Response title	**Abbreviation**	**Units**	**Description**
Temporal variation parameters			
Maximum total particle number concentration (PNC)	$PNC_{t,max}$	#/cm^3	The highest particle number concentration obtained during a single run. This indicates the level of total PNC immediately following the exhalation.
Total PNC after 5 s	$PNC_{t,5s}$	#/cm^3	Particle number concentration following the last peak after 5 sec. This indicates the rapid

			dispersion/transformation of particles.
Total PNC after 10 s	$PNC_{t,10s}$	$\#/cm^3$	Particle number concentration following the last peak after 10 sec. This indicates the rapid dispersion/transformation of particles.
Total PNC after 120 s	$PNC_{t,120s}$	$\#/cm^3$	Particle number concentration following the last peak after 120 sec. This indicates the long-term processes of particle transformations.
Concentration ratio: PNC after 5 s/PNC$_{max}$	$PNC_{t,5s}/PNC_{t,max}$	Non-dimensional	This indicates the decrease of PNC after 5 sec from the maximum, in relation to the maximum.
Concentration ratio: PNC after 10 s/PNC$_{max}$	$PNC_{t,10s}/PNC_{t,max}$	Non-dimensional	This indicates the decrease of PNC after 10 sec from the maximum, in relation to the maximum.
Concentration ratio: PNC after 120 s/PNC$_{max}$	$PNC_{t,120s}/PNC_{t,max}$	Non-dimensional	This indicates the decrease of PNC after 120 sec from the maximum, in relation to the maximum.
Concentration decrease rate from PNC$_{max}$ to PNC$_{5s}$	r_{max-5}	min^{-1}	This indicates the rate of the PNC decrease following the exponential decay kinetics and calculated as $(\ln PNC_{t,max}-\ln PNC_{t,5s})/(5/60)$.
Concentration decrease rate from PNC$_{max}$ to PNC$_{t,10s}$	r_{max-10}	min^{-1}	This indicates the rate of the PNC decrease following the exponential decay kinetics and calculated as $(\ln PNC_{t,max}-\ln PNC_{t,10s})/(10/60)$.
Particle size distribution parameters			
Mode at the maximum peak	M_{max}	nm	Particle size having the highest concentration during the $PNC_{t,max}$
Mode after 5 s	M_{5s}	nm	Particle size having the highest concentration during the $PNC_{t,5s}$
Mode after 10 s	M_{10s}	nm	Particle size having the highest concentration during the $PNC_{t,10s}$
Mode after 120 s	M_{120s}	nm	Particle size having the highest concentration during the $PNC_{t,120s}$

The experimental flow was conducted accordingly: the test chamber was purged before an experiment for at least 10 min. at a ventilation rate of 10 h^{-1} to reduce the background concentration of particles. A volunteer entered the chamber and used HTP or vaped the EC according to the planned regime, which was one puff every 30 seconds, five puffs in total. Each volunteer used separate devices (three Pulze and myblu devices in total), while the HTP device has been loaded with a fresh rod before every experiment. After puffing, a tobacco rod has been extinguished in a flask filled with water to prevent any further aerosolization of particles. A volunteer remained calmly seated for additional 30 minutes and exited the chamber afterwards.

2.3 Determination of HTP Generated Pollution in the Real Life Environment

The impact of HnB products on the IAQ in real-life settings has not been investigated so far. The aim of this study was to simulate the variation of IAQ, following the usage of the HnB product (namely, HTP) in a non-operating nightclub with varying amount of users and compare it to the typical IAQ in a nightclub under operation.

2.3.1 Description of Premises

The experiment was conducted in a nightclub in Kaunas, Lithuania, with the floor area of approx. 160 m^2 and the volume of approx. 880 m^3. The club premises may be subjectively divided into several areas, including wardrobe, bar area, DJ scene, the main dancing floor and the seating areas (Fig. 19).

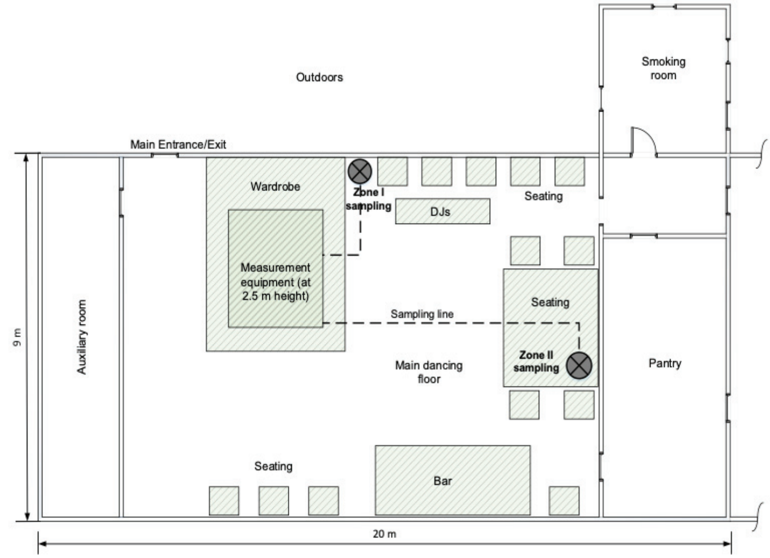


Figure 19. Floor plan and sampling-related information within the club premises [144]

The facility was equipped with a mechanical ventilation system, supplying the air via air handling unit, equipped with rotary heat exchanger, 9100 m^3 h^{-1} maximum airflow. However, this air handling unit was operated manually, while most of the time the club was being operated in the natural ventilation regime (0.5 ACH, as determined by the CO$_2$ concentration decay after the closure). The indoor of the club was a non-smoking area, following a smoking ban in public premises in Lithuania. The smoking room was located outdoors with an entrance from the main club area.

During the operation, the club was attended by 200–400 people at the time, while the smoking room may accommodate approx. 30 people. No cooking activities took place in a club, only drinks and occasionally cold appetizers were served.

2.3.2 Experimental Design, Analytics and Data Processing

The experiment was performed in February of 2018 during the course of two campaigns. The first campaign (Campaign #1) was carried out during the non-working hours of the club to evaluate the HTP-generated pollution while avoiding the presence of other active pollution sources over three days, each consisting of five measurement sessions: background; background with 10 humans present, but without HTP use; 10 humans simultaneously using HTP; background with 30 humans present, but without HTP use; 30 humans using HTP scattered all over the main club area. Each measurement session took 30 minutes. The experiments were conducted with only natural ventilation (air handling unit off), since it reflects the usual condition of a nightclub. After both IQOS usage sessions, the particle and CO_2 concentration were reduced to the background levels via purging indoor air by running the air handling unit at 75% capacity (equivalent to 7–8 ACH) for 30 minutes. The indoor air was sampled at the breathing height (1.5 m from the floor) at two locations, representing different types of occupancy: the main (dance floor) area of the club (Zone 1) and the sitting area (Zone 2) (see Fig. 19). Zone 2 was located under an internal balcony, thus restricting the airflows around it, presumably decreasing the dispersion rate of the exhaled aerosol.

The second campaign was carried out for three days (Campaign #2). During each day, 1 hour before the club opening (representing background) and 3 hours of club in operation were sampled in order to obtain the profile of IAQ, occurring during the typical club activities. During the third day, the real-time measurements were extended for 4 additional hours to obtain the variation of pollutant concentrations until and after the closure of the club. Similar sampling locations were employed as in Campaign #1. The ventilation during Campaign #2 was adjusted manually running air handling unit at 25% capacity (equiv. of 2.6 ACH) with no attendees to 100% capacity (equiv. of 10 ACH) with the maximum attendees (~400 people).

Analytics

The real-time particle number concentration (PNC), CO_2 concentration, relative humidity and temperature as well as off-line carbonyls (acetaldehyde and formaldehyde), off-line nicotine and 3-ethenylpyridine concentration were analysed.

The real-time size-segregated particle concentration have been measured by using the Electrical Low Pressure Impactor (ELPI+; Dekati, Inc., Finland) at a flow rate of 10 L min^{-1}. The real-time concentrations of aerosol samples have been registered in 1 Hz or 10 Hz intervals. The sampling airflow was switched between Zone 1 and Zone 2, using a 3-way switching valve every 5 minutes. The sample was delivered to particle sampler by using Tygon® formula E-3603 laboratory tubing. Such tubing has been confirmed as the best compromise considering the particle losses [178].

The values of carbon dioxide (CO_2), temperature and relative humidity have been continuously recorded by the air quality meters (IAQ-Calc 7545; TSI Inc., the USA).

The concentration of nicotine and 3-ethenylpyridine (3-EP) has been estimated following the ISO 18145 procedure, which is based on the collection of compounds by adsorption on a sorbent resin (4.7 × 70 mm in size, 2 sections, 40/80 mg sorbent, 20/40 mesh; XAD®; SKC Inc., the USA), extraction and determination by gas chromatography-mass spectrometry (GC-MS; GCMS-QP2010 Ultra; Shimadzu Corp., Japan). The concentration of formaldehyde and acetaldehyde has been estimated following the ASTM D5197-03 procedure based on the sample collection on a silica gel coated with 2,4-dinitrophenylhydrazine (DNPH) reagent (SKC Inc.). The DNPH derivatives were analysed for parent aldehydes, utilizing high-performance liquid chromatography (HPLC-DAD (UV)).

The data processing and analysis were carried out in the same way as described in sub-chater 2.1.3.

3 RESULTS AND DISCUSSION

3.1 The Assessment of Temporal and Spatial Variation of Exhaled HTP Aerosol in Chamber

3.1.1 Particle and Gaseous Pollutant Levels

General descriptive statistics of analytes that were investigated under various conditions is summarised in Table 3. The usage of HTP resulted in nicotine, acetaldehyde, particle number and mass concentrations significantly above the background. The highest coefficient of variation (suggesting a stronger influence of the experimental conditions), was observed in case of PM_{10}, $PM_{2.5}$, PNC as well CO_2, nicotine and acetaldehyde. The maximum 30 min. concentration of fine particulate matter as $PM_{2.5}$ (635.7 µg/m^3) and PNC (4.8E+05 #/cm^3) as well as 1 second maximum concentration of $PM_{2.5}$ (109.8 µg/m^3) and PNC (9.3E+06 #/cm^3) suggest that the intensive use of HTP in a confined space with limited ventilation might cause substantially elevated particle concentrations. However, such high maximum concentrations only occur for a very short period of time (5−7 s), as impacted by a directional exhalation from the user to a bystander. Moreover, due to the high volatility of particles (as described in subsequent sub-chapter), it is unlikely that the average particle mass concentration would be substantial as measured by the reference filter-based method. At the same time, the maximum concentrations of formaldehyde (16.3 µg/m^3) and acetaldehyde (12.4 µg/m^3) fell within the range of the mean concentrations that were observed in residential and public environments [4]. High maximum value of CO_2 indicated that numerous presence of users in the chamber caused increased CO_2 levels, which were not efficiently diluted by the air change that was applied.

Table 3. Descriptive statistics of air quality parameters during the use of HTP, summarizing 30 sampling runs under various conditions (usage intensity, distance, air change rate and relative humidity); the averaging time of particle number and mass concentrations corresponds to the sampling duration of gaseous compounds (30 min.)

	Background* (Mean±StDev)	***Mean***	***StDev***	***95% conf. interval***	***Median***	***Min value***	***5th percentile***	***25th percentile***	***75th percentile***	***95th percentile***	***Max value***
PNC, #/cm^3	8.5E+02±7.9E+02	2.1E+05	3.1E+05	2.7E+03	1.1E+05	8.7	1.4E+04	7.9E+04	3.0E+05	4.5E+05	4.8E+05
PM$_{2.5}$, µg/m^3	4.2±8.6	200.0	2339.4	19.8	11.4	0.0	2.6	5.3	32.5	119.4	635.7
PM$_{10}$, µg/m^3	43.2±117.1	1105	17000	144.2	19.3	0.0	3.1	5.9	35.5	188.7	874.9
CO$_2$, ppm	794±274	831	304	5.6	697	474	530	610	1008	1465	1960
CO, ppm	0.1±0.2	0.11	0.17	0.003	0.00	0.00	0.00	0.00	0.20	0.50	0.80
Formaldehyde, µg/m^3	12.0±2.8	11.0	3.7	1.4	10.9	3.6	5.0	7.9	14.4	15.7	16.3
Acetaldehyde, µg/m^3	3.2±1.2	5.7	2.8	1.1	4.9	1.4	2.0	3.6	7.1	10.9	12.0
Nicotine, µg/m^3	2.2±1.9	5.1	3.3	1.2	0.6	1.7	1.8	2.5	6.7	12.3	13.9
3-EP, µg/m^3	0.6±0.2	0.7	0.2	0.1	0.3	0.3	0.4	0.5	0.7	1.1	1.1

The smoking of the CC resulted in a significant increase above the background (i.e., background subtracted) of all measured analytes (Table 4), including formaldehyde (16.6 µg/m^3), acetaldehyde (25.3 µg/m^3), nicotine (28.0 µg/m^3), 3-EP (3.2 µg/m^3), CO (0.94 ppm), PM$_{2.5}$ (158.1 µg/m^3), PMC$_{10}$ (174.0 µg/m^3) and PNC (6.6E+05 #/cm^3). Such levels are higher (from several times to an order of magnitude) compared to the usage of HTP.

Table 4. Descriptive statistics for the air quality parameters during the use of HTP vs. conventional cigarette, under identical experimental conditions (single user, 0.5 ACH, 2 m distance to the bystander and 50% RH)

		Background* (Mean±StDev)	***Mean***	***StDev***	***95% conf. Interval***	***Median***	***Min value***	***5th percentile***	***25th percentile***	***75th percentile***	***95th percentile***	***Max value***
PNC, #/cm^3	THS	8.9E+02±5.4E+02	8.0E+04	3.2E+05	6.2E+02	8.6E+04	3.6E+02	2.6E+04	7.3E+04	9.7E+04	1.0E+05	1.1E+05
	CC	3.1E+02±2.7E+02	6.6E+05	2.6E+05	7.1E+03	7.8E+05	4.2E+02	3.2E+04	6.8E+05	8.2E+05	8.5E+05	9.2E+05
PM$_{2.5}$, μg/m^3	THS	3.3±4.1	5.7	7.6	0.2	5.6	0.4	2.6	3.5	6.8	8.7	311.5
	CC	1.3±8.4	159.4	57.1	1.5	169.8	0.2	22.8	148.9	192.8	225.5	796.1
PM$_{10}$, μg/m^3	THS	7.5±6.0	8.3	54.9	1.5	5.7	0.4	2.7	3.7	7.0	9.2	2352
	CC	5.8±21.1	179.8	191.2	5.1	171.5	0.2	28.7	150.9	195.4	232.1	4937
CO$_2$, ppm	THS	620±58	644	65	3.7	647	501	537	597	690	754	793
	CC	580±63	610	57	3.2	613	498	511	568	651	699	767
CO, ppm	THS	0.01±0.03	0.01	0.02	0.001	0.00	0.00	0.00	0.00	0.00	0.10	0.10
	CC	0.01±0.02	0.95	0.41	0.02	1.00	0.00	0.00	0.90	1.20	1.40	1.60
Formaldehyde, μg/m^3	THS	14.2±0.3	14.1	0.4	1.06	14.1**						
	CC	10.9±0.8	27.5	2.2	5.34	27.5**						
Acetaldehyde, μg/m^3	THS	3.3±0.1	3.6	0.4	0.94	3.6**						
	CC	3.6±0.8	28.9	2.6	6.51	28.9**						
Nicotine, μg/m^3	THS	1.4±0.1	1.81	0.14	0.35	1.81**						
	CC	1.6±0.3	29.6	1.8	4.39	29.6**						
3-EP, μg/m^3	THS	0.5±0.1	0.56	0.03	0.09	0.56**						
	CC	0.5±0.1	3.7	0.3	0.65	3.7**						

* human background

** median value was assumed to be equal to the mean value, assuming a normal distribution of the data

Aiming at the adequate comparison of measured concentrations with the results from the other studies, the measured concentrations were normalised as described in sub-chapter 2.1.3.. The calculations are summarized in Appendix 4. The normalised median nicotine concentration (0.5 μg/m^3) during the usage of HTP in this study was similar to the median concentrations (0.6, 0.7 and 0.6 μg/m^3) in the "Office", "Residential" and "Hospitality" simulations, representing different ventilation regimes [68]. The median concentration of PM$_{2.5}$ (2.9 μg/m^3) as well ranged similarly to Ruprecht et al. (2017) (2.3 μg/m^3). However, the mean PNC in this study was an order of magnitude higher (9.4E+04 #/cm^3) in comparison to the 9.8E+03 #/cm^3 as normalised from [71]. High variation of PNC among studies may be affected by multiple factors, including, but not limited to the differences in puffing topography, chamber size and measurement instrumentation. In a medium-sized room chamber, such as in this study, the distance from the emission source to the measurement site is shorter, preventing minimizing impacts of particle deposition and evaporation.

The normalised median concentration of formaldehyde (19.8 μg/m^3) during the smoking of CC was in good correspondence with the median concentrations (15.0, 13.7 and 16.2 μg/m^3) in the "Office", "Residential" and "Hospitality" simulations, respectively [68], and was in the same range with the mean concentration (21.9 μg/m^3) from [71]. A similar tendency has been observed for acetaldehyde with the median (30.2 μg/m^3) being close to 30.6, 32.3 and 30.6 μg/m^3 [68] as well as 38.9 μg/m^3 from [71]. The median nicotine concentration (33.4 μg/m^3) was higher than in the "Office" (18.1 μg/m^3) and "Residential" (11.2 μg/m^3), but close to "Hospitality" (32.0 μg/m^3) simulation [68]. The normalised mean PM$_{2.5}$ (188.7 μg/m^3) was slightly lower than the mean (273.8 μg/m^3) from [71], but PNC was an order of magnitude higher (7.8E+05 #/cm^3) than the mean (6.7E+04 #/cm^3) from [71], similarly to the HTP. Generally, the obtained data at well controlled conditions confirms the earlier findings in [68, 71] on the important pollutants related to the usage of the HTP and CC.

3.1.2 Highly Time Resolved Aerosol Concentration

The temporal variation patterns of aerosol concentration (as PNC) under varying conditions (distance to the bystander, varying intensity, relative humidity and product type), as measured during the experiments, are presented in Figure 20.

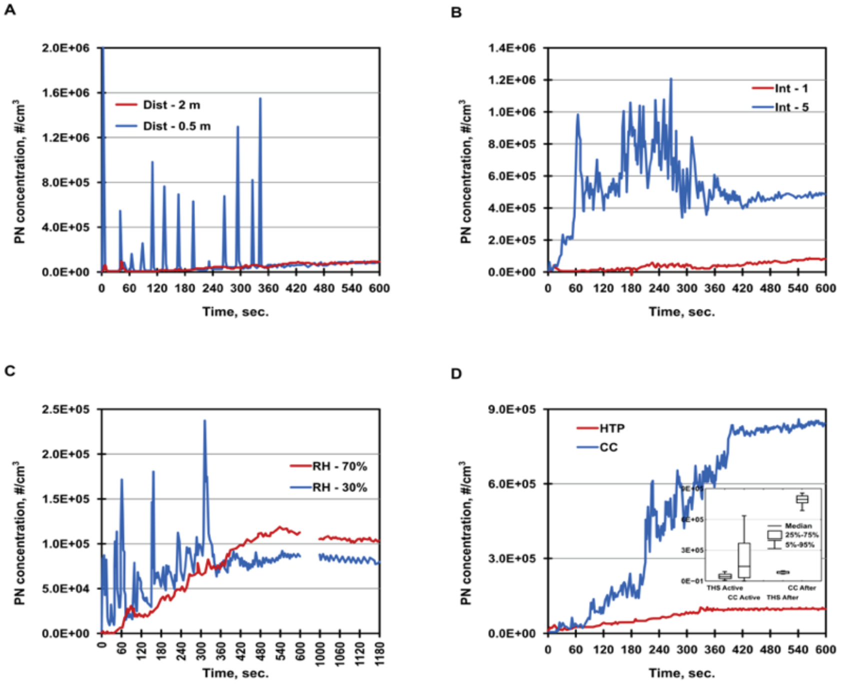
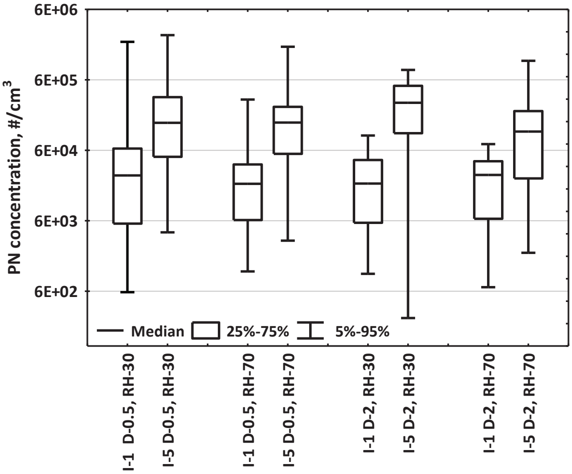


Figure 20. Temporal variation of particle number concentration while using the HTP: (A) the effect of distance from the bystander 0.5 m vs. 2 m (I-1, V-1, RH-70); (B) one user vs. five simultaneous users (D-2, V-0.2, RH-30); (C) RH 30% vs. 70% (D-2, I-1, V-1); (D) HTP vs. CC (I-1, D-2, V-0.5, RH-50); the inset of (D) represents the summarized distributions of PNC during the active usage and after the usage periods [73]

The graphs represent the time span of 10 minutes, starting at the beginning of the usage and including both "active" and "after" (post usage) period of each run. The distributions of the PNC as box plots under various experimental conditions are presented in Fig. 21.

A



B

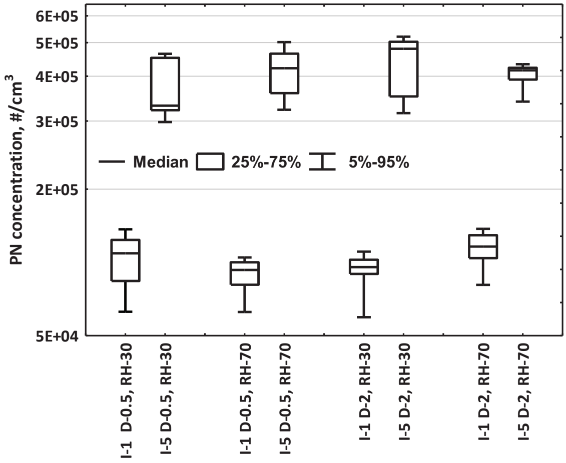


Figure 21. The effects on the distributions of particle number concentrations as a function of the number of varying intensity of the usage (number of users) during the "active" period (A) and the "after" period (B): I – for intensity (number of users), D – distance to the bystander (m), RH – relative humidity, % [73]

Distance

The usage of HTP at a close proximity (0.5 m) of a bystander resulted in a distinct pattern of aerosol variation, which included a rapid and significant increase in particle concentration by several orders of magnitude from the background and then dropping back at the same rate after several seconds (Fig. 20 (A)). The duration of the visible peaks was on average around 6 to 8 seconds, which corresponded to the duration of exhalation of the puff plus the time for the travel of aerosol to the sampler. Such pattern of temporal variation was evident only at a distance of 0.5 m from a bystander, where the bystander experienced direct puff into the breathing zone. Such pattern is similar to the one observed in the earlier study with electronic cigarettes [177].

The increase of distance from the bystander resulted in diminishing the visibility of peaks (puffs), which were not observable at a distance of 2 metres (with one user, ACH of 1 h^{-1} and RH of 30%). The distance factor resulted in a significantly higher mean PNC (4.2 times) during the active usage (9.7E+04 and 2.3E+04 for 0.5 and 2 m, respectively). At the same time, a longer distance had no significant effect on the PNC levels during the after the usage period (~370 sec. from the beginning of puffing), which were at comparable levels (7.0E+04 to 7.5E+04 for 0.5 and 2 m, respectively).

An evident decrease in PNC levels for 2 metres distance was possibly caused by several factors: a dispersion of particles in a room while travelling across, the deposition of aerosol particles on surrounding surfaces and partial evaporation of aerosol particles to gaseous phase compounds. The exposure of bystander was greatly influenced by the direction of the exhalation of the puff; however, for the longest tested distance, this factor had a minor impact.

Intensity

The number of parallel users of HTP (further referred to as the usage intensity) had a direct impact on the concentration of aerosol particles near the bystander and in the chamber. The PNC levels from five users were significantly higher compared with the HTP emission from one user (Fig. 20 (B)). The figure displays a comparison of users located at 2 metres from the bystander, while the ventilation and RH were at minimum (0.2 h^{-1} and 30%, respectively). The average PNC in case of five users was 27 times (mean of 2.0E+04 and 5.4E+05 #/cm^3) higher under “active” period and 8 times (mean of 5.9E+04 and 4.7E+05 #/cm^3) higher under “after” period. Such multiplied increase of the PNC generated by 5 users reflects a more rapid fill up of the chamber with the exhaled aerosol, thus a more effective transport of particles to the bystander. In case of one user, 2 m distance seems to be sufficiently long for the particles to disperse and not to reach the bystander, thus the concentration of PNC is naturally underestimated.

The usage intensity is as well a relatively ambiguous factor, since it depends on the topography of smoking/usage. It can vary with the inhaled puff duration, hold-in-body duration or exhaled puff duration. A quantitative influence of the usage topography may be attributed to a random error of the experiment, especially in case of the far distance from the bystander.

Relative Humidity

The temporal variations of PNC (from HTP) under two different relative humidity levels (30% vs. 70%) are presented in Fig. 20 (C). The figure reflects PNC variations in a case of a single user, ACH of 1 h^{-1} and 2 metres distance. The longer travel distance to the bystander was expected to provide sufficient time for the aerosol particles to react with the ambient moisture.

Having identical conditions at different levels of RH, the PNC levels under “active” period as well vary over the time in different patterns (Fig. 20(C)), indicating higher mean PNC levels (1.9 times, as from 7.2E+04 to 3.7E+04 for 30 and 70%, respectively) and fluctuations at 30% RH. This can be attributed to the fact that the particle deposition velocity increases with the relative air humidity [179]; therefore, at RH of 70%, it is not possible to identify the individual puffs that are clearly visible under RH of 30%. At the same time, the PNC levels under "after" period (which started from ~360 sec. and lasted until 1180 sec.) were 1.3 times higher at 70% RH and varied from 8.2E+04 to 1.1E+05 for 30 and 70%, respectively. The lower PNC levels at dry air conditions (30%) under "after" period could be observed due to faster aerosol particle evaporation at the relatively lower partial pressure of water vapour in the air.

Product Type: HTP vs. CC

The variation of PNC during the use of HTP was distinctly different as compared to the usage of CC under the identical conditions (intensity – 1 user, distance – 2 m, ACH – 0.5 h^{-1} and RH – 50%) (Fig. 20(D)). PNC was in the same range during 60 seconds from the start of the experiment. Then, at approx. 90 seconds from the start, PNC rapidly increased for CC, and it remained an order of magnitude higher compared to the HTP. The noise component of the time series was as well different, representing higher fluctuations in the case of CC, while HTP produced a relatively smooth curve. The coefficient of variation resulted in 0.5 to 0.82 during the "active" period for HTP and CC, respectively. During the entire "active" period, CC generated 5.6 times higher (2.8E+05 #/cm^3) concentration than HTP (4.9E+04 #/cm^3), while under "after" period, an even greater 8.5 times difference (8.0E+05 #/cm^3 vs. 9.5E+04 #/cm^3) was observed (Fig. 20(D)) inset as boxplots. Such patterns in aerosol concentration variations may be attributed to the volatility of particles. In case of HTP, the concentration does not rise similarly to the CC, since the particles evaporate (transfer from droplet to gaseous phase) faster than they accumulate in the air of the chamber.

The mainstream particles of the HTP have been shown as highly volatile [169], while exhaled aerosol is expected to have even lower content of non-volatile matter. This phenomena is similar to that registered with electronic cigarettes [43, 177]. At a closer distance, the real-time aerosol instrumentation is capable of registering peaks for both HTP and CC, but at a further distance, volatile particles may not reach the bystander, in case of the low usage intensity and low ambient humidity.

3.1.3 Particle Size Distribution

Generally, the particle size distribution (PSD) that was obtained during the usage of the tested HTP exhibited a unimodal distribution, having a mode at the smallest particle diameter of ELPI+. The shape of the distribution was comparable to both HTP and CC (Fig. 22). The main difference resulted from the overall particle levels with the CC producing order of magnitude higher particle concentrations across all measured size bins. Moreover, the CC as well produced higher amount of particles in the range 0.38–0.60 m, whilst in the case of HTP, no particles were registered during the period of active usage or the post-usage period.

