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Doctoral Thesis

Influence of Residential Characteristics and Ventilation Systems on Indoor VOC Concentrations

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Abstract

Volatile organic compounds (VOCs) are a large group of carbon-based chemicals that easily evaporate at room temperature. They are characterized by their high vapor pressure and low boiling point, which enable them to disperse quickly into the air. VOCs are commonly found in both natural and synthetic sources and are present in a wide range of indoor environments. VOCs in indoor can originate from numerous sources including building materials, furnishings, household products, and personal care items. Paints, varnishes, adhesives, and sealants often contain VOCs such as formaldehyde, benzene, and toluene, which can emit into the indoor air. Additionally, cleaning agents, air fresheners, and cosmetics can emit significant amounts of VOCs during use. Even activities such as cooking and smoking contribute to indoor VOC levels. Poor ventilation exacerbates the accumulation of VOCs indoors, as it limits the dilution and removal of these compounds from the indoor environment. Consequently, understanding and controlling the sources of VOCs, alongside ensuring adequate ventilation, are critical for maintaining healthy indoor air quality and reducing potential health risks associated with long-term exposure to these compounds. Twenty years after the issue of sick house syndrome in the 1990s, it is necessary to continuously monitor the impact of indoor VOCs on the health of residents in homes where people spend significant amounts of time. Additionally, there is a lack of data on various housing types and a paucity of research on the impact of indoor VOCs relative to different ventilation systems.

Chapter 1 provides a comprehensive overview of the study, beginning with the background and motivation for researching indoor VOCs. It delves into the physical and chemical characteristics of VOCs, outlining their common properties, sources, and potential health impacts. The chapter also reviews VOC guidelines and regulations in major countries around the world, highlighting the differences and similarities in standards aimed at protecting indoor air quality. A thorough summary and analysis of previous studies on VOC levels in both new and existing houses are presented, identifying trends, differences, and inconsistencies in the current body of research. Based on this literature review and the identified differences in research and data, the chapter explains the purpose of this study, which aims to fill these gaps (Characteristics, types and ventilation systems of housing) by providing new insights and comprehensive data on indoor VOC concentrations across various housing types and ventilation systems. Finally, the chapter concludes with an explanation of the structure of the paper, accompanied by a schematic diagram that outlines the organization and flow of the subsequent chapters, ensuring a clear and logical progression of the research findings and discussions.

Chapter 2: This study involved field measurements conducted in different types of housing (custom-made homes, built-for-sale homes, private rentals, and apartments) during both winter and summer to assess VOC concentrations. The study sampled indoor air quality in 116 houses in winter and 66 in summer, using data from questionnaires about housing type, residence period, and ventilation systems to understand factors influencing VOC concentrations. The findings revealed that 12% of the houses exceeded guideline values for formaldehyde and acetaldehyde. A notable observation was that houses with frequent alcohol consumption showed elevated acetaldehyde levels. While the living room generally had slightly higher VOC concentrations than the bedroom, no significant seasonal differences were found. However, there were statistical differences in VOC concentrations by housing type and ventilation system, with apartments showing the highest levels and a marked difference in concentrations depending on the ventilation balance and heating systems used.

Chapter 3: Building on the insights from Chapter 2, further surveys and field measurements focused on the impact of ventilation systems. The study differentiated between homes with balanced and unbalanced ventilation systems, measuring in 16 homes with balanced ventilation and 17 with unbalanced during both seasons. The results showed that both balanced and unbalanced ventilated homes measured lower VOCs than the guidelines, but balanced ventilation tended to have higher VOC concentrations than unbalanced ventilation. The frequency of ventilation was also lower in unbalanced systems on average than in balanced ventilation, due to the lower ventilation efficiency of unbalanced ventilation systems and the lower ventilation volume as lower temperatures in winter reduced the frequency and duration of window opening. When looking at the correlation between VOCs and air change rates in living rooms and bedrooms in winter and summer, certain VOCs varied in season and space, both positively and negatively, but there were no consistent results overall. Residential buildings with balanced ventilation systems tend to have more consistent and higher air change rates and are more effective at maintaining lower concentrations of VOCs, which helps manage indoor air quality.

Chapter 4: In Chapter 3, the VOC concentration in the unbalanced ventilation system was measured as higher than that in the balanced ventilation system. There are several factors, but given the possibility that VOC concentrations may flow into the room from enclosed spaces, the VOCs of the insulation material, one of the important materials for the airtight performance of the building, were evaluated. The emissions of VOCs and formaldehyde from various insulation materials were measured using the small chamber method. The materials tested included glass wool, mineral wool, expanded polystyrene (EPS), extruded polystyrene (XPS), phenolic foam, rigid polyurethane foam, cellulose, and cork. Specifically, EPS and cork exhibited higher emission rates for

toluene, styrene, and acetaldehyde. This indicates that in homes using only unbalanced ventilation, where indoor spaces are under negative pressure, VOCs can infiltrate from enclosed spaces and impact indoor air quality.

Chapter 5 (Conclusion): This study highlights the need for ongoing monitoring of VOCs in both new and existing homes to understand the variations in VOC concentrations across different housing types. Comparisons between balanced and unbalanced ventilation systems have shown that unbalanced systems often lead to higher VOC concentrations, necessitating further research into the impact of ventilation on indoor air quality. Additionally, in homes with unbalanced ventilation, VOCs can infiltrate from enclosed spaces, underscoring the importance of ensuring sufficient and appropriate ventilation.

Contents

Chapter 1. Introduction

1.1 Background

Many individuals spend a significant amount of time indoors, leading to an increased awareness of indoor air quality and potential exposure to volatile organic compounds (VOCs) [1-1,1-2,1-3,1-4]. VOCs have garnered attention since the 1990s owing to their association with "sick building syndrome (SBS)," which results in adverse health effects such as eye, mucous membrane, skin, and respiratory irritation, as well as fatigue and damage to the nervous system [1-5]. Notably, owing to their high toxicity and carcinogenicity, aromatic hydrocarbons, including benzene, toluene, ethylbenzene, xylene (collectively known as BTEX), and carbonyl compounds, require stringent control of indoor concentrations [1-6,1-7]. Recognizing these risks, the World Health Organization (WHO) issued guidelines that promote sustainable and healthy housing while outlining the dangers posed by pollutants such as benzene and formaldehyde [1-8]. The International Agency for Research on Cancer (IARC) classifies benzene and formaldehyde into group 1, human carcinogens. The Ministry of Health, Labor and Welfare (MHLW) set indoor pollutant concentration guidelines for 13 types of VOCs [1-9].

Indoor pollutant concentrations are influenced by the structural design and indoor sources of a dwelling, the ventilation system, and residents' lifestyles [1-10,1-11]. Indoor sources of BTEX and carbonyl compounds include construction materials such as wood furniture, panels, carpets, adhesives, paints, solvents, and household activities like smoking, cooking, printing, and scented candles [1-12,1-13,1-14]. In particular, a comprehensive evaluation and modeling of various variables affecting VOC emissions, including temperature and relative humidity, have been carried out using various building materials [1-15]. For example, press-wood products, such as particle boards and medium-density fiber plates, constitute significant indoor pollutants owing to their high formaldehyde emissions and widespread use in furniture and housing [1-16].

VOCs emitted from building materials are considered the most common cause of sick building syndrome in new homes. However, the use of VOC-free building materials and conducting bake-outs or flush-outs before occupants move in have improved the problem of sick building syndrome [1-17]. It is now important to focus on indoor air quality in existing homes as well as new construction. Even in existing homes, various sources of VOCs are present, affecting the health of the occupants [1-18]. For example, cooking, smoking, candles, hairspray, deodorants, room air fresheners, photocopiers, and laser printers can all contribute to increased indoor VOC concentrations [1-19]. The type of ventilation in a home and how it is used can also have a significant impact on

indoor air quality [1-20]. Unbalanced ventilation is much less efficient than balanced ventilation because it uses only exhaust fans, without supply fans, to locally move air from inside to outside. As a result, VOCs can build up and increase in concentration due to the lack of ventilation in the room. Balanced ventilation is definitely more efficient, but even if there is a balanced ventilation system, there are problems with noise and energy, and there are cases where people do not know that they have a ventilation system and do not use it [1-21].

In this study, VOC concentrations were determined according to the characteristics of existing houses. The characteristics of the houses were identified through a questionnaire survey, including the type of house (custom-made, built-for-sale, private rental, and apartment buildings), the residence period, and the type of ventilation system (balanced or unbalanced). VOC concentrations were analyzed according to the characteristics of the houses, and statistical analysis was performed to identify significant differences and correlations between substances and variables.

1.2 Characteristics of volatile organic compounds

1.2.1 Physical characteristics

VOCs are a diverse group of organic chemicals that share common physical and chemical properties, enabling them to evaporate easily at room temperature. Their high vapor pressure, which generally exceeds 0.01 kPa at 20 °C, and low water solubility facilitate their transition from liquid or solid states into the gaseous phase. This volatility is due to their molecular structure, typically comprising small to medium-sized molecules with a variety of functional groups such as alkanes, alkenes, aromatics, aldehydes, ketones, and others.

Scientifically, VOCs are characterized by their boiling points, which range from about 50 to 260 °C. This relatively low boiling point contributes to their rapid evaporation at ambient conditions. In terms of molecular weight, VOCs usually have a mass ranging from about 50 to 200 grams per mole. These compounds can interact with sunlight and other atmospheric chemicals, leading to the formation of secondary pollutants like ground-level ozone and particulate matter, which are key components of smog. VOCs are broadly categorized as follows,

VOCs can be classified based on their volatility into several categories: Very Volatile Organic Compounds (VVOCs), Volatile Organic Compounds (VOCs), Semi-Volatile Organic Compounds (SVOCs), and Microbial Volatile Organic Compounds (MVOCs). These categories are distinguished primarily by their boiling points and vapor pressures, which influence their behavior in the environment and their impact on indoor air quality.

VVOCs have extremely high vapor pressures and low molecular weights, which allow them to evaporate easily into the air. Their boiling points typically range from 0-50°C. Common examples of VVOCs include formaldehyde and acetaldehyde. These compounds are often found in building materials, furniture, and cleaning products, significantly affecting indoor air quality due to their rapid evaporation.

VOCs are the most widely recognized category, with boiling points ranging from 50-260°C. These compounds are commonly emitted from paints, solvents, adhesives, and aerosol sprays. Examples include benzene, toluene, and xylene (collectively known as BTEX compounds). VOCs readily evaporate into the air, impacting both indoor and outdoor air quality. They play a critical role in the formation of photochemical smog and ground-level ozone and can pose various health risks such as headaches, respiratory irritation, and, with prolonged exposure, more severe conditions [1-22].

SVOCs have lower vapor pressures and higher molecular weights, with boiling points ranging from 240-400°C.

These compounds are less likely to evaporate completely and often adhere to dust particles or surfaces. They are found in pesticides, plasticizers, flame retardants, and other materials [1-23]. Examples include polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) [1-24]. SVOCs can persist in the environment and accumulate over time, posing long-term exposure risks through inhalation or dermal contact.

MVOCs are produced by microorganisms such as mold and bacteria. These compounds typically emerge in moist environments and can significantly degrade indoor air quality. Common MVOCs include 1-octen-3-ol and 2-methylisoborneol, which are often associated with the musty odors of mold. MVOCs can cause respiratory issues and allergic reactions with prolonged exposure [1-25].

Table 1-1. Ministry of Health, Labor and Welfare's VOC guidelines substance characteristics

VOC (Molecular formula)	Molecular weight [g/mol]	Boiling point [°C]	Vapor pressure	Functional groups:	Uses/Sources
Formaldehyde (HCHO)	30.03	-19	3,890 mmHg at 25°C	Aldehyde	Building materials, tobacco smoke, combustion processes, disinfectants, and preservatives.
Acetaldehyde (CH3CHO)	44.05	20.2	740 mmHg at 20°C	Aldehyde	Alcoholic beverages, ripening fruits, combustion processes, and industrial processes.
*Benzene (C6H6)	78.11	80.1	95 mmHg at 25°C	Aromatic hydrocarbon	Industrial solvents, gasoline, cigarette smoke.
Toluene (C7H8)	92.14	110.6	28 mmHg at 25°C	Aromatic hydrocarbon	Paint thinners, adhesives, chemical manufacturing, and fuel additives.
Xylene	106.17	138.5	6 mmHg at	Aromatic	Solvents in the printing,

(C8H10)		(average for isomers)	25°C (average for isomers)	hydrocarbon	rubber, and leather industries, and paint thinners.
Ethylbenzene (C8H10)	106.17	136	9.6 mmHg at 25°C	Aromatic Hydrocarbon	Manufacturing of styrene, solvents, and paint thinners.
Styrene (C8H8)	104.15	145	5 mmHg at 25°C	Aromatic Hydrocarbon	Production of polystyrene plastics and resins, and as a precursor to other chemicals.
Paradichloro benzene (C6H4Cl2)	147.01	174	0.6 mmHg at 25°C	Aromatic Hydrocarbon	Moth repellents, air fresheners, and deodorants.
Tetradecane (C14H30)	198.39	253.5	0.011 mmHg at 25°C	Alkane	Calibration standards in laboratories, and as a component in fuels and lubricants.
Chlorpyrifos (C9H11Cl3NO3PS)	350.59	160 at 1.5 mmHg (decomposes)	1.87 x 10^-5 mmHg at 25°C	Phosphate ester Nitro Group	Insecticide used in agriculture, homes, and public health.
Fenobucarb (C12H17NO2)	207.27	185	1.3 x 10^{-5} mmHg at 25°C	Aromatic Hydrocarbon	Insecticide used in rice paddies and other agricultural applications.
Diazinon (C12H21N2O3PS)	304.35	125 at 0.1 mmHg	1.40 x 10^{-4} mmHg at 20°C	Aromatic Hydrocarbon	Insecticide used in agriculture, home gardens, and public health.
Di-n-butyl	278.35	340	9.7 x 10^{-5}	Aromatic	Plasticizer in plastics,

phthalate (DBP) (C16H22O4)			mmHg at 25°C	Hydrocarbon	solvents, and personal care products.
Di-2-ethylhexyl phthalate (DEHP) (C24H38O4)	390.56	385	3.4 x 10^{-8} mmHg at 25°C	Phthalate ester	Plasticizer in polyvinyl chloride (PVC) plastics, medical devices, and consumer products.

* Benzene is not included in the Ministry of Health, Labor and Welfare's VOC guidelines

1.2.2 Guidelines for VOCs

The World Health Organization (WHO) has guidelines for hazardous chemicals indoors. WHO guidelines for indoor air quality: selected pollutants provide guidelines for the following substances (benzene, carbon monoxide, formaldehyde, naphthalene, nitrogen dioxide, polycyclic aromatic hydrocarbons, radon, Trichloroethylene and Tetrachloroethylene) [1-26]. Among them, only the contents of VOC are summarized substances are as follows. Benzene is a known carcinogen with no safe level of exposure. It is found in both outdoor and indoor air, with indoor concentrations typically higher due to infiltration from outdoor sources and other indoor activities. Benzene exposure is primarily through inhalation, and its presence indoors necessitates stringent guidelines to minimize health risks. The WHO guidelines align with ambient air guidelines, emphasizing the reduction of indoor benzene levels to as low as possible through measures such as eliminating smoking and using low-emission building materials. Formaldehyde exposure predominantly occurs indoors and can cause sensory irritation at relatively low concentrations. The guideline value of 0.1 mg/m^3 for short-term (30-minute) exposure is intended to prevent irritation and is based on the lowest concentration causing eye irritation in humans. The guideline also aims to prevent long-term effects, including cancer, by ensuring indoor concentrations do not exceed 0.1 mg/m^3 at any time. Measures to reduce formaldehyde exposure include using low-emitting materials and improving ventilation. Naphthalene exposure can cause respiratory tract lesions and tumors, as well as hemolytic anemia. The guideline value of 0.01 mg/m^3 as an annual average is based on the lowest observed adverse effect level (LOAEL) from animal studies. The primary method of reducing exposure is to avoid using naphthalene-containing products like mothballs. For trichloroethylene (TCE), the guideline adopts a non-threshold approach due to its carcinogenic potential, with a unit risk estimate of 4.3×10^{-7} per μg/m^3. Recommended concentration limits aim to minimize

cancer risk. Tetrachloroethylene (PCE) guidelines focus on non-carcinogenic effects, such as neurobehavioral performance and renal changes, with a long-term exposure limit of 0.25 mg/m^3.

Japan has established guidelines for VOCs to ensure pleasant indoor air quality in the Ministry of Health, Labor and Welfare (MHLW) [1-27]. The guidelines set target concentration limits for different VOCs, particularly formaldehyde, toluene, and xylene, in indoor settings. These restrictions aim to mitigate health risks associated with VOC exposure, such as respiratory problems and other health issues. MHLW regularly updates these guidelines to reflect new scientific evidence. The following are the guidelines from 13 different VOCs presented by the Ministry of Health, Labor and Welfare.

Table 1-2. MHLW's guideline value of VOC in Japan

VOC	Guideline value μg/m³ (ppm)	Year of amendment and revision of the guidelines [year]
Formaldehyde	100 (0.08)	1997
Acetaldehyde	48 (0.03)	2002
Toluene	260 (0.07)	2000
Xylene	200 (0.05)	2000, 2019
Ethylbenzene	3800 (0.88)	2000
Styrene	220 (0.05)	2000
Paradichlorobenzene	240 (0.04)	2000
Tetradecane	330 (0.04)	2001
Chlorpyrifos	1 (0.07)	2000
Fenobucarb	33 (3.8)	2002
Diazinon	0.29 (0.02)	2001
Di-n-butyl phthalate	17 (1.5)	2000, 2019
Di-2-ethylhexyl phthalate	100 (6.3)	2001, 2019
TVOC	400	2000

The American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) has established comprehensive standards to ensure acceptable indoor air quality and the control of VOCs in various building environments. ASHRAE Standard 62.1, titled "Ventilation for Acceptable Indoor Air Quality," provides detailed guidelines on ventilation rates required to maintain indoor air quality in commercial and institutional buildings [1-28]. This standard emphasizes the importance of adequate ventilation to dilute and remove indoor pollutants, including VOCs, which can originate from building materials, furnishings, and occupant activities. It specifies minimum ventilation rates and outlines procedures for maintaining proper airflow to ensure that indoor environments are both comfortable and healthy for occupants. Additionally, ASHRAE Standard 189.1, known as the "Standard for the Design of High-Performance Green Buildings," extends these principles by incorporating advanced strategies for indoor air quality management. This standard promotes the use of sustainable building practices and materials with low VOC emissions to minimize environmental impact and enhance indoor air quality. It includes provisions for the selection of low-emitting materials, enhanced ventilation practices, and ongoing monitoring to ensure the continued health and well-being of building occupants. Together, these standards represent a comprehensive approach to managing indoor air quality through proper ventilation and the control of VOC concentrations, supporting both health and sustainability in building design and operation.

The U.S. Environmental Protection Agency (EPA) provides specific guideline values for various VOCs to protect indoor air quality and human health. For formaldehyde, the EPA recommends a concentration below 0.1 parts per million (ppm) or 100 parts per billion (ppb) to avoid irritation and potential cancer risk. Benzene exposure should be minimized as much as possible due to its carcinogenic nature, with no specific safe threshold established. Toluene has a reference concentration (RfC) of 5 milligrams per cubic meter (mg/m^3), approximately 1 ppm or 400 ppb, to prevent neurological and developmental effects. Xylene's RfC is 0.1 mg/m^3 or 100 micrograms per cubic meter ($\mu g/m^3$) to avoid respiratory and neurological issues. For ethylbenzene, the RfC is 1 mg/m^3 or 1,000 $\mu g/m^3$, aimed at protecting against respiratory and central nervous system effects. Trichloroethylene (TCE) has a concentration limit of 2 $\mu g/m^3$ due to its carcinogenic and non-carcinogenic effects. Tetrachloroethylene (PCE) has an RfC of 0.3 mg/m^3 or 250 $\mu g/m^3$ to prevent neurological damage and potential carcinogenic risks. Lastly, methylene chloride exposure should be minimized due to its high toxicity, with potential carcinogenic and central nervous system effects. These guidelines help maintain safe indoor air quality and reduce health risks associated with VOCs [1-29].

Table 1-3. Reference concentration in the U.S. EPA

VOC	RfC
Formaldehyde	<0.1 ppm
Acetaldehyde	9 μg/m³ (Rfc)
Benzene	Minimize exposure (no specific value due to its high toxicity)
Toluene	5 mg/m³
Xylene	0.1 mg/m³
Ethylbenzene	1 mg/m³
Trichloroethylene	2 μg/m³
Tetrachloroethylene	0.3 mg/m³
Methylene Chloride	Minimize exposure

Germany's Umwelt Bundesamt (Federal Environment Agency) is responsible for managing and implementing policies to ensure good indoor air quality. The agency focuses on identifying and mitigating indoor air pollutants that pose health risks. It conducts extensive research, develops guidelines, and sets standards for acceptable pollutant levels in indoor environments. The Umwelt Bundesamt collaborates with federal and state health authorities, providing scientific support and expertise. It also informs the public about potential indoor air hazards and best practices for maintaining healthy indoor environments. The agency's policies are aimed at protecting public health, especially in sensitive areas like homes, schools, and workplaces.

The German Committee on Indoor Air Guide Values (AIR) establishes health-based and hygienic guide values for assessing indoor air quality in public and private buildings. These values help determine safe concentrations of pollutants. The committee defines the indoor environment as homes, workplaces not covered by hazardous substance regulations, public buildings, and transport interiors. AIR's guide values include two levels: Value I (precautionary, indicating safe levels) and Value II (hazardous, requiring immediate action). These guidelines are used to protect public health from indoor pollutants. [1-30].

Table 1-4. Indoor air guide values derived by the German committee on indoor air guide values

VOC	Guideline Values [µg/m³]
Formaldehyde	100
Acetaldehyde	100
Toluene	300
Xylene	100
Ethylbenzene	200
Styrene	30

The French National Agency for Food, Environmental and Occupational Health and Safety (ANSES) develops indoor air quality guideline values (IAQGs) to protect public health from the harmful effects of VOCs. These guidelines are based on rigorous scientific assessments and aim to establish safe exposure levels for various pollutants in indoor environments. ANSES uses a comprehensive approach, incorporating toxicological data, epidemiological studies, and exposure assessments to determine these guideline values. The agency also emphasizes the importance of considering vulnerable populations, such as children and the elderly, in its assessments to ensure adequate protection for all segments of the population. Additionally, ANSES collaborates with other national and international bodies to harmonize standards and share scientific findings. The guidelines are continually reviewed and updated to incorporate the latest scientific knowledge and technological advancements in indoor air quality management. This ongoing review process ensures that the IAQGs reflect current understanding and provide robust protection against the adverse health effects of VOCs. Table 1-5 provides guidelines for long-term exposure to minimize health risks associated with chronic exposure to VOCs. The average annual concentration represents the maximum permissible level considered safe for human exposure over a year. These guideline values are air concentration limits, not direct inhalation limits. They specify the maximum amount of VOCs in the air that an individual can be exposed to without significant health risk, averaged over a one-year period [1-31].

Table 1-5. Long-term exposure guidelines for VOCs in France

VOC	Guideline Values [μg/m³]
Formaldehyde	10
Acetaldehyde	160 (Annual)
Benzene	2 (Long-term IAQG: for lifelong exposure corresponding to a risk level of 10^{-5})
Toluene	20,000 (Annual)
Ethylbenzene	1000 (Annual)
Trichloroethylene	20 (Long-term IAQG: for lifelong exposure corresponding to a risk level of 10^{-5})
Tetrachloroethylene	250 (Annual)

In South Korea, the Ministry of Environment has established stringent guidelines for indoor air quality specifically targeted at new apartment buildings to ensure a healthy living environment. These guidelines mandate that builders of new apartment complexes with 100 units or more must notify residents about the indoor air quality from 7 days before move-in to 60 days after move-in. This process involves active sampling techniques to measure the concentration of various VOCs, which include formaldehyde, benzene, toluene, ethylbenzene, xylene, and styrene. The specific guideline values set by the Ministry of Environment are formaldehyde at 210 μg/m³, benzene at 30 μg/m³, toluene at 1000 μg/m³, ethylbenzene at 360 μg/m³, xylene at 700 μg/m³, and styrene at 300 μg/m³ [1-32].

These values are derived from extensive health-based assessments and are intended to prevent health issues such as respiratory irritation, neurological effects, and cancer. The guidelines are part of the broader "Indoor Air Quality Management Act," which aims to tackle "sick house syndrome" caused by emissions from building materials and furnishings. Builders are required to use low-emission materials and proper ventilation systems to comply with these standards. Additionally, a mandatory labeling system for low VOC-emitting products helps consumers and builders make informed choices, promoting the use of healthier building materials. These measures collectively ensure that indoor environments in new apartment buildings meet high standards for air quality, thereby safeguarding the health of residents [1-33].

Table 1-6. Guideline values for VOCs by the ministry of environment in South Korea

VOC	Guideline value [μg/m³]	Year of amendment and revision of the guidelines [year]
Formaldehyde	210	2004, 2018
Benzene	30	
Toluene	1000	
Ethylbenzene	360	
Xylene	700	
Styrene	300	

In China, GB/T 18883-2022 Indoor Air Quality Standards is an official guideline value provided by the government of the People's Republic of China. It sets the standards for indoor air quality, including the permissible concentration limits for various VOCs and other pollutants in indoor environments [1-34]. This standard is issued by the National Health Commission of China and is enforced by the State Administration for Market Regulation and the Standardization Administration of China. The guidelines are designed to ensure the safety and health of individuals by regulating indoor air quality in residential and office buildings. Other indoor environments can refer to these standards to maintain air quality. The document includes detailed measurement methods, quality assurance protocols, and specific concentration limits for various pollutants to help maintain and improve indoor air quality.

Table 1-7. Guideline values for VOCs by the people's republic of China

VOC	Guideline value [μg/m³]	Averaging time	Year of amendment and revision of the guidelines [year]
Formaldehyde	≤ 80	1-hour average	2022
Acetaldehyde	≤ 54	1-hour average	
Benzene	≤ 30	1-hour average	
Toluene	≤ 200	1-hour average	
Xylene	≤ 200	1-hour average	
Trichloroethylene	≤ 6	8-hour average	
Tetrachloroethylene	≤ 120	8-hour average	
TVOC	≤ 600	8-hour average	

In Hong Kong, the Environmental Protection Department (EPD) has established comprehensive guidelines for indoor air quality (IAQ), particularly focusing on VOCs to protect public health. These guidelines are part of the "Indoor Air Quality Objectives for Office Buildings and Public Places" and classify air quality into "Excellent Class" and "Good Class," with the latter being the minimum acceptable standard [1-35]. The guideline values for VOCs in the "Good Class" include limits such as 100 μg/m³ for formaldehyde, 16 μg/m³ for benzene, 1000 μg/m³ for toluene, 220 μg/m³ for ethylbenzene, 870 μg/m³ for xylene, and 260 μg/m³ for styrene. These standards are derived from rigorous scientific assessments aimed at mitigating health risks like respiratory irritation, neurological impacts, and carcinogenic effects. The IAQ Certification Scheme further supports these guidelines by encouraging building owners to maintain high air quality standards, awarding IAQ Certificates to compliant buildings, and ensuring transparency and public awareness through regular monitoring and reporting. These guidelines are periodically reviewed and updated to incorporate the latest scientific findings and technological advancements, ensuring the continued protection of public health in indoor environments in Hong Kong.

Table 1-8. The VOC guideline values of the environmental protection department in Hong Kong

VOC	Guideline value [μg/m³]	Averaging time	Year of amendment and revision of the guidelines [year]
Formaldehyde	100	8-hour average	2019
Acetaldehyde	9		
Benzene	16		
Toluene	260		
Xylene	1,000		
Ethylbenzene	1,000		
Styrene	70		
TVOC	600		

In Singapore, the National Environment Agency (NEA) has established detailed guidelines to maintain indoor air quality, specifically targeting VOCs in office premises. These guidelines, outlined in the "Guidelines for Good Indoor Air Quality in Office Premises," aim to ensure a healthy indoor environment by regulating the concentrations of harmful VOCs [1-36]. The NEA specifies permissible limits for various VOCs commonly found in office settings. For instance, formaldehyde is limited to 100 μg/m³ to prevent health issues such as respiratory irritation and potential carcinogenic effects, while benzene is restricted to 5 μg/m³ due to its severe health risks. Toluene and xylene, which can cause neurological and respiratory problems, are limited to 300 μg/m³ and 200 μg/m³ respectively. These standards are based on comprehensive scientific research and align with international best practices to ensure a safe and comfortable working environment. The guidelines also emphasize the importance of regular monitoring and maintenance of ventilation systems to keep air quality within specified limits. Moreover, the NEA advocates for the use of low-emission materials and furnishings in office spaces to further reduce VOC levels. These comprehensive measures are part of Singapore's broader strategy to enhance public health and well-being by maintaining high indoor air quality standards in workplaces.

Table 1-9. The VOC guideline values of the national environment agency in Singapore

VOC	Guideline value [μg/m³]	Averaging time	Year of amendment and revision of the guidelines [year]
Formaldehyde	100	8-hour average	2019
Benzene	5		
Toluene	200		
Xylene	200		
Ethylbenzene	200		
Styrene	70		
TVOC	300		

1.2.3 Summary of previous research

Newly built residential buildings can be significant sources of VOCs, with higher levels found in new or renovated buildings compared to established ones. Researchers such as Rothweiler, Wäger, and Schlatter [1-42] and Brown [1-38] noted that VOC concentrations tend to be markedly higher immediately after construction or renovation. These elevated levels are primarily due to the off-gassing of materials used in the building process.

VOCs, including formaldehyde, are emitted from a variety of sources within a building. Building materials such as paints, adhesives, sealants, and varnishes are common emitters of VOCs. Additionally, new furniture, carpets, and household products can contribute significantly to indoor VOC levels [1-39, 1-41, 1-46]. The emission rates of these compounds can be influenced by factors such as temperature, humidity, and ventilation, which means that indoor air quality can vary widely between different buildings and even within different areas of the same building [1-43, 1-47].

The types and concentrations of VOCs in indoor environments can change over time due to the aging of materials and changes in usage patterns of the building. For instance, Yamaguchi and Hayashi have shown that the initial high levels of VOCs in new buildings tend to decrease over time as the rate of emission declines [1-47]. However, the persistence of some compounds, particularly those used in substantial quantities during construction or those with longer half-lives, can mean that elevated VOC levels remain a concern for several years.

Research has also highlighted the adverse health effects associated with prolonged exposure to VOCs. Symptoms can range from mild irritations of the eyes, nose, and throat to more severe impacts such as headaches, dizziness, and even long-term chronic conditions including respiratory diseases and cancer [1-39, 1-45]. Formaldehyde, in particular, is a well-known irritant and a potential human carcinogen, as classified by the International Agency for Research on Cancer (IARC).

Given the significant health risks associated with VOC exposure, effective measures are essential to mitigate their impact on indoor air quality. Proper ventilation is one of the most effective strategies. It can significantly reduce VOC concentrations by diluting indoor air with outdoor air, thus facilitating the removal of these pollutants from the indoor environment [1-42]. The use of low-VOC or VOC-free building materials and products is also strongly recommended to minimize initial emissions. Additionally, the implementation of air cleaning technologies, such as activated carbon filters and photocatalytic oxidation systems, can further help in reducing VOC levels indoors.

Established buildings can have persistent low levels of VOCs due to long-term emissions from building materials, as documented by Brown [1-38]. These compounds are emitted by a variety of sources including building materials, furniture, and consumer products, which was noted by Lagoudi, Loizidou, and Asimakopoulos [1-40]. The composition of these compounds can vary widely; some may become adsorbed onto material surfaces, complicating their behavior and persistence in indoor environments [1-37].

New or recently renovated buildings often exhibit significantly higher levels of VOCs, which can persist for several weeks post-construction or renovation. Wallace, Jungers, Sheldon, and Pellizzari observed that VOC concentrations are particularly elevated immediately following these activities, reflecting the extensive use of materials and products that emit these compounds. Liu, Shen, and Zhu found that concentrations of VOCs can be especially high in new apartment buildings, posing potential risks to the indoor air quality and occupant health during the initial occupancy period.

The emission rates of specific VOCs, such as toluene, are influenced by various factors including temperature and air exchange rate [1-44]. Higher temperatures can increase the volatility of these compounds, leading to greater emissions. Similarly, the rate of air exchange in a building can affect the concentration levels of VOCs; insufficient ventilation can result in the accumulation of these compounds, whereas proper ventilation can help to dilute and remove them from indoor air [1-42].

The persistence of VOCs in established buildings can be attributed to the continued off-gassing from building materials and furnishings, as well as from the use of consumer products. Studies by Hodgson and Lagoudi et al. have highlighted that even after the initial high emission period, VOCs can remain at detectable levels for extended periods, contributing to ongoing indoor air quality issues [1-39, 1-40]. These long-term emissions are often a result of slow diffusion processes from materials and the presence of semi-volatile organic compounds that can remain in the indoor environment longer than more volatile substances.

The health implications of prolonged exposure to low levels of VOCs have been a subject of extensive research. Symptoms associated with VOC exposure range from sensory irritations, such as eye, nose, and throat discomfort, to more severe effects like headaches, dizziness, and long-term respiratory conditions [1-39]. Formaldehyde, a common VOC found in many indoor environments, is particularly concerning due to its classification as a human carcinogen by the International Agency for Research on Cancer (IARC).

To mitigate the impact of VOCs on indoor air quality, several strategies have been recommended. Proper

ventilation is crucial; it helps to reduce the concentration of VOCs by introducing fresh outdoor air and expelling indoor air contaminants [1-42]. The use of low-VOC or VOC-free building materials and products during construction and renovation is also essential to minimize initial emissions [1-43]. Additionally, employing air purification technologies, such as activated carbon filters and photocatalytic oxidation, can be effective in removing VOCs from indoor air. Continuous monitoring of indoor air quality is vital to identify and address VOC sources promptly [1-49, 1-50].

Based on existing research, the focus has predominantly been on understanding VOC concentrations in newly built or renovated residential buildings. These environments typically exhibit higher VOC levels due to the off-gassing from new building materials such as paints, adhesives, sealants, and varnishes, as well as from new furniture and carpets. However, the sources of VOCs in these settings are often well-identified, allowing for targeted interventions.

Conversely, established residential buildings also present significant challenges in maintaining healthy indoor air quality due to the persistent emissions of VOCs from various sources. Over time, building materials and furnishings in older homes can continue to emit VOCs at lower but still detectable levels. Additionally, established homes may have accumulated VOCs from long-term use of consumer products, cooking, and other activities [1-51].

Research has highlighted the necessity of investigating VOC concentrations in older homes to identify diverse and potentially new sources of indoor air pollution. This approach is essential for several reasons:

(a). Long-Term Emissions: Materials in older homes, such as carpets, wood products, and paints, continue to emit VOCs even years after installation. These emissions, although lower in intensity compared to new materials, contribute to the overall indoor air quality problem.

(b). Secondary Sources: Older homes might have more complex interactions between various indoor pollutants. For instance, VOCs can adsorb onto surfaces and later desorb, leading to fluctuating concentrations that are harder to predict and control.

(c). Cumulative Exposure: Residents in established buildings may face cumulative exposure to VOCs over time, which could have different health implications compared to short-term high-level exposures in new buildings. Long-term exposure to low levels of VOCs has been associated with chronic health effects, including respiratory issues and cancer [1-48].

(d) Variety of Sources: Established homes might contain a wider variety of VOC sources, including accumulated products, furniture, and personal care items that have been used over many years [1-52]. This diversity requires comprehensive monitoring to identify all potential VOC contributors.

Environmental Changes: Factors such as changes in humidity, temperature, and ventilation can significantly affect VOC emission rates in older homes [1-53]. Seasonal changes and home modifications (like adding insulation or install HVAC systems) can also influence indoor VOC levels.

1.3 Objectives of study

In previous studies, VOC concentrations were measured and analyzed mainly for houses where new pollutants exist, such as new houses and renovated houses. It is also important to determine the VOC concentration in existing houses because additional pollutants occur in residents' lifestyle (ventilation method, drinking or smoking, cooking, and use of personal household goods). However, data on VOC concentrations in existing houses are very scarce. Based on these previous studies, Chapter 2 of this study classified existing single-family houses and apartment houses into custom-made, built-for-sale, apartment, and private rental, and reviewed the VOC concentration according to the characteristics and differences of VOC concentration by room (living room and bedroom), season (winter and summer), and type of ventilation system.

In addition, there are research results and data on the presence or absence of ventilation systems in houses, but studies on the difference between types of ventilation systems are very scarce. The indoor VOC concentration has a big impact because the ventilation efficiency of the house varies greatly depending on the type of the ventilation system. There are many existing studies on the difference in efficiency of ventilation systems, and there are data, but studies or data on whether it affects indoor VOC concentration are insufficient. Therefore, based on the results of Chapter 2, in Chapter 3, the difference in VOC concentration according to the type of ventilation system was clear, so the VOC concentration was additionally measured and compared for the group of houses using only balanced ventilation and the group of houses using only unbalanced ventilation.

As the airtightness of houses increases, the biggest problem related to the concentration of VOCs in houses that use only unbalanced ventilation is the problem of ventilation efficiency, but the use of unbalanced ventilation will sufficiently put the room in a negative pressure, and VOCs generated in confined spaces can penetrate the room. If there are many VOCs generated in confined spaces even if there are few or no pollutants indoors, the use of unbalanced ventilation devices can affect residents' health. Existing studies have shown that VOCs can enter confined spaces, but there are no specific studies on pollutants that generate VOCs in certain materials and how much. Therefore, in Chapter 4 of this study, VOC emission rates were identified for insulation materials, one of the materials in confined spaces. The purpose of this study is to provide basic data on the causes of one of the pollutants from various building materials in confined spaces by analyzing the data on VOCs from various insulation materials.

1.4 Composition of the paper

Figure 1 is the flowchart of the thesis. Chapter 1 provides the background to the study. It discusses how improvements in the airtightness and insulation performance of houses, coupled with insufficient ventilation, have led to problems with indoor air quality. The chapter also analyzes the physical and chemical properties of VOCs and the guideline values established by various countries. In addition, the purpose of this study is to provide new insights and comprehensive data on indoor VOC concentrations, considering various housing characteristics such as types, residence periods, and ventilation systems, while reviewing existing literature and previous studies.

In Chapter 2, field measurements were conducted in existing houses in Japan to determine the VOC concentration. Additionally, through a questionnaire survey, the basic survey of residents, life patterns and characteristics, the type of building, and the period of residence were identified. Based on this, it was verified whether there was a statistically significant difference between the VOC concentration and the characteristics of the house, and the correlation was identified. Subsequently, additional research was conducted in Chapter 3 based on these results.