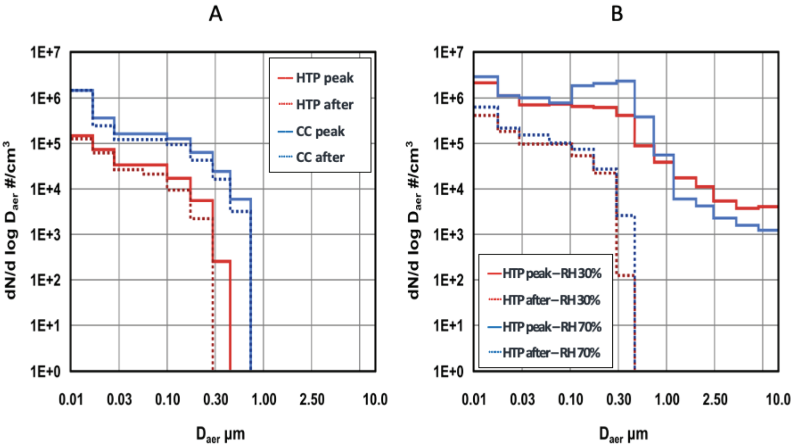


Figure 22. Particle size distribution (PSD) of indoor aerosol during the use of HTtp: (A) a comparison with a conventional cigarette (distance to the bystander 2 m, number of users 1, RH 50%, ventilation 0.5 ACH), (B) effect of the relative humidity in the room to the PSD (distance to the bystander 0.5 m, number of users 5, ventilation 0.5 ACH) [73]

The PSD of the aerosol was as well comparatively similar during the peak of the concentration, except to that in a channel 0.25–0.38 m, reflecting the presence of these "larger" particles in the directly exhaled aerosol, but not present later, possibly due to the high volatility of these particles. Figure 22 (A) presents the data on 2 m distance from the bystander to a user; thus, the particles have sufficient time to undergo transformations (evaporation, agglomeration, settling) while travelling across the room.

The PSDs obtained during this experiment are in line with that obtained during the earlier experiments with the usage of electronic cigarettes, with respect to a mode at 100–300 nm [180]. At the same time, a high concentration of nucleation mode particles is apparent. It was expected that ELPI+ is capable of registering particles, which rapidly evaporate from the accumulation mode to the nucleation mode, considering the high volatility of these particles (i.e., droplets).

Figure 22 (B) presents a specific case of PSDs as affected by a relative humidity (30% vs. 70%). These measurements were taken at the extreme conditions of five simultaneous users and close distance to the bystander (0.5 m). This resulted in a concentration that is an order of magnitude higher during the peak, when the direct exhalation of users had a substantial effect at a close distance. There, the larger particles are present as well. At the same time, the increase in RH resulted in a clearer mode at 0.10–0.38 m, representing the accumulation mode that is resulting in the growth of particles (or delayed shrinkage) due to the ambient humidity.

3.1.4 Screening the Factors That Are Affecting the Variation of Pollution Concentration

The experimental data were fitted to the regression model presented in (A5), thus providing information on the relationship and the significance of process factors to the various pollution estimates. Table 5 depicts the qualitative estimates on important factors affecting pollutant concentrations. Generally, the usage intensity (number of simultaneous users) prevailed as the most important factor that is positively affecting the pollutant variations. This was valid for the pollutants that have been associated with the usage of HTP [4, 68], including particles, nicotine and acetaldehyde, while 3-EP and formaldehyde were not impacted. During the “active” period, the difference between one and five users reached up to one order of magnitude, while in the “after” period, the difference was up to 4 times higher (Fig. 21). At the same time, the maximum particle mass concentration did not depend on the number of users, since a single user exhaling directly to the bystander may have caused higher variation than five users located further, as confirmed by the negative influence of the distance factor. Nicotine and acetaldehyde are among the gaseous pollutants, resulting from the usage of the HTP. At the same time, formaldehyde and 3-EP were not affected. The positive relationship of CO_2 and the number of users may be likely associated with an increase of exhaled CO_2 as related to the increased presence of humans in the room.

Table 5. Significance of the process factors to the responses (pollutant concentrations) during the usage of HTP in a chamber based on the interaction regression model: ++ denotes the most important positive factor (highest value among the obtained ones), + less important positive factor, - a negative factor, -- the most important negative factor, 0 – factor that is not statistically significant [73]

	PNC Mean (All)	**PNC Mean (After)**	**$PM_{2.5}$ Mean (All)**	**$PM_{2.5}$ Mean (After)**	**$PM_{2.5}$ Max**	**Nicotine**	**3-EP**	**Form-aldehyde***	**Acet-aldehyde**	**CO_2**
Air Change Rate (ACH)	-	-	0	0	0	0	0	0	0	0
Number of Simultaneous Users (Int)	++	++	+	++	0	++	0	0	++	+
Relative Humidity (RH)	0	0	0	+	0	0	++	+	+	0
Distance to the Bystander (Dist)	0	+	- -	0	-	-	0	0	0	0
ACH*Int	-	-	0	0	0	0	0	0	0	0
ACH*RH	0	0	0	0	0	+	+	0	0	0
ACH*Dist	0	0	0	0	0	+	+	0	0	0
Int*RH	0	0	+	+	0	0	0	0	0	0
Int*Dist	+	+	0	0	0	0	0	0	0	0
RH*Dist	+	0	0	0	0	0	+	0	0	0

* Low validity and low percent of the variation of response, according to the cross validation

The distance to a bystander was an important factor in case of $PM_{2.5}$ mean values: the further is the user, the lower is the concentration. At the same time, the increasing of distance resulted in higher PNC values in the "after" period, indicating the chamber-related dispersion patterns of fine aerosol, resulting in a more efficient transfer of small particles from the further distance after the active usage was over.

The combined effect of these two factors on the pollutant concentrations is presented in Figure 23. $PM_{2.5}$ concentration was directly affected by both distance and usage intensity. For example, a single user at a close distance to the bystander may generate an average concentration of ca. 200 $\mu g/m^3$, while at 2 m distance, the concentration is reduced more than four times to < 50 $\mu g/m^3$. However, five users at a further distance may increase the concentration twice (up to 100 $\mu g/m^3$), while at a closer distance, the increase may be eight-fold. A similar effect was applied to nicotine, where at a far distance, one user resulted in 2 $\mu g/m^3$, while five users at a close distance increased the potential exposure by 5 times. The PNC during the "after" period and acetaldehyde depended only on the number of users with potential exposure, increasing 2–3 fold, having one to five users in a room.

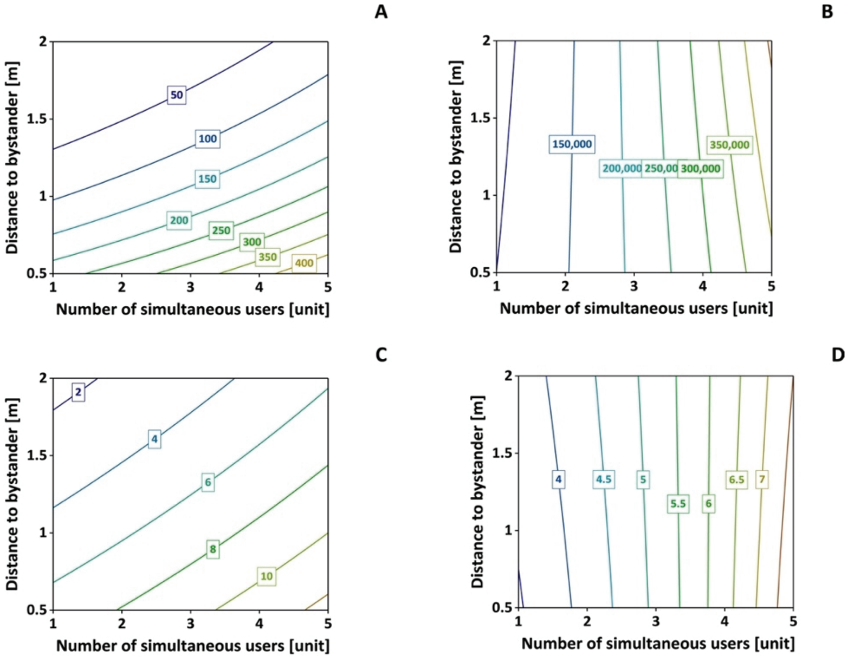


Figure 23. The joint effects of the number of simultaneous users and the distance to the bystander on pollutant concentrations at the scenario V = 0.2; RH = 30: (A) mean $PM_{2.5}$ concentration, μg/m^3, (B) mean PNC during "after" period, #/cm^3, (C) nicotine, μg/m^3, (D) acetaldehyde, μg/m^3 [73]

The air change rate appeared to be a not significant factor in the ranges where it operated (up to 1 ACH). Apparently, such ventilation intensity was not able to efficiently remove rather intensive emission of pollutants. The PNC was more sensitive to the changes in the ACH, since it is mainly determined by the presence of sub-micrometre particles, which efficiently travel with the ventilation air.

The relative humidity has significantly positively impacted $PM_{2.5}$ concentration variation in the "after" period. Increasing humidity resulted in higher vapour pressure around the exhaled volatile droplet particles, thus sustaining their existence and preventing rapid evaporation and shifting particle size to the accumulation range (thus increasing the particle mass), as confirmed by the increased mode of PSD in Fig 23 (B). At the same time, the effects of relative humidity to formaldehyde and acetaldehyde may be related to the measurement artefacts. The RH has been indicated as an important factor while sampling aldehydes by 2,4-DNPH impregnated silica cartridges [181]. While dry conditions prevent the efficient uptake of aldehydes, the increased humidity potentially results in the increased sorption on highly hydrophilic silica.

The interaction between the process parameters has not revealed a dramatic synergistic or antagonistic effect in the used setup. The explanation of the observed significant interactions is difficult to explain mechanistically. The positive effect of Intensity*Relative humidity to $PM_{2.5}$ concentration variations was apparently caused by the growth of particles at the increasing concentration. Intensity*Distance positively affected the submicrometric particle number variations, related to the particle transport and dispersion within the chamber.

3.1.5 Sub-Chapter summary

During the HTP use in these experiments, PM > 1 μm was found during the exhalation at close distance (with five simultaneous users in total). The distinct sawtooth pattern was observed in the online trace of PM when HTP was consumed at a distance of 0.5 m, and as the distance got longer, the intensity decreased, or the absence of peaks was observed. The “hidden” peaks in the online PM trace were as well noticed with an increase in the ambient humidity alongside the correlated increase in the measured aerodynamic diameter, which was likely related to the particle growth. Moreover, dry air conditions (30% RH) were a factor leading to a drop in the PNC after the HTP consumption. All these observations can be explained by the high volatility of the liquid droplets that were generated by the HTP use, their dispersion in approximately 6–8 s, and eventually, their lower probability of reaching the bystander at distances that exceeded 0.5 m at the low usage intensity and ambient humidity.

Quantitative experimental variables included HTP use intensity as a number of parallel users (1, 3 or 5), distance to the bystander (0.5, 1 or 2 m) as well as environmental conditions in a chamber: ventilation intensity as the air changes per hour (0.2, 0.5 or 1 h^{-1}) and relative humidity (RH, 30, 50 or 70%). The real-time particle number (PNC), CO and CO_2 concentrations as well as off-line acetaldehyde, formaldehyde, nicotine and 3-ethenylpyridine concentrations were measured during and after the active usage. The use of HTP resulted in a statistically significant increase of several analytes, including nicotine, acetaldehyde, $PM_{2.5}$ and the PNC, as compared to the background. The obtained levels were significantly lower (approximately 16, 8, 8 and 28 times for nicotine, acetaldehyde, PNC and $PM_{2.5}$, respectively) compared to the levels resulting from the conventional cigarette (CC) smoking under identical conditions. The maximum 30 min. concentration of the PNC (4.8E+05 #/cm^3) as well as the maximum concentration of the PNC (9.3E+06 #/cm^3) suggest that the intensive use of HTP in a confined space with limited ventilation might cause substantially elevated aerosol concentrations, although these particles appeared as highly volatile ones and evaporated within seconds. The concentrations of airborne nicotine and acetaldehyde showed a slight upward trend with an increasing number of simultaneous users; with regard to nicotine levels, the distance between the user and a bystander as well played a role in a significant drop in the indoor concentrations of nicotine with the increasing distance.

3.2 Comparison of Exhaled Aerosol, Following the Usage of HTP, EC and CC

3.2.1 Temporal Variations of Particle Number Concentrations During and After the Product Usage

The usage of tobacco products in the enclosed environment usually results in a very characteristic variation of particle concentration, which may be characterized by a sharp increase of PNC, following the exhalation of inhaled mainstream aerosol, and then an equally sharp decrease in the concentration, almost to a background, as it is quantitatively presented to be lower in this sub-chapter. Such pattern of temporal variability is valid in case of both conventional cigarette and electronic or heated tobacco products [73, 177]. Mostly depending on the distance from the user to a bystander, the sharpness of the peaks may be more or less pronounced.

There has been observed an expected variation of PNC in a bystander's position with clearly identifiable five peaks at a close proximity (0.5 m, Figure 24). The PNC variations of all devices and regimes are presented in same scale in Figure 25 deliberately to reflect the magnitude of variations. In case a user is close to the bystander, the puff does not have sufficient time to disperse in the volume of the chamber. At further distances, the exhaled puffs can still be registered, but the level of concentration is much lower, and the peaks are not as sharp.

At a close proximity, the EC produced a higher number of particles, compared to the HTP operating in both eco and standard mode. Across all runs, the HTP in Eco regime resulted in $PNC_{t,max}$ at 0.5 m distance, averaged at 1.14E+06 ± 3.91E+05 $\#/cm^3$, HTP in standard mode, 1.66E+06 ± 4.89E+05 $\#/cm^3$, while EC resulted in 4.26E+06 ± 1.05E+06 $\#/cm^3$, i.e., 2.6 and 3.7 times higher than HTP, which is a statistically significant difference ($p < 0.05$). Similar trend holds for the distances of 1 and 2 m with $PNC_{t,max}$ generated by the EC were 4.0–5.0 and 1.0–8.2 times higher than HTP, respectively. Such difference is determined by a more efficient aerosol generation within the electronic cigarette, which results in a higher mainstream aerosol concentration (10E+9 $\#/cm^3$ for EC [182]) vs. <10E+8 $\#/cm^3$ for HTP [169], and apparently, exhaled aerosol concentration.

HTP operating regime (eco vs. standard) did not yield significant differences in terms of the temporal variation of exhaled aerosol particles and the concentration levels and was within 95% confidence interval in all distances on average across all regimes. This may be due to the fact that other environmental factors play a more important role in the aerosol variation. Generally, a different temperature of heating may be expected to result in a substantially different mainstream aerosol generated by a device, but these differences are reduced once the aerosol is transformed within the human respiratory system. In several cases, such as at a closest proximity to the bystander, a significant difference between the product regimes emerged in terms of the indexes, representing the exhalation moment ($PNC_{t,max}$, $PNC_{t,5s}$) at a higher ventilation rate (1 h^{-1}). This shows that the standard regime may result in a higher aerosol concentration immediately after the exhalation.

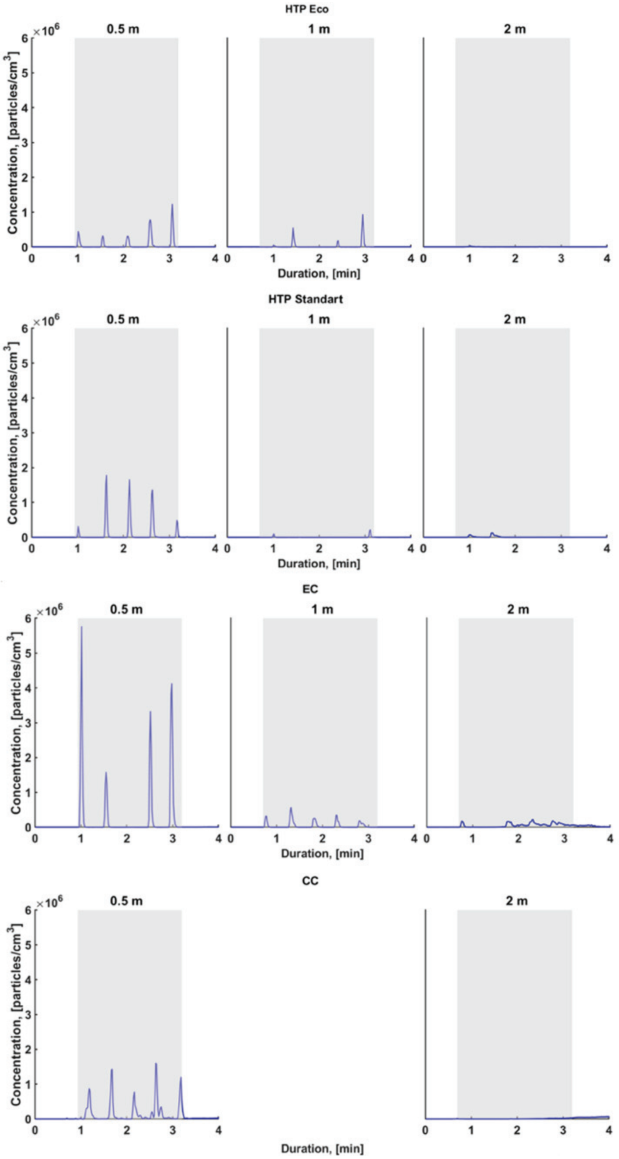


Figure 24. Temporal variation of total particle number concentration following the usage of tested HTP, EC and CC (the latter data is presented from Martuzevicius et al. [177]) products at various distances from the user to a bystander at the following environmental conditions: ventilation intensity 1 h^{-1}, relative humidity 30% [27]

The obtained concentration variation pattern and levels during the exhaled puff in this study are comparable to those obtained during the earlier studies with similar/other devices. For example, the vaping of Puritane (an early generation electronic cigarette device) resulted in peak concentrations from 1.2E+06 to 2.8E+06 #/cm^3 at 0.5 m distance [177], while using tobacco heating system IQOS yielded on average 0.5E+06–1.6E+06 #/cm^3, reaching 9.3E+06 #/cm^3 [73]. Conventional cigarette has resulted in a broader range of peak concentrations, ranging from 1.27E+06 to 2.62E+08 #/cm^3 at 0.5 m distance [177].

The peak concentration ($PNC_{t,max}$) is an important estimate of short term exposure to the particulate matter during the usage of either EC or HTP. However, these peaks are very short lived (up to 5 sec), and it is important to define the parameters, which reflect the aerosol behaviour after/between the peaks. The particles generated during the vaping of ECs (as opposed to those of conventional cigarettes) have a very volatile composition, namely, consisting of mostly water and propylene glycol/glycerol vapour, thus quickly dispersing in a room as well as evaporating [43, 73, 158, 169]. It has been hypothesized that after the last puff, the total PNC (PNC_t) should remain at the background level and not increase once the room volume is filled with exhaled particles, while the ventilation is not capable of removing those during the period of two minutes.

The characteristic behaviour of PNC_t following the last puff is presented in Figure 25. Once dropping to 10E+3 #/cm^3 level, it remains constant, such as in the case of HTP or slightly rises to 1E+4 #/cm^3 level, as in a case of EC. Generally, the EC resulted in higher $PNC_{t,120s}$ across all distances. This is somewhat unexpected, since the EC particles may have higher volatility due to being fully generated from a liquid, while HTP are tobacco-derived and thus may have a non-volatile core (due to the presence of aerosol fraction, referred to as nicotine-free dry particulate matter, NFDPM [183]). This phenomenon was verified by calculating the ratios of $PNC_{t,5s}$, $PNC_{t,10s}$ and $PNC_{t,120s}$ in comparison to the peak $PNC_{t,max}$, thus normalizing the residual concentration in comparison to the maximum. Such comparison for each tested product is presented in Figure 26 (A) as factor plots. While the large variation among multiple runs and conditions resulted in statistically insignificant differences, the averaged values indicate that the EC actually ends up in a lower concentration at both 5, 10 and 120 s after a puff peak; namely, the residual concentration in case of EC after 120 s comprises ca 0.5% of the $PNC_{t,max}$, while in case of HTP, this ratio is up to 1.5–2.5%.

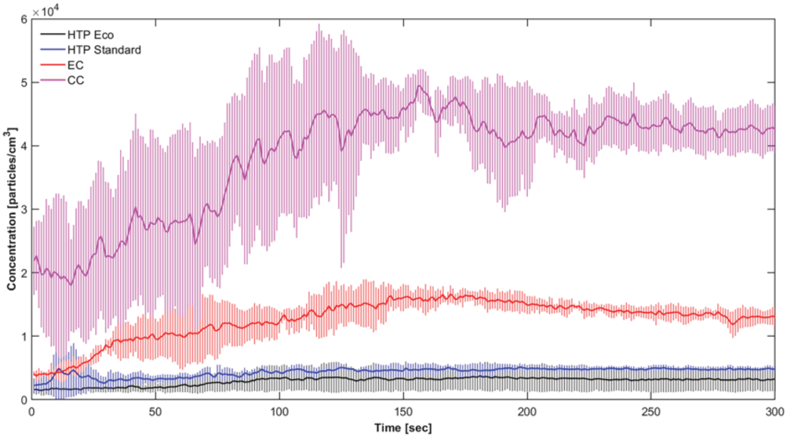
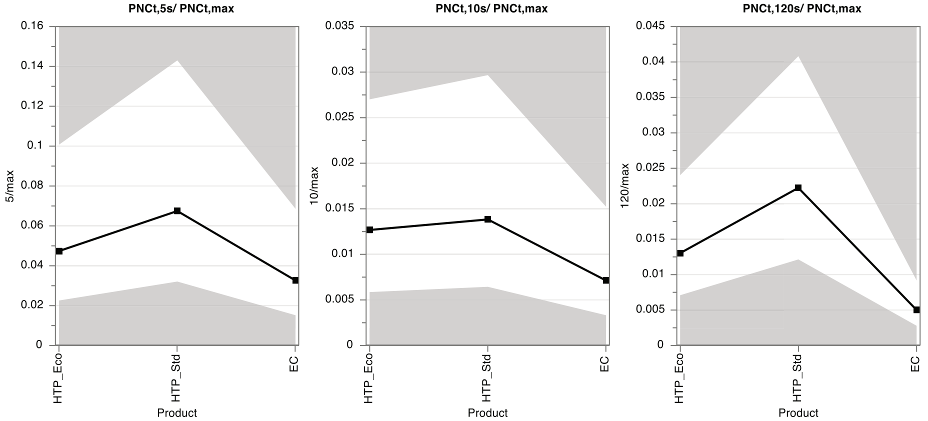


Figure 25. Temporal variation of PNC at the bystander position, starting at 20 seconds after the usage of HTP, EC (the average of three volunteers) and CC (the latter data is presented from Martuzevicius et al., 2019); the distance between the vaper and the bystander = 0.5 m, ventilation intensity = 1 h^{-1}, relative humidity 30% [27]

A)



B)

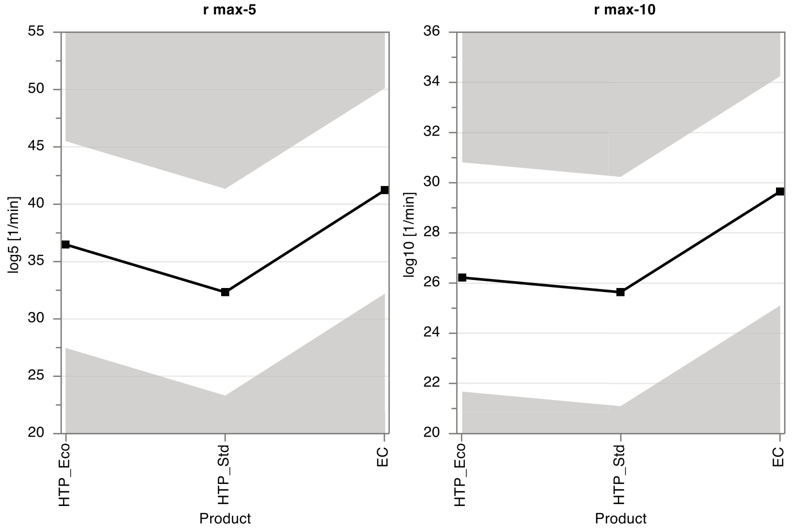


Figure 26. Temporal variation of PNC following the exhalation after the usage of tested products: (A) concentration after 5, 10 and 120 s in relation to the maximum, (B) concentration decrease rate from the maximum to 5 and 10 s [27]

Additionally, the concentration decrease (decay) rate following the peak during first 5 and 10 seconds has been calculated (Figure 26 (B)). Again, the EC resulted in a more rapid concentration decay (41.2±9.3 min^{-1} after 5 s and 29.7±4.7 min^{-1} after 10 s). This confirms that while the peak concentration of EC is higher than HTP, the particle removal/decay is as well faster, owing to a higher volatility of particles (droplets).

It must be emphasized that the processes following the exhalation of highly concentrated aerosol are very complex and go beyond evaporation only. The aerosol removal mechanisms include the discharge with ventilation air and gravitational settling. At the same time, the particle aggregation (conjugation of similarly sized particles) and coagulation (sticking to larger droplets) may occur. Furthermore, the particle removal as well as formation may take place, such as secondary organic aerosol, considering a relatively high emission of volatile organic species. The author of the dissertation does not anticipate strong contribution of the latter mechanism in a chamber environment due to the low concentration of ozone in supply air and the lack of UV radiation, but these processes may be more pronounced in real world indoor environments.

3.2.2 Particle Size Distributions

Particle size distribution (PSD) is a very important parameter in case of experimenting with highly dynamic sources of potential air pollution, such as vaping or using heated tobacco products. The particles that were generated during the vaping of electronic cigarettes (as opposed to those of conventional cigarettes) have a very volatile composition, namely, consisting of mostly water, propane-1,2-diol (propylene glycol) and propane-1,2,3-triol (glycerol) vapour, thus quickly dispersing in a room and evaporating. This results in rapid variations of the total concentrations of particles as well as the types of distributions. The mainstream aerosol generated by such devices has been reported as having tri-modal with a primary mode at 250 nm and two secondary modes at approximately 30–80 nm and 1 μm [184]. In exhaled aerosol PSD, the mode shifts towards 100 nm [169, 185]. At the same time, the rapid evaporation of these volatile particles results in the appearance of a nucleation mode at 2–10 nm, which indicates the transition of aerosol particles to the gas phase [143]. The shift from the accumulation mode to the nucleation mode of exhaled aerosol may be observed during highly time resolved measurements in case of both the electronic cigarette [177] and the heated tobacco product [73].

Throughout the study, it has been observed a broad variety of PSDs. A bimodal distribution was prevalent for both devices with the main mode usually located at approx. 60–80 nm for HTP and 115 nm for EC during the puff. At the same time, a secondary mode was apparent at the first channel of FMPS; however, most of the time, it was much lower. There was as well a substantial variation in PSD shape and mode allocations among various time periods, distances to the bystander and volunteers. Interestingly, the PSD changed many times among the puffs by the same volunteer, indicating that the transformations of an inhaled mainstream aerosol may compose an important factor that is worth exploring further.

Figure 27 presents a case of PSD at a close distance to the bystander during the puff and 5, 10 and 120 s after the last puff (average and standard deviation among five puffs and three volunteers). The variation among the volunteers was rather high and reflected by broad intervals of two standard deviations. This is especially noticeable in the case of HTP operating in standard mode, where the lower bound of error bar reached zero and could not be displayed in logarithmic axis in case of 5 and 10 seconds after a puff. The distributions of this case emerged as bimodal during the puff. The main mode was 93 nm for HTP in both eco and standard regimes, and 123 nm for EC. The latter as well represented higher concentration of particles compared to the HTP. Another mode was evident for all devices at 6 nm. After 5 seconds from the puff, the concentration got reduced by several orders of magnitude, while the main modes have shifted towards smaller particle sizes (52 nm in eco, 81 nm in standard, 107 nm for EC). In fact, 10 seconds after a puff, the main mode further shifts to smaller particle size, while the amount of larger particles (400–500 nm) dropped almost to zero. The relative amount of the smallest particles decreased as well, compared to the peak or 5 seconds after.

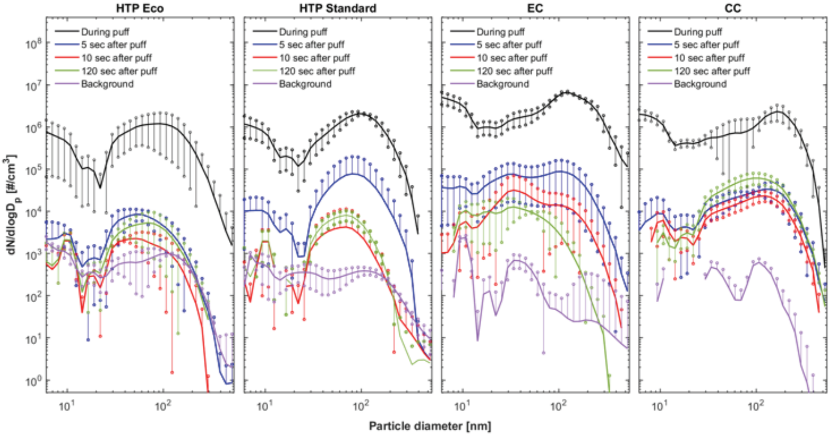


Figure 27. Characteristic particle size distributions (PSD) of ambient aerosol following the exhalation/puff after the usage of tested HTP, EC and CC (the latter data presented from [177]) during the puff, and 5, 10, 120 sec after the puff; the distance between the vaper and a bystander = 0.5 m, ventilation intensity = 1 h^{-1}, relative humidity 30%; the average and standard deviations of the three volunteers are presented [27]

The previously presented PSD patterns for HTP and EC devices were different from the conventional cigarette in the following aspects: a) the accumulation mode of PNC during the puff was larger for CC (165–200 nm, as opposed to 93 nm for the HTP and 123 nm for the EC); b) this mode has barely shifted to a smaller size range with time, 100–200 nm range as compared to the reduced modes (52–81 nm for HTP, 107 nm for EC). This confirms that the composition and thus the volatility of the exhaled aerosol particles are significantly different in cases of HTP and EC as opposed to the CC.