In Chapter 3, there was a significant result regarding the VOC concentration according to the ventilation system of the house, which is the result of Chapter 2, so further measurement was conducted. Measurements were conducted by dividing the houses into those using balanced ventilation and those using only unbalanced ventilation. Similarly, through a questionnaire survey, basic surveys and characteristics of the houses were identified. In addition to the comparison of the ventilation systems, the amount of ventilation was also considered important, so not only VOCs but also carbon dioxide were measured. The correlation with the VOC concentration was identified by determining the number of ventilation events in the house using the measured carbon dioxide.

In Chapter 4, further analysis was conducted based on the results of the study in Chapter 3, which showed that unbalanced ventilation housing had a higher VOC concentration than balanced ventilation housing. In the case of unbalanced ventilation, indoor air is forcibly exhausted to the outside, causing the room to likely become a negative pressure environment. If it becomes a negative pressure environment, VOCs generated from confined spaces of walls, floors, and roofs are more likely to flow into the room. Therefore, a small chamber method was conducted on insulation materials, which are key materials for energy conservation. VOCs emitted from eight types of insulation materials were identified.

Chapter 5 is the conclusion. It presents a summary of the results of this study based on the findings from Chapters 2, 3, and 4. Additionally, the thesis concludes by providing a presentation of the limitations and suggestions for

future studies.

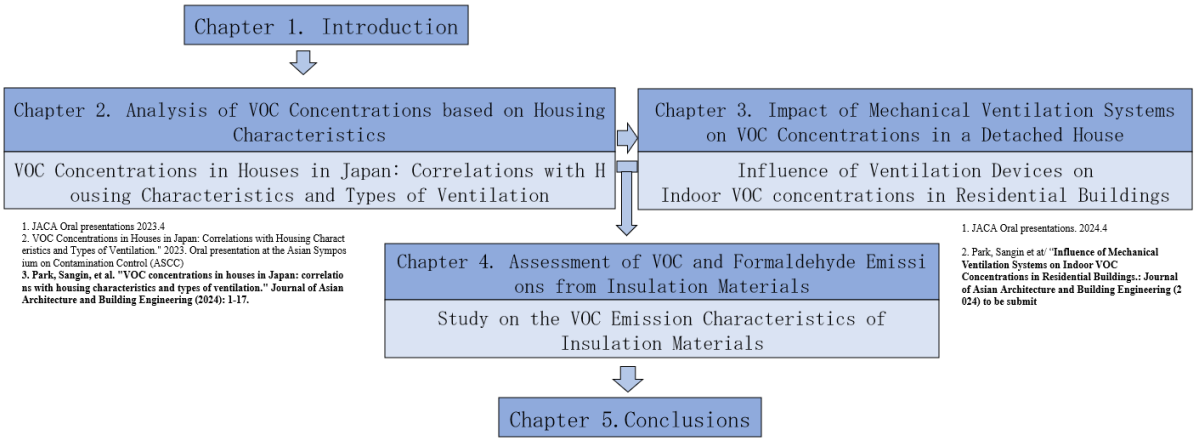

Figure 1. Flowchart of the thesis

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Chapter 2. Analysis of VOC concentrations based on housing characteristics

2.1 Previous study on VOC concentration in housing

In Chapter 1, we reviewed the existing literature on various housing and VOC pollution sources. The reviewed literature examined indoor VOC concentrations according to parameters such as seasonal differences, the year of construction of the house, the presence or absence of a garage, the influence of the atmospheric environment, and residents' living habits. The concentrations of VOCs emitted from building materials showed a consistent pattern, with VOC concentrations generally decreasing over time. However, some studies reported that indoor VOC concentrations were higher in summer than in winter [2-1, 2-2]. Conversely, other studies found that indoor VOC concentrations were higher in winter, indicating that a consistent pattern could not be identified due to the diverse causes and significant variables involved [2-3, 2-4]. In Chapter 2, we conducted field measurements to identify consistent patterns in VOC concentrations according to various residential characteristics. Our approach focused on several key factors: housing type (custom-made, built-for-sale, private rental, and apartment), residence period, and ventilation system (balanced and unbalanced) By incorporating these diverse factors into our field measurements, we aimed to provide a comprehensive understanding of the determinants of indoor VOC concentrations.

2.2 Field measurement of VOCs according to housing characteristics using the passive method

2.2.1 Measurement period and location

Field measurements were conducted in winter and summer. The measurement periods were January 28 to February 4, 2022; February 27 to March 9, 2022; and July 28 to August 26, 2022. The houses measured in the winter comprised 116 houses in 14 prefectures, including Tokyo. 49 of the 66 houses measured in the summer were the same as those measured in the winter (17 houses are not the same as winter.). One representative from each household responded to the survey. The survey included the age and sex of the residents, residential environment, lifestyle, health conditions, residential type, presence and type of ventilation system, and air supply.

Table 2-1. Location of housing where on-site measurements were conducted

Area	Winter	Summer (Excluding the same location as winter)
Tokyo	27	1
Kanagawa	18	-
Chiba	15	-
Saitama	14	6
Ibaraki	9	6
Miyagi	6	1
Iwate	6	1
Fukushima	5	-
Yamanashi	4	1
Tochigi	4	-
Akita	3	-
Gunma	3	1
Yamagata	2	-
Aomori	1	-
Total	116	17

2.2.2 Equipment

A data recorder (TR-72nw, T&D Corporation) was used to measure the temperature and relative humidity during the measurement period. Table 2-2 lists the analytical conditions for gas chromatography-mass spectrometry (GC/MS) and high-performance liquid chromatography (HPLC). VOC was used for sampling, and Tenax TA (60/80 mesh, Sigma-Aldrich Corporation) was used as the adsorption medium. The adsorbed component of Tenax TA was 2,6-diphenyl-p-phenylene oxide. In addition, carbonyl (formaldehyde and acetaldehyde) sampling and analyses were conducted. Aldehydes are organic compounds classified as very VOC (VVOC) and are very heat-unstable; therefore, it is difficult to proceed with the analysis as they are. Therefore, they are commonly analyzed as stable conductors using a derivation agent during sampling. DNPH passive tubes (SIBATA) were used to collect carbonyls, with 2,4-Dinitrophenylhydrazine (DNPH) as a carbonyl derivatization agent. The solution was analyzed using high-performance liquid chromatography (HPLC).

This study used auto-thermal desorption (ATD) as a sample introduction device for GC/MS. The ATD was designed to introduce the extracted components through thermal desorption into the GC/MS. First, the ATD device was subjected to a comprehensive leak test. The sample was purged to remove oxygen and moisture and then heated continuously to extract the volatile components. These volatile components were electronically cooled and concentrated using a cold trap with a low heat capacity. Subsequently, the samples were re-heated and introduced into the GC column through a heated transfer line. Concentrating the test components in this manner allows analysis without compromising the high separation performance of the column. In addition, we conditioned empty Tenax TA and glass tubes after analyzing quartz wool using ATD. It is assumed that the samples in the sampling process are fully desorbed by heating during the analysis; however, some residues may remain in cases of high sample concentrations. In this study, we achieved complete desorption of the samples through heat treatment at 330°C for 20 min using ATD.

The quantitative analysis details are as follows: Each peak area in the chromatogram corresponds to the quantity of each chemical component. The relationship between the peak area and component quantity depends on the type of detector, operating conditions, and components being detected. Quantitative analysis is typically based on the assumption that a relationship is proportional. The proportionality constant can be determined by introducing a standard sample solution of known concentration into the separation column to create a chromatogram and then generating a calibration curve that relates the peak area to the concentration. We extracted 45 VOCs at known

concentrations from mixed standard stock solutions (Kanto Chemical Co.). We used a microliter syringe to collect these samples, injected them into a sampling tube containing Tenax TA, and prepared calibration curves for the GC/MS analysis. We immediately analyzed the sampling tube to prevent potential volatilization of the standard solution after injection. All substances detected by GC/MS were quantitatively analyzed using a calibration curve established with toluene. The converted toluene has only a semi-quantitative value. However, this discrepancy does not affect the study's objectives, which involved comparing the relative concentrations in the measured houses.

Table 2-2. Analytical condition for GC/MS and HPLC

Category	Content
GC/MS	SHIMADZU GCMS-QP2010 SE
ATD	ParkinElmer TurboMatrix (automated thermal desorber)
Column	Inert cap 5MS/Sil, 5% phenyl methyl silicone, 0.25 mmφ x 60 m x 0.25 μm
Oven temperature	40°C(5 min) - (10°C/min) - 300°C(9 min)
Carrier	Helium
Gas Flow	0.7 ml/min
Analysis mode	SCAN(33-400)
Desorption time [min]	10
Cold trap temperature [°C]	-50
Cold trap heating temperature [°C]	300
Transfer line temperature [°C]	300
HPLC	SHIMADZU Prominence (DGU-20A, LC-20AB, SIL-20AC, CBM-20A, SPC-M20A, CTO-20AC)
Column	Kinetex C18, 5.0 μm, 250 mm × 4.6 mm
Injection	20 μL
Column temperature	40°C
Flow	1.2 mL/min
Eluent	water / acetonitrile (4 : 6)
Detection wavelength	360 mm, 254 nm

Figure 2-1. Gas Chromatography-Mass Spectrometry

Figure 2-2. High-performance liquid chromatography

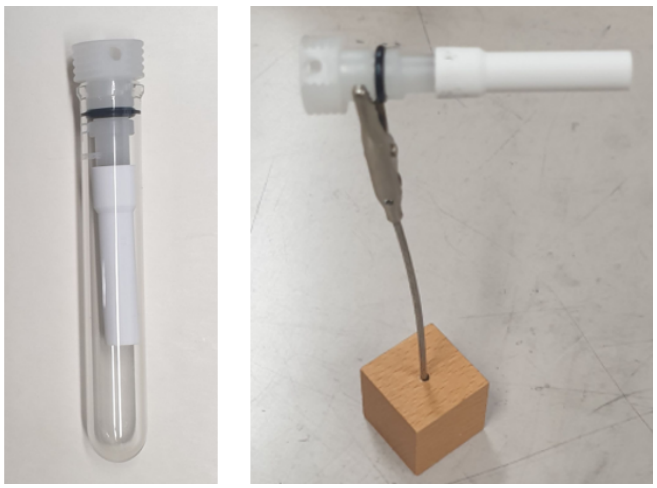

Figure 2-3. Passive Sampler of VOC

2.2.3 Methods for calculating VOC concentrations

The passive method requires the sampling rate (SR) of each substance to calculate the concentration. This rate refers to the speed at which the substance diffuses into the sample and is adsorbed onto the sorbent material, analogous to the suction speed of the pump in the active method.

SR varies depending on factors such as the shape of the sample, but the SR for some VOCs is provided based on the passive sampling mechanism. SR calculation methods for unknown substances are mainly divided into two types: experimental and actual measurements, and estimations based on theoretical formulas.

In this study, SR was estimated using a theoretical formula, and the concentration was calculated using this value. The theoretical formula for SR used in this study is expressed in Equations 2.1 to 2.6.

Equation 2.1 defines the material inflow amount J [ng/cm²/sec] corresponding to unit time t [sec] and unit area s [cm²].

$$J = D \times \left(\frac{C}{Z}\right) \quad 2.1$$

where: D is the diffusion coefficient, representing the rate at which the VOCs diffuse through the air [cm²/sec], C is the characteristic distance or thickness over which diffusion occurs [cm].

By multiplying both sides of Equation 2.1 by the sampling area A [cm²], the absorption rate W/t [ng/min] is obtained.

$$J \times A = D \times \left(\frac{C}{Z}\right) \times A = {}^{W}/_{t} \quad 2.2$$

Equation 2.3 establishes that the concentration C [ng/mL] is proportional to the absorption rate. When the exposure time t is expressed in minutes and the collection amount W is expressed in nanograms, the proportionality constant a [min/mL] can be obtained as shown in Equation 2.4.

$$C = a \times \left({}^{W}/_{t}\right) \quad 2.3$$

$$a = \left(\frac{1}{60}\right) \times \left(\frac{1}{D}\right) \times \left(\frac{Z}{A}\right) \quad 2.4$$

Since SR can be considered the same as the suction speed of the active method pump, this is expressed in Equation 2.5.

$$W/_t = SR \times C \quad (2.5)$$

According to Equations 2.3 and 2.5, SR is the reciprocal of a, which leads to Equation 2.6.

$$SR = \frac{60 \times A \times D}{Z} \quad (2.6)$$

For the diffusion area and diffusion length of the sample required in the above SR calculation equation, measurement values for various passive samples from observations and measurements using a scanning electron microscope from existing literature were used. The VOC-TD passive sampler used in this study had the same values as the VSD (diffusion area was 1.532 cm², diffusion length was 0.157 cm).

Additionally, the diffusion coefficient in Equation 2.6 can be obtained using the estimation equation. There are various equations for estimating the diffusion coefficient, but Equation 2.7 was used here. The critical temperature [K] and critical pressure [atm] used in the formula may be indicated in chemical engineering manuals, etc., but can be obtained using Equations 2.7 to 2.9 below.

$$D = \frac{0.00067 \times T^{1.83}}{[(\frac{Tc_1}{Pc_1})^{1/3} + (Tc_2/Pc_2)^{1/3}]^3} \times \sqrt{\frac{1}{M_1} + \frac{1}{M_2}} \quad (2.7)$$

For here, T : Temperature at the time of exposure [K], Tc_1: Critical temperature of air [K], Pc_1 : Critical pressure of air [atm], M_1 : Molecular weight of air [g/mol], Tc_2 : Critical temperature of the target substance [K], Pc_2 : Critical pressure of the target substance [atm], M_2 : Molecular weight of the target substance [g/mol]

$$Tc_2 = \frac{T_b}{0.567 + \sum \Delta T + (\sum \Delta T)^2} \quad (2.8)$$

$$Pc_2 = \frac{M}{(0.34 + \sum \Delta P)^2} \quad (2.9)$$

For here, T_b : Boiling point of the target substance [K], ΔT: Temperature difference [K], ΔP: Pressure difference [atm]; Addition factors for calculating critical constants.

The SR calculated in this study is shown in Table 2-3.

Table 2-3. SR of VOC

VOC	SR [m^3/h]
Toluene	0.002652547
Ethylbenzene	0.002403943
Xylene	0.002286322
Styrene	0.002364318
P-dichlorobenzene	0.00229253
Tetradecane	0.001215113
α-pinene	0.001808307
D-limonene	0.001863029

In this study, the substances for which sampling, analysis, and concentration calculation were performed using DNPH were formaldehyde and acetaldehyde. To calculate the concentration, the uptake rates of these substances were 5.1 and 5.6 [μg/(ppm·h)], respectively.

The following shows how to calculate the concentration of the substance sampled using the DNPH passive gas tube. The HPLC used in this study provides concentration results in [μg/mL] as the basic output of data. After extraction with 5 mL of acetonitrile, the sampling amount [μg] was calculated using Equation 2.10 before proceeding with the HPLC analysis.

$$W' = C_0 \times 5 \quad 2.10$$

For here, W'= Sampling amount [μg], C_o : Concentration [μg/mL]

The concentration C_{ppm} [ppm] can be obtained by multiplying the uptake rate (UR) by the sampling time t [h] and dividing by the sampling amount obtained in Equation 2-10.

$$C_{ppm} = W' \div (UR \times t) \quad 2.11$$

For here, C_{ppm} : Concentration[ppm], UR : Uptake rate [μg/(ppm·h)], t : Sampling time

Since the desired concentration unit in this study is μg/m^3, it was converted as shown in Equation 2-12.

Additionally, the temperature at the time of exposure was the average temperature of the measured house, and the atmospheric pressure at the time of exposure was 1013 Pa.

$$C' = C_{ppm} \times \frac{M_2}{22.4} \times \frac{273.15}{T} \times \frac{P}{1013} \times 10^3 \quad 2.12$$

For here, C' : Concentration[μg/m³] , M_2 : Molecular weight of the target substance [g/mol], T : Temperature at the time of exposure [K], P : Barometric pressure during exposure [Pa]

2.2.4 Statistical analysis

Statistical analyses were performed using the SPSS software (IBM SPSS Statistics Version 27.0.1) and Excel (2016). Differences in the parameters measured by the survey from residents, such as detached and apartment houses (type of house), residence period, and type of ventilation system, were tested using both parametric (two-sample t-test) and non-parametric (Wilcoxon Mann-Whitney rank sum test) tests. These tests were conducted on all the available data for each variable.

2.3 Results

2.3.1 Characteristics of houses

Table 2-4 shows the characteristics of the houses and the survey results. The number of custom-made houses was the highest, with 56 houses (48.3%) in winter and 38 houses (57.5%) in summer. Next, built-for-sale houses were the second most common in winter, with 20 houses (17.2%), followed by 19 private rental houses (16.3%), 18 apartments (15.5%), and 3 public corporation rental houses (2.5%). In summer, there were 11 apartments (16.7%), 10 built-for-sale houses (15.2%), 6 private rental houses (9.1%), and 1 public corporation rental house (1.5%). We surveyed the residents regarding their residence period. In winter, residences of 10–15 years were most common, with 40 cases (34.4%), followed by 26 cases (22.4%) for 3–5 years, and 19 cases (16.3%) for 16–19 years. In houses measured in summer, there were 17 cases (26.1%) for 10–15 years, 16 cases (24.6%) for 16–19 years, and 12 cases (18.4%) for 6–9 years. In addition, we investigated the daily usage of mechanical ventilation fans through a survey. In winter, 33 out of 100 participants (33.0%) answered that they always used a bathroom fan, followed by 18 (18.0%) who used it for more than 4 hours, 16 (16.0%) who used it for less than 30 minutes, and 10 (10.0%) who did not use it at all. For toilets, 37 cases (40.2%) responded that they always used them, followed by 26 cases (28.2%) in which they used them for less than 30 minutes, and nine cases (9.8%) in which they used them for 30 minutes to an hour. Other responses included "rarely used," "use it for a few minutes only when it smells," and "sensor-activated." It has been confirmed that most of these systems are either always used or used as needed, except in houses that lack proper ventilation. For bathroom fan usage time in summer, 15 cases (26.3%) reported using it all the time, and 13 cases (22.8%) reported using it for more than 4 hours, indicating that they were frequently used by more than half of the houses. For toilet fans, most respondents admitted to always using them or using them for at least 30 minutes (95.8%), except for two cases (4.1%) who said they did not use them. For ventilation fans in the living room and bedroom, 11 cases (50.0%) reported always using them, and three cases reported opening windows and ventilating without using ventilation fans (13.6%).

We surveyed the living rooms and wall-finishing materials of houses measured during the winter. The floor materials in the living room used in the surveys were typically divided into flooring, carpet, and straw mats. Consequently, 85.3% of houses had flooring as the main finishing material, 9.5% had carpets, and 5.2% had straw mats. The walls were investigated by dividing them into vinyl cross, wood products, paint, and diatomaceous earth. Vinyl cross was the main finishing material for the walls (58.6%), followed by wood products (31.0%).

Only 10% of the walls were finished with paint, diatomaceous earth, and other materials.