The phenomenon of volatile particle transformations after exhalation is yet to be researched systematically. It is obvious that the transformation of exhaled particle follows a different mechanism as opposed to the inhaled mainstream aerosol. The mainstream aerosol of HTP has been estimated to contain 75–85% of water [186], while EC contains 73–83% of glycerol [162]. This is much different from the traditional cigarette mainstream emissions, which only contain 17–27% of water.

The high temperature mainstream aerosol particles first cool and then hygroscopically grow in size during the inhalation in water-saturated respiratory airways. In such environment, the CC particle growth is slower compared to the EC [166], and presumably, to the HTP. Then, the cooled particles and enlarged particles, which do not get adsorbed on the airway tissue, start shrinking in size already in the airways and then continue evaporating in a relatively dry indoor air (compared to the airway environment). Thus, a very fast evaporation of EC and HTP particles is observed, which were indicated to have a half-life of 11 s (for EC) as opposed to 19–20 min. of CC [158]. While CC particles lose a relatively small amount of adsorbed water, leaving a low volatility core of primary particles, the EC and HTP particles lose both the adsorbed water and volatile constituents; thus, the observed evaporation is much faster and shorter.

3.2.3 Factors Affecting the Dynamics of Aerosol

The presented data in this manuscript has been obtained from full factorial experimental design, allowing the assessment of statistical significance of the process factors. The data presented in the previous sub-chapters have already indicated some trends that were quantitatively confirmed by the model. Generally, the model suffered from relatively low statistical power (R^2 ranging from 0.07 M_{5s} to 0.64 for $PNC_{t,max}$), yet statistically significant, based on Q^2 value. Such outcome was partially unexpected, since the experiments have been performed under strictly controlled conditions. The largest uncertainty has been brought with three volunteers, having different topographies for the product usage.

The significant experimental variables are listed in Table 6. It is apparent that the exhaled aerosol from the two tested products, i.e., HTP and EC, have significant differences in terms of variation of particle concentration and diameter, depending on the conditions. The effect of these variables was usually the opposite, such as in case of $PNC_{t,max}$, the HTP_Eco and HTP_Std coefficient values were negative, while EC values were positive, indicating that the latter contributed to the larger values of PNC. In case of M_{120s}, this trend was the opposite, and HTP resulted in positive coefficients, while EC in negative. This agrees well with the observations in the previous sub-chapters, where the presented data show that the EC results in higher PNC, but the particle shrinkage is faster; however, the decay rate (r) was not significant for various products. Although these products can be distinguished statistically in a controlled experiment under chamber conditions, the real-world environment may be more challenging, if at all possible.

Table 6. Significant process variables: * denotes a statistically significant model term, $p < 0.05$ (specific values are listed in the parenthesis), 0 denotes terms having $p > 0.05$

Response	**Experimental variables**					
	Product	**Distance**	**Ventilation**	**RH**	**Volunteer**	**Interaction terms**
$PNC_{t,max}$	*	*	0	0	* (v1)	0
$PNC_{t,5s}$	*	0	0	0	* (v3)	HTP_Eco*Vent, EC*Vent
$PNC_{t,10s}$	*	*	0	0	0	EC*Vent
$PNC_{t,120s}$	* (HTP_Eco, EC)	0	0	0	* (v1)	0
$PNC_{t,5s}$/ $PNC_{t,max}$	0	*	0	0	0	EC*Vent.
$PNC_{t,10s}$/ $PNC_{t,max}$	0	*	0	0	0	0

PNC$_{t,120s}$/ PNC$_{t,max}$	* (HTP_Std, EC)	*	0	0	*(v1, v3)	EC*Dist.
r$_{max-5}$	0	*	0	0		EC*Vent.
r$_{max-10}$	0	*	0	0	0	0
PNC$_{6nm,5s}$	* (HTP_Eco, EC)	*	0	0	* (v1)	HTP_Eco* v1
PNC$_{6nm,10s}$	* (EC)	0	0	0	*(v1, v3)	0
PNC$_{6nm,120s}$	0	0	0	0	*	HTP_Eco* v2
M$_{max}$	* (HTP_Std, EC)	*	0	0	0	HTP_Std* Dist.
M$_{5s}$	0	0	0	0	0	0
M$_{10s}$	0	*	0	0	*(v1, v3)	EC*v3
M$_{120s}$	*	0	0	0	0	Dist.*RH

The distance from a bystander is another important parameter affecting both potential exposure level and the particle size. This is especially evident in a case of puff (peak concentration) or immediately afterwards. After 120 s, the distance has less effect, since due to a substantial time of aerosol travel and dispersion within the room, the transformation processes managed to reach more or less stationary conditions.

The ventilation intensity and relative humidity (RH) did not yield significant contributions to the model, thus rejecting the hypotheses about their importance. The limited impact of ventilation has been demonstrated in the earlier chamber studies, researching the EC and HTP products [73, 177]. This is due to the fact that the source intensity of exhaled aerosol and the transformations of aerosol are substantially faster and overwhelming processes, compared to a relatively slow process of air replacement up to 1 change per hour. The effects of RH on particle condensation/evaporation processes are important [166, 177], but apparently, the rapidness of particle dispersion and transformations overshadows the potential impacts of RH. A significant positive effect of RH has been registered when considering the mode of particles after 120 s in interaction with the distance. Possibly, this time span was sufficient to observe the impact of RH on the condensation of ambient water vapour particle size growth after the exhalation.

The effect of volunteer, i.e., the topography of usage, is the most ambiguous factor in this experiment. Only volunteers 1 or 3 have been associated with the significant effect to the particle variation. Moreover, this effect has been the opposite: whenever both V1 and V3 were significant, V1 was positive and V3 was negative (such as in PNC$_{6nm,10s}$, M$_{10s}$) or vice versa (PNC$_{t,120s}$/PNC$_{t,max}$). It is difficult to interpret such findings based on the actual processes during the usage. While the volunteers have been instructed about the purpose of the experiment and were given general instructions about the product usage regime, it seems that a large variation occurred during the course of experiment, thus obstructing the clear effect of the volunteer on the process as well as bringing in much variation in the entire data set and diminishing the associations that would allow establishing stronger models. At the same time, this illustrates the fact that the usage of HTP or EC may be largely subjected to the usage topography, and the variation among the users may hinder, underlying the differences between products.

Several plots as predicted from the model, depicting the main quantitative associations between the aerosol parameters, such as $PNC_{t,max}$, $PNC_{t,120s}$, M_{max} and M_{120s}; the distance to the bystander, ventilation intensity and tested products are presented in supplementary material (A 8.). Even if the models lack statistical strength (especially with the ventilation intensity variable), they provide insights on the quantitative associations between the variables in this particular setting. The case of $PNC_{t,max}$ demonstrates the limited effect of ventilation intensity and establishes the relationships between the distance to the bystander and $PNC_{t,max}$. The surface plots confirm the observations that have been presented in the previous sub-chapters on a higher overall concentration levels while using EC, when these levels vary from 0.5E+06 #/cm^3 at 2 m distance and gradually increase to 6E+06 #/cm^3 at a 0.5 m distance. HTPs follows the same variation trend but with lower levels of PNC. In case of $PNC_{t,120s}$, the distance and ventilation seems to have an equal role in the variation of concentration. Although the ventilation variable lacks statistical significance, this indicates that during longer time spans, the ventilation may play an important role in reducing the concentration levels. These two engineering measures may be coupled together in managing potential exposure to the bystanders if these products are consumed in a closed environment. For example, these measures at the maximum settings are capable of reducing PNC levels by 20%, while each of them contributes equally by 10%.

A similar trend of both factors' influence is valid when predicting the prevailing particle size distribution after 120 s. Increasing the ventilation and distance from the bystander results in shrinking the particles as an outcome of evaporation of volatile substances. Within the tested boundaries, it was not possible to register the accumulation of HTP particles in a room, when the evaporation diminishes and the concentrations starts increasing, considering that some exhaled HTP particles have a non-volatile core. It may take more users to observe this effect [73]. For the reported conditions, it has been concluded that the dynamics of these particles follow rather similar patterns to the exhaled EC particles.

3.2.4 Sub-Chapter Summary

The comparison of the aerosol dynamics following the exhalation after using heated tobacco product (HTP) and vaping product (electronic cigarette – EC) indicates both the similarities and differences between the two products, and both are very distinct from the conventional cigarettes.

In this sub-chapter, the dynamics of the generated exhaled aerosols following the use of a new HTP (branded as "Pulze", operating in eco and standard modes) and EC ("myblu") in a chamber environment by three volunteers, controlling the distance to the bystander, the ventilation intensity and microclimate were assessed. The HTP and EC data was compared with the CC data. HTP generally resulted in lower aerosol number concentration during the puffs, reaching 1.66E+06 #/cm^3 at 0.5 m from a bystander, compared to EC (averaging 4.3E+06 #/cm^3) and CC (1.47E+08 #/cm^3). No significant difference was observed between the "eco" and "standard" modes of HTP. At the same time, the EC concentration decrease after the puffs was as well faster, indicating the higher volatility of particles. The EC as well featured higher mode during the puff (120 nm) compared to the HTP (90 nm), which was significantly different from the conventional cigarette (165–200 nm). The evaporation/shrinkage of particles has been observed within 10 sec after the puff with the HTP and EC. The distance to the bystander was shown to be as a significant factor, affecting the aerosol dynamics; however, the ventilation intensity and relative humidity did not have a statistically significant effect. As discussed throughout this sub-chapter, the aerosols generated by a heating process, as in a case of the HTP or ECs, have different physicochemical properties than smoke, which is a result of biomass combustion.

3.3 Evaluation of Generated Pollution in the Real-Life Environments

3.3.1 Time Resolved Variation of Aerosol Concentrations

Highly time resolved aerosol concentration has been measured across the entire experiment, including both the controlled usage of IQOS (Campaign #1) as well as the uncontrolled operation of the club (Campaign #2). The time series of PNC during the two measurement campaigns are shown in Fig. 28.

One of the main tasks was to measure the impact of the HTP use on the air quality in a nightclub during the non-operating hours. Since the specific HTP brand, IQOS, was used in the experiment, this brand name will be mentioned instead of the general abbreviation HTP. The controlled experiment (Campaign #1) started with the background, having the median particle concentration of 9.4E+3 #/cm^3 and 9.8E+3 #/cm^3 in Zones 1 and 2, respectively. This is a moderate concentration of particles that is observable in many general indoor living environments, usually ranging within 1E+3 and 9E+4 #/cm^3 [187]. This background was measured in the premises where no continuous activities were present after several days of workweek closure before the opening at the weekend. The entry of 10 volunteers did not affect the background concentration, with the median concentrations remaining at 9.2E+3 #/cm^3 and 9.9E+3 #/cm^3 in Zones 1 and 2, respectively. Moreover, no major variation of particle concentration has been registered. The human movement may have resulted in resuspension of dust, but such super-micrometer particles were not reflected well in the total particle concentration variation, possibly due to the losses in the sampling line transport.

Once the 10 volunteers started using IQOS devices, the concentration of particles has increased, as reflected by the first peak (Fig. 28(A)). The maximum has been registered at 1.2E+5 #/cm^3, while the median concentrations were at 3.6E+4 #/cm^3 and 3.5E+4 #/cm^3 in Zones 1 and 2, respectively. This is a statistically significant increase ($p < 0.05$). Such level of particles is comparable to the simulation measurements in a room-scaled chamber with 1 volunteer and the ventilation rate of 0.5 [73]. At the same time, this is a relatively low-level compared to the other hospitality environments, even with no smoking areas. In Germany, very high median PNCs of 2.2E+5 #/cm^3 were measured during 4 main visiting hours in the non-smoking areas in four cafés/restaurants, 1.1E+5 #/cm^3 in two bars and 2.9E+5 #/cm^3 in seven discos [188].

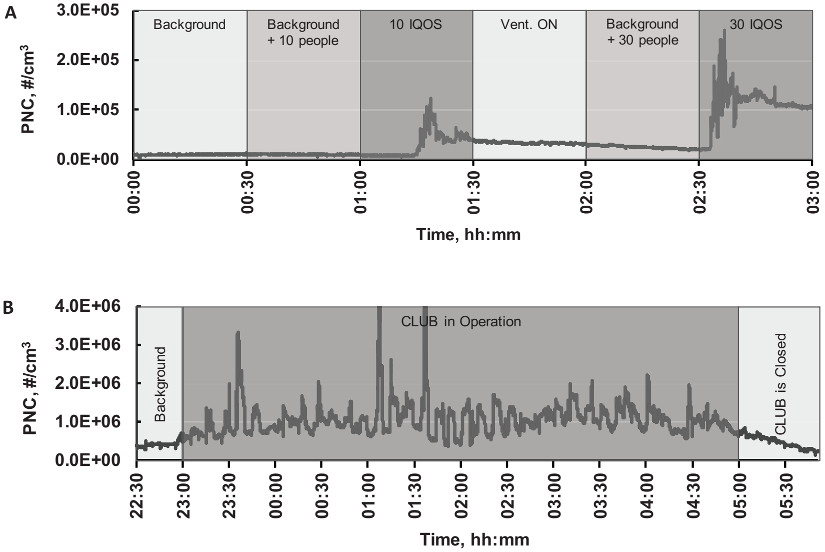


Figure 28. Variations of PNC (#/cm^3) during the controlled use of simultaneous 10 and 30 IQOS devices (A) and during the uncontrolled operation of the club (B) [144]

Once the usage of 10 IQOSs stopped, the concentration started decaying due to the natural ventilation and further due to the purging with forced ventilation to a level of 4E+4 #/cm^3 (CO_2 level between 460–480 ppm), which served as a background concentration for 30 volunteers that were present in the premises.

Thirty IQOS users resulted in another significant increase of PNC to the maximum value of 1.5E+5 #/cm^3 and median of 1.2E+5 #/cm^3 in Zone 1 and 1.3E+5 #/cm^3 in Zone 2. Such levels are comparable to the one registered in a chamber with 5 volunteers using IQOS [73] and in the higher range of hospitality environments, such as German restaurants, pubs and discotheques, ranging from 1.2E+5 #/cm^3 to 2.1E+5 #/cm^3 [188]. The concentration has not decreased quickly to the background. This did not agree with the obtained findings from the simulations in a chamber, where was observed a rapid decay of concentrations to the background levels, and it was attributed to the short life span of the volatile particles. Such difference from the chamber findings is probably due to a much more intensive emission, having 30 simultaneous users, as well as lower ventilation, having air handling unit off (air exchange rate approx. 0.5 ACH) during the test.

The preparations for the club opening (Campaign #2) already resulted in high background values (2.7E+5 #/cm^3 in Zone 1 and 2.9E+5 #/cm^3 in Zone 2, Fig. 28(B)). Yet, the club operation resulted in the highest PNC (max.: 1.7E+7 #/cm^3; median:

7.3E+5 #/cm^3 in Zone 1 and max.: 6.7E+6 #/cm^3; median: 8.0E+5 #/cm^3 in Zone 2), which is statistically significantly higher compared to the both backgrounds of the same day, but more importantly, all the scenarios involving IQOS usage. Such levels and variations are the results of the local particle sources, among most important were the operation of the artificial fog machine as well as fugitive emissions from the adjacent smoking room, as confirmed further by the concentrations of CC markers. The particle levels were generally high, compared to those registered in a chamber during the use of CC [73], and higher than in the German hospitality environments, involving smoking of tobacco [188].

A general trend comparing the results of the PNC increase from both measurement campaigns was as follows: background at 1E+4 #/cm^3, 10 IQOS at 5E+4 #/cm^3, 30 IQOS at 1E+5 #/cm^3 and club in operation at 1E+6 #/cm^3 to 1E+7 #/cm^3. This represents an increase by an order of magnitude in each subsequent scenario.

3.3.2 Particle Mass Concentrations

The particle mass concentrations (PMCs), as calculated from real-time ELPI+ measurements, were grouped based on the measurement scenarios (Fig. 29). Generally, $PM_{2.5}$ and PM_{10} displayed similar variations between the measurement scenarios, except that PM_{10} indicated longer tails of the distribution (represented by both 5th and 95th percentile whiskers), suggesting that there was a higher variation of concentration. This is due to the fact that PM_{10} is more influenced by the re-suspended dust, associated with human activities, while $PM_{2.5}$ is associated with primary and secondary particles, resulting from the thermal aerosol release (such as fog machines, cigarette smoking or exhalation of cigarette aerosol).

The scenarios from the background to using 30 IQOS devices did not result in a significant increase in PMC with the median $PM_{2.5}$ ranging from 2.7 µg/m^3 at Zone 1 and 2.8 µg/m^3 at Zone 2 in case of the background to 11.4 µg/ m^3 (Zone 1) and 12.3 µg/m^3 (Zone 2) in case of 30 IQOS. Such comparatively low impact of IQOS usage to the indoor PMC may have resulted due to the fact that the majority of particles was located in sub-micrometer size range (as discussed in the following sub-chapter), thus not carrying much mass. Another issue may be associated with the particle mass loss during the transport in sampling lines, since the particles may have partially evaporated.

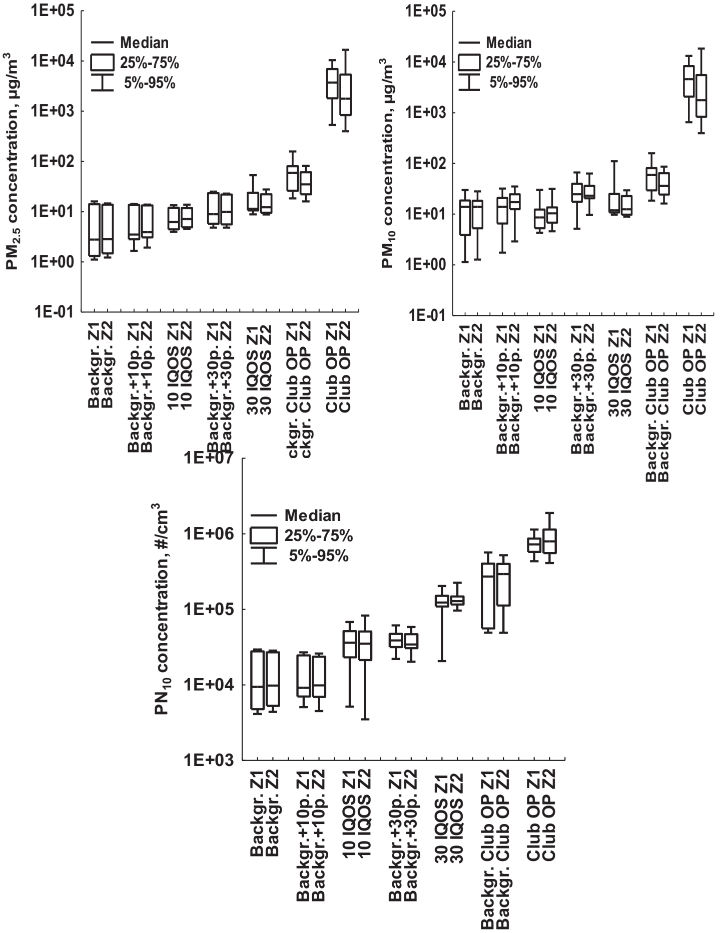


Figure 29. Particle mass ($PM_{2.5}$ and PM_{10}) and number (PN_{10}) concentrations in Zone 1 and Zone 2 under controlled conditions (10 IQOS and 30 IQOS) and club in operation (Club OP): Backgr. + 10 p.: background with 10 humans present but without the HTP use; Backgr. + 30 p.: background with 30 humans present but without the HTP use [144]

The operating club resulted in significantly higher concentrations compared to the 30 IQOS users. The preparations for the club opening already resulted in higher $PM_{2.5}$ concentrations, this time having spatial variations between Zone 1 (59.0 μg/ m^3) and Zone 2 (35.0 μg/ m^3). This may be associated with more intensive staff moving and re-suspending particles from the surfaces as well as potential secondary tobacco aerosol brought by the staff from the smoking room.

Further on, the club in operation resulted in almost several orders of magnitude higher median $PM_{2.5}$ (3715 μg/m^3 at Zone 1 and 1770 μg/m^3 at Zone 2) and PM_{10} (4590 μg/m^3 at Zone 1 and 2470 μg/m^3 at Zone 2) concentrations.

These levels are significantly higher than $PM_{2.5}$ concentration in hospitality venues (36–869 µg/m^3) as reviewed by Fromme et al. [187] in the smoking areas of Californian casinos, during the periods of the highest occupancy (median: 44–110 µg/m^3) [189] or German discotheques (median: 599.2 µg/m^3) during the principal business hours before the implementation of partial smoking ban [190] and are closer to the mean $PM_{2.5}$ and PM_{10} concentrations (619.1 µg/ m^3 and 1156.6 µg /m^3) during the rush working hours (17–21) at the combined water-pipe/CC smoking cafes in Tehran [191]. It must be noted that such differences in the case of the measurements possibly result from deriving of the mass concentration from the real-time PNC measurements, as opposed to the filter-based collection. In the latter method, the filters are conditioned before the gravimetric analysis, thus losing a significant portion of the mass in the case of volatile aerosol, which seems to be the case in the obtained measurements, as indicated by the PSD analysis in the following sub-chapter.

3.3.3 Particle Size Distributions

PSDs based on the PNC indicated that the major part of particles were of sub-micrometer size range, and more importantly, sub-100 nm range (Fig. 30(a)). It may be expected that the aerosol in premises with thermal sources would have a mode at 80–100 nm range, indicating accumulating particles [188]. However, the ultrafine mode indicates that the major portion of the particles was in the nucleation size range for all measurement scenarios. There has been observed such phenomena in a chamber for the exhaled HTP aerosol, and such distribution was attributed to the rapidly evaporating volatile matter of the exhaled aerosol, which may reach 95% of the total mass. Moreover, the measured RH levels during the IQOS experiment averaged between 36–43%, which has been indicated as a favourable range for the particle evaporation [73]. The club in operation yielded similar PSD, possibly reflecting the presence of highly volatile aerosol from the fog machine, while RH stayed relatively low (29–35%).

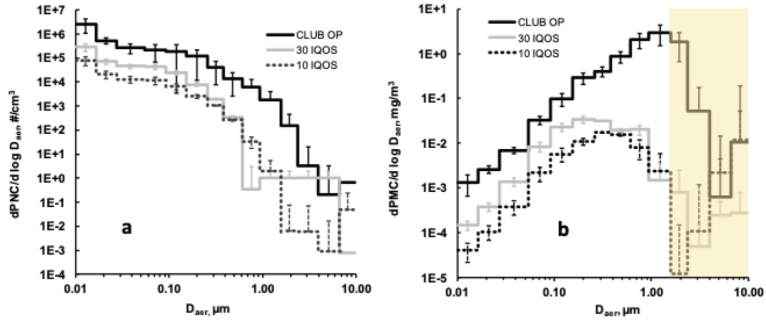


Figure 30. Particle size distribution of (A) particle number concentration and (B) particle mass concentration during the use of HTP under controlled conditions (10 IQOS and 30 IQOS) and club in operation [144]

The PSD based on the mass concentration (Fig. 30 (B)) indicated the mode at 200–300 nm during the use of IQOS and at ~1 µm during the club in operation. The latter is associated with the fact that particle number concentration was significantly higher in that range, compared to IQOS. The PSD in the range of 2–10 µm had much uncertainty associated with the particle losses during the sampling and thus could not be estimated quantitatively with high accuracy, but expectedly, it should feature a mode in particle mass, caused by the re-suspended dust from the moving personnel and visitors.

3.3.4 Gaseous Pollutants

The club featured relatively high background concentrations of gaseous pollutants (Fig. 31 and Table S1). The mean background concentration of formaldehyde (37.4 µg m^{-3}) and acetaldehyde (41.1 µg m^{-3}) were higher than typically found in the residential and public environments [4]. The mean background concentration of nicotine (3.4 µg m^{-3}) and 3-EP (1.7 µg m^{-3}) were similar to those measured in non-smoking areas of Finnish nightclubs and discos during the peak hours (geometric mean: 2.9 µg m^{-3} and 0.8 µg m^{-3}, respectively) [192]. Presumably, the fugitive emissions from the smoking room as well as exhaled cigarette smoke and emissions from clothing and hair from the returning smokers to the main club area resulted in the substantial adsorption of tobacco combustion products and subsequent re-emission, called the third-hand smoke.

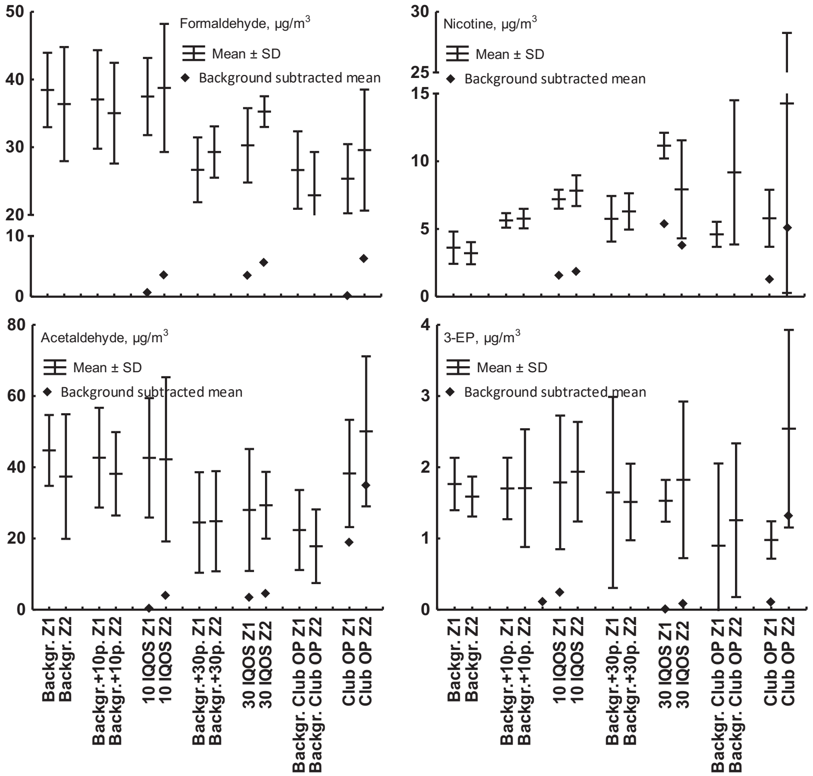


Figure 31. Concentration of gaseous pollutants under different scenarios: 10 volunteers using IQOS (10 IQOS), 30 volunteers using IQOS (30 IQOS) and club in operation (CLUB OP); Z1: Zone 1, Z2: Zone 2, the background subtracted values are indicated as dots [144]

The impact of the usage of HTP in non-operating club to the gaseous pollutant concentrations was much lower, if at all significant, compared to the impact on the particle concentrations (Fig. 31). The usage of 30 IQOS caused a slight increase of the concentrations as compared to 10 IQOS; however, only an increase of formaldehyde and acetaldehyde concentrations was statistically significant in Zone 1 ($p < 0.05$). Hypothetically, the usage of IQOS in Zone 2 should have resulted in higher concentrations in comparison to Zone 1 due to the restricted dispersion. This hypothesis has been partially confirmed for formaldehyde, acetaldehyde and nicotine (10 IQOS), but the differences did not appear to be statistically significant.

The club in operation resulted in either similar levels of pollutants to 30 IQOS (formaldehyde and nicotine in Zone 2) or higher pollutant levels (3-EP and statistically significantly acetaldehyde). Although the club is a non-smoking environment, Zone 2 was most likely affected by fugitive emissions of tobacco smoke from the adjacent smoking room. This is evidenced by the increased concentrations of tobacco-specific markers, i.e., nicotine and 3-EP. The elevated levels of acetaldehyde may be attributed to the consumption of alcohol-containing beverages, as acetaldehyde is a product of ethanol metabolism and is present in the exhaled breath [193].