Table 2-4. The characteristics of houses

Season	Winter		Summer	
Parameter	N	(%)	N	(%)
Type of house				
Custom-made house (detached house)	56	(48.3)	38	(57.5)
Built-for-sale house (detached house)	20	(17.2)	10	(15.2)
Apartment	18	(15.5)	11	(16.7)
Private rental house	19	(16.3)	6	(9.1)
Public corporation rental house	3	(2.5)	1	(1.5)
Period of residence				
1–2	7	(6.0)	0	(0.0)
3–5	26	(22.4)	10	(15.3)
6–9	9	(7.7)	12	(18.4)
10–15	40	(34.4)	17	(26.1)
16–19	19	(16.3)	16	(24.6)
≥ 20	15	(12.9)	10	(15.3)
Type of ventilation				
Balanced ventilation	39	(33.6)	23	(34.9)
Unbalanced ventilation	77	(66.4)	43	(65.1)
The frequency of resident's drink				
Almost every day	33	(28.5)	17	(25.8)
4 or 5 days a week	11	(9.5)	8	(12.1)
2 or 3 days a week	22	(19.0)	16	(24.2)
Once a week	14	(12.1)	10	(15.1)
2 or 3 days a month	9	(7.8)	1	(1.5)
One day a month~	1	(0.9)	2	(3.0)
Less than one day a month	10	(8.6)	1	(1.5)
No drink	16	(13.8)	11	(16.7)
Flooring finishing materials (living room)				
Flooring	99	(85.3)	-	-
Carpet	11	(9.5)	-	-
Straw mat	6	(5.2)	-	-
Wall-finishing materials (living room)				
Vinyl cloth	68	(58.6)	-	-
Woody material	36	(31.0)	-	-
Paint	8	(6.9)	-	-
Diatomaceous earth, plaster	2	(1.7)	-	-
Other	2	(1.7)	-	-
The most used heating device				
Spaces	L	B	L	B
Air conditioner	43	63	(37.1)	(54.3)	-	-
Fan heater (use kerosene)	22	12	(19.0)	(10.3)	-	-
Fan heater (use gas)	6	2	(5.2)	(1.7)	-	-
FF type stove	5	1	(4.3)	(0.9)	-	-
Floor heating	10	2	(8.6)	(1.7)	-	-
Heated tables and electric blankets	15	0	(12.9)	(0.0)	-	-
Electric fan heater	0	8	(0.0)	(6.9)	-	-
No heating device	0	22	(0.0)	(19.0)	-	-
Other	15	6	(12.9)	(5.2)	-	-

Daily ventilation fan usage time Spaces	BT	T	L.B	B	T	L.B	B	T	L.B	B	T	L.B
Always use (24 hours)	33	37	13	(33.0)	(40.2)	(39.4)	15	24	11	(26.3)	(50.0)	(50.0)
Don't use	10	6	8	(10.0)	(6.5)	(24.2)	95	2	2	(8.8)	(4.2)	(9.1)
Less than 30 minutes	16	26	5	(16.0)	(28.2)	(15.2)	5	12	2	(8.8)	(25.0)	(9.1)
30 minutes to 1 hour	8	9	3	(8.0)	(9.8)	(9.1)	8	4	1	(14.0)	(8.3)	(4.6)
1 to 2 hours	5	6	2	(5.0)	(6.5)	(6.1)	4	3	0	(7.0)	(6.3)	(0.0)
2 to 4 hours	8	2	2	(8.0)	(2.2)	(6.1)	7	1	2	(12.3)	(2.1)	(9.1)
More than 4 hours	18	3	0	(18.0)	(3.3)	(0.0)	13	1	1	(22.8)	(2.1)	(4.6)
Others	2	3	0	(2.0)	(3.3)	(0.0)	0	1	3	(0.0)	(2.1)	(13.6)

1. BT is bathroom, T is toilet, L and B are living room and bedroom, respectively, and 2. " indicates no data.

Table 2-5. The guideline values for VOCs set by MHLW

VOCs	MHLW's guideline values [μg/m^3]
Formaldehyde	100
Acetaldehyde	48
Toluene	260
Xylene	200
Ethylbenzene	3800
Styrene	220
P-dichloro benzene	240
Tetradecane	330
Chlorpyrifos	1
Fenobucarb	33
Diazinon	0.29
Dibutyl phthalate	17
Di-2-ethylhexyl phthalate	100

2.3.2 Temperature and relative humidity

Due to the effects of indoor temperature and relative humidity on the emissions of indoor VOCs, temperature and humidity were measured during the measurement period. The measured temperature and relative humidity were the average values for all measurements (approximately 2-3 days). Figure 2-4 shows the results of the living room and bedroom measurements in winter and summer, respectively. In winter, the living room temperature was 17.2°C on average, and the relative humidity was 42.2%. The average bedroom temperature was 15.0°C, and the relative humidity was 47.3%. The average indoor temperature was 2.2°C higher in the living room than in the bedroom, and the relative humidity was 5.1% higher in the bedroom than in the living room. In summer, the average indoor temperature was 27.3°C and 27.8°C in the living room and bedroom, which did not differ significantly. For the indoor average relative humidity, those in the living room and bedroom were 61.4% and 61.0%, respectively, with no significant differences in relative humidity.

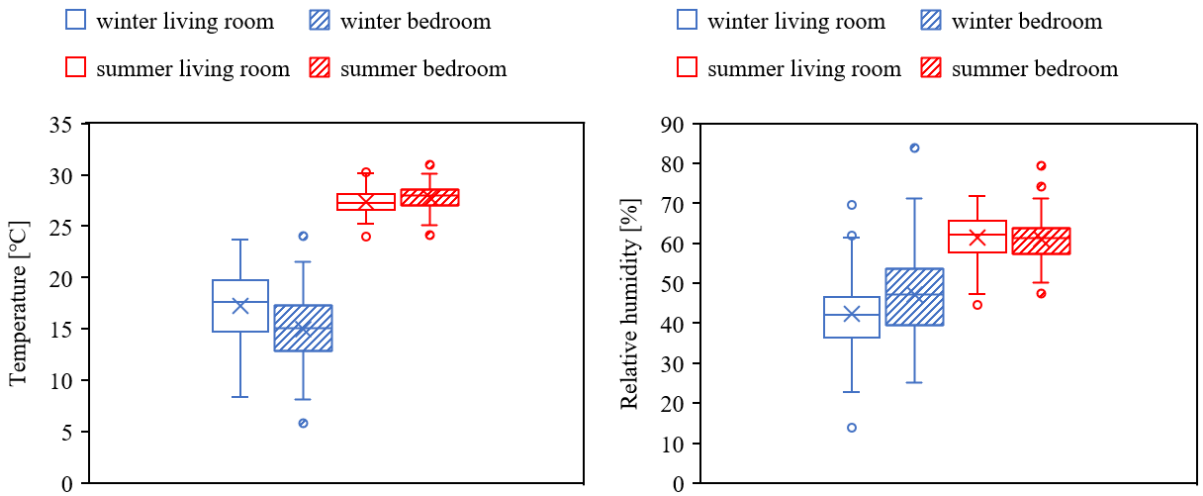

Figure 2-4. Temperature and relative humidity in living room and bedroom in winter and summer

2.3.3 VOC concentrations

Figure 2-5 shows the concentrations of formaldehyde and VOCs in the living rooms and bedrooms in winter (left figure) and summer (right figure). The formaldehyde concentration in winter was lower than the MHLW's guidelines in Japan (Table 2-5), except in one living room and bedroom. The concentration of acetaldehyde exceeded the MHLW's guideline values of 48 μg/m^3 in 17 living rooms and 15 bedrooms out of 116 households. As for the VOC concentrations, all other VOCs, including toluene, were lower than the guidelines indicated by the MHLW. In addition, for xylene ($P<0.01$ in winter, $P<0.05$ in summer), p-dichlorobenzene ($P<0.01$ in winter and summer), and α-pinene ($P<0.05$ in winter only), a significant difference between the concentration of the living room and the bedroom was confirmed. In summer, the concentration of formaldehyde in the living room exceeded the guideline values in two cases: formaldehyde in the bedroom exceeded three bedrooms, and acetaldehyde exceeded two bedrooms. For VOCs (toluene, ethylbenzene, xylene, decane, p-dichlorobenzene, 2-ethyl-1-hexanol, d-limonene, and α-pinene) similar to winter, no houses exceeded the guideline values, and there were no significant differences between VOC concentrations in living rooms and bedrooms. In the previous study, the concentration of formaldehyde in houses in Japan had a median value of 63.6 μg/m^3 [2-10], while another study reported an average formaldehyde concentration of 69.16 μg/m^3 [2-11] and 13.0 μg/m^3 [2-12]. Additionally, the average formaldehyde concentration in 160 homes in Australia was 28.0 μg/m^3 (equivalent to 22.8 ppb) [2-13]. The average formaldehyde concentration was 25.8 μg/m^3 in 15 Hungarian houses, 15.4 μg/m^3 in 72 French houses, and 22.2 μg/m^3 in 833 English houses [2-14,2-15,2-16]. The average formaldehyde concentration in this study was 13.8 μg/m^3 in winter and 27.2 μg/m^3 in summer, which was somewhat lower than the previous studies

or showed no significant difference.

First, we explain the characteristics and differences in the VOC concentrations measured during the winter and summer. In winter, the concentrations of formaldehyde and acetaldehyde in the living room did not exhibit significant differences with temperature changes, and no distinct patterns were observed (R^2 = 0.02 and 0.01). Bedrooms also exhibited data points for formaldehyde and acetaldehyde that followed linear patterns; however, these relationships were neither strong nor explanatory, and the predictions from the trend lines did not closely match the actual data points. Even for the concentration of d-limonene (R^2 = 0.05), which had the highest R-squared value in the bedroom, the relationship was weak, and the forecast from the trend line was not close to the actual data points. In the summer, the concentrations of formaldehyde and acetaldehyde in the living room decreased as the temperature increased. However, the R^2 values were 0.02 and 0.01, respectively, indicating that the data points followed some linear patterns but were not highly explanatory. Other VOCs showed slight increases or decreases depending on temperature, but no distinct characteristics were observed. The change in VOC concentrations did not significantly differ between winter temperatures of 8-24°C (with an average of 17.3°C ± 3.1 and 15.1°C ± 3.1 in the living room and bedroom) and summer temperatures of 24-31°C (with an average of 27.4°C ± 1.2 and 27.7°C ± 1.3 in the living room and bedroom). A study in Germany reported seasonal characteristics by continuously measuring the indoor VOC concentrations in ten apartments. This study reported that the pattern of annual seasons was the most dominant in apartments in Leipzig and Munich. They also reported that the characteristics of seasonal VOC changes were stronger than those of the locality. This study reports that seasonality and outdoor environmental conditions, such as the number and activity of residents, ventilation and wind velocity, and increased air pollution, are important variables [2-8]. Kinney et al. reported seasonal differences in indoor VOC concentrations compared with outdoor concentrations. One reason for the large changes in the concentration of VOCs due to seasonal characteristics is the air exchange rate, which is important for residents to open and close windows. Also, indoor temperature and humidity changes are seasonal, affecting indoor VOCs [2-17].

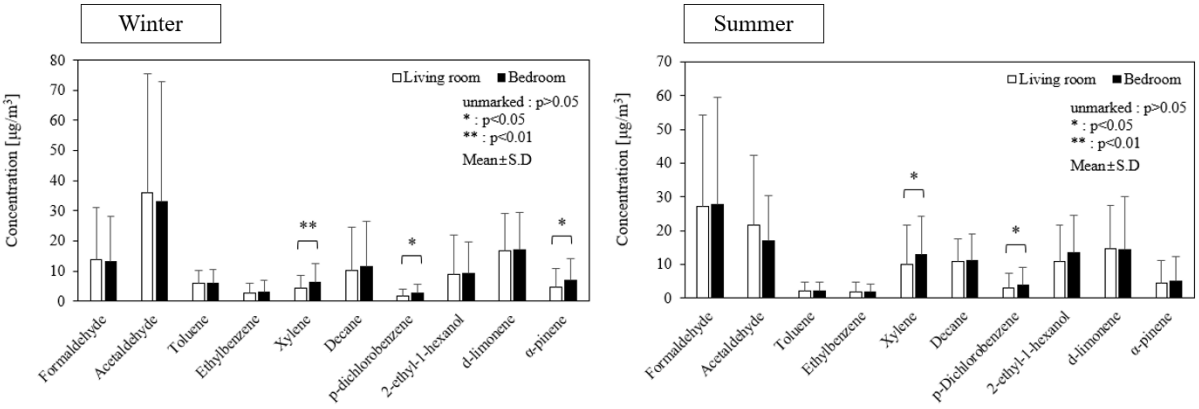

Figure 2-5. Average concentration of VOCs measured in winter and summer

2.3.4 Characteristics of housing and VOC concentrations

We reviewed the relationships among house type, residence period, type of ventilation equipment, and indoor VOC concentrations. Figure 2-6 compares the concentrations of formaldehyde and VOCs in winter according to house type. The analysis excluded corporate public rental houses because of their small sample size. The concentrations of formaldehyde, acetaldehyde, and toluene were higher in apartments (multifamily houses) in the living room and bedroom, whereas the concentrations in built-for-sale houses were the lowest. The concentration of d-limonene in private rental houses was the highest, whereas that in custom-made houses was the lowest. There was a statistically significant difference in the average concentration of acetaldehyde in bedrooms between custom-made houses and apartments ($P<0.05$). There was a statistically significant difference in the average concentrations of toluene in the living rooms of custom-made and built-for-sale houses ($P<0.05$). There was a statistically significant difference in the average concentration of xylene in both the living room and bedroom between built-for-sale houses and apartments. There were statistically significant differences between custom-made houses and private rental houses ($P<0.05$) and between apartments and private rental houses ($P<0.01$) in the living room.

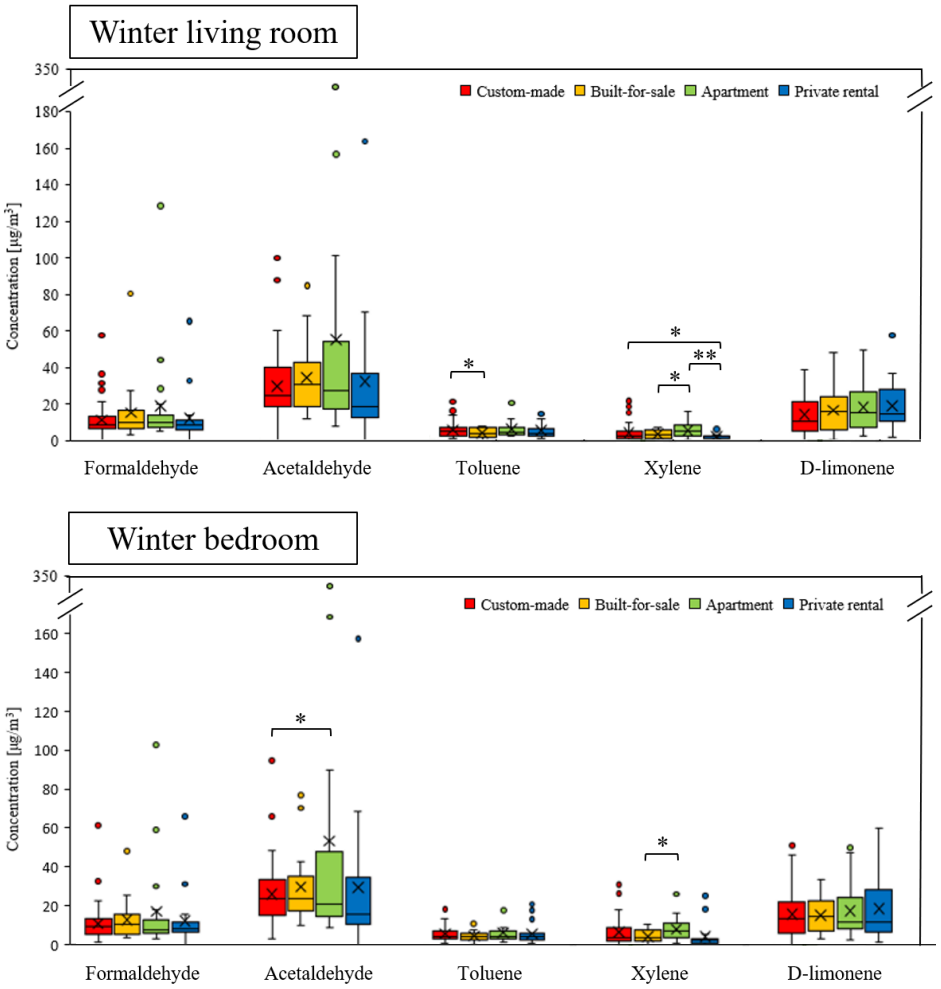

Figure 2-6. Comparison of VOC concentrations in living room and bedroom by type of house in winter

Figure 2-7 compares formaldehyde and VOC concentrations according to the type of house in summer. For formaldehyde and toluene, the concentrations in apartments were the highest, while for acetaldehyde and d-limonene, the concentrations in built-for-sale houses were the highest. In winter and summer, the indoor concentration of apartments was slightly higher than that of other house types. This increase may be due to the different building types, building materials used, airtightness depending on the type of house, and differences in resident ventilation [2-18]. The average concentration of acetaldehyde in the living room was statistically significant (custom-made and private rental houses; $P<0.01$). There was a statistically significant difference in the average concentration of toluene in the living rooms between custom-made houses and apartments ($P<0.05$). There was a statistically significant difference in the average concentration of xylene in the living rooms between custom-made and privately rented houses ($P<0.05$).

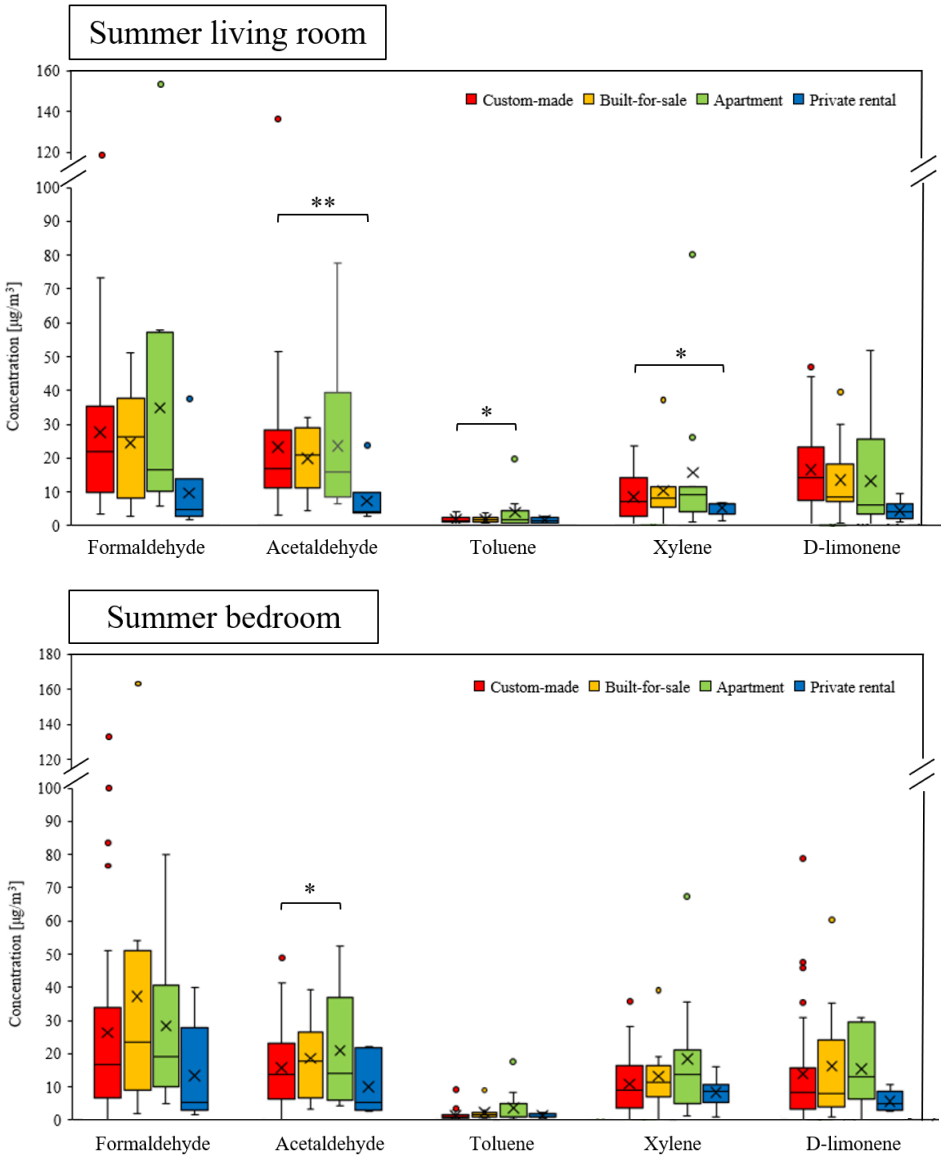

Figure 2-7. Comparison of VOC concentrations in living room and bedroom by type of house in summer

Figure 2-8 shows indoor aldehyde and VOC concentrations in winter according to the residence period, and the residence period was investigated because residents often do not know the exact year of construction. The average value was calculated as the survey was conducted during different residence periods. In general, if the concentration of VOCs was followed up for a long period in the same house, the concentration of VOCs gradually decreased, as in previous studies. However, because the same house was not measured, it was not possible to confirm whether the longer the residence period, the lower the concentration of VOCs. The average concentration of acetaldehyde in the bedroom during winter was higher when the residence period was 20 years than 1.5 years. (16 μg/m^3 vs. 50 μg/m^3) In three of these houses, the concentrations of 48.5 μg/m^3, 66.4 μg/m^3, and 338.9 μg/m^3

were confirmed, exceeding the MHLW's guideline value of acetaldehyde, 48 μg/m^3. According to the survey results, these houses only had bathrooms and toilets with ventilation fans installed, and the openings were closed, even though they were facing the outer wall. It has also been reported that the residents drink daily. In addition, most of the measured VOC concentrations did not show significant differences according to the residence period, nor did they show any distinct characteristics.