The observed levels of nicotine during the use of 30 IQOS and club in operation were at same range compared to the concentrations in the smoking areas of Finnish nightclubs and discos during the peak hours (geometric mean: 11.0 $\mu g/m^3$ and 2.4 $\mu g/m^3$, respectively; [192]), Las Vegas casinos where smoking is still not banned (6.7 $\mu g/m^3$; [194]) and New York City hookah bars during the hours of the highest occupancy (4.2 $\mu g/m^3$ [195]).

The penetration of pollutants from smoking areas to non-smoking areas has been reported in multiple earlier research campaigns. South Australian pubs, clubs and cafes having separately enclosed smoking areas had lower mean levels of nicotine (4.4 $\mu g/m^3$) compared to the unenclosed ones (7.5 $\mu g/m^3$), while the air of smoking areas contained nicotine concentration of 15 $\mu g /m^3$ that was measured over a period of 2–4 hours during a normal to busy dinner or lunch sessions [196]. The pollutant concentrations were significantly higher in Spanish hospitality venues with outdoor smokers close to the entrance than in those without outdoor smokers measured during the same range of hours (morning/afternoon) [197]).

3.3.5 Sub-Chapter Summary

In this sub-chapter, the pollution generated by the HTP in a hospitality venue was assessed. The volunteers used the HTP in a nightclub during the non-operating hours. Additionally, the indoor air quality of the club was evaluated during the operating hours. The usage of HTP use in the nightclub increased the background particle number and mass concentrations by an order of magnitude; nevertheless, these values remained an order of magnitude lower than those measured during the club's operating hours, which were as well influenced by the fugitive emissions from the adjacent smoking room and the fogging machines.

The particles generated by both the HTP and the club in operation exhibited high volatility, as indicated by the sizable nucleation-mode fraction of the particle distribution. However, the HTP did not significantly affect the majority of the gaseous pollutant concentrations in a club, which displayed high background values for tobacco-smoke-related pollutants. The real-time aerosol particle concentration and the off-line carbonyl, nicotine and 3-ethenylpyridine concentrations were measured. The observed particle number concentrations were 1E+4, 5E+4, 1E+5 and 1E+6 to 1E+7 $\#/cm^3$ for the background, 10 users, 30 users and the club during operation, respectively, representing an increase by an order of magnitude for each subsequent scenario. The club featured relatively high background concentrations of gaseous pollutants, presumably due to the third-hand smoke, and the use of HTP in a club during the non-operating hours did not significantly affect the majority of these concentrations, nicotine being the only exception. Despite the increase in the background particle number and mass concentrations due to the HTP use, these values were still an order of magnitude lower than during the operating hours.

The mean airborne acetaldehyde levels that have been noted during the club operation exceeded those that were recorded in all other experiments. Such results can possibly be explained by the simultaneous consumption of alcohol-containing beverages. The airborne nicotine levels went higher than the average during both sessions with the HTP use; however, 3-ethenylpyridine remained unchanged: it neither exceeded the background levels nor dropped below them. It is important to mention that despite the fact that the club is officially considered a non-smoking environment, the indoor concentrations of nicotine and 3-ethenylpyridine during the standard operating hours were either in a similar range to those in the experiments with HTP or, in some cases, exceeded them (one such case was observed). These findings were explained by cross-contamination with various other pollutants from the neighbouring smoking area. Due to this discovery, it seems reasonable to assume that cigarette smoking remains a very important pollution factor even when a certain closed space (such as a club) is officially defined as a non-smoking area.

4 GENERAL DISCUSSION AND RECOMMENDATIONS

4.1 Discussion

Following the adoption of WHO Framework Tobacco Control Convention in 2003, smoking bans in public places have been introduced in many countries [198]. A wide range of regulatory responses, ranging from no regulation to complete bans, were as well applied to the harm-reduction products [199]. The bans against the use of nicotine-containing products in public spaces in many countries are possibly the main reason why evidence of the passive exposure to the vapour released or exhaled from the harm-reduction products under real conditions is still scarce.

Secondhand and thirdhand smoking is a sensitive issue that has been ignored largely. Nowadays, it is known that the smoke particles left on clothes may be very harmful. Even if there are no smokers in the family, the family members may be passive smokers and will not be able to change it, because the smoke particles are permeable, spread through ventilation, stay on clothing and last for a long time.

While there is rapid market transition to the new nicotine containing products, the data on these devices is very sparse so far. That was the reason for making a comparative analysis of results of the existing scientific publications in order to determine the impact of heated tobacco products on the indoor air levels of VOCs and PM, originating from various indoor sources (including micro-environments, such as kitchens and transport cabins) with the aim to compare the pollution levels with those generated by the HTP and provide a quantitative indication on possible hazard caused by the HTP versus common everyday life environments. For the first time, the quantitative indications were provided to compare possible hazards caused by the EC and HTP versus the majority of researched pollution sources (CC, water pipe, incense, mosquito coils). Consequently, the authors quoted in the review concluded that the persons in proximity to such non-nicotine combustion products might be exposed to the greater levels of pollutants than persons standing near the EC or HTP user [4]. Moreover, the measured acetaldehyde concentrations fell within the typical range of the mean concentrations that were observed in the residential and public environments [2, 4, 73]. It is important to note that most of the studies differed in the used residential and public environments as well as the list of constituents measured in the air, even if most of them measured formaldehyde, benzene, toluene and $PM_{2.5}$. Thus, it is difficult to draw conclusions on the basis of the reported overall findings. An additional point of concern is that most of these studies simply assumed that the regulatory norms for occupational exposure in terms of adequate ventilation were respected and did not systematically verify the ventilation rates.

Three experiments were conducted in order to determine the differences of HTP, EC and CC generated aerosol dynamics, their impact on IAQ, the impact of the number of simultaneous smokers on PNC and other characteristics.

In the first experiment, the HTP use intensity as the number of parallel users, distance to the bystander as well as environmental conditions in a chamber were evaluated. The concentrations of airborne nicotine and acetaldehyde showed a slight upward trend with increasing numbers of simultaneous users; with regard to nicotine levels, the distance between the user and a bystander played a role as well in a significant drop in the indoor concentrations of nicotine with increasing distance.

Following the first experiment, another one was conducted aiming to compare the pollution levels generated by the HTP, EC and CC. Since these nicotine containing products have different working principles, their generated aerosols have different physicochemical properties, especially when EC and HTP are compared with CC. The aerosols generated by a heat-not-burn process are more volatile, the ventilation rate has bigger impact on their PNC, and the environmental conditions, such as temperature, humidity and other, have greater impact on their dynamics. In case of CC, the chemical composition of generated smoke is very broad with more than 4000 compounds and differs depending on the CC brand. In CC smoke, there are more solid particles, much less water vapour and volatile organic compounds that are found in the HTP and EC vapour, as glycerol and propylene glycol. This makes CC smoke less affected by the environmental conditions in comparison to the HTP and EC. Some similarities were observed as well, e.g., the distance to the bystander is a significant factor that is affecting the aerosol dynamics in all cases.

The third experiment was conducted in real-life environment. Only a minor increase in the indoor concentrations of $PM_{2.5}$ and PM_{10} and similar to that in the case of the PNC was observed when HTP products were used in the night club during the non-operational hours. This increase did not exceed or go below the usual range of background variations before the club opening, and it was not measured to be substantially below the levels noted at other times during the regular nightclub operation conditions. The assessment of particle size distribution in all measurement scenarios showed an ultrafine mode, where the major portion of particles was within the nucleation size range. These data were on the same level with those found in the first experiment of environmental aerosols of HTP in the exposure chamber [73] and can be explained by a speedy evaporation of the liquid droplets that are generated when the HTPs are used. As far as formaldehyde and acetaldehyde are concerned, generally, the indoor concentrations of these compounds during the HTP use stayed in the same range as the background variations. Only one exception was noticed with a minor increase in one of the measurement zones when 30 volunteers participating in the study used HTP simultaneously.

4.2 Recommendations

Based on the research results, the following recommendations can be suggested.

1. It is important to bear in mind that aerosols produced by HTP and EC leads to passive smoking, although to a significantly lower level compared to the CC aerosol. The number of parallel users has a direct impact on the concentration of aerosol particles near the other users. The experiments were conducted in a room with a volume of 35.8 m^3 and a floor area of 13 m^2. In the case of five users, compared with one user, the average PNC was 27 times higher during an "active" period and 8 times higher in the "after" period. The rate of the air exchange up to 1 ACH is not capable of efficiently removing the air pollutants during a 30-minute period. The intensive use of HTPs and ECs in a confined space with limited ventilation can

substantially increase the exposure of the bystanders to the secondhand aerosols; therefore, the regulations on the ventilation rates in smoking rooms should be put in place.

2. The distance from the NCP user is another important parameter affecting both the potential exposure level and the particle size. This is especially evident in the case of puff (peak concentration) or immediately afterwards. However, highly volatile liquid droplets generated by the HTP and EC disperse relatively fast and have a lower probability of reaching the bystander at a distances that exceed 0.5 m. The presence in close proximity (< 0.5 M) of a NCP user should be avoided.

3. The concentration of airborne nicotine and acetaldehyde increase in line with the following factors: higher number of HTP parallel users (when the number of users is increased 3 times, the PNC is raised approximately by 3.6 times), shorter distance to the bystander (when this distance is increased from 0.5 to 2 m, the PNC is lowered approximately by 4.2 times), lower ventilation intensity and lower relative humidity (when RH is increased from 30% to 70%, the PNC lowers approximately by 1.9 times). It is recommended that the places in which NCP are used should be adapted in line with these parameters.

4. A literature review revealed that both the international organisations and individual countries do not have a limit value for nicotine for indoor air and only occupational limit values are determined. It is recommended to set a limit value for nicotine for indoor air in residential and public environments analogously to many other indoor pollutants.

5. Thirdhand smoking, as the experiments have shown, is caused by the increased air pollution from the adjacent smoking area. Even if the NCPs are not used in a room, the concentration of nicotine can still be high enough to give rise to the thirdhand smoke. It is recommended that the thirdhand smoke prevention approaches should be assessed and developed through further research, e.g., ozonation and air cleaners with HEPA filters. Smoking room must have an independent ventilation system in order to prevent secondhand and thirdhand exposure to non-smoking areas.

6. According to the results of this thesis, it is recommended that a new device or technology on a market should undergo a thorough investigation in a well-controlled environment, aiming at the accurate characterization of the generated aerosol and thus potential impacts on the air quality and bystander exposure. Perhaps, it is necessary to develop a set of specific HPHCs in order to represent HTP environmental aerosol from the matrix of indoor pollutants.

7. It can be recommended to do computer modeling of particle dynamics. The Computational Fluid Dynamics (CFD) could be used for this purpose. The comparison of modelling results with the empirical data would verify the model and serve for the better understanding of the aerosol dynamics. The developed model would allow predictions of pollution levels in different scenarios of NCP use intensity, space as well as micro-climatic conditions.

It is fundamental to emphasize that all nicotine-containing products must be used with caution near other people, especially pregnant women and children. The composition of mainstream as well as environmental HTP and EC generated aerosol shows that it might be a much safer option than CC; however, it is still not known if there are no undiscovered negative consequences of HTP and EC in the long-term usage. The best method of protection from passive smoke is to shield them from the exposure to active smoke sources, if shielding is impossible, at least both passive and active smokers should be aware of lucid and scientifically proven consequences of the usage of new nicotine containing products.

CONCLUSIONS

1. The HTP use has been commonly linked to lower or comparable concentrations of indoor air pollutants, compared with other conventional indoor sources or environments, in most cases distinguishable above the background and potentially associated with more health effects at prolonged exposures than any other source of artificial air pollution.

Among the greater number of researched pollution sources that were researched (conventional cigarettes, water pipes, incense and mosquito coils), the lowest concentrations of formaldehyde, benzene, toluene and $PM_{2.5}$ were produced with the use of HTP (as well as an electronic cigarette) in a controlled environment. Significantly higher levels of pollution of benzene, toluene and formaldehyde may occur in public environments, especially the micro-environments relating to the transport.

These findings show that the levels of the most important indoor air pollution markers, in the case of HTP environmental aerosol, may be too low to distinguish from the background. The data that has been obtained raised additional challenges for the epidemiological studies, seeking to assess the second-hand exposure in real-life environments. Perhaps, it is necessary to develop a set of specific HPHCs in order to represent HTP environmental aerosol from the matrix of indoor pollutants.

2. The HTP use produced a statistically significant increase of several analytes, including nicotine, acetaldehyde, PN and $PM_{2.5}$ concentrations. Compared to the levels produced by the conventional cigarette (CC) smoking under identical conditions, the obtained levels were significantly lower (approximately 16, 8, 8 and 28 times for nicotine, acetaldehyde, PN and $PM_{2.5}$ concentrations, respectively). The maximum concentrations of formaldehyde (16.3 $\mu g/m^3$) and acetaldehyde (12.4 $\mu g/m^3$) fell within the range of mean concentrations found in the residential and public environments. A maximum 30-minute concentration of PNC (4.8E+05 $\#/cm^3$) and maximum concentration of PNC (9.3E+06 $\#/cm^3$) suggest that the intensive HTP used in a confined space with limited ventilation may cause substantially elevated concentrations of aerosol, despite of the fact that these particles turned out to be highly volatile, evaporating within seconds. As confirmed by the highest concentration of particles at nucleation mode, the exhaled HTP aerosol particles (in contrast to the CC) evaporated faster than they accumulated in the chamber air.

The intensity of usage (number of simultaneous users) prevailed, for the most part, as the most important factor affecting pollutant variations positively. The distance to the bystander was another important factor. Under identical conditions, compared to one use, mean PNC in the case of five users was 27 times higher during the usage and eight times higher after the usage. The distance factor produced a significantly higher mean PNC (4.2 times) at a distance of 0.5 m in comparison to 2 m during the active usage. However, a longer distance had no significant effect on the PNC levels during the after-usage period, which were at comparable levels for both distances. The rate of air change turned out to be less significant factor in the ranges, in which it was operated (up to 1 ACH). This demonstrated that such ventilation intensity was not capable of efficiently removing the intensive emission of pollutants during a 30-minute period, requiring prolonged ventilation at the maximum regimes.

The EC puffs showed a higher total PNC, which as well decayed more rapidly. What is more, these particles, owing to their more volatile nature, shrank more quickly. The similarities between the EC and the HTP aerosol in terms of their particle size distributions and indoor dispersion factors, despite the differences in concentration or removal rate, indicated the existence of shared physical mechanisms for transport and transformation (e.g., particle shrinkage due to the evaporation). These mechanisms were distinct from those for tobacco-burning products, such as conventional cigarettes. The usage topography introduced large uncertainties into these processes. Thus, in future investigations, it must be either strictly standardised or recognised as a major variable. The distance to the user turned out to be the primary parameter for controlling short-term exposure. This particular factor and the ventilation intensity should be managed in tandem to make the long-term exposure less severe.

3. In the nightclub, the HTP use increased the number and mass concentrations of the background particles by an order of magnitude. However, these values remained an order of magnitude lower than the ones measured during the club's operating hours, and these were as well influenced by the fugitive emissions from an adjacent smoking room and fogging machines. As indicated by a sizable nucleation-mode fraction of particle distribution, the particles generated by both the HTP and the club in operation were highly volatile. However, the HTP did not significantly affect the majority of gaseous pollutant concentrations in the club, which displayed high background values for tobacco-smoke-related pollutants.

The intensive HTP use (e.g., 30 simultaneous users) in indoor hospitality environments may be associated with deteriorating air quality and increased exposure to nicotine and acetaldehyde as well as particles. However, the traditional cigarettes, despite the partial smoking ban, potentially remain the largest factor in the exposure to guests.

SUMMARY

Vartojamos sąvokos ir trumpiniai:

Dalelių dydžio pasiskirstymas (DDP) (angl. *particle size distribution*) – rodo, kokio dydžio aerozolio dalelės (pvz., µm) ir koks jų kiekis yra tam tikrame oro mėginio tūryje (pvz., cm^3).

Dalelių skaitinė koncentracija (DSK) (angl. *particle number concentration*) – bendras dalelių skaičius oro tūrio vienete (pvz., m^3).

Elektroninės cigaretės (EC) (angl. *electronic cigarettes*) – elektroniniai prietaisai, turintys kaitinamąjį elementą, skirtą išgarinti specialiam tirpalui, kad susidarytų įkvepiamasis aerozolis, kurio sudėtyje paprastai yra nikotino, kvapiųjų medžiagų ir kitų junginių.

Įprastinės cigaretės (ĮC) (angl. *conventional cigarettes*) – rūkymui skirtas gaminys iš plonai pjaustytų džiovintų tabako lapų, suvyniotų į popierinį cilindrą su filtru. Naudojimo metu tabakas lėtai rusena, o susidarančius dūmus įkvepia rūkantysis.

Įkvepiamasis aerozolis (angl. *mainstream aerosol*) – tai nikotino turinčių produktų specialiai sukuriamas ir vartotojo įkvepiamas aerozolis šių produktų vartojimo metu.

Iškvepiamasis aerozolis (angl. *exhaled aerosol*) – tai nikotino turinčių produktų vartotojo iškvepiamas aerozolis šių produktų vartojimo metu (po įkvepiamojo aerozolio įkvėpimo).

KD$_X$ (angl. *particulate matter*) – kietųjų dalelių, kurių aerodinaminis skersmuo mikrometrais mažesnis ar lygus X, masės koncentracija, µg/m^3.

Kaitinamojo tabako produktai (KTP) (angl. *heated tobacco products*) – kaitinamas tikras tabakas siekiant išgauti nikotino turintį aerozolį, degimo procesas nevyksta.

Oro kaitos dažnis (OKD) (angl. *air change rate*) – rodo, kiek kartų per valandą patalpose keičiasi oras.

Patalpų aerozolis (angl. *environmental aerosol*) – tai patalpose besisklaidantis aerozolis, kurį daugiausia sudaro iškvepiamas aerozolis, o įprastinių cigarečių (ĮC) atveju – dar ir šalutinis aerozolis. Kvėpavimas patalpų aerozoliu vertinamas kaip antrinis rūkymas ar nikotino turinčių produktų vartojimas.

Šalutinis aerozolis (angl. *sidestream aerosol/smoke*)

– tai įprastinių cigarečių ar kitų atviro tabako smilkimo produktų vartojimo metu susidarantis aerozolis, kuris nėra sukuriamas specialiai įkvėpti produktų vartotojui. Kaitinamojo tabako produktai (KTP) bei elektroninės cigaretės (EC) nesukuria šalutinio aerozolio.

1 ĮŽANGA

Per pastarąjį dešimtmetį buvo sparčiai kuriami alternatyvūs nikotino turintys produktai, siekiant pakeisti tabako deginimu / pirolize paremtus produktus mažiau kenksmingais. Garinimo principu veikiantys produktai, pvz., elektroninės cigaretės (EC), arba kaitinamojo tabako produktai (KTP) suteikia rūkantiems asmenims naujų pasirinkimo galimybių. Nors abiem šiomis technologijomis siekiama sumažinti kancerogeninių junginių susidarymą įkvepiamame aerozolyje, jų veikimo principai skiriasi, o ilgalaikio vartojimo poveikis sveikatai dar nėra žinomas. Taip pat trūksta duomenų apie naujų nikotino turinčių produktų įtaką patalpų oro kokybei, lyginant su kitais oro taršos šaltiniais.

Tik nedaugelyje paskelbtų mokslinių straipsnių nagrinėjamas naujų nikotino turinčių produktų (NNTP) vartojimo poveikis patalpų oro kokybei. Kadangi šiuose produktuose tabakas nėra deginamas, pagrindinis taršos šaltinis yra vartojimo metu iškvepiamas aerozolis. Jo sudėtis iš esmės skiriasi nuo įkvepiamo aerozolio: dauguma iškvepiamų dalelių yra vandens pagrindo, o diduma nikotino yra absorbuojama vartotojo kūne [1, 2]. Vis dėlto keletas tyrimų parodė, kad EC naudojimas ir, mažesniu mastu, KTP poveikis turi apčiuopiamos įtakos patalpų oro kokybei ir potencialiai prisideda prie pasyviojo rūkymo.

Kaitinant tabaką, o ne deginant, kyla mažesnė rizika sveikatai, jei lyginsime su įprastomis cigaretėmis (ĮC). Kaitinant tabaką susidaro ne toks sudėtingas aerozolis [3], be to, tyrimai, susiję su kaitinimo be degimo (angl. *heat-not-burn*) tabako gaminiais (dar vadinamais „tabako kaitinimo sistemomis“ [TKS] arba naujais tabako produktais [NTP]), parodė, kad kenksmingų ir potencialiai kenksmingų sudedamųjų dalių kiekis aerozolyje yra žymiai mažesnis.

KTP vartojimas dažniausiai siejamas su mažesne patalpų oro tarša, lyginant su kitais įprastais patalpų oro taršos šaltiniais, tačiau KTP gali daryti poveikį sveikatai, jei vartojami ilgai, kaip ir bet kuris kitas dirbtinis oro taršos šaltinis.

Daktaro disertacijos tikslas – ištirti aerozolių, susidarančių vartojant naujus nikotino turinčius produktus, dinamiką laike ir erdvėje, taip pat šių aerozolių cheminę sudėtį ir poveikį patalpų oro kokybei.

Uždaviniai

1. Atlikti lyginamąją ankstesnių tyrimų rezultatų analizę siekiant nustatyti, kokią įtaką patalpų oro kokybei daro kaitinamojo tabako produktai palyginus su kitais galimais oro taršos šaltiniais, pvz.: įprastomis cigaretėmis, smilkalais, vabzdžius atbaidančiomis spiralėmis.
2. Siekiant nustatyti svarbius oro kokybės valdymo parametrus patalpose, kuriose vartojami nikotino turintys produktai, atlikti fizinį modeliavimą kontroliuojamomis sąlygomis ir įvertinti KTP, EC bei ĮC patalpų aerozolio sklaidą kambario ore remiantis dalelių koncentracijos, dydžio pasiskirstymo matavimais.

Ištirti galimus oro kokybės pokyčius, vartojant naujus nikotino turinčius produktus neveikiančiame naktiniame klube, kai vartotojų skaičius skirtingas, ir palyginti su oro kokybės rodikliais tame pačiame naktiniame klube jam veikiant.

Mokslinis naujumas

Nors rinkoje sparčiai pereinama prie naujų nikotino turinčių produktų, duomenų apie šių produktų vartojimo metu išskiriamą į aplinką aerozolį vis dar yra nedaug. Šiame darbe pirmą kartą pateiktos kiekybinės charakteristikos, kuriomis remiantis nesunku įvertinti galimus KTP pavojus ir palyginti su dauguma kitų taršos šaltinių, pvz., įprastų cigarečių, kaljano, smilkalų, uodų atbaidymo spiralių, keliama grėsme.

Aerozolio dispersijos kitimas laike patalpoje, esant įvairioms aplinkos sąlygoms, yra dar mažai ištirta tema; tą parodė mokslinės literatūros analizė. Šiame tyrime, remiantis didelio dažnio dalelių koncentracijos ir dalelių dydžio pasiskirstymo matavimais KTP iškvepiamojo aerozolio atveju, buvo palyginti iškvepiamojo KTP, EC ir ĮC aerozolio pokyčiai laike ir erdvėje tiriamajame kambaryje.

KTP poveikis patalpų oro kokybei imituojant realias sąlygas anksčiau nebuvo tirtas. Dėl to atliktas tyrimas realiame naktiniame klube. Šis tyrimas taip pat papildė duomenis, gautus kambaryje kontroliuojamomis sąlygomis.

Disertacijos struktūra

Disertaciją sudaro šie skyriai: padėkos, įvadas, literatūros apžvalga, metodai ir matavimai, rezultatai ir aptarimas, bendra diskusija ir rekomendacijos, išvados, literatūros sąrašas, publikacijų sąrašas ir priedai. Darbą sudaro 170 puslapiai, 31 paveikslas, 6 lentelės ir 8 priedai.

Tyrimo rezultatų paskelbimas

Remiantis disertacijos tyrimų rezultatais, paskelbti keturi moksliniai straipsniai tarptautiniuose žurnaluose: trys straipsniai – žurnaluose, registruotuose *CA Web of Science* duomenų bazėje, vienas straipsnis – žurnale su „Scimago Journal & Country Rank“ (SJR) rodikliu (*Scopus*). Eksperimentiniai rezultatai buvo pristatyti trijose tarptautinėse konferencijose.

Darbo praktinė vertė

Tyrimui atlikti buvo sukurta metodika KTP poveikiui patalpų oro kokybei vertinti. Literatūros apžvalga parodė, kad KTP dažniausiai siejami su gerokai mažesniu taršos lygiu, tačiau disertacijos tyrimo rezultatai atskleidė, kad EC ir KTP patalpų aerozoliai prisideda prie pasyviojo rūkymo. Tiriamojo kambario sąlygomis ir imituojant realias gyvenimo sąlygas buvo nustatyti keli svarbūs kriterijai, turintys įtakos aerozolių dalelių koncentracijos dinamikai. Atsižvelgiant į šiuos kriterijus, galima sukurti sveikesnę aplinką naujų nikotino turinčių produktų vartotojams ir jų aplinkoje esantiems žmonėms.

Autorės indėlis

Disertacijos tyrimą sudaro 4 logiškai susieti eksperimentai, atlikti teoriškai literatūros apžvalgos metu ir empiriškai. Eksperimentai atlikti kartu su mokslininkų komanda, o eksperimentų rezultatai pateikti keturiuose moksliniuose straipsniuose. Autorės indėlis į kiekvieną straipsnį aprašytas toliau.

1 straipsnyje (*A Review of the Impacts of Tobacco Heating System on Indoor Air Quality versus Conventional Pollution Sources – Tabako kaitinimo sistemos poveikio patalpų oro kokybei palyginimas su įprastais oro taršos šaltiniais*) autorė suplanavo bei atliko duomenų rinkimą ir dalyvavo rengiant straipsnio rankraštį.

2 straipsnyje (*Impact of Exhaled Aerosol from the Usage of the Tobacco Heating System to Indoor Air Quality: A Chamber Study – Tabako kaitinimo sistemos naudojimo metu iškvėpto aerozolio poveikis tiriamojo kambario oro kokybei*) autorė padėjo planuoti ir atliko eksperimentus tiriamajame kambaryje, išanalizavo duomenis ir dalyvavo rengiant straipsnio rankraštį.

3 straipsnyje (*Impact of Using a Tobacco Heating System (THS) on Indoor Air Quality in a Nightclub – Tabako kaitinimo sistemos (TKS) naudojimo poveikis patalpų oro kokybei naktiniame klube*) autorė suplanavo ir atliko eksperimentus, skirtus patalpų oro kokybei vertinti panaudojus TKS neveikiančiame naktiniame klube esant skirtingam TKS naudotojų skaičiui, ir palygino jį su įprastai veikiančio naktinio klubo patalpų oro kokybe. Atliko duomenų analizę ir dalyvavo rengiant straipsnio rankraštį.

4 straipsnyje (*The Dynamics of Exhaled Aerosol Following the Usage of Heated Tobacco Product, Electronic Cigarette, and Conventional Cigarette – Iškvėptojo aerozolio dinamika pavartojus kaitinamojo tabako produktus, elektronines cigaretes ir įprastas cigaretes*) autorė suplanavo ir atliko visus eksperimentus, skirtus palyginti KTP, EC ir ĮC naudojimo metu išsiskiriančių dalelių koncentracijas, dydžius ir dinamiką; išanalizavo duomenis ir nubraižė grafikus. Remdamasi eksperimentinių duomenų analize autorė parengė straipsnio rankraštį.

2 LITERATŪROS APŽVALGA

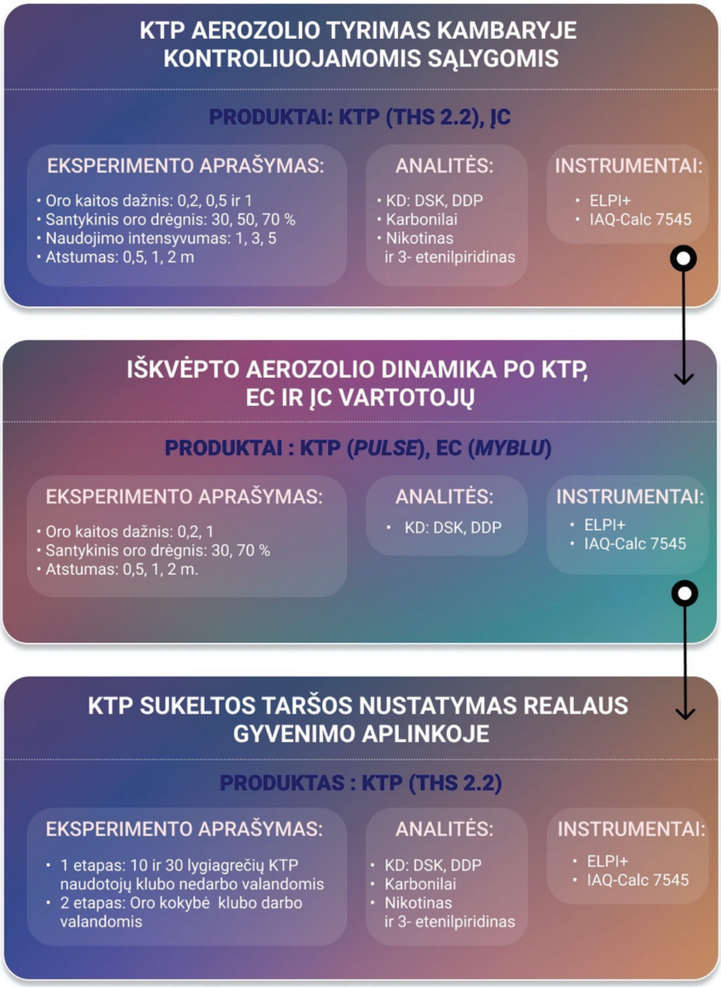
Disertacijos 1.1 poskyrio „Nikotino turinčių produktų įtaka patalpų oro kokybei“ santrauka: apibendrinus išanalizuotą literatūrą paaiškėjo, kad EC ir KTP vartojimas daro mažesnį neigiamą poveikį patalpų orui, palyginti su ĮC, kaljanais, taip pat daugeliu kitų oro taršos šaltinių, pvz., smilkalų deginimu ir uodų atbaidymo ritėmis. Tai dar labiau patvirtina kelių kitų tabako gaminių, t. y. cigarų ir pypkių, tyrimų rezultatai. Svarbiausia, kad EC ir KTP aplinkos aerozolių sudėtis ir kokybinės bei kiekybinės savybės labai skiriasi nuo tabako dūmų savybių. Disertacijoje pateikta naujausių KTP, EC ir ĮC poveikio patalpų oro kokybei tyrimų santrauka rodo, kad nors vartojant KTP ir EC patalpų oro kokybė blogėja daug mažiau palyginus su ĮC, iškvepiamas aerozolis vis dėlto yra labai dinamiška sistema, kuriai įtakos turi daug veiksnių.

Disertacijos 1.2 poskyrio „Kaitinamojo tabako produktų poveikio patalpų oro kokybei palyginimas su įprastiniais taršos šaltiniais“ santrauka: išanalizavus mokslinę literatūrą nustatyta, kad KTP vartojimas yra silpnas taršos šaltinis, vertinant pagal karbonilų, monoaromatinių angliavandenilių, nikotino ir smulkių kietųjų dalelių koncentraciją; lyginant su tradicinėmis cigaretėmis ($p < 0{,}05$) ir kaljanais, KTP statistiškai nedaug didesnis taršos šaltinis už EC ($p > 0{,}05$). Tarp nikotino neturinčių oro taršos šaltinių didesnius aldehidų, lakiųjų organinių junginių ir $KD_{2,5}$ kiekius skleidžia smilkalai ir uodų atbaidymo spiralės, nors formaldehido atveju žvakių oro tarša buvo tokia pati ($p > 0{,}05$) kaip KTP. Tai rodo, kad smilkalų ir ypač uodų spiralių deginimas patalpose, jei jos naudojamos tokiu pačiu periodiškumu kaip ir nauji nikotino turintys produktai, gali sukelti didesnę oro taršą nei stovint šalia aktyvaus EC ar KTP vartotojo.

Disertacijos 1.3 poskyrio „Tabako produktų išskirto aerozolio dinamika“ santrauka: šio poskyrio tikslas buvo pateikti apibendrintą aerozolių dinamikos, fizikinių ir cheminių charakteristikų apžvalgą. Kai naudojama EC ir KTP, proceso metu išsiskiria submikroniniai skysčio lašeliai, sudaryti iš itin lakių ingredientų. Esant skirtingiems ventiliacijos parametrams, patalpos tūriui ir naudojimo intensyvumui, gali atsirasti tam tikrų skysčių lašelių aerodinaminio skersmens skirtumų. Tokie lašeliai išsisklaido per kelias sekundes, todėl ultrasmulkiųjų dalelių poveikis trunka labai trumpą laiką ir jį gali jausti tik asmenys, esantys arti KTP vartotojų. Tai labai skiriasi nuo tabako dūmų smulkiųjų dalelių savybių. Tabako degimo metu susidarantis aerozolis yra daug stabilesnis ir ilgai išlieka uždaroje aplinkoje, todėl labai padidėja patalpų tarša, ypač tais atvejais, kai rūkymo dažnis yra didelis, o ventiliacijos lygis žemas. Aerozolių dinamika ir gravitacinis nusėdimas priklauso nuo daugelio veiksnių, pvz., aerozolį sukuriančio produkto tipo (EC arba KTP), aerozolio dalelių parametrų, pvz., lakumo, taip pat nuo aplinkos sąlygų, veikiančio oro kondicionieriaus, transportavimo atstumo, vėdinimo intensyvumo ir kt.

3 METODAI IR MATAVIMAI

Pagrindinė informacija apie tris empirinius eksperimentus yra pateikta 1 pav. Išsamesnis kiekvieno eksperimento aprašymas pateiktas tolesniuose 3.1, 3.2, 3.3 poskyriuose.

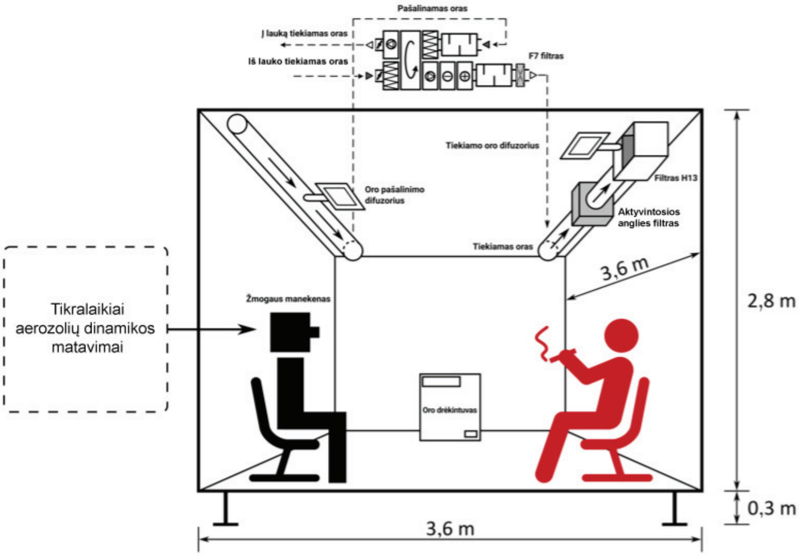


1 pav. Pagrindiniai atliktų eksperimentų etapai

3.1 KTP patalpų aerozolio tyrimas kontroliuojamomis sąlygomis

Tiriamajame kambaryje, esant kontroliuojamoms aplinkos sąlygoms, buvo lyginamas KTP ir ĮC poveikis patalpų oro kokybei identiškomis eksperimentinėmis sąlygomis. Matavimai buvo atliekami keičiant aplinkos sąlygas: OKD, santykinį oro drėgnį, KTP naudojimo intensyvumą ir atstumą iki nikotino produktų vartotojų.

Tiriamasis kambarys, imituojantis standartinę tyrimų kamerą, įrengtas iš įprastų statybinių medžiagų (grindų plotas – 13 m^2, patalpos tūris – 35,8 m^3 (žr. 2 pav.). Tiriamajame kambaryje galima kontroliuoti aplinkos parametrus (oro apykaitos greitį, drėgmę), taip pat atlikti aerozolio bei dujinių teršalų koncentracijos matavimus. Kameroje buvo sumontuotas stačiakampės geometrijos „manekenas“, imituojantis sėdintį žmogų – stebėtoją. Aerozolio mėginiai buvo imami pro manekeno varinį įleidimo vamzdelį ir analizuojami. Santraukoje vietoj žodžio „manekenas“ vartojamas terminas „stebėtojas“.



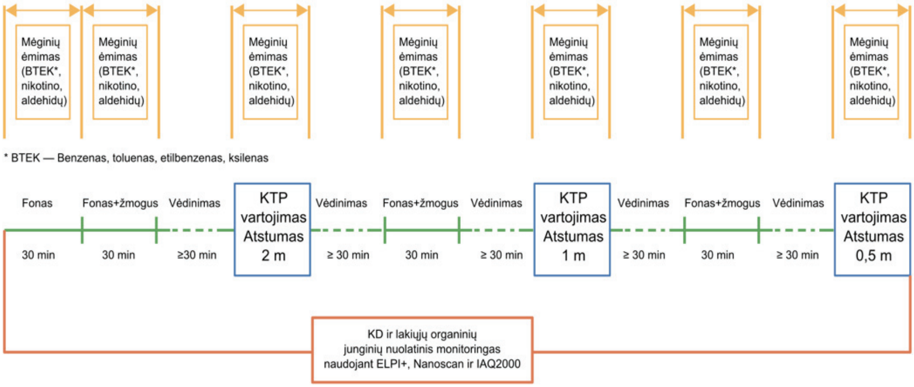
2 pav. Tiriamasis kambarys [73]

DSK buvo nustatoma ELPI+ (*Dekati Ltd.*, Suomija), esant 10 l/min srautui. Aerozolių mėginių koncentracijos buvo registruojamos kas sekundę. Dalelių dydis nuo 10 nm iki 420 nm buvo registruojamas SMPS (*TSI Inc.*, JAV). Dalelių dydis registruojamas kas sekundę 1 l/min srauto greičiu.

Formaldehido ir acetaldehido koncentracijos aerozolyje buvo vertinamos pagal ASTM D5197-03 procedūrą. Aerozolis buvo traukiamas pro silikagelio vamzdelį su 2,4-dinitrofenilhidrazino (DNPH) reagentu (*SKC Inc.*, JAV). DNPH dariniai buvo analizuojami skysčių chromatografijos metodu.

Nikotino ir 3-etenilpiridino koncentracijos aerozolyje buvo vertinamos pagal ISO 18145 procedūrą. Analitės adsorbuojamos sorbentu (XAD®-4, *SKC Inc.*, JAV), o vėliau ekstrahuojamos ir nustatomos dujų chromatografijos bei masių spektrometrijos metodais.

Eksperimento struktūra pavaizduota 3 pav.



3 pav. Eksperimento struktūra [73]

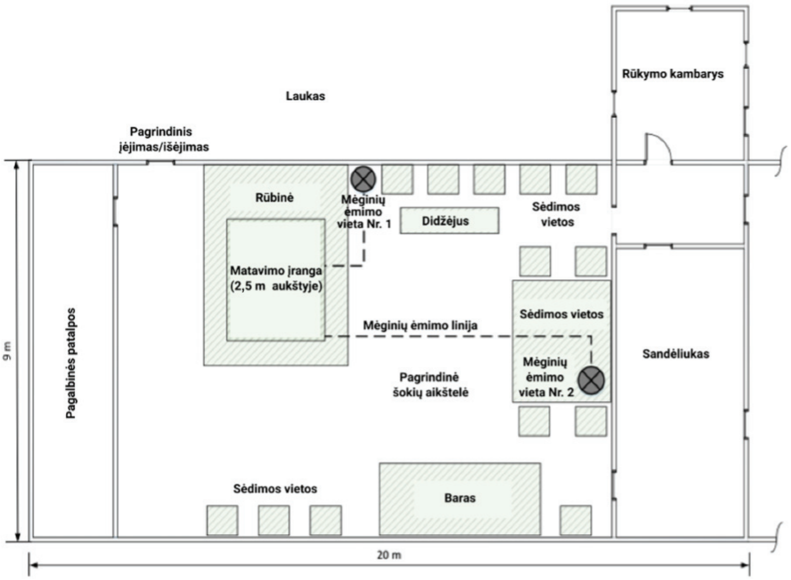
3.2 KTP, EC ir ĮC patalpų aerozolio dinamika kontroliuojamomis sąlygomis

Kitas eksperimentas, sudarytas iš 108 bandymų, buvo atliktas tame pačiame tiriamajame kambaryje (2 pav.), siekiant palyginti iškvepiamuosius aerozolius, kai vartojami KTP, EC ir ĮC. Buvo matuojama dalelių koncentracijos dinamika ir dalelių dydžių pasiskirstymas. Ištirti ir kiekybiškai nustatyti svarbūs veiksniai, turintys įtakos aerozolių dinamikai kambaryje.

DSK ir DDP buvo kas sekundę matuojami FMPS spektrometru (*TSI Inc.*, JAV) dydžių diapazone nuo 5,6 nm iki 560 nm. Santykinis drėgnis ir temperatūra buvo matuojami IAQ-Calc 7545 (*TSI Inc.*, JAV) kas 30 sek.

3.3 KTP poveikio oro kokybei realaus gyvenimo aplinkoje vertinimas

Eksperimentas buvo atliktas naktiniame klube Kaune (Lietuva). Klubo plotas apie 160 m^2. Klubo patalpų bendras tūris apie 880 m^3 (4 pav.). Eksperimento tikslas buvo nustatyti KTP vartojimo poveikį oro kokybei neveikiančiame naktiniame klube (1 etapas) ir palyginti su oro kokybe klubui veikiant (2 etapas). Šiam tikslui pasiekti 10 ir vėliau 30 savanorių vienu metu naudojo KTP prietaisus neveikiančio naktinio klubo patalpose. Veikiančio naktinio klubo patalpose rūkyti draudžiama, tačiau teršalų dalelių sklaida vyko iš gretimos rūkymo patalpos ir rūko aparato, todėl veikiančiame ir neveikiančiame naktiniame klube buvo matuojamos kietųjų dalelių skaitinės bei masės koncentracijos ir imami lakiųjų organinių junginių bandiniai.



4 pav. Naktinio klubo patalpų planas, parodantis su mėginių ėmimu susijusią informaciją

DSK buvo nustatoma ELPI+ (*Dekati Ltd.*, Suomija), esant 10 l/min srautui. Aerozolių mėginių koncentracijos buvo registruojamos kas sekundę. Formaldehido ir acetaldehido bei nikotino ir 3-etenilpiridino koncentracijos buvo vertinamos naudojant sorbcinius vamzdelius, kaip aprašyta 6.3.1 skyriuje.

4 REZULTATŲ APTARIMAS

4.1 KTP patalpų aerozolio pokyčių laike ir erdvėje vertinimas tiriamajame kambaryje

4.1.1 Dalelių ir dujinių teršalų koncentracija

Buvo išskirti svarbūs veiksniai, turintys įtakos galimai teršalų sklaidai. Vartojant KTP, ore statistiškai reikšmingai padidėjo keleto analičių: nikotino, acetaldehido, dalelių skaitinės koncentracijos ir $KD_{2,5}$. Apibendrinus, vartojimo intensyvumas (vienu metu nikotino produktus vartojančių asmenų skaičius) buvo svarbiausias veiksnys, labiausiai veikiantis teršalų koncentracijų pokyčius. Kitas svarbus veiksnys – atstumas tarp nikotino produktų vartotojo ir stebėtojo. Anksčiau tokie rezultatai nebuvo skelbti. Gauti duomenys padeda geriau suprasti, kaip valdyti oro kokybę patalpose, kuriose vartojami nikotino turintys produktai.

Bendra aprašomoji įvairiomis sąlygomis tirtų analičių statistika apibendrinta 1 lentelėje. KTP patalpų aerozolyje nikotino, acetaldehido, dalelių skaičiaus ir masės koncentracija gerokai viršijo foną, tačiau didžiausios formaldehido (16,3 µg/m^3) ir acetaldehido (12,0 µg/m^3) koncentracijos pateko į vidutinių koncentracijų gyvenamosiose ir visuomeninėse patalpose ribas [4].

1 lentelė. Įvairiomis sąlygomis atliktų 30 bandymų apibendrinanamoji oro kokybės parametrų statistika KTP patalpų aerozolyje

	Fonas* (Vidurkis ±SD)	Vidurkis	SN	95 % PI	Mediana	Minimali reikšmė	5-asis procentilis	25-asis procenti-lis	75-asis procentilis	95-asis procentilis	Maksimali reikšmė
DSK, #/cm³	88,5E+02± 7,9E+02	2,1E+05	3,1E+05	2,7E+03	1,1E+05	8,7	1,4E+04	7,9E+04	3,0E+05	4,5E+05	4,8E+05
$KD_{2,5}$, µg/m³	4,2±8,6	200,0	2339,4	19,8	11,4	0,0	2,6	5,3	32,5	119,4	635,7
KD_{10}, µg/m³	43,2±117, 1	1105	17000	144,2	19,3	0,0	3,1	5,9	35,5	188,7	874,9
CO_2, ppm	794±274	831	304	5,6	697	474	530	610	1008	1465	1960
CO, ppm	0,1±0,2	0,11	0,17	0,003	0,00	0,00	0,00	0,00	0,20	0,50	0,80
Formaldehidas, µg/m³	12,0±2,8	11,0	3,7	1,4	10,9	3,6	5,0	7,9	14,4	15,7	16,3
Acetaldehidas, µg/m³	3,2±1,2	5,7	2,8	1,1	4,9	1,4	2,0	3,6	7,1	10,9	12,0
Nikotinas, µg/m³	2,2±1,9	5,1	3,3	1,2	0,6	1,7	1,8	2,5	6,7	12,3	13,9
3-EP, µg/m³	0,6±0,2	0,7	0,2	0,1	0,3	0,3	0,4	0,5	0,7	1,1	1,1

ĮC naudojimas gerokai padidino visų išmatuotų analičių koncentracijas (žr. 2 lentelę).

2 lentelė. Aprašomoji oro kokybės parametrų statistika KTP ir ĮC patalpų aerozolyje, esant vienodoms eksperimentinėms sąlygoms

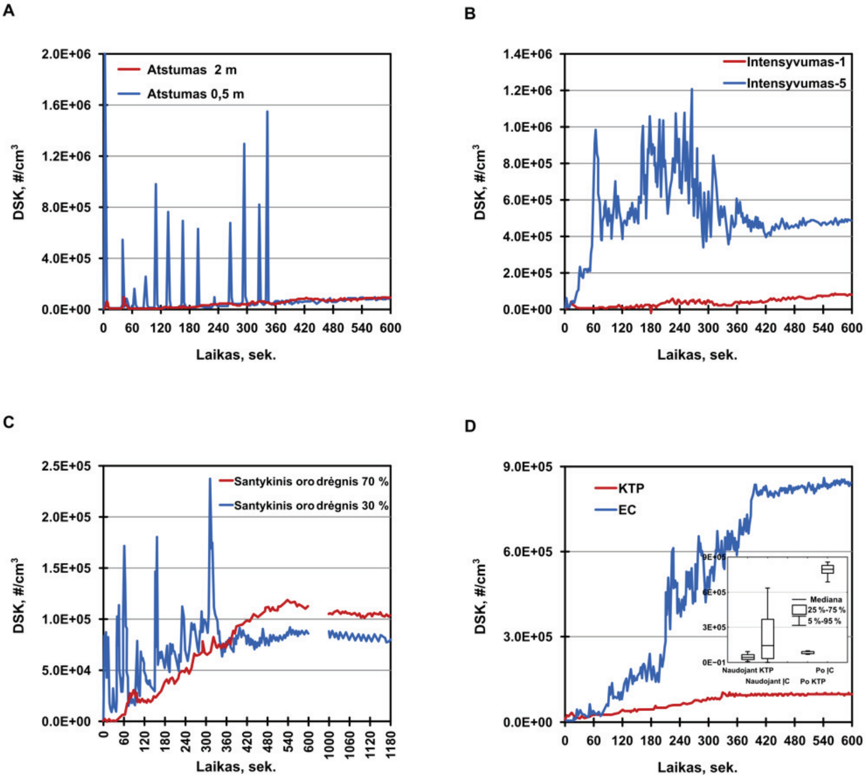
		Fonas* (Vidurkis±SD)	Vidurkis	SN	95 % PI	Mediana	Minimali reikšmė	5-asis procentilis	25-asis procentilis	75-asis procentilis	95-asis procentilis	Maksimali reikšmė
PNC, #/cm³	KTP	8,9E+02±5,4E+02	8,0E+04	3,2E+05	6,2E+02	8,6E+04	3,6E+02	2,6E+04	7,3E+04	9,7E+04	1,0E+05	1,1E+05
	ĮC	3,1E+02±2,7E+02	6,6E+05	2,6E+05	7,1E+03	7,8E+05	4,2E+02	3,2E+04	6,8E+05	8,2E+05	8,5E+05	9,2E+05
$PM_{2.5}$, µg/m³	KTP	3,3±4,1	5,7	7,6	0,2	5,6	0,4	2,6	3,5	6,8	8,7	311,5
	ĮC	1,3±8,4	159,4	57,1	1,5	169,8	0,2	22,8	148,9	192,8	225,5	796,1
PM_{10}, µg/m³	KTP	7,5±6,0	8,3	54,9	1,5	5,7	0,4	2,7	3,7	7,0	9,2	2352
	ĮC	5,8±21,1	179,8	191,2	5,1	171,5	0,2	28,7	150,9	195,4	232,1	4937
CO_2, ppm	KTP	620±58	644	65	3,7	647	501	537	597	690	754	793
	ĮC	580±63	610	57	3,2	613	498	511	568	651	699	767
CO, ppm	KTP	0,01±0,03	0,01	0,02	0,001	0,00	0,00	0,00	0,00	0,00	0,10	0,10
	ĮC	0,01±0,02	0,95	0,41	0,02	1,00	0,00	0,00	0,90	1,20	1,40	1,60
Formaldehidas, µg/m³	KTP	14,2±0,3	14,1	0,4	1,06	14,1**						
	ĮC	10,9±0,8	27,5	2,2	5,34	27,5**						
Acetaldehidas, µg/m³	KTP	3,3±0,1	3,6	0,4	0,94	3,6**						
	ĮC	3,6±0,8	28,9	2,6	6,51	28,9**						
Nikotinas, µg/m³	KTP	1,4±0,1	1,81	0,14	0,35	1,81**						
	ĮC	1,6±0,3	29,6	1,8	4,39	29,6**						
3-EP, µg/m³	KTP	0,5±0,1	0,56	0,03	0,09	0,56**						
	ĮC	0,5±0,1	3,7	0,3	0,65	3,7**						

* žmogaus fonas

** daroma prielaida, kad duomenys atitinka normalųjį skirstinį, mediana yra lygi vidutinei vertei

4.1.2 Realiojo laiko aerozolių dalelių koncentracijos nustatymas

Aerozolių DSK pokyčiai laike išmatuoti eksperimentų metu įvairiomis sąlygomis, keičiant: atstumą iki vartotojo, intensyvumą – vienu metu nikotino produktus vartojančių asmenų skaičių, santykinį oro drėgnį ir gaminio tipą, pateikti 5 pav.



5 pav. Dalelių skaitinės koncentracijos pokytis laike KTP patalpų aerozolyje, keičiant: A – atstumą; B – vartojimo intensyvumą; C – santykinį oro drėgnį; D – produkto tipą [4]

Atstumas

Kai atstumas nuo KTP vartotojo iki stebėtojo yra trumpas, nustatytos ryškios dalelių koncentracijos smailės (5 pav., A). Ši tendencija yra panaši į stebėtą ankstesniame tyrime naudojant EC [177]. Akivaizdų dalelių skaitinės koncentracijos lygio sumažėjimą 2 metrų atstumu galėjo lemti keli veiksniai: dalelių sklaida kambaryje judėjimo metu; aerozolio dalelių nusėdimas ant aplinkinių paviršių ir dalinis aerozolio dalelių garavimas. Didelę įtaką dalelių sklaidai iki šalia KTP vartotojo esančio stebėtojo turėjo aerozolio iškvėpimo kryptis. Tačiau šio veiksnio įtaka mažėja didėjant atstumui.

Vartojimo intensyvumas

KTP vartojimo intensyvumas turėjo tiesioginės įtakos aerozolių dalelių koncentracijai tiriamajame kambaryje. Esant penkiems aktyviems vartotojams, KTP dalelių skaitinė koncentracija patalpų aerozolyje buvo daug didesnė, lyginant su vieno vartotojo atveju (5 pav., B). Vartojimo intensyvumas vis dėlto yra gana abejotinas veiksnys, nes jis priklauso nuo rūkymo / naudojimo topografijos. Jis taip pat gali skirtis pagal įkvėpimo trukmę, aerozolio užlaikymo trukmę ar iškvėpimo trukmę.

Santykinis oro drėgnis

Dalelių skaitinių koncentracijų KTP patalpų aerozolyje kitimas laike, esant dviem skirtingiems santykinio oro drėgnio lygiams (30 % ir 70 %), pateiktas 5 pav., C. Paveikslas atspindi dalelių skaitinių koncentracijų skirtumus vieno vartotojo atveju, oro kaitos dažnis (OKD) – 1 h^{-1}, esant 2 metrų atstumui. Buvo tikimasi, kad ilgesnis kelias iki stebėtojo suteiks pakankamai laiko aerozolių dalelėms reaguoti su aplinkos drėgme. Dalelių nusėdimo greitis didėja didėjant oro santykinei drėgmei [179]; todėl, esant 70 % santykiniam drėgniui, neišryškėjo atskirų išpūtimų smailės, kurios aiškiai matomos esant 30 % santykiniam drėgniui.

Produkto tipas: KTP ir ĮC

Dalelių skaitinės koncentracijos kitimas KTP patalpų aerozolyje ir ĮC patalpų aerozolyje esant toms pačioms sąlygoms (intensyvumas – 1 vartotojas, atstumas – 2 m, OKD – 0,5 h^{-1} ir santykinis oro drėgnis – 50 %) pavaizduotas 5 pav., D. Triukšmo lygis laiko ašyje ĮC naudojimo atveju buvo didesnis, matomi didesni svyravimai, o KTP atveju matoma palyginti sklandi kreivė. Variacijos koeficientas „aktyviojoje“ vartojimo fazėje buvo 0,5 KTP ir 0,82 ĮC naudojimo atvejais. Per visą „aktyviąją“ fazę ĮC patalpų aerozolyje dalelių koncentracija buvo 5,6 karto didesnė (2,8E+05 #/cm^3) už dalelių koncentraciją (4,9E+04 #/cm^3) KTP aerozolyje, „pasyviojoje“ fazėje, matomas dar didesnis, 8,5 karto, skirtumas (8,0E+05 #/cm^3, palyginti su 9,5E+04 #/cm^3) tarp koncentracijų ĮC ir KTP patalpų aerozolyje.

4.1.3 Veiksnių, turinčių įtakos teršalų koncentracijos pokyčiui, atranka

Eksperimentiniams duomenims buvo pritaikytas regresijos modelis, šitaip nustatytas proceso veiksnių tarpusavio ryšys ir įtaka taršos lygiui. Apibendrinant, vienas svarbiausių veiksnių, turinčių teigiamą poveikį teršalų sklaidai, yra KTP vartojimo intensyvumas [4, 68].

Atstumas nuo KTP vartotojo iki stebėtojo turėjo įtakos $KD_{2,5}$ vidutiniams įverčiams – kuo toliau vartotojas, tuo mažesnė kietųjų dalelių koncentracija. Oro kaitos dažnis neturėjo reikšmingos įtakos jo vartojimo diapazone (nuo 0,2 iki 1 OKD). Santykinio oro drėgnio didinimas buvo teigiamai susijęs su $KD_{2,5}$ koncentracijos kitimu „pasyviojoje“ fazėje. Didesnė drėgmė lėmė didesnį garų slėgį, tokiu būdu juos stabilizavo ir neleido greitai išgaruoti lakiosioms aerozolio dalelėms. Dalelių dydis, skersmuo ir masė taip pat didėjo didėjant santykiniam drėgniui. Santykinis drėgnis buvo svarbus veiksnys, darantis įtaką aldehidų koncentracijai, mėginius imant iš 2,4-DNPH impregnuotų silicio dioksido kasečių [181].