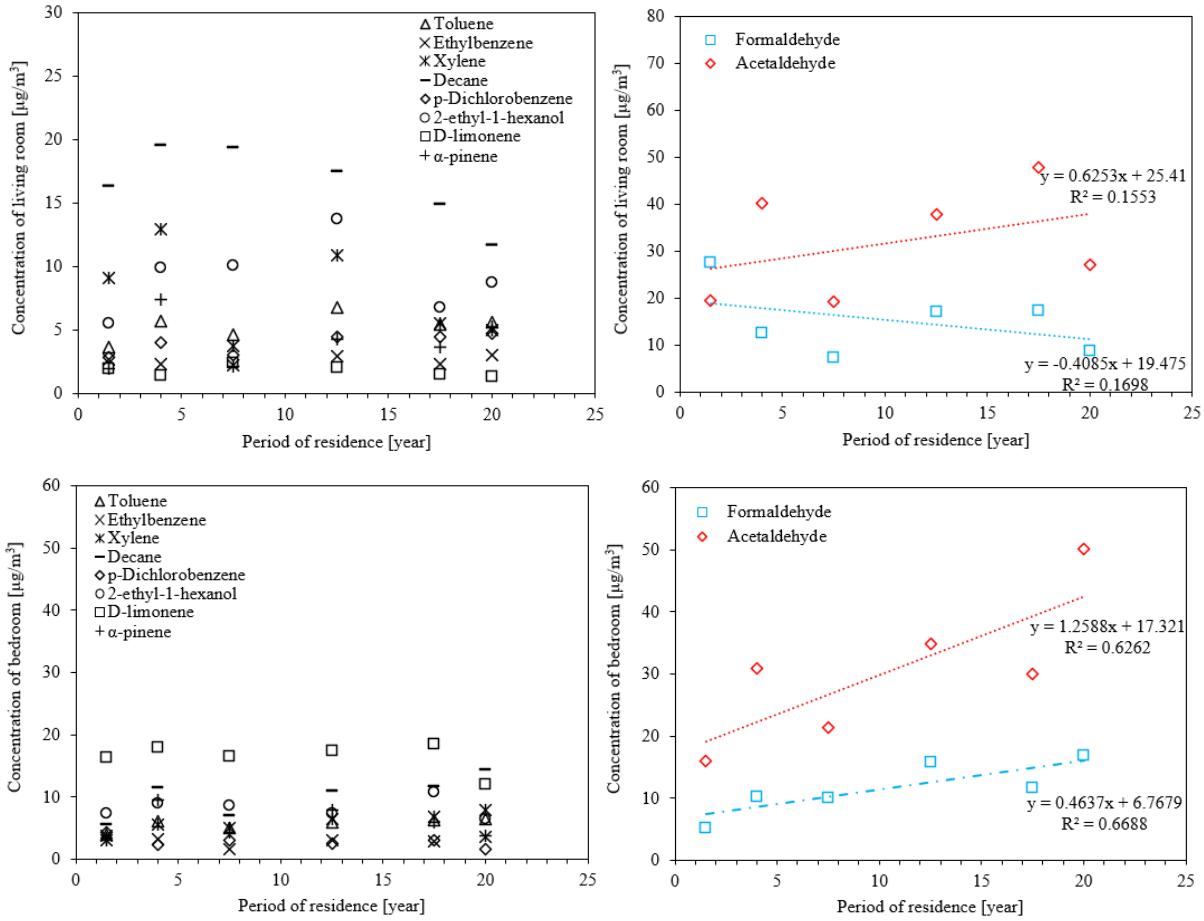

Figure 2-8. Comparison of VOC concentrations by residence period in living rooms and bedrooms in winter

Figure 2-9 shows the concentrations of VOCs according to the summer residence period. Most substances did not show much difference according to the residence period. For formaldehyde concentration in the bedroom alone, houses with a residence period of 1.5 years were measured the highest at 40.4 μg/m^3, followed by houses with a residence period of 20 years with 30.6 μg/m^3, the second highest average formaldehyde concentration. Formaldehyde is often used in adhesive resins for complex wood products, such as plywood, medium-density fiberboard, and hardwood plywood paneling, and is commonly used for furniture cabinet countertops and flooring because there are several pollutants such as insulation materials, carpets, household cleaning products, cosmetics,

and preservatives, and the characteristics of concentration according to the increase or decrease in the residence period could not be found [2-20].

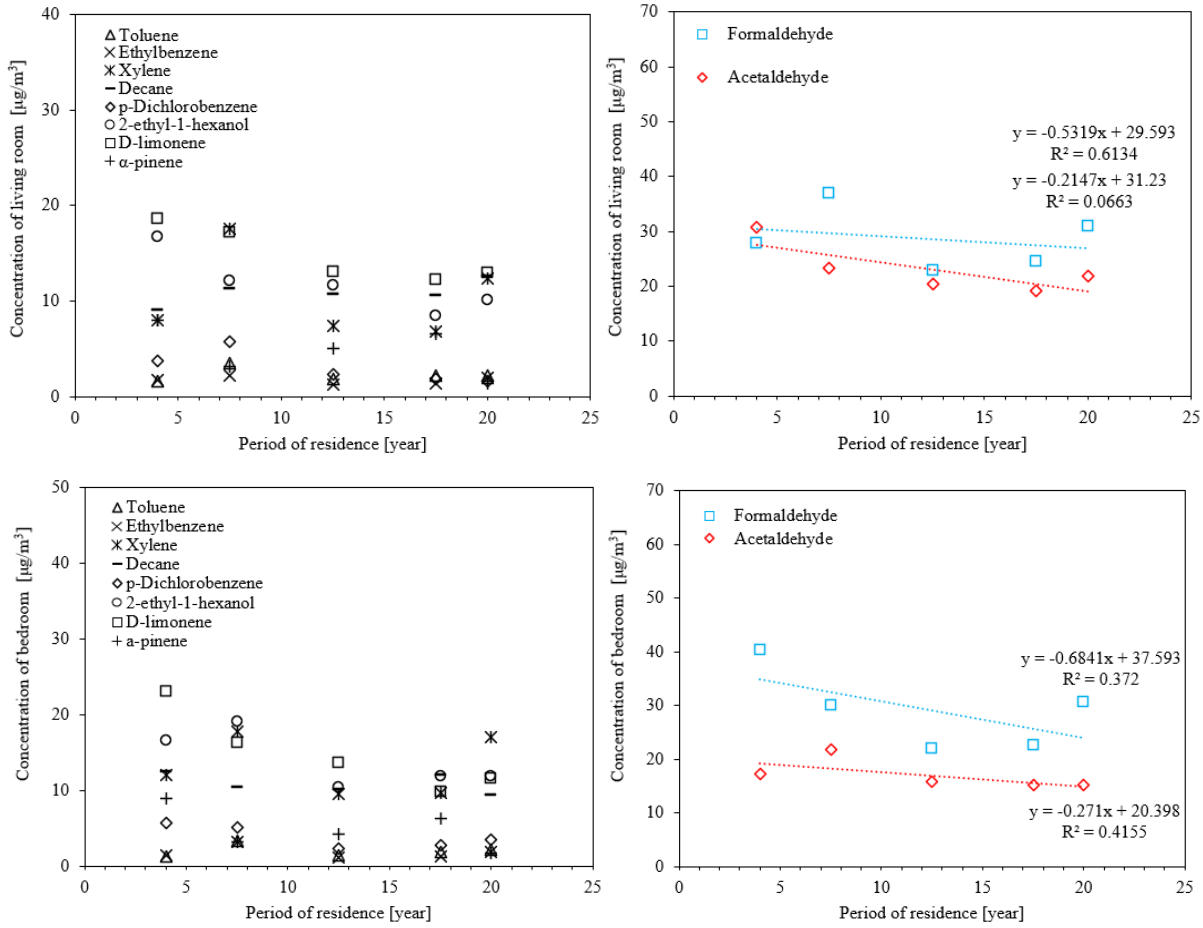

Figure 2-9. Comparison of VOC concentrations by residence period in living rooms and bedrooms in summer

Considering the type of ventilation equipment, it was difficult for residents to identify the type of mechanical ventilation. Thus, we classified balanced and unbalanced ventilation based on a combination of factors: installation location, supply air location, and fan presence. Cases with a fan in the bathroom and washroom and an air supply port installed on the outer wall surface were classified as having unbalanced ventilation. Balanced ventilation occurs when fans are attached to the living room and bedroom and when the air supply does not face the outer wall or ceiling, even if the fans are not attached to the living room and bedroom. Thus, the concentration of each pollutant was compared between balanced and unbalanced ventilation.

Figures 2-10 and 2-11 compare carbonyl and VOC concentrations according to the type of ventilation in winter and summer. In winter, all substances, except ethylbenzene, in the living room were 1.01 to 1.71 times higher

under unbalanced ventilation than under balanced ventilation. All substances except α-pinene in the bedroom had concentrations 1.01 to 1.63 times higher than balanced ventilation. For summer living rooms, only toluene, xylene, p-dichlorobenzene, and d-limonene α-pinene were high in unbalanced ventilation, and there was no consistency. Most VOCs, including toluene, were high under unbalanced ventilation in the bedroom. Formaldehyde and acetaldehyde were 1.24 times higher during the summer under balanced ventilation than under unbalanced ventilation. It was confirmed that the concentrations during winter were higher than those during summer, and the concentrations under unbalanced and balanced ventilation were higher than those in previous studies.

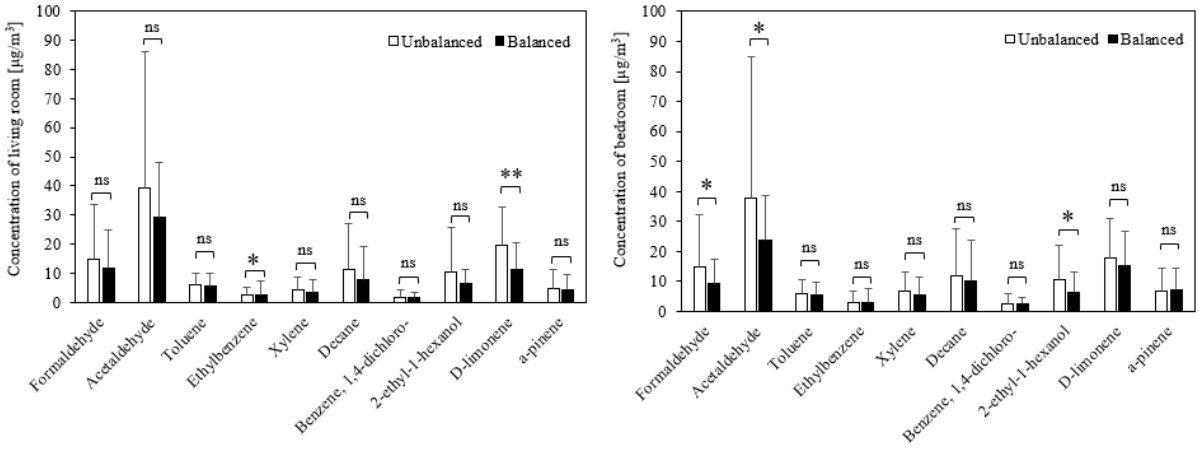

Figure 2-10. VOC concentrations in winter: unbalanced ventilation vs. balanced ventilation.

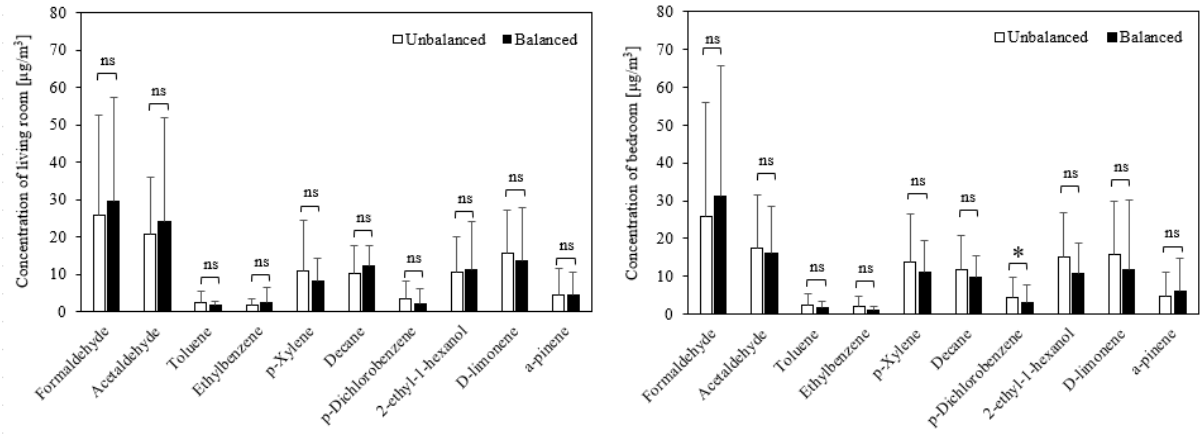

Figure 2-11. VOC concentrations in summer: unbalanced ventilation vs. balanced ventilation.

According to the survey results (Figure 2-12), 22 and 12 houses were equipped with fan heaters (kerosene) for heating the living rooms and bedrooms, respectively, during the winter. In the living room with a fan heater (kerosene), the average decane was 17.5 μg/m^3. The average decane concentration in the living room using other heating devices was 9.6 μg/m^3. In the bedrooms, the average concentration of decane using a fan heater (kerosene)

was 28.1 μg/m^3. The average decane concentration in the bedroom using other heating devices was 10.6 μg/m^3. (All concentrations were rounded to the second decimal place.) The average concentration of decane in living rooms and bedrooms using fan heaters (kerosene) was 57.8% to 136.4% higher than in homes using other heating devices. There was no statistically significant difference in the decane concentration between the living room and bedroom. Decane is sourced from fossil fuels, especially oil [2-19]. In previous studies, the decane concentration of the house where we lived was approximately three times higher than that of a vacant house, so it is largely related to human activities [2-7]. The category of homes that used kerosene only included those that reported using fan heaters (kerosene) and did not include FF-type stoves (Forced draught balanced flue). Therefore, the higher concentrations of decane observed in the "no kerosene" homes can be attributed to the use of FF-type stoves.

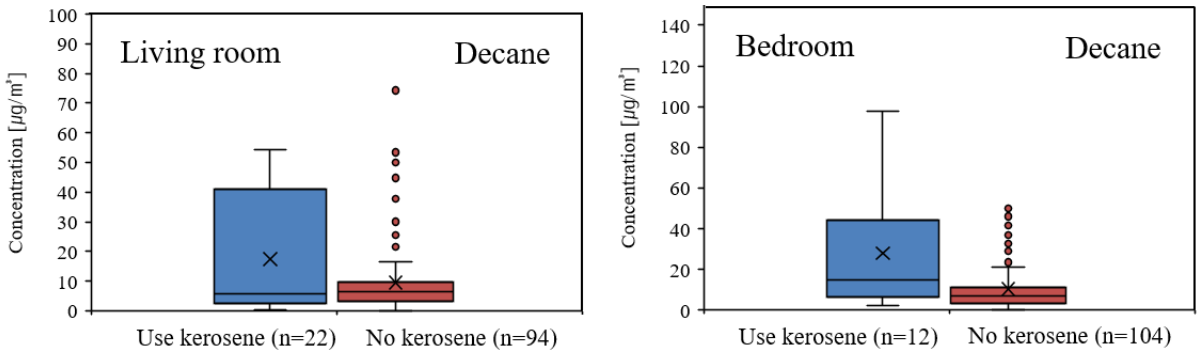

Figure 2-12. Comparison of decane concentrations in living room and bedroom in winter

2.4 Discussion

2.4.1 VOC Concentrations in different types of houses

We measured the concentration of VOCs in detached houses and apartments where the residents live in winter and summer. In addition, the characteristics of houses and residents were identified using a questionnaire. The survey results classified the type of house, residence period, and ventilation system as parameters. Houses were classified as custom-made, built-for-sale, apartments, and private rentals. Except for apartments, all three types of houses were detached. Each house had the following characteristics: Custom-made houses are tailored to homeowners' specific requirements and preferences, resulting in wide variation in the materials used. Custom-made houses typically emphasize high-quality materials and customization. They may sometimes incorporate traditional materials based on each country's characteristics and newer materials like steel, concrete, and advanced insulation for energy efficiency [2-20,2-21]. Built-for-sale houses are constructed for the general market, and most adhere to standardized designs and common material choices. Building materials are selected to balance cost, efficiency, and quality. These houses utilize various building materials, similar to custom-made houses, and often feature wood and steel structures with various finishes, such as synthetic sidings and brick veneers [2-22,2-23]. When selecting building materials, apartments are typically designed to be cost-effective and durable. Reinforced concrete and steel constructions are common in high-rise apartments. Inner finishing materials may include dry walls, laminate flooring, and ceramic tiles [2-24]. This study observed significant differences in toluene and xylene concentrations in houses during both winter and summer ($P<0.05$). Specifically, significant differences were observed in acetaldehyde concentrations between custom-made houses and apartments in winter, particularly in the bedrooms. Various building materials, including paint, coatings, flooring, and other construction materials, are used based on the type of building, and variations in these materials can impact indoor air quality. Additionally, statistically significant differences were identified for toluene and xylene in both winter and summer, confirming that the association with building type significantly influenced the indoor concentrations of these substances. In winter, statistically significant differences in acetaldehyde concentrations were observed, particularly in the bedrooms of custom-made houses and apartments, suggesting that building type, especially in winter, can influence the indoor concentrations of these substances. Furthermore, the choice of building materials in various house types can lead to variations in indoor pollutant concentrations, as confirmed by statistical analyses, with significant differences observed for certain substances.