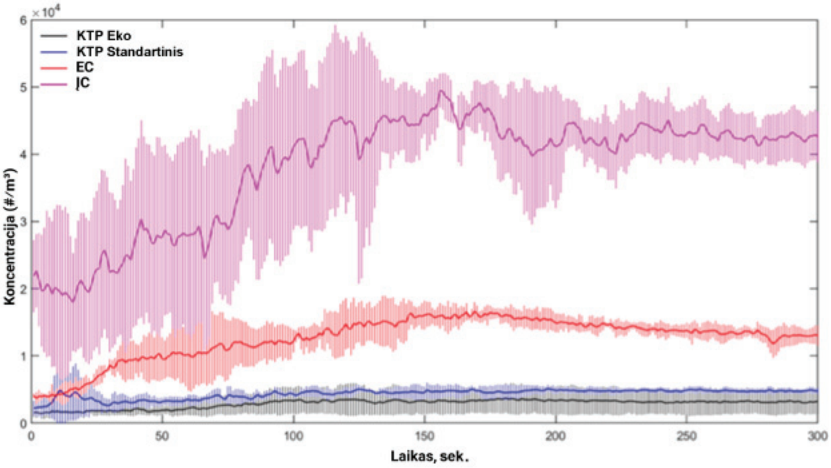
4.2 KTP, EC ir ĮC patalpų aerozolio palyginimas

Šiame skyriuje aprašoma KTP („Pulze“, veikiančio EKO ir standartiniais režimais) ir EC („myblu“) patalpų aerozolio dinamika, kai buvo keičiamas atstumas nuo stebėtojo iki trijų šių produktų vartotojų tiriamajame kambaryje, kuriame taip pat buvo kontroliuojamas vėdinimo intensyvumas ir mikroklimatas. KTP ir EC naudojimo metu gauti duomenys buvo lyginami su ĮC duomenimis. KTP patalpų aerozolyje dalelių skaitinė koncentracija vidutiniškai buvo mažesnė, palyginti su EC (4,3E+06 #/cm^3) ir ĮC (1,47E+08 #/cm^3), ir siekė 1,66E+06 #/cm^3 esant 0,5 m atstumui nuo vartotojo iki manekeno. Didelio skirtumo tarp EKO ir standartinio KTP veikimo režimų nepastebėta. Nustatyta, kad EC patalpų aerozolyje skaitinė dalelių koncentracija, išpūtus aerozolį, sumažėdavo greičiau, veikiausiai dėl didesnio dalelių lakumo. EC patalpų aerozolyje taip pat užfiksuota didesnė moda (120 nm), lyginant su KTP patalpų aerozoliu (90 nm), ir labai skyrėsi nuo ĮC (165–200 nm). KTP ir EC patalpų aerozolyje dalelių garavimas / sumažėjimas pastebėtas per 10 sekundžių po išpūtimo. Nustatyta, kad atstumas iki stebėtojo buvo svarbus veiksnys, turintis įtakos aerozolių dinamikai, o vėdinimo intensyvumas ir santykinis drėgnis neturėjo statistiškai reikšmingo poveikio. KTP ir EC patalpų aerozoliai pasižymi kitokiomis fizikinėmis ir cheminėmis savybėmis nei ĮC patalpų aerozolis, todėl skiriasi ir jų dinamika.

4.2.1 Dalelių skaitinės koncentracijos pokyčiai nikotino turinčių produktų vartojimo metu ir po vartojimo

Maksimali dalelių skaitinė koncentracija (DSK_t, *max*) KTP ir EC patalpų aerozolyje yra svarbi trumpalaikio kietųjų dalelių poveikio įvertinimui. Tačiau maksimalių koncentracijų smailės yra labai trumpalaikės (iki 5 sekundžių), todėl svarbu apibrėžti parametrus, atspindinčius aerozolio dinamiką prieš ir po šių smailių susidarymo. EC patalpų aerozolio dalelės, skirtingai negu ĮC patalpų aerozolyje, yra labai lakios, daugiausia sudarytos iš vandens ir propilenglikolio / glicerolio garų, todėl jos greitai išsisklaido patalpoje ir išgaruoja [43, 73, 158, 169]. Buvo iškelta hipotezė, kad po nikotino produkto vartotojo paskutinio aerozolio iškvėpimo bendra DSK (DSK_t) turėtų likti fono lygyje ir nedidėti, kai kambario tūris yra pripildytas dalelių, o ventiliacija negali pašalinti jų per dvi minutes.

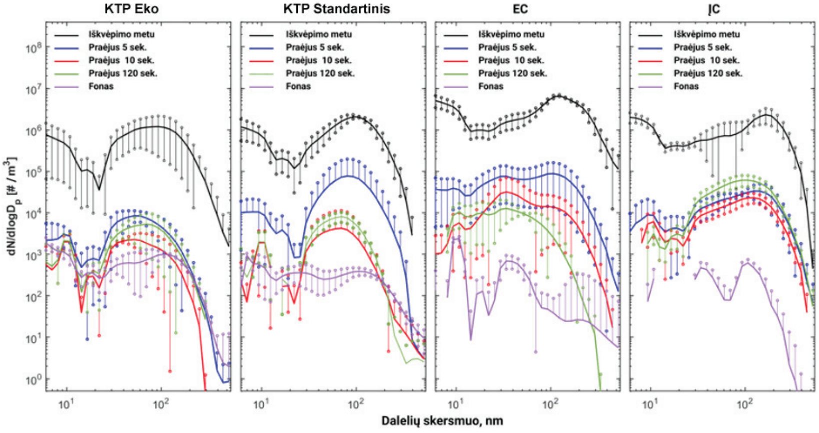
Būdinga DSK_t elgsena po nikotino produkto vartotojo paskutinio aerozolio iškvėpimo parodyta 6 paveiksle. Kai koncentracija nukrinta iki 10E + 3 #/cm^3 lygio, ji išlieka pastovi KTP atveju arba šiek tiek pakyla iki 1E + 4 #/cm^3 lygio EC atveju. Apskritai, EC naudojimas lėmė didesnes $DSK_{t,\ 120\ s}$, esant įvairiems atstumams iki stebėtojo. Šie rezultatai buvo netikėti, kadangi EC aerozolių skystosios dalelės yra lakesnės, o KTP aerozolių dalelės susidaro kaitinant tabako ir tirpiklio mišinį, todėl gali turėti nelakios medžiagos branduolį [183]. Šis fenomenas buvo patikrintas, apskaičiuojant $DSK_{t,\ 5\ s}$, $DSK_{t,\ 10\ s}$ ir $DSK_{t,\ 120\ s}$ santykius su $DSK_{t,\ max}$ maksimalia koncentracija, taip normalizuojant likutinę koncentraciją.



6 pav. DSK kitimas laike, matuojant iš stebėtojo pozicijos, pradedant nuo 20 sekundžių panaudojus KTP, EC (trijų savanorių vidurkis) ir ĮC naudojimo (duomenys paimti iš [177]). Atstumas tarp aktyvaus naudotojo ir stebėtojo 0,5 m, oro kaitos dažnis = 1 h^{-1}, santykinis oro drėgnis 30 % [27]

4.2.2 Dalelių dydžio pasiskirstymas

Dalelių dydžio pasiskirstymas (DDP) yra svarbus parametras atliekant eksperimentus su labai dinamiškais oro taršos šaltiniais, pvz., EC ir KTP. 7 paveiksle pateikti DDP duomenys, esant artimam atstumui (0,5 m) tarp stebėtojo ir nikotino produkto vartotojo aktyvaus vartojimo metu bei praėjus 5, 10 ir 120 s po nikotino produkto vartotojo paskutinio aerozolio iškvėpimo. Pateikti penkių iškvėpimų / išpūtimų vidutinis ir standartinis nuokrypiai, esant trims nikotino produktų vartotojams.



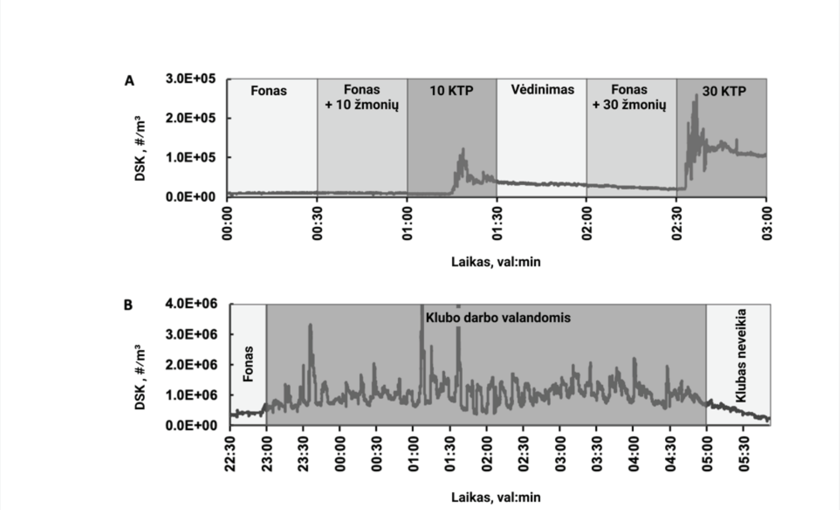
7 pav. Aerozolių dalelių dydžio pasiskirstymai (DDP) po iškvėpimo / išpūtimo panaudojus KTP, EC ir ĮC (šie duomenys paimti iš [177]), praėjus 5, 10 ir 120 sek. Atstumas tarp vartotojo ir stebėtojo yra 0,5 m, OKD = 1 h^{-1}, santykinis oro drėgnis 30 % [27]

KTP ir EC produktų DDP kreivės skyrėsi nuo įprastų cigarečių kreivių šiais aspektais: a) DDP augimas aerozolio išpūtimo metu buvo didesnis ĮC (165–200 nm) atveju, lyginant su KTP (93 nm) ir EC (123 nm); b) ĮC atveju DDP, bėgant laikui, beveik nepasikeitė ir neperėjo į mažesnių 100–200 nm dydžių diapazoną, o KTP (52–81 nm) ir EC (107 nm) atvejais stebėtas sumažėjimas. Tai patvirtina, kad KTP ir EC patalpų aerozolių dalelių sudėtis ir lakumas labai skiriasi nuo ĮC patalpų aerozolio.

4.3 Patalpų aerozolio vertinimas realaus gyvenimo aplinkoje

4.3.1 Aerozolio koncentracijos kitimas laike

Viso eksperimento metu naktiniame klube realiuoju laiku buvo matuojama aerozolio dalelių koncentracija. Eksperimentas atliktas dviem etapais: vartojant KTP (IQOS) neveikiančiame naktiniame klube (1 etapas) ir nevartojant KTP, bet matuojant oro teršalų koncentracijas veikiančiame naktiniame klube (2 etapas). Dalelių skaitinių koncentracijų kitimas laike abiem etapais parodytas 8 pav.



8 pav. Dalelių skaitinės koncentracijos pokyčiai, esant 10 aktyvių IQOS naudotojų neveikiančiame naktiniame klube (viršuje) ir klubo veikimo valandomis, kai patalpose nėra rūkoma (apačioje) [144]

Dalelių skaitinės koncentracijos KTP patalpų aerozolyje buvo 1E+4, 5E+4, 1E+5 #/cm^3, esant 10 ir 30 vartotojų neveikiančiame klube, ir 1E+6 iki 1E+7 #/cm^3 klubo veikimo metu. Klubo veikimo laikotarpiu (2 etapas) buvo gana didelė foninė dujinių teršalų koncentracija, greičiausiai dėl teršalų pernašos iš rūkomojo kambario. O KTP vartojimas neveikiančio klubo patalpose (1 etapas) neturėjo reikšmingos įtakos daugumos analičių koncentracijai, išskyrus nikotiną. Nepaisant padidėjusio dalelių skaičiaus ir masės koncentracijos KTP patalpų aerozolyje, šios vertės vis tiek buvo mažesnės nei klubo darbo valandomis.

4.3.2 Dujiniai teršalai

Klubo veikimo metu nustatyta vidutinė acetaldehido koncentracija buvo didesnė nei atliekant kitus eksperimentus. Tai gali būti paaiškinta alkoholinių gėrimų vartojimu veikiančiame naktiniame klube, nes acetaldehidas yra alkoholio metabolizmo produktas. Vartojant KTP abiem atvejais, kai klubo patalpose buvo 10 ar 30 žmonių, nikotino koncentracija padidėjo, tačiau 3-etenilpiridino koncentracija išliko nepakitusi. Nors naktinio klubo patalpose rūkyti yra draudžiama, klubo veikimo metu (2 etapas) nikotino ir 3-etenilpiridino koncentracijos buvo panašios į stebėtas eksperimentų su KTP metu (1 etapas) arba net viršijo jas. Tokie rezultatai gali būti paaiškinti užteršimu dujiniais teršalais iš šalia esančio rūkomojo. Remiantis gautais rezultatais, galima teigti, kad ĮC rūkymas išlieka labai svarbiu taršos šaltiniu net tais atvejais, kai patalpose nėra rūkoma.

5 REKOMENDACIJOS

1. Asmenims, esantiems šalia nikotino produkto vartotojo, rekomenduojama turėti omenyje, kad KTP, EC ir ĮC lemia pasyvųjį rūkymą. Nikotino produktų vartotojų skaičius turi tiesioginės įtakos aerozolių dalelių koncentracijai. Eksperimentai atlikti patalpoje, kurios tūris 35,8 m^3, o grindų plotas – 13 m^2. Lyginant DSK, esant penkiems aktyviems vartotojams arba vienam vartotojui, vidutinė DSK buvo 27 kartus didesnė aktyvaus vartojimo metu ir 8 kartus didesnė laikotarpiu po aktyvaus vartojimo, esant didesniam vartotojų skaičiui. Oro kaitos dažnis, 1 pasikeitimas per valandą, nepajėgus efektyviai pašalinti oro teršalų per 30 minučių. Intensyvus KTP ir EC naudojimas uždaroje erdvėje su ribota ventiliacija gali žymiai padidinti antrinio rūkimo pavojų, todėl turėtų būti nustatytos rūkymo patalpų vėdinimo normos.
2. Atstumas nuo nikotino produkto vartotojo iki stebėtojo yra dar vienas svarbus parametras, turintis įtakos oro teršalų koncentracijai. Tai ypač akivaizdu iškvėpimo / išpūtimo metu, kai pasiekiama didžiausia teršalų koncentracija. Antra vertus, lakūs skysčių lašeliai, susidarantys naudojant KTP ir EC, išsisklaido palyginti greitai ir turi mažesnę tikimybę pasiekti stebėtoją, kai atstumas tarp nikotino produkto vartotojo ir stebėtojo yra didesnis už 0,5 m.
3. Ore sklindančio nikotino ir acetaldehido koncentracija didėja dėl šių veiksnių: KTP aktyvių vartotojų skaičiaus (kai vartotojų skaičius padidinamas 3 kartus, DSK padidėja maždaug 3,6 karto), atstumo iki kito asmens (kai atstumas padidėja nuo 0,5 iki 2 m, DSK sumažėja maždaug 4,2 karto), vėdinimo intensyvumo ir santykinio oro drėgnio (kai santykinis drėgnis padidėja nuo 30 % iki 70 %, DSK sumažėja maždaug 1,9 karto). Vietas, kuriose vartojami NTP, rekomenduojama pritaikyti pagal šiuos parametrus. Rūkymui skirtas vietas rekomenduojama pritaikyti atsižvelgiant į šiuos parametrus.
4. Literatūros apžvalga atskleidė, kad tarptautinės organizacijos ir atskiros šalys neturi nustatytos ribinės nikotino vertės gyvenamųjų patalpų ore. Nikotino ribinę vertę gyvenamosiose ir viešosiose patalpose rekomenduojama nurodyti analogiškai kaip ir daugelio kitų patalpų oro teršalų.
5. Tretinis rūkymas, kaip parodė eksperimentai, atsiranda dėl oro taršos iš gretimų rūkymo patalpų. Net jei NTP patalpoje nenaudojami, nikotino koncentracija gali būti pakankama, kad atsirastų tretinio rūkymo pavojus. Rekomenduojama įvertinti ir plėtoti tretinio rūkymo prevencijos metodus, atliekant tolesnius oro valymo tyrimus, pvz., ozonavimą ir oro valytuvų su HEPA filtrais naudojimą. Rūkymo patalpose turi būti atskiros vėdinimo sistemos, kad būtų išvengta antrinio ir tretinio rūkymo pavojaus.
6. Remiantis šio darbo rezultatais rekomenduojama, kad rinkoje esantys NTP būtų kruopščiai ištirti kontroliuojamoje aplinkoje, siekiant tiksliai apibūdinti susidarančio aerozolio charakteristikas ir galimą poveikį oro kokybei bei žmonių sveikatai. Galbūt reikėtų sukurti specifiškų NTP kenksmingų ir potencialiai kenksmingų medžiagų (analičių) sąrašą, kad būtų galima išskirti KTP aplinkos aerozolį iš patalpų teršalų matricos.

7. Rekomenduotina atlikti kompiuterinį dalelių dinamikos modeliavimą. Šiuo tikslu būtų galima remtis skaičiavimo skysčių / fluidų dinamika (*CFD – Computational Fluid Dynamics*). Modeliavimo rezultatų palyginimas su empiriniais duomenimis padėtų geriau suprasti aerozolio dinamiką. Sukurtas modelis leistų numatyti taršos lygius vartojant įvairiau nikotino turinčius produktus, esant skirtingam jų naudojimo intensyvumui, erdvei ir mikroklimato sąlygoms.

Svarbu pabrėžti, kad visi nikotino turintys produktai turi būti vartojami atsakingai šalia kitų žmonių, ypač nėščiųjų ir vaikų. KTP ir EC patalpų aerozolių sudėtis rodo, kad tai gali būti daug saugesnis pasirinkimas už ĮC, tačiau vis dar nežinoma, ar ilgalaikis KTP ir EC naudojimas neturi neigiamų pasekmių. Geriausias būdas apsisaugoti nuo pasyviojo rūkymo – būti toliau nuo taršos šaltinio. Visais atvejais aktyvieji ir pasyvieji rūkaliai / NTP vartotojai turėtų būti supažindinti su moksliškai pagrįstomis nikotino turinčių produktų naudojimo pasekmėmis.

6 IŠVADOS

1. KTP vartojimas paprastai siejamas su mažesne oro tarša, panašia į kitų įprastinių taršos šaltinių sukeliamą taršą. Daugeliu atvejų KTP taršą galima atskirti nuo foninės, ir ji potencialiai siejama su ilgalaikiu poveikiu sveikatai kaip ir bet kuris kitas oro taršos šaltinis.

Tarp daugumos analizuotų taršos šaltinių, įskaitant KTP, EC, ĮC, kaljanus, smilkalus, uodų rites ir kitus, mažiausios formaldehido, benzeno, tolueno ir $KD_{2,5}$ koncentracijos buvo užfiksuotos naudojant KTP ir EC kontroliuojamoje aplinkoje. Daug didesnė tarša benzenu, toluenu ir formaldehidu pasitaiko viešojo naudojimo patalpose, ypač transporto mikroaplinkoje.

Tyrimų rezultatai rodo, kad svarbiausių KTP aerozolio oro taršos rodiklių koncentracijos gali būti per mažos, kad jas būtų galima atskirti nuo fono. Tokie duomenys kelia papildomų iššūkių epidemiologiniams tyrimams, skirtiems įvertinti nikotino produktų įtaką pasyviajam rūkymui realaus gyvenimo aplinkose. Manoma, kad yra būtina sukurti kenksmingų ir potencialiai kenksmingų rodiklių rinkinį, kuriuo remiantis būtų galima atskirti KTP taršą nuo kitų oro taršos šaltinių.

2. KTP vartojimas statistiškai reikšmingai padidino nikotino ir acetaldehido, (DSK) ir $KD_{2,5}$ koncentracijas patalpų aerozolyje lyginant su fonu. Palyginus su ĮC sukeliamu taršos lygiu tomis pačiomis sąlygomis, KTP aerozolyje nikotino, acetaldehido, DSK ir $KD_{2,5}$ koncentracijos buvo maždaug 16, 8, 8 ir 28 kartus mažesnės. Didžiausios nustatytos formaldehido (16,3 $\mu g/m^3$) ir acetaldehido (12,4 $\mu g/m^3$) koncentracijos atitinko vidutinę koncentraciją gyvenamosiose ir viešo naudojimo patalpose. Didžiausia 30 minučių DSK (4,8E+05 $\#/cm^3$) ir apskritai didžiausia DSK (9,3E+06 $\#/cm^3$) rodo, kad intensyvus KTP vartojimas uždaroje erdvėje su ribota ventiliacija gali reikšmingai padidinti aerozolio koncentraciją, nepaisant to, kad aerozolio dalelės yra labai lakios ir išgaruoja per kelias sekundes. Kaip patvirtina didžiausia dalelių koncentracija branduolio susidarymo stadijoje, KTP patalpų aerozolio dalelės, skirtingai nei ĮC, patalpų aerozolyje išgaruoja greičiau ir nespėja akumuliuotis tiriamojo kambario ore.

Vartojimo intensyvumas (ir kartu vartojančių skaičius) dominavo kaip svarbiausias veiksnys, lemiantis padidėjusias teršalų koncentracijas. Atstumas tarp naudotojo ir stebėtojo yra dar vienas svarbus veiksnys. Vienodomis sąlygomis vidutinė DSK, esant penkiems KTP vartotojams, buvo 27 kartus didesnė nikotino produkto „aktyviojoje“ fazėje ir 8 kartus didesnė „pasyviojoje“ fazėje, lyginant su vieno vartotojo atveju. Atstumo įtaka akivaizdi, analizuojant vidutinę DSK, kuri buvo 4,2 karto didesnė 0,5 m atstumu, lyginant su 2 m atstumu „aktyviojoje“ fazėje. Tačiau didesnis atstumas neturėjo reikšmingo poveikio DSK „pasyviojoje“ fazėje, kai atstumai yra 0,5 m ar 2 m. Oro kaitos dažnis pasirodė esąs mažiau svarbus veiksnys tirtame diapazone (iki 1 OKD). Tai rodo, kad tokio vėdinimo intensyvumo nepakako veiksmingam oro teršalų pašalinimui per 30 minučių, todėl reikėtų ilgesnio vėdinimo intensyvesniu režimu.

EC patalpų aerozolyje DSK buvo didesnė, vis dėlto taip pat greičiau ir sumažėdavo. Šio aerozolio dalelių dydis taip pat greičiau sumažėdavo dėl dalelių lakumo. EC ir KTP patalpų aerozolių panašumai, lyginant dalelių dydžio pasiskirstymą (DDP) ir dispersijos faktorius erdvėje, nepaisant koncentracijų ar pašalinimo greičio skirtumų, parodė, kad fiziniai aerozolių pernašos ir transformacijos mechanizmai yra panašūs (pvz., dalelių dydžio sumažėjimas dėl garavimo). Šie mechanizmai skyrėsi nuo tabako deginimo produktų, pvz., ĮC patalpų aerozolio, mechanizmų.

Vartojimo topografija įnešė daug neapibrėžties. Atliekant tyrimus ateityje ji turi būti griežtai standartizuota arba pripažinta pagrindiniu kintamuoju. Atstumas iki stebėtojo pasirodė esąs pagrindinis trumpalaikio poveikio kontrolės parametras. Siekiant sumažinti ilgalaikį NTP vartojimo poveikį, atstumas ir vėdinimo intensyvumas turėtų būti valdomi kartu.

3. KTP vartojimas naktiniame klube padidino dalelių skaitinę ir masės koncentracijas. Tačiau koncentracijos KTP patalpų aerozolyje neveikiančiame klube buvo mažesnės nei nustatytos klubo darbo valandomis. Rezultatai klubui veikiant rodo teršalų sklaidą iš gretimos rūkymo patalpos ir rūko mašinų. Sprendžiant pagal branduolio susidarymo stadijos dalelių frakciją, tiek KTP patalpų aerozolio neveikiančiame klube, tiek ir patalpų aerozolio veikiančiame klube dalelės buvo labai lakios. Tačiau KTP neturėjo reikšmingos įtakos daugumos dujinių teršalų koncentracijoms, kurių foninės vertės buvo gan didelės, veikiausiai dėl ilgalaikio tabako dūmų poveikio.

Intensyvus KTP vartojimas (pvz., 30 vartotojų) viešo naudojimo patalpose gali pabloginti oro kokybę dėl padidėjusių nikotino, acetaldehido bei kietųjų dalelių koncentracijų. Tačiau ĮC, nepaisant dalinio draudimo rūkyti, išlieka potencialiai didžiausias minėtų teršalų šaltinis viešo naudojimo patalpose.

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Appendix 1. List of references and summarized parameters in simulated indoor environments (chambers)

Test product	Pollutants taken for comparison	Analytical methods	Chamber type	Chamber volume, m^3	Air exchange, h^{-1}	Purification of supply air	Number of test products	Delay of sampling from the start of product use, h	Sampling duration, h	Reference
THS **Conventional cigarette**	benzene toluene formaldehyde acetaldehyde nicotine	active sampling on Anasorb sorbent tubes, GC MS active sampling on DNPH cartridges, GC MS active sampling Tenax TA sorbent tubes, GC MS	walk-in room imitating residential conditions	72.3	1.2	background reported	3/h 2 smokers	1	4	Mitova et al., 2016
Incense with spice fragrances	benzene toluene formaldehyde acetaldehyde	HS-PTR-MS, active sampling Tenax TA sorbent		32.3	0.8	the room was flushed during 12 h with outdoor air in order to reach the	1	0.5	1	Manoukian et al., 2013

		tubes, thermal desorption, GC MS active sampling on DNPH cartridges, HPLC-UV				atmospheric background level for VOCs and particles				
Incense traditional, aromatic, church incense	benzene toluene formaldehyde acetaldehyde $PM_{2.5}$	grab samples, GC MSD active sampling on DNPH cartridges, HPLC-UV dust-track air monitors	environmental test chamber	18.26 stainless steel	0.5	the chamber was purged by blower air, which was passed through a clean air system with activated charcoal particle filters and high-efficiency particulate air (HEPA) filters	3 at a time, 10 different tested	0 when burning 0.5 after the incense has extinguished	~0.5–1 depending on an incense type	Lee, Wang, 2004
Scented and unscented candles	benzene toluene formaldehyde	active sampling Tenax TA sorbent	ISO 9001:2000 registered	26	0.5 1	after each experiment, the testing chambers	1 3	2 6	0.5	Petry et al., 2014

	acetaldehyde PM$_{2.5}$	tubes, thermal desorption, GC MS active sampling on DNPH cartridges, HPLC-UV continuous particle concentration monitor, gravimetric method	indoor air quality laboratory, stainless steel and aluminium surfaces			were cleaned by flushing with HEPA filtered ambient air for at least 6 h				
Mosquito coils scented candles	benzene toluene formaldehyde acetaldehyde PM$_{2.5}$	grab samples, GC MSD active sampling on DNPH cartridges, HPLC-UV dust-track air monitors	environmental test chamber	18.26 stainless steel	0.5	the chamber was purged by blower air, which was passed through a clean air system with activated charcoal particle filters and high-efficiency	1 at a time, 5 different mosquito coils and 5 different candles tested	0 when burning 0.5 after the tested specimens have extinguished	1 for mosquito coils ~1.5–2 depending on a candle type	Lee, Wang, 2006)

						particulate air (HEPA) filters				
THS, e-cigarette, conventional cigarette	formaldehyde acetaldehyde $PM_{2.5}$	active sampling on DNPH cartridges, HPLC PM mass monitor particle counter	sitting room of a flat owned by habitual smokers	48	1.54		10 iQOS with menthol and 14 without menthol in sequence 13 EC in sequence 9 CC in sequence smokers	0	3 iQOS ~2–3 EC 1.5	Ruprecht et al., 2017
E-cigarette, conventional cigarette	benzene formaldehyde acetaldehyde $PM_{2.5}$	active sampling on Tenax GR, thermal desorption, GC MS active sampling on DNPH cartridges, HPLC LAS	stainless-steel emission test chamber	8	0.3		3 at a time, 3 smokers, 3 e-cigarette liquids tested	at puff 4	2	Schripp et al., 2013

E-cigarette	formaldehyde acetaldehyde benzene nicotine	active sampling on DNPH cartridges, HPLC active sampling on Tenax GR, thermal desorption, GC MS	office room	45	0.56	background reported	3 at a time, 3 nicotine free and 3 nicotine containing liquids tested	0	2	Schober et al., 2014
E-cigarette	nicotine	active sampling on Tenax TA tubes, GC–MS	walk-in environmental chamber	30	0.5	the chamber was filled with ultra-clean pre-dried and filtered air	1 at a time, 6 replicates for each of 7 liquids smoking machine	0		Geiss et al., 2011
E-cigarette	nicotine $PM_{2.5}$	active sampling on XAD-7 tubes, GC light-scattering integrating		0.4425	1.53	in order to improve the reliability of the experimental results, the system was purged with pure air	2 at a time, 2 puffing intensities and 2 liquids tested			Lee et al., 2017

		nephelometer								
Water pipe	benzene nicotine $PM_{2.5}$	active sampling on Tenax GR, thermal desorption, GC MS LAS WRAS	office room	57	not indicated	background reported	4 tablets in sequence, 4 smokers	0	4	Fromme et al., 2009

Appendix 2. List of references and summarized parameters on the “real-life” indoor environments

Environment type, location	Number of investigated environments	Pollutants taken for the comparison	Analytical methods	Air exchange, h^{-1}	Reference
suburban houses, mostly detached Perth, Australia	study 1: 68 houses study 2: 250 houses study 3: 69 houses	formaldehyde benzene toluene	DNPH treated glass filters exposed for 24 h, HPLC passive diffusion over 7 days (study 1, 2) active sampling on Radiello and Custom (study 3) samplers (study 3), GCMS	not specified	Maisey et al., 2013
urban houses, mostly detached Edmonton, Canada	26 houses in summer and winter	formaldehyde acetaldehyde benzene toluene	7 consecutive 24-h periods passive collection into Summa canisters, GCMS HPLC for carbonyls	not specified	Bari et al., 2015
low VOC emitting and conventional new houses, detached California, USA	9 houses with controlled ventilation 13 houses with low VOC emitting materials	formaldehyde acetaldehyde	active sampling on DNPH cartridges, HPLC	0.2; 0.4; 0.8 0.08–0.48	[86]
suburban houses in Melbourne, Australia	31 detached 9 semi-detached or flat	formaldehyde acetaldehyde benzene toluene	active sampling on DNPH cartridges, HPLC active sampling on multi-sorbent tubes, GC MS	closed-up state 0.5 typical open state 5.7	[87]