2.4.2 Temporal changes in VOC concentrations

According to previous research, because pollutants such as building materials that affect indoor VOC concentrations vary significantly over time, the concentration characteristics according to the residence period were identified [2-6]. P. Wolkoff tested five building materials for 250 days at varying temperatures (23°C, 35°C, and 60°C) and humidity (0%, 50%). Most VOC concentrations were reported to decrease sharply after four weeks [2-25]. Previous studies have shown that the level of VOCs in new houses steadily increases during the initial months but decreases significantly after one year. It has been reported that the level of VOCs in existing houses did not change significantly over the past three years [2-9]. Other studies have identified VOC concentrations in homes, schools, and offices in China, with a reported decrease between 2000 and 2021 [2-33]. However, the results of this study did not indicate a tendency for the concentration to decrease with the duration of residence, which is likely because the measurements were not taken over several years in the same house. However, there were cases in which the concentration of acetaldehyde was high depending on the residence period, suggesting that factors or variables such as cleaning, ventilation, product use, changes in building materials and furniture, renovations, smoking, and drinking may play a role [2-26].

2.4.3 Seasonality and VOC concentrations

We investigated the change in VOC concentrations according to season (room temperature). The results of this study did not reveal the characteristics of the VOC concentrations with temperature. The R^2 values of the temperature and VOC trendlines were low. Although the data for VOCs and concentrations (R^2 = 0.01-0.06) follow some linear patterns, they are not strong or explanatory, and the trend line predictions are not closely aligned with the actual data points. Additionally, in the living room, the concentrations of toluene, ethylbenzene, d-limonene, α-pinene, and acetaldehyde were higher in the winter than in the summer. In the bedroom, the toluene, ethylbenzene, decane, d-limonene, α-pinene, and acetaldehyde concentrations were also higher in the winter. Mentese et al. measured VOCs in homes, offices, kindergartens, and elementary schools for five days during the winter and summer in Ankara, Turkey. Their results showed that TVOC, benzene, toluene, ethylbenzene, xylene, and hexane concentrations were higher in winter than in summer, except for office TVOC, which showed no detection [2-27]. Wolkoff et al. measured the VOCs for one year in two Danish twin apartments. VOCs generated from building materials generally decreased, but formaldehyde concentrations increased again during the fall. Human activity has been suggested to contribute to several indoor VOCs. Hexanal also increased during the warm

season, and TVOC was reported to be approximately 50 percent higher during the winter [2-7]. Seifert et al. studied the seasonal changes in VOC concentrations in German houses. They measured VOC concentrations in 12 houses in Berlin over one year using a manual sampler 26 times for two weeks each. In most houses, it has been reported that the total VOC concentration in winter was 2-3 times higher than that in summer [2-28]. Weihui et al. conducted regular VOC measurements for 18 months after the completion of the internal construction of a new apartment in Beijing, China. Immediately after internal construction, the VOC concentrations were very high and showed a noticeable decline in the first few months. Subsequently, the pattern changed, and more diverse VOCs with higher concentrations were observed in the summer than in the winter. They reported that the rate of emissions from building materials may have increased because of the nature of new apartments [2-5].

2.4.4 Influence of ventilation systems on VOC concentrations

We also assessed concentrations based on the type of ventilation system used. Previous studies have reported that balanced ventilation systems ventilate homes more than unbalanced systems, resulting in higher air exchange rates [2-29]. In Godish's study, reductions in indoor formaldehyde concentration were measured in a urea-formaldehyde foam-insulated house and a mobile home using a heat exchange ventilation system. The air exchange rate ranged from 0.45 to 1.49 ACH, resulting in a 57-67% decrease in formaldehyde [2-30]. Additionally, homes with balanced ventilation systems had lower VOC concentrations than those with unbalanced ventilation systems [2-31]. Huang et al. conducted indoor air quality monitoring in eight different buildings and found that buildings with mechanical ventilation systems had lower levels of indoor TVOC and formaldehyde than those with natural ventilation, demonstrating the effectiveness of mechanical ventilation in reducing indoor pollutant concentrations [2-32]. Consistent with previous research, we observed that homes with balanced ventilation had lower VOC concentrations than those with unbalanced ventilation. Specifically, we found statistically significant differences in the concentrations of d-limonene and ethylbenzene during winter and formaldehyde, acetaldehyde, and 2-ethyl-1-hexanol during summer between balanced- and unbalanced-ventilation houses. These findings suggest that balanced ventilation systems provide higher air exchange rates, leading to more effective removal of indoor contaminants.

2.4.5 Heating appliances and decane

The sources of decane generated indoors can vary widely. In addition to combustion appliances, paints, varnishes, adhesives, and sealants release decane over time due to the solvents used in their manufacturing processes [2-33,

2-34]. Household cleaning products and air fresheners emit decane into the air during use [2-35], and newly purchased furniture and carpeted textile products also release decane over extended periods as a result of the chemicals used in their production [2-36, 2-37]. Electronic devices emit decane from internal components and plastic casings, and cooking processes that use gas or oil generate decane along with various hydrocarbons [2-38,2-39]. Tobacco smoke contains various VOCs, including decane. Lastly, automotive maintenance products such as gasoline, engine oil, and solvents, when stored indoors or in garages, can release decane into indoor air [2-40,2-41]. In this study, we compared the use of fan heaters (kerosene) with other cases. Higher concentrations of decane were observed in homes where residents reported using FF-type stoves (Forced draught balanced flue), suggesting that these stoves may be one of the sources of decane. In this study, as a result of comparing houses that use fan heaters (kerosene) and houses that are not used in winter, the concentration of decane in houses that use fan heaters (kerosene) was higher than in houses that are not used.

2.4.6 Health risk assessment

This study currently focuses on surveying the actual VOC concentration conditions in Japanese houses. However, ultimately, it is necessary to evaluate the effect of VOC concentration on health. To achieve this, there is a way to utilize the human health risk assessment model proposed by previous studies while conducting a long-term follow-up on VOC concentration and health status [2-42]. Given that the subject of this survey is housing in specific areas of Japan, with surveys conducted at one point in each winter and summer season, it is necessary to carefully review the representativeness of the survey results and subsequently evaluate health risks. Therefore, the health risk assessment will be considered in future studies.

2.5 Summary

The novelty of the study in Chapter 2 is that it reveals the characteristics of VOC concentration according to various regions and types of houses targeting houses actually occupied by residents in Japan, and evaluates the VOC concentration of balanced ventilation and unbalanced ventilation equipment as a factor.

This study measured VOCs and carbonyls in the living rooms and bedrooms of 116 Japanese houses in winter and 66 Japanese houses in summer. Most of the substances were present at very low concentrations. However, 12% of the measured houses exceeded the guideline values for formaldehyde and acetaldehyde. Although the number of samples was small, the characteristic of the three houses where the concentration of acetaldehyde exceeded the guideline value was that they drank almost every day. In addition, when comparing the concentrations in the living room and bedroom, although each substance was different, the concentrations in the living room were slightly higher. However, there were no significant seasonal differences in the concentrations of VOCs between the living rooms and bedrooms. As a result of identifying the concentration by housing type, the concentrations were highest in apartments, and the concentration in built-for-sale houses tended to be higher than that in custom-made houses. In winter, there were statistically significant differences ($P<0.01$) in the average xylene concentrations between apartments and private rental houses. Significant differences ($P<0.05$) were also observed between custom-made and private rental houses and between built-for-sale houses and apartments. In summer, there were statistically significant differences ($P<0.01$) in the average acetaldehyde concentrations between custom-made and private rental houses. Significant differences ($P<0.05$) were found between built-for-sale and private rental houses and between apartments and private rental houses. In the winter and summer, the concentration of VOCs was determined according to the residence period in the bedroom and living room. As a result, most of the VOCs, except acetaldehyde, did not show any difference in concentration or distinct characteristics according to the residence period. Comparing the concentrations of VOCs in houses using balanced and unbalanced ventilation systems, houses with unbalanced ventilation had a higher average concentration of most pollutants in winter than those with balanced ventilation. In summer, houses with unbalanced ventilation had a higher average concentration of pollutants than houses with balanced ventilation, except for formaldehyde. In winter, the average concentration of decane in living rooms and bedrooms using fan heaters (kerosene) was 57.8% to 136.4% higher than in homes using other heating devices. If the concentration of decane was high in a house that did not use kerosene, it was a house that responded that FF-type stores were used.

To sum up the important results, the study revealed that while VOC and carbonyl concentrations in most Japanese houses were low, 12% exceeded guidelines for formaldehyde and acetaldehyde, particularly in homes with frequent alcohol consumption. Living rooms generally had higher VOC levels than bedrooms, regardless of season. Apartments and built-for-sale houses exhibited the highest concentrations, with significant differences observed between housing types for xylene and acetaldehyde levels. Unbalanced ventilation systems were associated with higher pollutant concentrations compared to balanced systems, especially in winter. Additionally, the use of kerosene fan heaters significantly increased decane concentrations in winter. These findings indicate that housing type, ventilation system, and heating method are critical factors affecting indoor air quality, necessitating tailored strategies to mitigate VOC exposure in residential settings.

The data in this study are valuable for understanding the levels of VOC and carbonyl concentrations in various houses in Japan during winter and summer. Also, residents were surveyed on the type of house, residence period, and type of ventilation, and the relationship between indoor VOC and carbonyl was analyzed. The results of this study provide recent data on indoor VOC and carbonyl concentrations in Japanese houses and may offer better information on ventilation strategies. Finally, this study will be a useful foundation for conducting health risk assessments in future studies.

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Chapter 3. Impact of mechanical ventilation systems on VOC concentrations in a detached house

3.1 Previous study on the ventilation system and VOC concentration in housing

In Chapter 2, VOC concentrations were identified according to the characteristics of various houses. As a result, there were significant differences in concentration in houses using balanced ventilation compared to unbalanced ventilation. The VOC concentration in houses using unbalanced ventilation was higher than that in houses using balanced ventilation. Therefore, in Chapter 3, additional measurements were conducted for houses using only balanced ventilation and unbalanced ventilation. The papers related to the ventilation system are as follows. Here's a review of studies on VOC concentrations in mechanically ventilated homes that examined indoor air quality in California residences with mechanical ventilation systems. During the measurement period, all houses operated their mechanical ventilation systems with closed windows for one week. The measured pollutants included formaldehyde, nitrogen dioxide, carbon dioxide, and particulate matter. The study reported a 44% decrease in the average concentration of formaldehyde compared to previous California study results, with a higher ventilation rate. Most measured pollutants were found to be below health guidelines, indicating compliance with indoor air quality standards in California houses equipped with residential mechanical ventilation. However, data on the time and frequency of ventilation were insufficient during the initial visits to the measuring houses, as only 26% of the houses were using ventilation systems [3-1]. Research on existing houses, rather than newly built ones, is lacking. Even in existing houses, various sources of VOCs must be identified to assess the indoor environment. Houses without balanced ventilation, in particular, may suffer from inadequate ventilation.

Studies on natural and mechanical ventilation indicate that houses with mechanical ventilation have higher ventilation rates and lower VOC concentrations than those with only natural ventilation [3-2]. Houses with only natural ventilation may lack continuous and systematic ventilation due to the influence of residents' subjective ventilation habits. If houses with mechanical ventilation systems are not in use, ventilation may be inadequate, leading to higher concentrations [3-3]. Verniers et al. explored the differences in particulate matter (PM) and VOC levels under various ventilation conditions, including natural and mechanical ventilation, during everyday activities like cooking and vacuuming. It was found that mechanical ventilation systems (including heat recovery types) were more effective at reducing VOC concentrations compared with natural or no ventilation, especially during high emission activities like cooking [3-4]. In another study, researchers examined VOC emissions from typical household activities and found that mechanically controlled environments could better manage and reduce VOC levels compared to natural ventilation settings. Controlled ventilation effectively mitigated the impact of

high-emission activities like cooking [3-5]. Hernandez et al. assessed VOC emissions from new furnishings in settings with different ventilation strategies. It highlighted that mechanical ventilation significantly lowers VOC concentrations faster and more efficiently than natural ventilation in newly furnished homes [3-6]. There is data on the comparison of VOC concentrations in houses with and without mechanical ventilation, but studies on VOCs according to the type and method of mechanical ventilation are lacking. Additionally, previous studies have relatively large datasets on VOCs and ventilation systems for commercial buildings or educational facilities, rather than houses [3-7, 3-8].

Previous studies have primarily focused on the qualitative analysis of VOC concentrations across various residential settings, often overlooking the impact of mechanical ventilation types and operational habits on indoor air quality.

The novelty of this study lies in its examination of VOC concentrations in various regions and types of residences in actual inhabited homes in Japan. Specifically, it evaluates the VOC concentrations of balanced ventilation and unbalanced ventilation systems as one of the influencing factors. In this study, the hypothesis was set that houses with balanced ventilation systems would have stable ventilation rates, whereas houses with unbalanced ventilation systems would have higher VOC concentrations due to insufficient ventilation rates or the induction of VOCs into the indoor environment due to negative pressure. Traditionally, ventilation systems have been mandated as a countermeasure for sick house syndrome, but there have been very few studies on the effects of different ventilation methods (balanced and unbalanced) and even fewer studies on VOC concentrations in houses using a whole-house ventilation system. Furthermore, the study aims to investigate not only how ventilation rates determine indoor VOC concentrations but also how the ventilation systems affect indoor air quality through interactions with wall structures, as highlighted in this study, in actual inhabited homes.

Therefore, in this study quantitatively measured the concentration of VOCs in living rooms and bedrooms, specifically considering the type of mechanical ventilation—balanced (Group A) versus unbalanced (Group B)—used in houses occupied by residents during the contrasting climatic conditions of winter and summer. Furthermore, a comprehensive questionnaire survey was conducted to gain deeper insights into behavioral and environmental factors affecting VOC levels. This survey gathered data on the year of construction, the status of air inlets (open or closed), and detailed user engagement with the ventilation systems, including usage time, frequency, and the reasons behind their preferences. Additionally, this study also measured indoor carbon dioxide levels to approximate the air change rate, exploring its relationship with VOC concentrations. By integrating

quantitative measurements with user behavioral data, this study provides a more holistic understanding of the factors influencing indoor air quality in residential settings.

3.2 Measurement of VOC concentration in houses with balanced and unbalanced ventilation systems

3.2.1 Measurement overview

Field measurements were conducted in single-family homes in Northern Tokyo (Ibaraki Prefecture), Japan, where current occupants reside. These homes were distinctly categorized based on their ventilation systems, with separate measurements taken in homes using balanced ventilation (Group A) and those using unbalanced ventilation (Group B). In this study, balanced ventilation refers to a central air conditioning system that manages the air quality and temperature throughout the entire building. This means the system not only cools or heats the air but also ensures a constant flow of fresh air by supplying fresh air from outside and removing stale air from inside in equal amounts, creating a balanced and comfortable indoor environment. Balanced ventilation is a feature added to the central air conditioning system, not the central air conditioning system itself. The single-family homes were characterized as having glass wool insulation. Homes with balanced ventilation were built between 2011 and 2021, with an average construction year of approximately 2018. Homes with unbalanced ventilation were built between 1995 and 2020, with an average construction year of approximately 2006. VOCs in the measured homes were sampled in the living room and bedroom during winter and summer. The field measurement periods were from January 20 to March 4, 2022, and from July 17 to September 30, 2022.

3.2.2 Equipment

The analytical conditions for GC/MS and HPLC are listed in Table 2-2 in Chapter 2, which are the same as Table 3-1. VOCs were measured using the active method in accordance with the Manual for Measurement of Indoor Airborne Chemical Substances by the Ministry of Health, Labor and Welfare. All openings facing the outside air in the measurement house were opened and the house was ventilated for 30 minutes. Then, the room was sealed and left closed for a period of five hours before measurements were taken. VOCs were sampled in a Tenax Tube (Sigma-Aldrich Co.) using a mini-pump (MP-Σ30NII, SIBATA Co.) at a height of 1.2m-1.5m in the center of the living room for 30 minutes (flow rate : 0.3 L/min, total: 9 L). Aldehydes were also sampled with a DNPH cartridge (SIBATA) at a flow rate of 1 L/min, accumulating a total volume of 30 liters. In this study, an auto-thermal desorption (ATD) system was utilized as a sample introduction device for GC/MS. ATD is a device designed to thermally deliver extracted components to GC/MS. Initially, the ATD unit underwent a comprehensive leak test. After purging the sample to remove oxygen and moisture, the volatile components were extracted through

continuous heating. The extracted components were then cooled using a low-temperature capacity cold trap and concentrated, subsequently reheated and transferred to the GC column via a heated line. This process effectively concentrated the components, allowing them to be analyzed while maintaining the separation capabilities of the column. Prior to sampling, the quartz was treated using ATD, and the Tenax TA and glass tubes underwent a conditioning process. While the experiments assumed nearly complete desorption of the sample during analysis, some residue may remain if the sample concentration is high. To ensure complete desorption, samples were heat-treated at 300°C for 20 minutes. The area of each chromatogram peak represents the amount of that chemical component, and the relationship between peak area and component amount can vary depending on the detector type, operating conditions, and the component being detected. Quantitative analysis is typically based on the proportional relationship of components. The proportionality constant is determined from a calibration curve generated by introducing a mixed standard sample solution of known concentration into the chromatograph and comparing the peak areas and concentrations. This calibration process involved extracting a standard sample solution (Kanto Chemical Co.) containing known concentrations of 45 VOCs. The standard solution was injected into a sampling tube containing Tenax TA for GC/MS analysis, and the analysis was performed immediately after injection to minimize volatility. All detected substances were quantitatively analyzed using the established toluene calibration curve. The analysis of the carbonyls was conducted using a high-performance liquid chromatography (HPLC) system configured with a Shimadzu Prominence setup, which includes modules such as DGU-20A, LC-20AB, SIL-20AC, CBM-20A, SPC-M20A, and CTO-20AC. The chromatographic separation was achieved on a Kinetex C18 column, measuring 250 mm by 4.6 mm with a 5.0 μm particle size. For the injections, a volume of 20 μL was used. The column temperature was maintained at 40°C throughout the analysis. A flow rate of 1.2 mL/min was employed, utilizing an eluent mixture of water and acetonitrile in a 40:60 ratio. Detection of analytes was performed at wavelengths of 360 nm and 254 nm, providing the necessary sensitivity for identifying and quantifying the components in the samples. These wavelengths were selected based on their ability to effectively absorbance characteristics of carbonyl compounds. This setup was chosen for its efficiency and reliability in separating complex mixtures, ensuring precise analytical results. A data recorder (TR-76Ui T&D Corporation) was used to record the temperature, relative humidity, and CO2 concentration during the measurement period.

Table 3-1. Analytical condition for GC/MS and HPLC

Category	Content
GC/MS	SHIMADZU GCMS-QP2010 SE
ATD	ParkinElmer TurboMatrix (automated thermal desorber)
Column	Inert cap 5MS/Sil, 5% phenyl methyl silicone, 0.25 mmφ x 60 m x 0.25 μm
Oven temperature	40°C(5 min) - (10°C/min) - 300°C(9 min)
Carrier	Helium
Gas Flow	0.7 ml/min
Analysis mode	SCAN(33-400)
Desorption time [min]	10
Cold trap temperature [°C]	-50
Cold trap heating temperature [°C]	300
Transfer line temperature [°C]	300
HPLC	SHIMADZU Prominence (DGU-20A, LC-20AB, SIL-20AC, CBM-20A, SPC-M20A, CTO-20AC)
Column	Kinetex C18, 5.0 μm, 250 mm × 4.6 mm
Injection	20 μL
Column temperature	40°C
Flow	1.2 mL/min
Eluent	water / acetonitrile（4 : 6）
Detection wavelength	360 mm, 254 nm

3.2.3 Statistical analysis

In this study, SPSS statistical software (IBM SPSS Statistics, Version 27) and Microsoft Excel were used to evaluate the differences and conduct the correlation analysis of VOC concentrations within residential environments. The normality of the data was assessed using the Shapiro-Wilk test. Since the data were found to be non-normally distributed, the Mann-Whitney U test was subsequently used to determine statistically significant differences between the two groups.

3.3 Results

3.3.1 Questionnaire survey

Table 3-2 shows the results of the survey. A total of 33 houses were measured, with 16 (48.5%) using balanced ventilation and 17 (51.5%) using unbalanced ventilation.

Twelve (70.6%) of the houses with unbalanced ventilation had the air inlet opening located in the living room. Of these, 9 (52.9%) reported that the air inlet opening was always open, and 3 (17.7%) reported that it was closed. The remaining 5 (29.4%) homes were unsure. A total of 11 homes indicated that the air intake was located in the bedrooms, with 8 (64.7%) reporting that it was always open and 3 (17.7%) reporting that it was always closed. In the results of a survey on the use of ventilation systems by season, 15 households (93.8%) in Group A and 13 households (76.5%) in Group B responded that they use the ventilation system all the time for 24 hours in the summer. One household in Group A said that the ventilation system is used only when necessary. In Group B, 1 household (6.3%) responded as 'Other.' In winter, 16 households (100%) in Group A and 13 households (76.5%) in Group B responded that they use it 24 hours a day. Four households (23.5%) in Group B responded that they do not use it. In the spring and fall responses, 15 households (93.8%) in Group A and 13 households (76.5%) in Group B responded that they use it 24 hours a day. One household (6.3%) in Group A responded that they only use it when necessary, and four households (23.5%) in Group B responded that they do not use it.

In addition to the existing ventilation systems (balanced and unbalanced), we investigated whether other ventilation methods were used. The survey results for the winter season showed that Group A opened windows or doors in the living room in 6 houses (37.5%), used an air cleaner in 5 houses (31.3%), and did not use an air cleaner in 5 houses (31.3%). Group B had open windows or doors in 12 houses (57.1%), used an air cleaner in 8 houses (38.1%), and did not use one in 1 house (4.7%). In the bedroom, Group A opened a window in 5 houses (33.3%), used an air cleaner in 4 houses (26.7%), and did not use one in 6 houses (40.0%). Group B opened a window in 13 houses (65.0%), used an air cleaner in 5 houses (25.0%), and did not use one in 2 houses (10.0%). In the dressing room and washroom, Group A opened a window or door in 1 house (6.3%), used a bathroom exhaust fan in 7 houses (43.8%), and never used it in 8 houses (50.0%). Group B opened a window or door in 10 houses (43.5%), used a bathroom exhaust fan in 9 houses (39.1%), used an air cleaner in 1 house (4.4%), and never used it in 3 houses (13.0%). In the living room, 1 house (20.0%) used the air cleaner for less than 1-3 or 6-9 hours, and 3 houses (60.0%) used it for 21-24 hours. In the bedroom, 2 houses (25.0%) used it for 15-18 or 2-

24 hours, and 1 house (12.5%) used it for 1-3, 3-6, 6-9, or 18-21 hours. When asked about the reasons for ventilation, Group A had the highest number of responses at 9 houses (34.6%) for comfort improvement, followed by 5 houses (19.2%) for having a family member with hay fever, and 4 houses (15.4%) for infection control measures. In Group B, comfort improvement was the highest at 11 houses (34.4%), followed by room temperature control at 6 houses (18.8%), cooking and living odors and infection control measures at 4 houses (12.5%), and dustiness at 3 houses (9.4%).

Here are the survey results for the summer months. In the living rooms surveyed for ventilation other than the ventilation system, Group A opened windows or doors in 4 houses (26.7%), used an air cleaner in 4 houses (26.7%), and did not use an air cleaner in 6 houses (40.0%). In Group B, 8 houses (53.3%) opened a window or door, 5 houses (33.3%) used an air cleaner, and 2 houses (13.3%) did not use an air cleaner. In the bedroom, Group A had 5 houses (38.5%) with windows or doors open, 4 houses (30.8%) with air cleaners in use, and 4 houses (30.8%) without. Group B had 10 houses (71.4%) with windows or doors open, 2 houses (14.3%) with air cleaners in use, and 2 houses (14.3%) without. As for the duration of air cleaner use in the living room, Group A had 2 houses (50.0%) using it for 18-21 hours, 1 house (25.0%) for less than 1 hour, and 1 house (25.0%) for 6-9 hours. Group B had 4 houses (80.0%) using it for 21-24 hours and 1 house (20.0%) for 1-3 hours. In the bedroom, Group A had 3 houses (60.0%) using it for 21-24 hours, 1 house (20.0%) for less than 1 hour, and 1 house (20.0%) for 6-9 hours. Group B had 2 houses (100.0%) using it for 21-24 hours. In terms of reasons for ventilation, Group A had 9 houses (30.0%) for comfort improvement, 7 houses (23.3%) for cooking and living odors, and 4 houses (13.3%) for room temperature control and infection control measures. Group B had 8 houses (22.9%) for comfort improvement, 5 houses (14.3%) for room temperature control and cooking and living odors, and 3 houses (8.6%) for high humidity in the room and infection control measures.

Table 3-3 shows the survey results of heating devices used in winter. (Multiple responses allowed.) In the living room, 16 homes with balanced ventilation used central air conditioning systems, 2 homes using an electric heated table covered with a blanket, and no other heating devices. Conversely, unbalanced ventilation in living rooms included 4 homes using an Forced Flue (FF) type warm air heater, 2 homes with an electric stove/fan heater, 4 homes with an electric carpet, 5 homes using an electric heated table covered with a blanket, 11 homes with an air conditioner, and 1 home with another type of heating device. In the bedrooms, Group A was applied in 5 homes with a central air conditioning system and 11 homes with an air conditioner. For bedrooms in houses with unbalanced ventilation, there was 1 home each using a kerosene stove/fan heater, an electric stove/fan heater, and

an electric oil heater, while 12 homes were using an air conditioner. No other heating devices were reported in the bedrooms.

Table 3-4 shows the results of a survey on the wall and floor finishing materials of the living room and bedroom, dressing room and wash room in balanced ventilation and living room ventilation housing. Wallpaper (vinyl) and Wallpaper (paper) were the most investigated in each room of balanced ventilation and unbalanced ventilation housing. As for the floor finish material, wood flooring was the most common in each room of balanced ventilation and unbalanced ventilation housing, and most of the dressing and washrooms were Vinyl chloride sheets.

Table 3-2. Summary of the survey (Ventilation)

	Group A (%) Balanced	Group B (%) Unbalanced
Type of Ventilation	16(48.5)	17(51.5)
Air supply port (Unbalanced only)		
Living room		
Open	-	9(52.9)
Close	-	3(17.6)
No air supply port	-	-
Don't know	-	5(29.4)
Bedroom		
Open	-	8(64.7)
Close	-	3(17.7)
No air supply port	-	-
Don't know	-	6(35.2)
Other rooms		
Open	-	8(47.0)
Close	-	3(17.6)
No air supply port	-	-
Don't know	-	6(35.2)
Daily ventilation system usage time		
Summer		
Always (24 hour)	15(93.8)	13(76.5)
Morning	-	-
Day	-	-
Night	-	-
Only when necessary	1(6.3)	-
Not used	-	-
Other	-	4(23.5)
Winter		
Always (24 hour)	16(100.0)	13(76.5)
Morning	-	-
Day	-	-
Night	-	-
Only when necessary	-	-
Not used	-	4(23.5)
Other	-	-
Spring or Autumn		
Always (24 hour)	15(93.8)	13(76.5)

Morning	-		-	
Day	-		-	
Night	-		-	
Only when necessary	1(6.3)		-	
Not used	-		4(23.5)	
Other	-		-	
Use other than ventilation systems (multiple responses)	Winter (%)	*Summer (%)	Winter (%)	*Summer (%)
Living room				
Opening of a window or door	6(37.5)	4(26.7)	12(57.1)	8(53.3)
Air cleaner	5(31.3)	4(26.7)	8(38.1)	5(33.3)
Not used	5(31.3)	6(40.0)	1(4.7)	2(13.3)
Other	-	1(6.7)	-	-
Bedroom				
Opening of a window or door	5(33.3)	5(38.5)	13(65.0)	10(71.4)
Air cleaner	4(26.7)	4(30.8)	5(25.0)	2(14.3)
Not used	6(40.0)	3(23.1)	2(10.0)	2(14.3)
Other	-	1(7.7)	-	-
Dressing and washroom				
Opening of a window or door	1(6.3)	2(12.5)	10(43.5)	8(50.0)
Ventilation fan (bathroom exhaust fan)	7(43.8)	5(31.3)	9(39.1)	7(43.8)
Air cleaner	-	-	1(4.4)	-
Not used	8(50.0)	8(50.0)	3(13.0)	1(6.3)
Other	-	1(6.3)	-	-
Air cleaner usage time				
Living room				
Less than 1 hour	-	1(25.0)	-	-
Less than 1-3 hour	1(20.0)	-	-	1(20.0)
Less than 3-6 hours	-	-	1(12.5)	-
Less than 6-9 hour	1(20.0)	1(25.0)	1(12.5)	-
Less than 9-12 hour	-	-	1(12.5)	-
Less than 12-15 hour	-	-	-	-
Less than 15-18 hour	-	-	2(25.0)	-
Less than 18-21 hour	-	-	1(12.5)	-
21-24 hour	3(60.0)	2(50.0)	2(25.0)	4(80.0)
Bedroom				
Less than 1 hour	-	1(20.0)	-	-
Less than 1-3 hour	-	-	-	-
Less than 3-6 hours	-	-	-	-
Less than 6-9 hour	-	1(20.0)	3(60.0)	-
Less than 9-12 hour	-	-	1(20.0)	-
Less than 12-15 hour	-	-	-	-
Less than 15-18 hour	-	-	-	-
Less than 18-21 hour	-	-	-	-
21-24 hour	-	3(60.0)	1(20.0)	2(100.0)
Reasons for ventilation (multiple responses)				
Room temperature control	1(3.9)	4(13.3)	6(18.8)	5(14.3)
Comfort improvement	9(34.6)	9(30.0)	11(34.4)	8(22.9)
Smell of cigarette	-	-	-	1(2.9)
Building material odor	-	-	-	-
Cooking and living odors	1(3.9)	7(23.3)	4(12.5)	5(14.3)
Body odor	-	-	-	1(2.9)
Moldy smell	-	-	-	1(2.9)
Dusty	1(3.9)	1(3.3)	3(9.4)	2(5.7)
High humidity in the room	2(7.7)	-	-	3(8.6)
Sense of dryness in the room	1(3.9)	1(3.3)	-	1(2.9)

Have a family member with hay fever	5(19.2)	3(10.0)	2(6.3)	2(5.7)
Have a family member with allergies	2(7.7)	1(3.3)	2(6.3)	2(5.7)
Infection control measures	4(15.4)	4(13.3)	4(12.5)	3(8.6)
Other	-	-	-	1(2.9)

*Two houses in summer, Group A and Group B, are missing responses.

Table 3-3. Summary of the survey (heating)

	Balanced ventilation	Unbalanced ventilation
Heating device in Living room (multiple responses)		
Central air conditioning system	17	-
FF type warm air heater	-	4
Kerosene stove/fan heater	-	-
Gas stove/fan heater	-	-
Electric stove/fan Heater	-	2
Electric carpet	-	4
Electric panel heater	-	-
Electric heated table covered with a blanket	2	5
Electric hot air heater	-	-
Electric oil heater	-	-
Underfloor heating	-	-
Air conditioner	-	11
Other	-	1
No heating	-	-
Heating device in Bedroom (multiple responses)		
Central air conditioning system	5	-
FF type warm air heater	-	-
Kerosene stove/fan heater	-	1
Gas stove/fan heater	-	-
Electric stove/fan Heater	-	1
Electric carpet	-	-
Electric panel heater	-	-
Electric heated table covered with a blanket	-	-
Electric hot air heater	-	-
Electric oil heater	-	1
Underfloor heating	-	-
Air conditioner	11	12
Other	-	-
No heating	-	-

Table 3-4. Summary of the survey (finishing material)

	Balanced ventilation			Unbalanced ventilation		
	Living room	Bedroom	Dressing and washroom	Living room	Bedroom	Dressing and washroom
Wall finishing material						

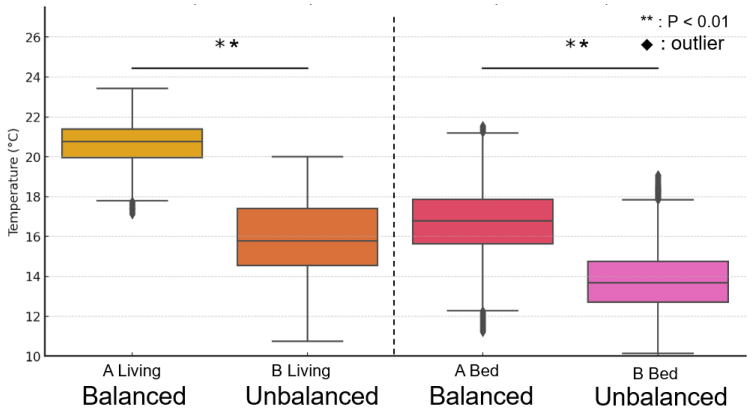
Woody coating material (boarding)	-	-	-	-	-	-
Tile	3	-	2	-	-	-
Wallpaper (vinyl)	8	8	9	7	7	8
Wallpaper (paper)	7	7	6	10	10	9
Painted walls (plaster, diatomaceous, etc.)	-	-	-	-	-	-
Other	-	-	-	-	-	-
Floor finishing material						
Wood flooring	16	16	1	16	14	3
Carpet	3	-	-	2	2	-
Vinyl chloride sheet (cushion floor)	-	-	11	-	-	11
P-tile	-	-	2	-	-	1
Cork	-	-	-	-	-	1
Other	-	-	2	1	-	1

3.3.2 Temperature and relative humidity

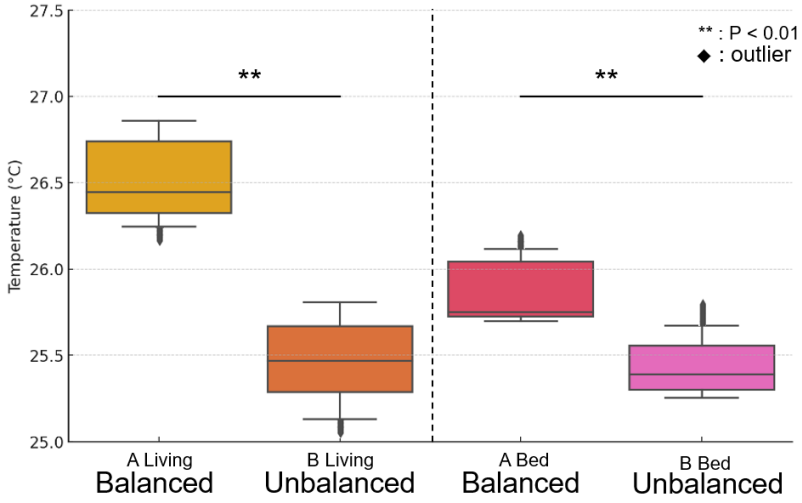
Figure 3-1 (a) presents a box plot comparison of average temperatures in living rooms and bedrooms between two groups during the winter. Temperature and relative humidity data were rounded to the second decimal place (average value of box plots). In living rooms, Group A recorded a significantly higher average temperature of 21.0°C compared to 15.9°C in Group B, with a statistically significant difference (p-value < 0.01). Similarly, in bedrooms, the average temperature for Group A was significantly higher at 17.7°C compared to 14.1°C for Group B, also showing statistical significance (p-value < 0.01). These results suggest that the differences in indoor temperature levels between the groups may be due to the air conditioning systems.

Figure 3-1 (b) presents a detailed comparison of average temperatures between two groups in living rooms and bedrooms during the summer season. In living rooms, Group A exhibited a higher mean summer temperature of 26.5°C with a relatively low standard deviation of 1.1°C, suggesting consistent temperature control within the

group. Conversely, Group B's living room mean summer temperature was lower at 25.5°C, with a higher standard deviation of 1.9°C, indicating greater variability in temperature regulation. Similarly, in bedrooms, Group A maintained an average summer temperature of 25.8°C with a low standard deviation of 1.0°C, compared to Group B's average of 25.4°C with a standard deviation of 1.2°C. These results indicate that Group A consistently achieves higher indoor temperatures with less variability compared to Group B during the summer. Statistical analysis confirmed the significance of these mean differences, with p-values < 0.01 as denoted by asterisks in the box plot. As with the winter results, these results suggest that the differences in indoor temperature levels between the groups during the summer may also be due to the air conditioning systems.



(a) Winter



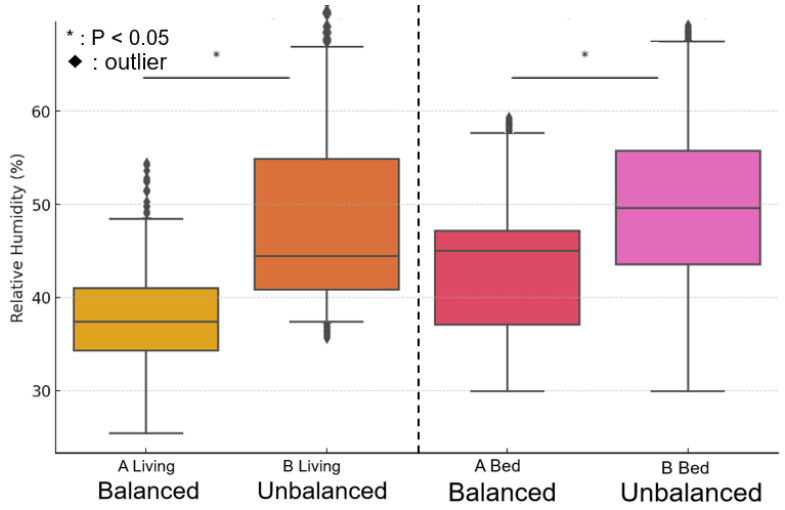
(b) Summer

Figure 3-1. Comparison of temperature between group A and Group B in living rooms and bedrooms during winter and summer

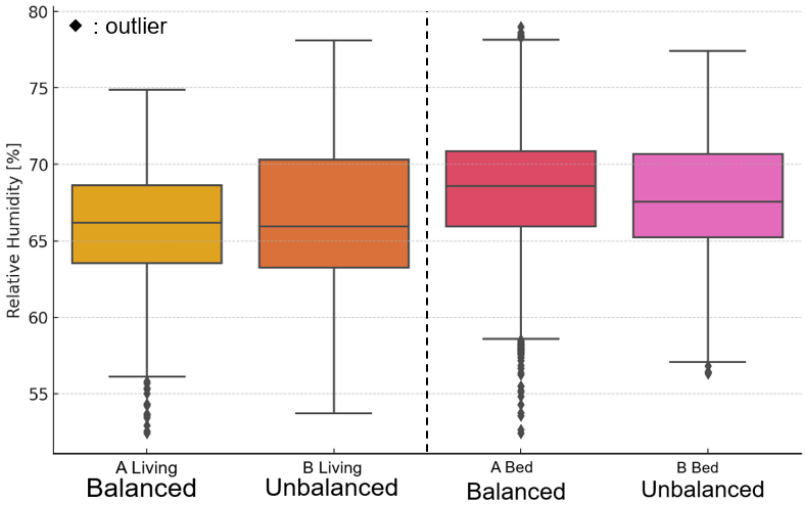
Figure 3-2 (a) presents a comparative analysis of relative humidity levels between Group A and Group B in living rooms and bedrooms during the winter season. The box plot highlights the differences in humidity control between

the groups. In living rooms, Group A exhibited a mean relative humidity of 37.4% with a standard deviation of 6.7%, indicating a relatively narrow range of humidity variation. In contrast, Group B's living rooms had a higher mean relative humidity of 47.0% with a larger standard deviation of 8.6%, suggesting greater variability in humidity levels. This difference is statistically significant with a p-value < 0.01. For bedrooms, Group A maintained a mean relative humidity of 42.9% with a standard deviation of 8.9%, whereas Group B had a mean relative humidity of 53.9% and a standard deviation of 8.7%. The difference in the mean bedroom humidity levels between the groups is also statistically significant with a p-value < 0.01. For bedrooms, Group A maintained a mean relative humidity of 42.9% with a standard deviation of 8.9%, whereas Group B had a mean relative humidity of 53.9% and a standard deviation of 8.7%. The difference in bedroom humidity levels between the groups is also statistically significant with a p-value < 0.01. The statistical analysis indicates a disparity in humidity control between the two groups. Group A, which employs a balanced ventilation system, consistently maintained lower and more stable humidity levels compared to Group B, which uses an unbalanced ventilation system.