			7 days sampling		
high-performance houses, California, USA	24 new or deeply retrofitted houses	formaldehyde acetaldehyde	active sampling on DNPH Sep-Pak cartridges, HPLC 6 days sampling	~0.3	Less et al., 2015
residential houses nationwide, Japan	602 in winter and summer	formaldehyde acetaldehyde benzene toluene	passive sampling on DSD-BPE/DNPH cartridges, HPLC passive sampling on VOC-SD, GC MS 24 h sampling	not specified	[201]
new energy-efficient houses, detached Centre, Pays-de-la-Loire, Ile-de-France and Rhône-Alpes, France	7 in winter and summer	formaldehyde acetaldehyde benzene toluene	passive sampling by DNPH coated Florisil cartridges, HPLC passive sampling by Carbograph 4 adsorbents, GC MS, FID 7 days sampling	0.02–1	Derbez et al., 2014
new apartment houses with low emitting materials, Helsinki, Turku, Finland	14 apartments in 8 houses	formaldehyde benzene toluene	active sampling into a sulphuric acid-solution, spectrometric acetyl-acetone method active sampling on Tenax TA tubes, GC MS, FID	0.79–1.61	Järnström et al., 2006

residential houses nationwide, France	567	formaldehyde acetaldehyde benzene	passive sampling by DNPH coated Florisil cartridges, HPLC passive sampling by Carbograph 4 adsorbents, GC MS, FID 7 days sampling	0.65 night time	[92]
new low energy houses, detached Lithuania	11	formaldehyde benzene toluene	passive sampling by DNPH coated Florisil cartridges, HPLC passive sampling by charcoal adsorbents, GC MS 7 days sampling	0.08–0.69	[203]
residential apartment houses, Dalian, China	53 in summer 100 in winter	formaldehyde benzene toluene	passive sampling on Sep-Pak DNPH-Silica cartridge, HPLC passive sampling on activated charcoal cartridges, GC MS 24 h sampling	not specified	[95]
residential houses, public buildings and schools/ kindergartens in eleven European cities	103 private homeplaces, 182 different working environments (offices, classrooms, waiting halls)	formaldehyde acetaldehyde benzene toluene	passive sampling by DNPH coated cartridges, HPLC passive sampling on activated charcoal cartridges, GC FID	not specified	[96]

			7 days sampling		
low-energy school buildings, northern and eastern France	10	benzene toluene formaldehyde acetaldehyde	passive sampling on activated charcoal cartridges, GC FID passive sampling by DNPH coated cartridges, HPLC 7 days sampling	2.05–3.33	[204]
early childhood education facilities, California, the USA	40	formaldehyde acetaldehyde	passive sampling on Sep-Pak DNPH-Silica cartridge, HPLC	0.28–5.63	[98]
schools, Porto, Portugal	73 classrooms	formaldehyde acetaldehyde benzene toluene	passive sampling by DNPH coated cartridges, HPLC passive sampling on Tenax TA tubes, GC MS, FID 5 days sampling	not specified	Madureira et al., 2016
office, suburb of Athens, Greece	1	formaldehyde benzene toluene	active sampling on DNPH cartridges, HPLC active sampling on Tenax TA tubes, GC FID	not specified	Saraga et al., 2011
shopping mall, four cities, Western China	4 malls, 67 sampling sites	formaldehyde	analyser Formaldehyde 400	not specified	[102]

shopping mall, suburbs of Bari, Italy	1st campaign: 96 samples 2nd campaign: 146 samples	benzene toluene	passive sampling on Carbograph 4 cartridges, GC FID 3 x 48 h sampling	not specified	[93]
coffee shops, libraries, copy centres, pharmacies, newspaper stands, offices, gymnasiums, hairdressers, restaurants and supermarkets, Bari, Italy	24 different environments	benzene toluene	passive sampling on Carbograph 4 cartridges, GC MS 7 x 24 h sampling	not specified	Bruno et al., 2008
used cars, driven, Varese, Northern Italy	23 car cabins	formaldehyde acetaldehyde benzene toluene	passive sampling by DNPH coated cartridges, HPLC passive sampling on activated charcoal cartridges, GC FID 7 days sampling	not specified, some of the driven cars had open windows, some had air conditioning	Geisset al., 2009
new cars, unoccupied, Beijing, China	monitoring was conducted in 802 car cabins in an underground ventilated parking garage	formaldehyde benzene toluene	active sampling by hydroxybenzene solution, spectrophotometry active sampling on activated charcoal cartridges, GC FID	< 0.01–0.63	[99]
new and used cars, Poland	10 new cars, unoccupied 2 cars, driven at the mileage of	benzene toluene	active sampling on Carbograph 1TD cartridges, GC FID, MS	not specified	Faberet al., 2013

	2100 to 20200 km				
trams, cars, Ghent, Belgium	144 samples (3 days x 4 times/day x 6 sections x 2 modes of transport)	benzene toluene	active sampling on Tenax TA tubes, GC MS	not specified	[106]
buses highways, Taiwan	16 one-way journeys	formaldehyde benzene toluene	formaldemeter active sampling on Tenax TA tubes, GC FID 2 h sampling	not specified	[107]

Appendix 3. The experimental plan with variables and responses (THS: Exp. No. 1–30; CC: Exp. No. 31–33)

Exp. No.	Run order	Air change rate	Intensity	Humidity	Distance to the bystander	Product type
1	12	0.2	1	30	0.5	THS
2	15	0.5	1	30	0.5	THS
3	8	1	1	30	0.5	THS
4	25	0.2	5	30	0.5	THS
5	19	0.5	5	30	0.5	THS
6	28	1	5	30	0.5	THS
7	11	0.2	1	70	0.5	THS
8	9	0.5	1	70	0.5	THS
9	27	1	1	70	0.5	THS
10	21	0.2	5	70	0.5	THS
11	1	0.5	5	70	0.5	THS
12	30	1	5	70	0.5	THS
13	18	0.2	1	30	2	THS
14	7	0.5	1	30	2	THS
15	4	1	1	30	2	THS
16	26	0.2	5	30	2	THS
17	13	0.5	5	30	2	THS
18	14	1	5	30	2	THS
19	3	0.2	1	70	2	THS
20	24	0.5	1	70	2	THS
21	17	1	1	70	2	THS
22	22	0.5	5	70	0.5	THS
23	2	1	5	30	0.5	THS
24	10	0.2	5	70	0.5	THS
25	16	0.5	3	50	1	THS
26	6	0.5	3	50	1	THS
27	23	0.5	3	50	1	THS
28	29	0.5	1	50	2	THS
29	20	0.5	1	50	2	THS
30	5	0.5	1	50	2	THS
31	31	0.5	1	50	2	Conv. cig.
32	32	0.5	1	50	2	Conv. cig.
33	33	0.5	1	50	2	Conv. cig.

Appendix 4. Comparison of normalised concentrations at (I-1, V-0.5, D-2, RH-50) with the results from other studies

Pollutant	Mean, μ/m^3	Normalised mean, μ/m^3	Median, μ/m^3	Normalised median, μ/m^3	Volume of the chamber, m^3	Air ex-change, 1/h	Number of used products	Reference
THS								
Formaldehyde	2.7	1.0			48	1.54	14	Ruprecht et al., 2017
	no statistical difference from the background							Mitova et al., 2016
	no statistical difference from the background							This study
Acetaldehyde			3.7	1.9	72.3	2.16	20	Office, Mitova et al., 2016
			5.1	2.0	72.3	1.2	15	Residential, Mitova et al., 2016
			1.4	1.3	72.3	7.68	40	Hospitality, Mitova et al., 2016
			3.5	1.2	48	1.54	14	Ruprecht et al., 2017
	no statistical difference from the background							This study
Nicotine			1.1	0.6	72.3	2.16	20	Office, Mitova et al., 2016
			1.8	0.7	72.3	1.2	15	Residential, Mitova et al., 2016
			0.7	0.6	72.3	7.68	40	Hospitality, Mitova et al., 2016
			0.4	0.5	35.8	0.5	1	This study
$PM_{2.5}$	6.5	2.3			48	1.54	14	Ruprecht et al., 2017
	2.4	2.9			35.8	0.5	1	This study

PNC	27745	9766			48	1.54	14	Ruprecht et al., 2017
	79110	94405			35.8	0.5	1	This study
CC								
Formaldehyde	40.0	21.9			48	1.54	9	Ruprecht et al., 2017
			28.9	15.0	72.3	2.16	20	Office, Mitova et al., 2016
			35.5	13.7	72.3	1.2	15	Residential, Mitova et al., 2016
			17.5	16.2	72.3	7.68	40	Hospitality, Mitova et al., 2016
			16.6	19.8	35.8	0.5	1	This study
Acetaldehyde	71.0	38.9	71.0	38.9	48	1.54	9	Ruprecht et al., 2017
			58.8	30.6	72.3	2.16	20	Office, Mitova et al., 2016
			83.8	32.3	72.3	1.2	15	Residential, Mitova et al., 2016
			33.1	30.6	72.3	7.68	40	Hospitality, Mitova et al., 2016
			25.3	30.2	35.8	0.5	1	This study
Nicotine			34.7	18.1	72.3	2.16	20	Office, Mitova et al., 2016
			29.1	11.2	72.3	1.2	15	Residential, Mitova et al., 2016
			34.6	32.0	72.3	7.68	40	Hospitality, Mitova et al., 2016
			28	33.4	35.8	0.5	1	This study
$PM_{2.5}$	500.0	273.8			48	1.54	9	Ruprecht et al., 2017

	158.1	188.7			35.8	0.5	1	This study
PNC	122672	67170			48	1.54	9	Ruprecht et al., 2017
	659690	787230			35.8	0.5	1	This study

Appendix 5. Air quality parameters as an input to the regression model during the use of THS (Exp. No. 1–30) and CC (Exp. No. 31–33)

Exp. No.	PNC mean (All), #/cm^3	PNC mean (After), #/cm^3	PM2.5 mean (All), µg/m^3	PM2.5 mean (After), µg/m^3	PM2.5 max, µg/m^3	CO dummy, ppm	CO2 dummy, ppm	Formaldehyde, µg/m^3	Acetaldehyde, µg/m^3	Nicotine, µg/m^3	4-EP, µg/m^3
1	1.5E+05	1.0E+05	199.7	7.5	4.4E+04	0.0	765	11.1	4.6	6.2	1.0
2	1.3E+05	9.0E+04	294.2	11.2	9.5E+04	0.0	665	4.9	1.8	6.1	1.1
3	1.5E+05	7.4E+04	417.9	6.5	9.4E+04	0.0	633	3.6	1.4	3.6	0.4
4	4.5E+05	4.0E+05	525.4	37.1	7.3E+04	0.0	1124	10.7	6.6	11.8	0.8
5	3.2E+05	2.7E+05	503.6	34.2	5.0E+04	0.0	1170	10.5	9.4	12.8	0.5
6	3.0E+05	2.8E+05	339.5	26.0	4.9E+04	0.0	1030	5.6	3.8	4.7	0.5
7	6.8E+04	7.9E+04	57.5	8.2	3.0E+04	0.0	613	14.4	5.6	2.5	0.6
8	8.8E+04	8.5E+04	149.9	5.4	5.0E+04	0.3	637	14.5	4.6	3.0	1.0
9	7.5E+04	7.3E+04	149.0	5.9	3.9E+04	0.0	617	6.3	7.6	2.7	0.6
10	4.0E+05	4.3E+05	401.6	46.7	7.4E+04	0.0	1132	15.3	11.3	6.9	0.7
11	3.7E+05	3.7E+05	635.7	58.4	7.7E+04	0.4	1136	16.3	12.0	13.9	1.0
12	3.1E+05	3.0E+05	576.4	43.7	1.0E+05	0.5	963	15.3	9.5	7.1	0.7
13	7.0E+04	8.5E+04	5.7	6.0	3.5 x101	0.0	571	6.9	3.3	2.1	0.5
14	5.9E+04	7.9E+04	5.2	5.5	3.8 x101	0.0	585	7.4	3.5	2.0	0.3
15	8.0E+04	8.2E+04	9.4	5.2	1.5E+03	0.2	584	7.6	2.2	2.3	0.4
16	4.8E+05	4.6E+05	82.2	37.1	2.2E+03	0.3	1027	10.4	7.3	7.0	0.7
17	3.7E+05	4.3E+05	39.3	32.5	2.8E+03	0.3	1027	11.0	6.3	4.6	0.5
18	2.7E+05	2.9E+05	31.1	22.8	2.0E+03	0.0	1050	5.0	9.6	7.4	0.7
19	8.2E+04	9.7E+04	5.8	4.0	4.3E+02	0.0	635	13.2	4.5	1.9	0.6
20	7.2E+04	8.7E+04	4.3	3.8	5.1E+02	0.1	634	13.8	4.4	2.6	1.1
21	9.0E+04	1.0E+05	6.1	6.9	6.0 x101	0.0	608	16.0	4.6	8.5	1.1
22	3.5E+05	3.6E+05	448.4	77.5	4.9E+04	0.3	1188	15.3	10.3	6.1	0.6
23	3.5E+05	3.2E+05	306.7	30.1	1.1E+05	0.0	-	9.0	5.7	5.3	0.5

24	3.1E+05	3.4E+05	344.6	87.0	3.7E+04	0.3	1309	12.8	3.6	5.8	0.7
25	1.9E+05	2.1E+05	85.4	21.6	1.8E+04	0.0	814	10.2	5.4	3.7	0.5
26	2.4E+05	2.4E+05	160.1	23.1	2.5E+04	0.1	809	10.7	5.2	3.4	0.5
27	2.7E+05	2.7E+05	184.9	25.4	2.1E+04	0.2	821	10.7	5.8	4.6	0.6
28	6.8E+04	7.5E+04	3.4	3.3	3.3 x101	0.0	641	14.5	3.4	1.7	0.6
29	9.1E+04	1.0E+05	7.5	7.0	3.1E+02	0.0	602	14.2	3.4	1.8	0.6
30	8.2E+04	9.1E+04	6.1	6.3	3.9 x101	0.0	688	13.6	4.0	2.0	0.5
31	7.0E+05	8.1E+05	155.3	171.8	6.0E+02	1.0	625	25.2	31.9	30.7	4.0
32	6.2E+05	7.7E+05	168.4	197.6	3.7E+02	1.0	599	29.5	28.1	30.5	3.8
33	6.6E+05	7.8E+05	154.6	172.6	8.0E+02	0.9	608	27.7	26.9	27.5	3.5

Appendix 6. Summary table of general air quality parameters at various experiment conditions and different days in a nightclub

Backgr.+10p. – background with 10 humans present but without THS use; Backgr.+30p. - background with 30 humans present but without THS use; 10 IQOS - 10 humans simultaneously using THS; 30 IQOS - 30 humans simultaneously using THS; Z1 – zone 1; Z2 – zone 2

Date	**Experiment conditions**	**PNC, #/cm^3**	**PM$_{2.5}$, µg/m^3**	**PM$_{10}$, µg/m^3**	**Nicotine, µg/m^3**	**Formaldehyde, µg/m^3**	**Acetaldehyde, µg/m^3**	**3-EP, µg/m^3**	**CO$_2$, ppm**	**T, °C**	**RH, %**
2018-01-30	Backgr. Z1	4.6E+03	1.2	1.5	3.6	32.2	43.7	1.6	616	14	43
	Backgr. Z2	4.9E+03	1.3	3.1	3.0	26.7	30.4	1.5	552	14	43
	Backgr.+10p. Z1	6.5E+03	2.3	4.1	5.9	28.7	38.4	1.5	732	14	43
	Backgr.+10p. Z2	5.9E+03	2.8	10.1	6.6	27.3	32.9	1.5	661	14	43
	10 IQOS Z1	3.4E+04	6.1	6.6	7.7	30.9	37.9	1.4	896	15	43
	10 IQOS Z2	1.7E+04	6.9	7.9	8.8	27.8	33.0	1.6	841	15	43
	Backgr.+30p. Z1	3.9E+04	5.7	52.5	7.6	21.7	22.8	1.2	925	15	41
	Backgr.+30p. Z2	3.3E+04	5.2	48.5	7.7	26.0	24.0	1.3	857	15	41
	30 IQOS Z1	1.2E+05	10.8	11.0	10.1	24.5	25.6	1.4	1208	16	43
	30 IQOS Z2	1.2E+05	9.2	9.3	9.3	33.1	30.2	1.6	1126	16	42
2018-01-31	Backgr. Z1	9.5E+03	2.8	14.4	2.4	42.4	41.4	1.7	597	18	38
	Backgr. Z2	9.8E+03	2.8	16.2	2.5	40.5	37.2	1.6	542	18	37
	Backgr.+10p. Z1	8.8E+03	3.4	15.6	5.0	40.9	40.6	1.7	734	18	38
	Backgr.+10p. Z2	9.8E+03	3.7	20.8	5.5	42.2	39.7	1.6	671	18	37

	10 IQOS Z1	3.7E+04	4.4	4.5	7.5	40.6	39.7	2.1	850	18	38
	10 IQOS Z2	4.3E+04	4.7	4.9	8.1	44.3	42.2	2.0	792	19	38
	Backgr.+30p. Z1	2.6E+04	8.8	10.7	4.4	27.1	19.9	2.2	786	18	34
	Backgr.+30p. Z2	2.8E+04	9.7	19.4	5.0	28.4	19.7	1.6	716	19	34
	30 IQOS Z1	1.1E+05	11.0	11.2	11.5	31.0	22.7	1.5	1058	19	36
	30 IQOS Z2	1.2E+05	12.3	12.3	10.7	37.6	25.2	1.5	963	20	35
2018-02-01	Backgr. Z1	2.8E+04	14.8	17.1	4.8	40.8	49.2	1.9	635	18	38
	Backgr. Z2	2.8E+04	14.0	14.7	4.1	41.9	44.5	1.7	574	18	38
	Backgr.+10p. Z1	2.6E+04	13.8	17.7	6.0	41.7	49.1	1.9	747	18	39
	Backgr.+10p. Z2	2.5E+04	13.5	18.3	5.2	35.6	41.9	2.1	682	18	38
	10 IQOS Z1	5.2E+04	12.0	12.0	6.4	40.9	50.4	1.9	849	18	39
	10 IQOS Z2	5.1E+04	12.1	12.1	6.6	44.2	51.5	2.2	777	18	39
	Backgr.+30p. Z1	5.2E+04	23.8	24.7	5.3	31.2	30.8	1.5	923	18	39
	Backgr.+30p. Z2	4.8E+04	22.0	22.3	6.2	33.5	30.9	1.6	820	18	38
	30 IQOS Z1	1.5E+05	24.5	24.9	11.9	35.4	35.8	1.7	1202	18	41
	30 IQOS Z2	1.5E+05	23.3	23.3	3.8	35.1	32.6	2.3	1074	18	41
2018-02-02	Backgr. Club OP Z1	5.9E+04	20.1	23.8	3.5	32.3	26.0	1.4	829	18	39
	Backgr. Club OP Z2	5.1E+04	18.5	19.9	4.4	29.8	22.1	1.7	724	18	40
	Club OP Z1	6.4E+05	7301	8743	4.2	28.0	42.8	1.3	1590	21	37
	Club OP Z2	8.3E+05	1145	1174	6.6	35.1	57.7	3.7	1432	21	37

2018-02-03	Backgr. Club OP Z1	3.1E+05	63.8	63.9	5.2	26.8	23.8	0.8	596	19	36
	Backgr. Club OP Z2	3.1E+05	66.6	66.6	14.9	21.6	17.5	1.3	574	19	37
	Club OP Z1	7.7E+05	2748	3001	6.8	23.4	35.1	4.0	1625	21	36
	Club OP Z2	4.9E+05	7859	8862	22.9	28.1	50.5	2.2	1474	21	36
2018-02-09	Backgr. Club OP Z1	4.2E+05	68.0	68.0	5.0	20.8	17.3	0.5	863	17	28
	Backgr. Club OP Z2	4.1E+05	33.2	33.3	8.2	17.2	13.8	0.8	771	17	28
	Club OP Z1	9.2E+05	2084	1977	6.3	24.6	36.9	0.7	1477	20	29
	Club OP Z2	9.8E+05	1718	1738	13.4	25.6	42.1	1.8	1331	20	29

Appendix 7. Experimental worksheet

Run number	Device	Distance, m	ACH, h^{-1}	RH, %	Volunteer's No.	Conc max	Conc 5 s (total)	Conc 10 s	Conc 120 s	Conc 5 s (6 nm)	Conc 10 s (6 nm)	Conc 120 s (6 nm)	Mode Max	Mode 5 s	Mode 10 s	Mode 120 s	5/max	10/max	120/max	(lnMax-Ln5)/(5/60)	(lnMax-Ln10)/(10/60)
1	HTP_Eco	0.5	0.2	30	v1	1.01E+05	7.01E+03	8.41E+02	8.11E+02	6.71E+02	3.45E+01	6.97E+01	9.31E+01	6.04E+00	8.06E+01	9.31E+01	6.94E-02	8.33E-03	8.03E-03	3.20E+01	2.87E+01
2	HTP_Std	0.5	0.2	30	v1	2.88E+06	2.98E+04	5.03E+02	1.81E+03	1.39E+02	1.49E+01	1.08E+02	1.43E+02	9.31E+01	4.53E+01	9.31E+01	1.03E-02	1.75E-04	6.27E-04	5.49E+01	5.19E+01
12	EC	0.5	0.2	30	v1	2.47E+06	1.95E+04	1.64E+04	7.38E+03	3.05E+02	5.10E+02	1.33E+02	1.24E+02	3.40E+01	3.40E+01	2.94E+01	7.89E-03	6.64E-03	2.99E-03	5.81E+01	3.01E+01
28	HTP_Eco	1	0.2	30	v1	8.66E+05	9.69E+04	1.02E+04	2.06E+03	4.87E+03	2.20E+02	6.25E-02	1.24E+02	8.06E+01	8.06E+01	4.53E+01	1.12E-01	1.18E-02	2.38E-03	2.63E+01	2.66E+01
26	HTP_Std	1	0.2	30	v1	1.51E+06	1.14E+05	2.31E+04	2.03E+03	2.73E+03	9.08E+02	6.25E-02	1.24E+02	8.06E+01	8.06E+01	6.04E+01	7.55E-02	1.53E-02	1.35E-03	3.10E+01	2.51E+01
3	EC	1	0.2	30	v1	1.74E+06	1.88E+05	4.26E+03	3.79E+03	1.20E+04	2.60E+02	1.20E+02	1.43E+02	6.04E+00	3.92E+01	3.40E+01	1.08E-01	2.45E-03	2.18E-03	2.67E+01	3.61E+01
25	HTP_Eco	2	0.2	30	v1	9.99E+03	2.83E+03	2.32E+03	5.00E+03	1.00E-02	1.00E-02	6.25E-02	6.98E+01	6.04E+01	6.98E+01	5.23E+01	2.83E-01	2.32E-01	5.00E-01	1.51E+01	8.76E+00
20	HTP_Std	2	0.2	30	v1	1.66E+04	9.80E+03	4.82E+03	1.03E+04	3.23E+02	1.14E+02	1.08E+02	6.04E+01	5.23E+01	5.23E+01	5.23E+01	5.90E-01	2.90E-01	6.21E-01	6.32E+00	7.42E+00
9	EC	2	0.2	30	v1	2.79E+04	1.09E+04	1.09E+04	1.16E+03	1.56E+02	6.16E+02	5.92E+01	1.08E+02	3.40E+01	3.40E+01	2.55E+01	3.91E-01	3.91E-01	4.17E-02	1.13E+01	5.64E+00
19	HTP_Eco	0.5	1	30	v1	1.64E+06	7.82E+03	2.26E+03	5.19E+03	3.77E+02	1.05E+02	1.21E+02	8.06E+01	6.04E+01	6.04E+01	6.98E+01	4.77E-03	1.38E-03	3.16E-03	6.41E+01	3.95E+01
11	HTP_Std	0.5	1	30	v1	1.79E+06	6.98E+03	1.90E+02	3.16E+03	5.56E+02	4.31E+01	9.34E+01	9.31E+01	6.98E+01	6.04E+00	6.98E+01	3.90E-03	1.06E-04	1.76E-03	6.66E+01	5.49E+01
27	EC	0.5	1	30	v1	5.77E+06	7.02E+03	6.16E+02	1.21E+04	1.00E-02	1.00E-02	6.25E-02	1.08E+02	3.40E+01	1.08E+01	3.40E+01	1.22E-03	1.07E-04	2.10E-03	8.05E+01	5.49E+01
4	HTP_Eco	1	1	30	v1	9.45E+05	3.54E+03	1.79E+03	2.14E+03	8.12E+01	9.59E+01	1.05E+02	9.31E+01	6.98E+01	6.98E+01	6.98E+01	3.75E-03	1.89E-03	2.26E-03	6.70E+01	3.76E+01
17	HTP_Std	1	1	30	v1	3.81E+05	1.39E+04	1.24E+03	5.18E+03	4.05E+02	8.29E+01	9.67E+01	1.08E+02	6.98E+01	6.04E+01	5.23E+01	3.65E-02	3.25E-03	1.36E-02	3.97E+01	3.44E+01
18	EC	1	1	30	v1	2.41E+06	1.21E+05	1.15E+04	3.89E+03	2.89E+03	2.24E+02	8.41E+01	6.04E+00	9.31E+01	3.40E+01	2.94E+01	5.02E-02	4.77E-03	1.61E-03	3.59E+01	3.21E+01
10	HTP_Eco	2	1	30	v1	2.67E+04	1.09E+04	4.13E+03	1.21E+03	1.08E+02	1.00E-02	7.62E+01	6.04E+01	5.23E+01	6.04E+01	3.92E+01	4.08E-01	1.55E-01	4.54E-02	1.08E+01	1.12E+01

35	HTP_Std	2	1	30	v1	1.32E+05	5.43E+04	1.67E+04	2.73E+03	3.04E+02	4.86E+02	6.25E-02	9.31E+01	9.31E+01	9.31E+01	6.98E+01	4.11E-01	1.27E-01	2.07E-02	1.07E+01	1.24E+01
36	EC	2	1	30	v1	3.19E+05	8.35E+04	1.07E+04	3.21E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+01	9.31E+01	2.94E+01	1.08E+01	2.62E-01	3.35E-02	1.01E-02	1.61E+01	2.04E+01
7	HTP_Eco	0.5	0.2	70	v1	2.15E+06	1.27E+04	4.70E+02	1.98E+03	1.00E-02	5.44E+01	9.24E+01	1.24E+02	9.31E+01	8.06E+01	6.04E+01	5.91E-03	2.18E-04	9.20E-04	6.16E+01	5.06E+01
8	HTP_Std	0.5	0.2	70	v1	1.60E+06	7.54E+03	7.19E+02	4.39E+03	2.95E+02	6.93E+01	5.15E+01	1.66E+02	1.08E+02	8.06E+01	6.98E+01	4.71E-03	4.50E-04	2.74E-03	6.43E+01	4.62E+01
21	EC	0.5	0.2	70	v1	5.10E+06	5.47E+04	5.04E+03	3.42E+03	1.18E+03	2.67E+02	8.63E+01	9.31E+01	9.31E+01	3.40E+01	1.91E+01	1.07E-02	9.88E-04	6.70E-04	5.44E+01	4.15E+01
13	HTP_Eco	1	0.2	70	v1	4.71E+05	4.01E+04	3.30E+03	2.98E+03	1.94E+03	1.56E+02	8.43E+01	1.08E+02	8.06E+01	6.98E+01	6.04E+01	8.51E-02	7.01E-03	6.32E-03	2.96E+01	2.98E+01
14	HTP_Std	1	0.2	70	v1	7.76E+05	1.21E+05	2.84E+04	2.72E+03	6.88E+00	1.02E+03	2.34E+01	1.08E+02	1.08E+02	8.06E+01	6.04E+01	1.56E-01	3.66E-02	3.50E-03	2.23E+01	1.98E+01
33	EC	1	0.2	70	v1	1.06E+06	3.65E+03	9.58E+01	5.74E+03	1.67E+02	1.00E-02	6.25E-02	1.43E+02	9.31E+00	1.08E+01	3.40E+01	3.44E-03	9.04E-05	5.42E-03	6.81E+01	5.59E+01
34	HTP_Eco	2	0.2	70	v1	1.62E+05	5.41E+04	1.25E+04	2.10E+03	1.00E-02	5.23E+02	6.25E-02	1.08E+02	1.08E+02	8.06E+01	5.23E+01	3.34E-01	7.72E-02	1.29E-02	1.32E+01	1.54E+01
32	HTP_Std	2	0.2	70	v1	5.11E+03	4.79E+03	4.65E+03	4.04E+03	1.00E-02	1.00E-02	6.25E-02	6.98E+01	6.98E+01	6.98E+01	6.98E+01	9.37E-01	9.10E-01	7.90E-01	7.76E-01	5.66E-01
15	EC	2	0.2	70	v1	4.80E+05	6.77E+04	3.78E+04	3.65E+03	1.57E+03	1.22E+03	8.49E+01	1.08E+02	9.31E+01	9.31E+01	2.94E+01	1.41E-01	7.88E-02	7.61E-03	2.35E+01	1.52E+01
22	HTP_Eco	0.5	1	70	v1	8.46E+05	8.10E+02	4.35E+02	2.30E+03	9.16E+01	7.40E+01	1.11E+02	6.04E+00	6.04E+00	6.04E+00	5.23E+01	9.57E-04	5.14E-04	2.71E-03	8.34E+01	4.54E+01
29	HTP_Std	0.5	1	70	v1	1.71E+06	1.19E+03	3.33E+02	1.25E+03	1.00E-02	1.00E-02	6.25E-02	1.08E+02	1.08E+01	1.08E+01	1.08E+01	6.96E-04	1.95E-04	7.31E-04	8.72E+01	5.13E+01
30	EC	0.5	1	70	v1	3.86E+06	3.99E+05	4.27E+04	4.59E+03	1.00E-02	2.08E+03	6.25E-02	1.24E+02	1.24E+02	9.31E+01	1.08E+01	1.03E-01	1.11E-02	1.19E-03	2.72E+01	2.70E+01
31	HTP_Eco	1	1	70	v1	1.15E+06	8.38E+02	5.31E+02	4.32E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+01	1.08E+01	1.08E+01	5.23E+01	7.29E-04	4.62E-04	3.76E-03	8.67E+01	4.61E+01
23	HTP_Std	1	1	70	v1	5.53E+03	5.03E+03	4.77E+03	4.13E+03	1.28E+02	7.17E+01	5.97E+01	6.04E+01	6.98E+01	6.98E+01	6.04E+01	9.10E-01	8.63E-01	7.47E-01	1.14E+00	8.87E-01
24	EC	1	1	70	v1	3.08E+06	1.23E+05	1.94E+04	5.59E+03	1.00E-02	1.56E+03	1.01E+01	1.43E+02	1.24E+02	6.04E+00	3.40E+01	3.99E-02	6.30E-03	1.81E-03	3.86E+01	3.04E+01
16	HTP_Eco	2	1	70	v1	1.50E+05	9.17E+03	6.71E+02	1.29E+03	5.45E+02	4.09E+01	1.64E+02	9.31E+01	6.98E+01	6.98E+01	6.04E+00	6.11E-02	4.47E-03	8.63E-03	3.35E+01	3.25E+01
5	HTP_Std	2	1	70	v1	2.76E+04	3.03E+03	4.33E+03	3.53E+03	7.02E+01	5.34E+01	9.65E+01	6.04E+00	6.98E+01	6.98E+01	6.04E+01	1.10E-01	1.57E-01	1.28E-01	2.65E+01	1.11E+01

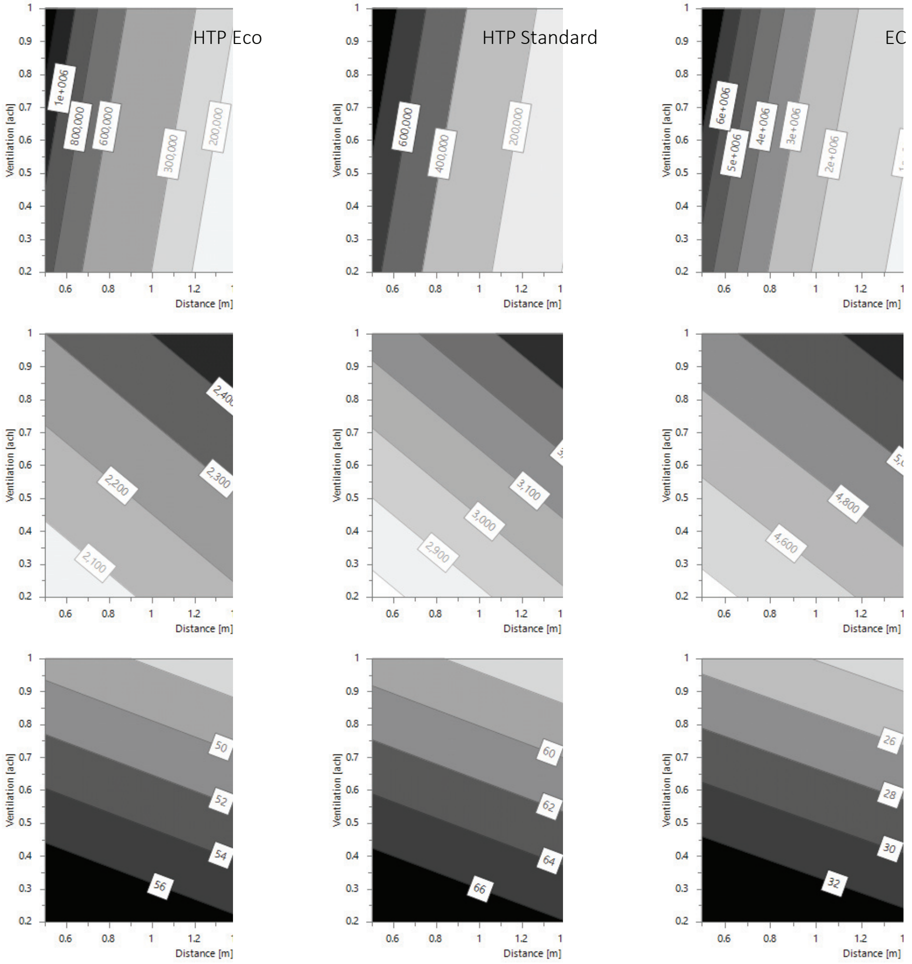
6	EC	2	1	70	v1	1.19E+06	2.36E+05	9.63E+04	1.37E+04	1.00E-02	1.61E+03	7.59E+01	1.43E+02	1.43E+02	1.24E+02	3.40E+01	1.98E-01	8.09E-02	1.15E-02	1.94E+01	1.51E+01
37	HTP_Eco	0.5	0.2	30	v2	1.38E+06	1.48E+04	1.41E+03	1.65E+03	1.00E-02	1.00E-02	6.25E-02	3.92E+01	5.23E+01	4.53E+01	4.53E+01	1.07E-02	1.02E-03	1.19E-03	5.44E+01	4.13E+01
38	HTP_Std	0.5	0.2	30	v2	2.08E+04	4.82E+02	7.21E+02	5.54E+03	1.00E-02	1.00E-02	6.25E-02	3.92E+01	1.08E+01	1.08E+01	6.04E+01	2.32E-02	3.47E-02	2.66E-01	4.52E+01	2.02E+01
48	EC	0.5	0.2	30	v2	2.65E+06	4.50E+05	1.50E+05	2.11E+04	4.82E+03	1.17E+03	6.25E-02	9.31E+01	9.31E+01	1.65E+01	3.40E+01	1.70E-01	5.66E-02	7.97E-03	2.13E+01	1.72E+01
64	HTP_Eco	1	0.2	30	v2	2.27E+05	3.27E+03	2.79E+03	4.47E+03	1.00E-02	1.00E-02	6.25E-02	6.04E+01	5.23E+01	4.53E+01	5.23E+01	1.44E-02	1.23E-02	1.97E-02	5.09E+01	2.64E+01
62	HTP_Std	1	0.2	30	v2	1.45E+04	9.29E+03	1.06E+04	8.99E+03	1.00E-02	1.00E-02	6.25E-02	5.23E+01	4.53E+01	5.23E+01	4.53E+01	6.41E-01	7.31E-01	6.20E-01	5.34E+00	1.88E+00
39	EC	1	0.2	30	v2	6.61E+05	3.61E+04	1.25E+03	3.16E+03	1.24E+03	1.00E-02	6.25E-02	1.24E+02	3.40E+01	4.53E+01	1.08E+01	5.46E-02	1.89E-03	4.78E-03	3.49E+01	3.76E+01
61	HTP_Eco	2	0.2	30	v2	9.62E+04	5.82E+03	4.59E+03	4.81E+03	1.00E-02	1.00E-02	6.25E-02	6.04E+01	5.23E+01	5.23E+01	3.92E+01	6.05E-02	4.77E-02	5.00E-02	3.37E+01	1.83E+01
56	HTP_Std	2	0.2	30	v2	1.37E+05	7.91E+03	1.18E+03	2.09E+03	8.68E+01	1.00E-02	6.25E-02	8.06E+01	8.06E+01	1.08E+01	1.08E+01	5.77E-02	8.61E-03	1.53E-02	3.42E+01	2.85E+01
45	EC	2	0.2	30	v2	5.20E+05	2.40E+05	2.03E+05	1.76E+03	1.00E-02	5.66E+03	6.25E-02	1.08E+02	1.08E+02	9.31E+01	1.08E+01	4.62E-01	3.90E-01	3.38E-03	9.28E+00	5.64E+00
55	HTP_Eco	0.5	1	30	v2	1.24E+06	3.54E+03	1.70E+03	4.07E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+01	6.98E+01	5.23E+01	6.98E+01	2.85E-03	1.37E-03	3.28E-03	7.03E+01	3.96E+01
47	HTP_Std	0.5	1	30	v2	1.18E+06	1.13E+05	6.49E+03	5.60E+03	1.22E+03	1.00E-02	6.25E-02	1.08E+02	8.06E+01	6.98E+01	6.98E+01	9.58E-02	5.50E-03	4.75E-03	2.82E+01	3.12E+01
63	EC	0.5	1	30	v2	5.67E+06	1.31E+05	4.06E+04	1.42E+04	3.14E+03	1.92E+02	5.07E+01	1.24E+02	3.40E+01	3.40E+01	3.40E+01	2.31E-02	7.16E-03	2.50E-03	4.52E+01	2.96E+01
40	HTP_Eco	1	1	30	v2	1.18E+05	2.23E+03	1.91E+03	7.19E+03	1.00E-02	1.00E-02	6.25E-02	6.04E+01	3.40E+01	3.40E+01	4.53E+01	1.89E-02	1.62E-02	6.09E-02	4.76E+01	2.47E+01
53	HTP_Std	1	1	30	v2	2.21E+05	5.54E+03	1.03E+03	2.86E+03	1.00E-02	1.00E -02	6.25E-02	6.04E+00	8.06E+01	6.04E+01	6.98E+01	2.51E-02	4.66E-03	1.29E-02	4.42E+01	3.22E+01
54	EC	1	1	30	v2	1.53E+06	1.50E+05	2.11E+03	2.30E+03	6.11E+02	1.00E-02	6.25E-02	9.31E+01	1.91E+01	2.94E+01	1.08E+01	9.80E-02	1.38E-03	1.50E-03	2.79E+01	3.95E+01
46	HTP_Eco	2	1	30	v2	1.02E+05	5.73E+04	4.61E+04	4.84E+03	1.00E-02	2.07E+02	6.25E-02	8.06E+01	8.06E+01	8.06E+01	6.04E+01	5.62E-01	4.52E-01	4.74E-02	6.92E+00	4.76E+00
71	HTP_Std	2	1	30	v2	1.55E+04	1.29E+03	3.88E+03	4.11E+03	1.00E-02	6.27E+01	2.63E+00	8.06E+01	1.08E+01	8.06E+01	6.98E+01	8.32E-02	2.50E-01	2.65E-01	2.98E+01	8.31E+00
72	EC	2	1	30	v2	2.24E+05	8.95E+04	4.96E+04	7.59E+03	3.09E+02	1.00E-02	6.25E-02	3.40E+01	3.40E+01	3.40E+01	3.40E+01	4.00E-01	2.21E-01	3.39E-02	1.10E+01	9.05E+00

44	HTP_Eco	0.5	0.2	70	v2	2.04E+06	3.66E+03	5.64E+02	1.22E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+01	6.98E+01	1.08E+01	1.08E+01	1.79E-03	2.77E-04	5.96E-04	7.59E+01	4.92E+01
43	HTP_Std	0.5	0.2	70	v2	1.31E+06	5.27E+03	5.79E+02	3.27E+03	1.46E+02	1.00E-02	6.25E-02	1.24E+02	8.06E+01	1.08E+01	6.04E+01	4.02E-03	4.42E-04	2.50E-03	6.62E+01	4.63E+01
57	EC	0.5	0.2	70	v2	2.53E+06	1.25E+04	9.87E+02	2.86E+03	5.63E+02	1.00E-02	1.38E+01	1.43E+02	9.31E+00	1.08E+01	1.08E+01	4.94E-03	3.90E-04	1.13E-03	6.37E+01	4.71E+01
49	HTP_Eco	1	0.2	70	v2	4.26E+05	7.14E+04	9.72E+04	5.13E+03	1.00E-02	1.85E+03	6.25E-02	1.08E+02	9.31E+01	9.31E+01	6.98E+01	1.68E-01	2.28E-01	1.21E-02	2.14E+01	8.87E+00
50	HTP_Std	1	0.2	70	v2	3.66E+05	9.57E+04	1.92E+04	1.40E+03	2.04E+03	1.00E-02	6.25E-02	1.24E+02	1.24E+02	1.43E+02	9.31E+01	2.61E-01	5.25E-02	3.83E-03	1.61E+01	1.77E+01
69	EC	1	0.2	70	v2	1.91E+06	6.25E+04	2.36E+03	6.46E+03	4.08E+03	1.00E-02	6.25E-02	1.43E+02	6.04E+00	3.40E+01	3.40E+01	3.27E-02	1.24E-03	3.38E-03	4.10E+01	4.02E+01
70	HTP_Eco	2	0.2	70	v2	3.62E+04	3.02E+04	1.45E+04	2.38E+03	6.63E+02	2.11E+02	6.25E-02	1.08E+02	1.08E+02	9.31E+01	6.98E+01	8.34E-01	4.01E-01	6.57E-02	2.17E+00	5.49E+00
68	HTP_Std	2	0.2	70	v2	9.36E+03	8.41E+03	8.29E+03	8.85E+03	1.00E-02	1.00E-02	1.85E+01	5.23E+01	5.23E+01	5.23E+01	5.23E+01	8.99E-01	8.86E-01	9.46E-01	1.28E+00	7.28E-01
51	EC	2	0.2	70	v2	7.00E+03	4.15E+03	2.00E+03	1.64E+03	1.00E-02	1.00E-02	6.25E-02	1.08E+02	1.08E+01	1.08E+01	1.08E+01	5.93E-01	2.86E-01	2.34E-01	6.27E+00	7.52E+00
58	HTP_Eco	0.5	1	70	v2	8.73E+05	4.06E+03	2.13E+03	1.37E+03	1.00E-02	6.52E+01	6.25E-02	1.24E+02	8.06E+01	9.31E+00	1.08E+01	4.65E-03	2.44E-03	1.57E-03	6.44E+01	3.61E+01
65	HTP_Std	0.5	1	70	v2	2.75E+06	7.01E+03	5.68E+02	4.06E+03	3.12E+02	1.00E-02	6.95E+00	1.08E+02	8.06E+01	1.08E+01	5.23E+01	2.55E-03	2.06E-04	1.48E-03	7.17E+01	5.09E+01
66	EC	0.5	1	70	v2	6.76E+06	6.09E+04	2.76E+03	1.08E+04	2.86E+03	2.01E+01	9.46E+01	1.08E+02	9.31E+01	1.08E+01	1.08E+01	9.01E-03	4.08E-04	1.59E-03	5.65E+01	4.68E+01
67	HTP_Eco	1	1	70	v2	2.90E+04	7.06E+02	1.01E+03	4.31E+03	1.00E-02	1.00E-02	6.25E-02	6.04E+01	1.08E+01	1.08E+01	5.23E+01	2.43E-02	3.48E-02	1.49E-01	4.46E+01	2.01E+01
59	HTP_Std	1	1	70	v2	2.60E+05	1.40E+04	1.55E+03	3.02E+03	7.90E+02	1.00E-02	3.69E+01	1.24E+02	9.31E+01	1.08E+01	8.06E+01	5.38E-02	5.96E-03	1.16E-02	3.51E+01	3.07E+01
60	EC	1	1	70	v2	9.76E+05	3.27E+05	3.34E+04	5.61E+03	8.87E+03	8.18E+02	5.49E+00	1.08E+02	1.08E+02	3.40E+01	3.40E+01	3.35E-01	3.42E-02	5.75E-03	1.31E+01	2.02E+01
52	HTP_Eco	2	1	70	v2	1.25E+05	7.09E+04	1.98E+04	1.44E+03	9.96E+02	9.14E+02	6.25E-02	1.08E+02	9.31E+01	6.98E+01	3.92E+01	5.67E-01	1.58E-01	1.15E-02	6.80E+00	1.11E+01
41	HTP_Std	2	1	70	v2	2.46E+04	3.83E+03	1.76E+03	7.16E+03	1.00E-02	1.00E-02	6.25E-02	8.06E+01	5.23E+01	3.92E+01	6.04E+01	1.56E-01	7.15E-02	2.91E-01	2.23E+01	1.58E+01
42	EC	2	1	70	v2	4.98E+05	1.48E+05	8.11E+03	1.17E+04	1.01E+04	1.00E-02	7.01E+01	1.24E+02	6.04E+00	3.40E+01	3.40E+01	2.97E-01	1.63E-02	2.36E-02	1.46E+01	2.47E+01
73	HTP_Eco	0.5	0.2	30	v3	1.14E+06	1.28E+04	7.09E+02	2.00E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+01	6.98E+01	5.23E+01	5.23E+01	1.12E-02	6.22E-04	1.76E-03	5.39E+01	4.43E+01

74	HTP_Std	0.5	0.2	30	v3	2.35E+06	4.10E+03	1.18E+03	5.44E+03	1.29E+01	1.00E-02	6.25E-02	1.08E+02	6.98E+01	6.98E+01	6.98E+01	1.74E-03	5.02E-04	2.31E-03	7.62E+01	4.56E+01
84	EC	0.5	0.2	30	v3	5.17E+06	1.13E+04	2.80E+03	9.93E+03	5.13E+01	1.00E-02	3.52E+01	1.08E+02	2.94E+01	3.40E+01	3.40E+01	2.19E-03	5.42E-04	1.92E-03	7.35E+01	4.51E+01
100	HTP_Eco	1	0.2	30	v3	8.26E+03	7.74E+03	7.94E+03	7.24E+03	1.00E-02	1.00E-02	6.25E-02	5.23E+01	5.23E+01	5.23E+01	5.23E+01	9.37E-01	9.61E-01	8.77E-01	7.80E-01	2.37E-01
98	HTP_Std	1	0.2	30	v3	1.58E+04	1.17E+04	4.19E+03	5.34E+03	1.00E-02	1.00E-02	6.25E-02	6.04E+01	5.23E+01	5.23E+01	4.53E+01	7.41E-01	2.65E-01	3.38E-01	3.61E+00	7.96E+00
75	EC	1	0.2	30	v3	2.83E+06	3.78E+05	3.39E+03	5.12E+03	1.58E+04	6.54E+01	6.25E-02	1.24E+02	9.31E+01	3.40E+01	1.08E+01	1.34E-01	1.20E-03	1.81E-03	2.42E+01	4.04E+01
97	HTP_Eco	2	0.2	30	v3	6.70E+03	6.24E+03	6.31E+03	6.11E+03	1.00E-02	1.00E-02	6.25E-02	3.92E+01	3.92E+01	3.92E+01	3.92E+01	9.31E-01	9.42E-01	9.12E-01	8.54E-01	3.60E-01
92	HTP_Std	2	0.2	30	v3	3.04E+03	1.67E+03	1.63E+03	1.58E+03	1.00E-02	1.00E-02	6.25E-02	1.08E+01	1.08E+01	1.08E+01	9.31E+00	5.49E-01	5.36E-01	5.19E-01	7.19E+00	3.74E+00
81	EC	2	0.2	30	v3	5.35E+05	3.89E+05	1.13E+05	1.41E+04	2.50E+03	1.41E+03	6.25E-02	1.24E+02	1.08E+02	1.08E+02	3.40E+01	7.27E-01	2.11E-01	2.64E-02	3.82E+00	9.33E+00
91	HTP_Eco	0.5	1	30	v3	1.35E+05	5.48E+03	1.15E+03	1.08E+03	3.54E+01	1.00E-02	6.25E-02	5.23E+01	3.92E+01	1.08E+01	1.08E+01	4.06E-02	8.52E-03	8.00E-03	3.85E+01	2.86E+01
83	HTP_Std	0.5	1	30	v3	1.32E+06	4.17E+03	5.93E+02	3.76E+03	1.20E+02	1.00E-02	6.25E-02	9.31E+01	8.06E+01	1.08E+01	6.98E+01	3.16E-03	4.49E-04	2.85E-03	6.91E+01	4.62E+01
99	EC	0.5	1	30	v3	4.36E+06	1.37E+05	1.97E+04	1.25E+04	3.78E+03	1.00E-02	6.25E-02	1.24E+02	1.08E+02	3.40E+01	3.40E+01	3.14E-02	4.52E-03	2.88E-03	4.15E+01	3.24E+01
76	HTP_Eco	1	1	30	v3	2.70E+05	8.32E+02	1.20E+03	3.24E+03	1.00E-02	1.00E-02	6.25E-02	6.04E+01	1.08E+01	1.08E+01	5.23E+01	3.08E-03	4.44E-03	1.20E-02	6.94E+01	3.25E+01
89	HTP_Std	1	1	30	v3	9.49E+04	3.29E+03	4.09E+02	1.10E+03	1.54E+01	1.00E-02	6.25E-02	6.98E+01	6.04E+01	6.04E+01	6.04E+01	3.47E-02	4.31E-03	1.16E-02	4.03E+01	3.27E+01
90	EC	1	1	30	v3	5.77E+05	7.23E+04	1.23E+03	1.82E+03	4.17E+03	1.00E-02	6.25E-02	1.08E+02	2.94E+01	3.40E+01	1.08E+01	1.25E-01	2.13E-03	3.16E-03	2.49E+01	3.69E+01
82	HTP_Eco	2	1	30	v3	7.45E+03	2.66E+03	2.50E+03	3.64E+03	1.00E-02	1.00E-02	6.25E-02	8.06E+01	5.23E+01	5.23E+01	5.23E+01	3.57E-01	3.36E-01	4.88E-01	1.24E+01	6.55E+00
107	HTP_Std	2	1	30	v3	2.13E+05	8.28E+02	5.80E+02	7.97E+03	1.00E-02	1.00E-02	6.25E-02	6.04E+01	1.08E+01	1.08E+01	5.23E+01	3.89E-03	2.72E-03	3.74E-02	6.66E+01	3.54E+01
108	EC	2	1	30	v3	3.41E+05	9.17E+04	5.93E+04	1.48E+04	1.00E-02	1.00E-02	6.25E-02	1.08E+02	1.08E+02	3.40E+01	3.40E+01	2.69E-01	1.74E-01	4.33E-02	1.58E+01	1.05E+01
79	HTP_Eco	0.5	0.2	70	v3	1.73E+06	3.73E+03	9.89E+02	2.06E+03	2.41E+01	1.00E-02	6.25E-02	9.31E+01	6.04E+01	1.08E+01	4.53E+01	2.16E-03	5.72E-04	1.19E-03	7.37E+01	4.48E+01
80	HTP_Std	0.5	0.2	70	v3	5.31E+05	9.89E+02	6.11E+02	2.91E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+01	1.08E+01	1.08E+01	6.04E+01	1.86E-03	1.15E-03	5.47E-03	7.54E+01	4.06E+01

93	EC	0.5	0.2	70	v3	5.67E+05	3.12E+04	3.58E+03	1.88E+03	7.20E+02	1.00E-02	6.25E-02	1.08E+02	3.40E+01	3.40E+01	1.08E+01	5.50E-02	6.31E-03	3.31E-03	3.48E+01	3.04E+01
85	HTP_Eco	1	0.2	70	v3	1.48E+05	1.32E+03	7.54E+02	1.87E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+01	1.08E+01	1.08E+01	4.53E+01	8.92E-03	5.09E-03	1.27E-02	5.66E+01	3.17E+01
86	HTP_Std	1	0.2	70	v3	6.45E+03	8.18E+02	7.50E+02	3.91E+03	1.00E-02	1.00E-02	6.25E-02	9.31E+00	1.08E+01	1.08E+01	6.98E+01	1.27E-01	1.16E-01	6.07E-01	2.48E+01	1.29E+01
105	EC	1	0.2	70	v3	1.55E+06	2.80E+03	1.98E+03	1.11E+04	1.00E-02	1.00E-02	6.25E-02	1.24E+02	1.08E+01	1.08E+01	3.40E+01	1.81E-03	1.28E-03	7.17E-03	7.58E+01	4.00E+01
106	HTP_Eco	2	0.2	70	v3	1.60E+04	5.32E+03	9.27E+03	5.36E+03	1.00E-02	1.00E-02	4.46E+01	6.04E+01	6.04E+01	6.04E+01	5.23E+01	3.33E-01	5.79E-01	3.35E-01	1.32E+01	3.27E+00
104	HTP_Std	2	0.2	70	v3	8.62E+03	5.88E+03	6.06E+03	6.24E+03	1.00E-02	1.00E-02	6.25E-02	3.92E+01	6.04E+01	6.04E+01	5.23E+01	6.82E-01	7.03E-01	7.24E-01	4.59E+00	2.11E+00
87	EC	2	0.2	70	v3	1.45E+06	4.95E+03	9.17E+03	8.26E+03	1.00E-02	3.58E+02	6.25E-02	1.24E+02	1.08E+01	9.31E+00	1.08E+01	3.41E-03	6.32E-03	5.70E-03	6.82E+01	3.04E+01
94	HTP_Eco	0.5	1	70	v3	4.25E+05	4.83E+03	4.84E+02	2.58E+03	1.00E-02	1.00E-02	7.03E+00	1.08E+02	6.98E+01	9.31E+00	5.23E+01	1.14E-02	1.14E-03	6.08E-03	5.37E+01	4.07E+01
101	HTP_Std	0.5	1	70	v3	2.44E+06	2.05E+04	1.07E+03	6.81E+03	3.30E+02	1.00E-02	6.25E-02	9.31E+01	6.98E+01	1.08E+01	5.23E+01	8.40E-03	4.39E-04	2.79E-03	5.74E+01	4.64E+01
102	EC	0.5	1	70	v3	6.15E+06	9.49E+04	6.24E+03	1.20E+04	7.62E+03	1.00E-02	6.25E-02	1.08E+02	6.04E+00	3.40E+01	3.40E+01	1.54E-02	1.01E-03	1.95E-03	5.01E+01	4.14E+01
103	HTP_Eco	1	1	70	v3	4.05E+05	3.01E+03	7.35E+02	2.67E+03	5.93E+01	1.00E-02	6.25E-02	8.06E+01	9.31E+00	1.08E+01	3.92E+01	7.43E-03	1.82E-03	6.58E-03	5.88E+01	3.79E+01
95	HTP_Std	1	1	70	v3	3.87E+05	6.96E+03	1.67E+03	3.06E+03	9.30E+01	3.86E+01	6.25E-02	9.31E+01	8.06E+01	9.31E+00	6.04E+01	1.80E-02	4.32E-03	7.91E-03	4.82E+01	3.27E+01
96	EC	1	1	70	v3	1.77E+06	4.90E+04	1.23E+03	7.20E+03	2.98E+03	1.00E-02	6.25E-02	1.24E+02	6.04E+00	1.08E+01	3.40E+01	2.77E-02	6.95E-04	4.07E-03	4.30E+01	4.36E+01
88	HTP_Eco	2	1	70	v3	3.51E+03	2.90E+03	2.91E+03	2.59E+03	1.00E-02	1.00E-02	6.25E-02	5.23E+01	5.23E+01	5.23E+01	5.23E+01	8.26E-01	8.29E-01	7.37E-01	2.29E+00	1.12E+00
77	HTP_Std	2	1	70	v3	6.61E+03	4.20E+03	4.35E+03	4.28E+03	1.00E-02	1.00E-02	6.25E-02	1.08E+01	6.04E+01	6.04E+01	6.04E+01	6.35E-01	6.58E-01	6.47E-01	5.44E+00	2.51E+00
78	EC	2	1	70	v3	5.18E+05	4.43E+05	2.73E+05	1.41E+04	6.49E+02	1.60E+03	6.25E-02	1.24E+02	1.24E+02	1.24E+02	3.40E+01	8.55E-01	5.27E-01	2.73E-02	1.88E+00	3.84E+00

Appendix 8. Predicted key parameters of aerosol particle properties, following the exhalation after the usage of tested products: $PNC_{t,max}$ (during the puff, a), $PNC_{t,120s}$ (after the puff, b), the mode of PSD, 120 s after the puff (c)



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1. KAUNELIENĖ, V., MEIŠUTOVIČ-AKHTARIEVA, M., MARTUZEVIČIUS, D. A review of the impacts of tobacco heating system on indoor air quality versus conventional pollution sources. *Chemosphere*. 2018, 206. DOI 10.1016/j.chemosphere.2018.05.039.
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