Figure 3-2 (b) presents a comparative analysis of relative humidity levels between Group A and Group B in living rooms and bedrooms during the summer season. In living rooms, Group A exhibited a mean relative humidity of 66.0% with a standard deviation of 3.5%, indicating a relatively narrow range of humidity variation. In contrast, Group B's living rooms had a slightly higher mean relative humidity of 66.7% with a larger standard deviation of 4.8%, suggesting greater variability in humidity levels. There was no statistically significant difference between the two groups. For bedrooms, Group A maintained a mean relative humidity of 68.4% with a standard deviation of 3.4%, whereas Group B had a mean relative humidity of 67.9% with a standard deviation of 3.8%. Again, there was no statistically significant difference between the two groups.



(a) Winter



(b) Summer

Figure 3-2. Comparison of relative humidity between group A and group B in living rooms and bedrooms during winter and summer

3.3.3 CO_2 and air change rate

Figure 3-3 represents CO_2 data measured during winter. The unit for CO_2 is ppm, and the values were rounded to the one decimal place (average value of box plots). The analysis of CO_2 concentrations in the living rooms and bedrooms for groups A and B reveals notable differences. From the next sentence onward, a comparison of the average value of all rooms between the groups is described. Group A's living rooms have CO_2 concentrations of 542 ppm with a standard deviation of 79 ppm, while Group B's living rooms exhibit a significantly higher mean of 924.9 ppm and a much larger standard deviation of 391 ppm. This indicates that CO_2 concentrations in Group

B's living rooms are both higher and more variable. Similarly, in the bedrooms, Group A has CO_2 concentrations of 662 ppm with a standard deviation of 210 ppm, whereas Group B shows a higher mean of 1159 ppm with a standard deviation of 652 ppm. This pattern suggests that Group B experiences higher and more fluctuating CO_2 concentrations in both living rooms and bedrooms compared to Group A. Notably, in winter, there were a total of 4 houses in Group B that used oil stoves and fan heaters in their living rooms. The average CO_2 concentration in these 4 houses was 1524 ppm, with maximum values ranging from 3042 to 9968 ppm. Additionally, there were 13 houses that did not use oil stoves and fan heaters. The average CO_2 concentration in these 13 houses was 740 ppm, with a maximum value of 2636 ppm. During winter, in the living rooms, houses using oil stoves and fan heaters had an average CO_2 concentration approximately twice as high as those that did not use these appliances.

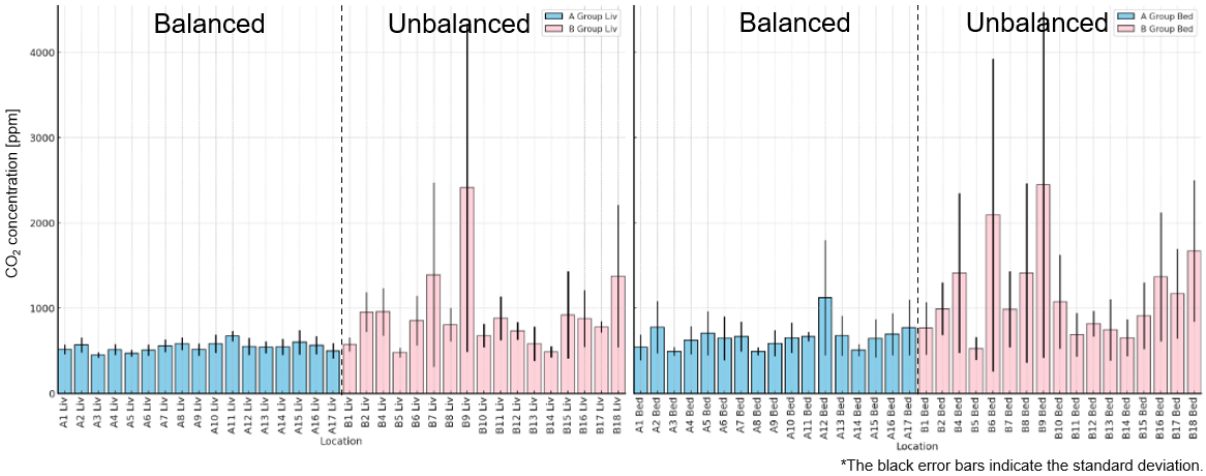


Figure 3-3. Comparison of winter CO_2 concentrations in groups A and B

Figure 3-4 shows CO_2 data measured during summer. The analysis of CO_2 concentrations in the living rooms and bedrooms for groups A and B during this period reveals notable differences. Group A's living rooms have CO_2 concentrations of 552 ppm with a standard deviation of 65, while Group B's living rooms have a mean of 575 ppm with a standard deviation of 89 ppm. This indicates that CO_2 concentrations in Group B's living rooms are slightly higher and more variable compared to those in Group A. Similarly, in the bedrooms, Group A has CO_2 concentrations of 645 ppm with a standard deviation of 162 ppm, whereas Group B shows a higher mean of 801 ppm with a standard deviation of 256 ppm. This pattern suggests that Group B experiences higher and more fluctuating CO_2 concentrations in both living rooms and bedrooms compared to Group A during the summer. These differences may be attributed to seasonal behaviors, such as the likelihood of opening windows during summer and heating-related combustion activities in winter, impacting the environmental conditions between the

two groups.

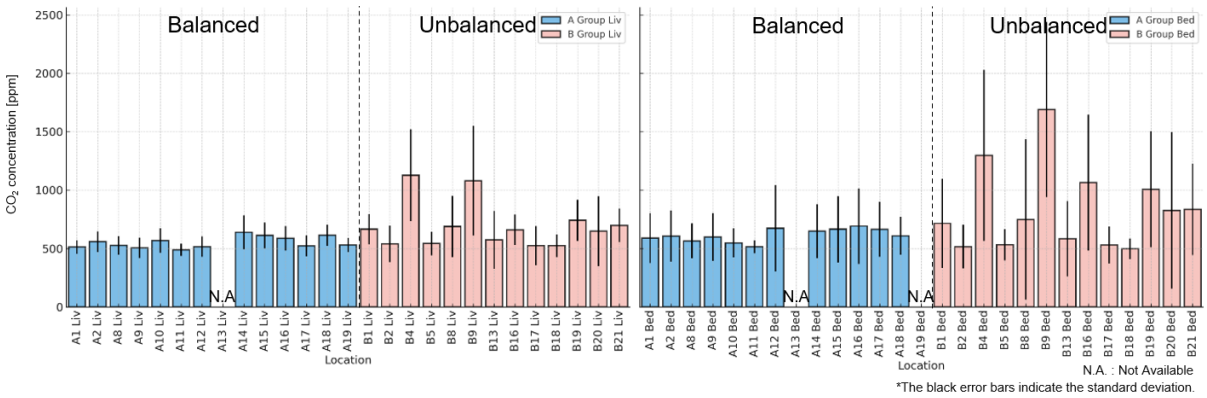


Figure 3-4. Comparison of summer CO_2 concentrations in groups A and B

Figure 3-5 shows the air change rates in the living rooms and bedrooms for Groups A and B during winter and summer, with standard deviations indicated by error bars. The calculation of the air change rate was performed using the concentration decay method during periods when it was confirmed that the occupants were absent. Within these periods, calculations were made starting from the times when CO_2 concentrations were initially high. The ambient air concentration was assumed to be 400 ppm, and the mean value of the three sections was used. In winter, the average air change rate for Group A was 0.96 h^{-1} (standard deviation: 0.4 h^{-1}) in the living room and 0.72 h^{-1} (standard deviation: 0.33 h^{-1}) in the bedroom. For Group B, the average winter air change rate was 0.67 h^{-1} (standard deviation: 0.23 h^{-1}) in the living room and 0.37 h^{-1} (standard deviation: 0.25 h^{-1}) in the bedroom. During summer, the average air change rate for Group A was 0.88 h^{-1} (standard deviation: 0.18 h^{-1}) in the living room and 0.82 h^{-1} (standard deviation: 0.36 h^{-1}) in the bedroom. For Group B, the average summer air change rate was 0.33 h^{-1} (standard deviation: 0.26 h^{-1}) in the living room and 0.27 h^{-1} (standard deviation: 0.09 h^{-1}) in the bedroom. The average ventilation rate for Group A exceeded the required air change rate of 0.5 h^{-1} in both the living room and bedroom during both seasons. In contrast, the average ventilation rate for Group B's bedrooms was below 0.5 h^{-1} in both winter and summer. Additionally, Group B exhibited more variation in ventilation rates between winter and summer compared to Group A.

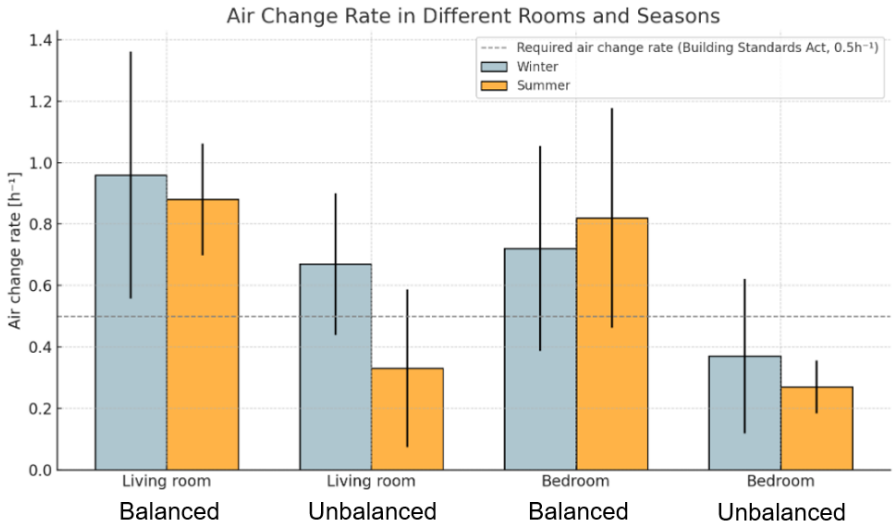
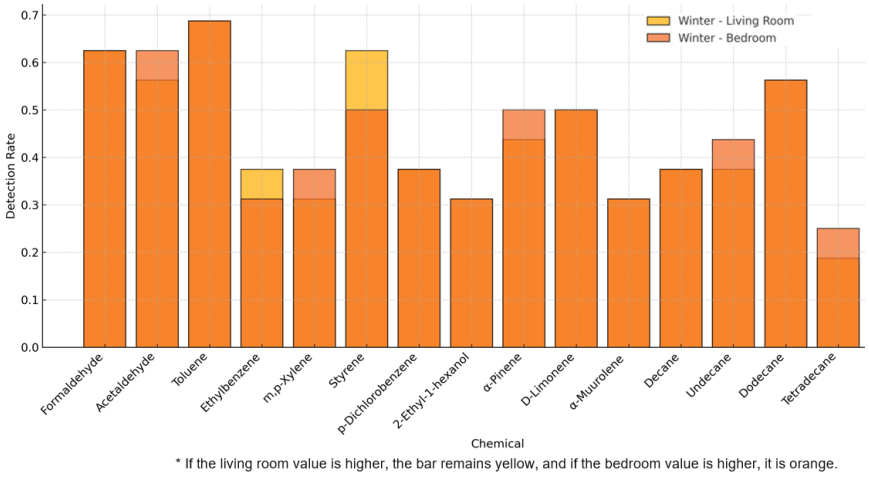


Figure 3-5. Air change rate in different rooms and seasons.

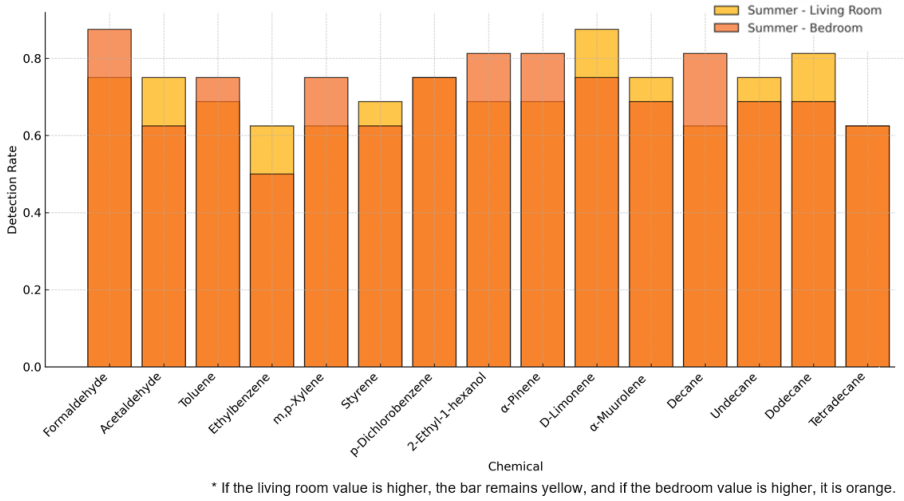
3.3.4 VOCs concentration

Figure 3-6 (a) shows the analysis of VOC detection rates across different seasons and rooms, revealing notable patterns. In the winter, the detection rates of chemicals such as formaldehyde and acetaldehyde are relatively high in both living rooms and bedrooms, indicating a consistent presence across these indoor environments. Specifically, formaldehyde shows a detection rate of approximately 62.5% in both spaces, while acetaldehyde is detected at 56.3% in living rooms and 62.5% in bedrooms.

During the summer, the detection rates of VOCs generally increase, as shown in Figure 3-6 (b). Formaldehyde reaches a detection rate of up to 75% in living rooms and 87.5% in bedrooms. This seasonal rise is also observed in other compounds like toluene and ethylbenzene, which exhibit higher detection rates in the summer compared to winter.



(a) Winter



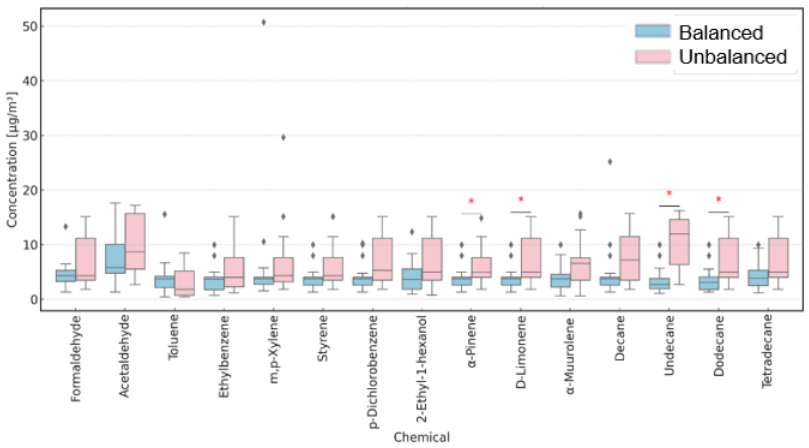
(b) Summer

Figure 3-6. Seasonal detection rates of VOCs in living rooms and bedrooms

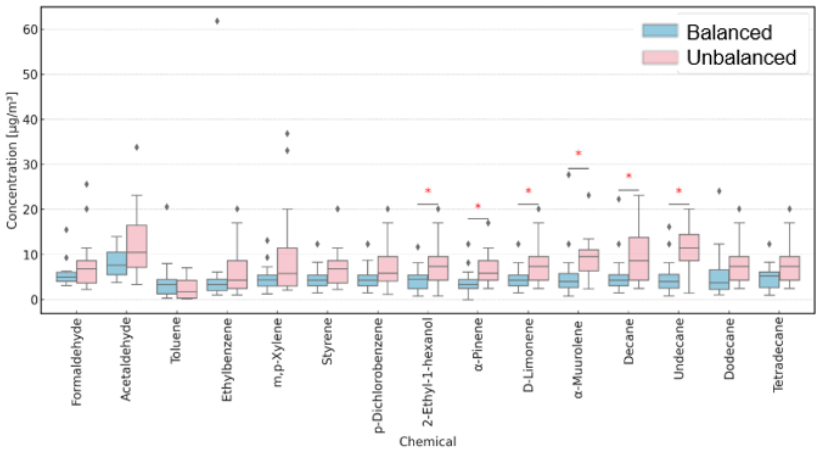
Figure 3-7 (a) shows the comparison of VOC concentrations in the living room between Group A and Group B, indicating that Group B generally has higher concentrations of most chemicals. From the following sentence, the concentrations listed for each substance represent the minimum to maximum concentrations. For formaldehyde, Group B ranges from 7.7 μg/m³ to 11.5 μg/m³, which is higher than Group A's range of 3.2 μg/m³ to 5.0 μg/m³. Similarly, for acetaldehyde, Group B ranges from 11.5 μg/m³ to 23.2 μg/m³, while Group A ranges from 4.3 μg/m³ to 10.5 μg/m³. For toluene, Group A ranges from 0.8 μg/m³ to 15.6 μg/m³, higher than Group B's 0.3 μg/m³ to 3.3 μg/m³. For α-pinene, Group B ranges from 2.0 μg/m³ to 4.0 μg/m³, higher than Group A's 1.0 μg/m³ to 3.0 μg/m³, with a statistically significant difference. Similarly, d-limonene in Group B ranges from 7.0 μg/m³ to 15.0 μg/m³, higher than Group A's 3.0 μg/m³ to 6.0 μg/m³, also showing a statistically significant difference. For decane and

undecane, Group B ranges from 2.0 μg/m³ to 6.0 μg/m³ and 5.0 μg/m³ to 10.0 μg/m³ respectively, higher than Group A's concentrations. Dodecane in Group B ranges from 4.0 μg/m³ to 10.0 μg/m³, while Group A ranges from 2.0 μg/m³ to 5.0 μg/m³, showing a statistically significant difference. Finally, tetradecane in Group B ranges from 2.0 μg/m³ to 4.0 μg/m³, higher than Group A's 1.0 μg/m³ to 3.0 μg/m³. In summary, Group B has higher concentrations for most chemicals, with statistically significant differences observed for α-pinene, d-limonene, undecane, and dodecane.

Figure 3-7 (b) shows the comparison of VOC concentrations in the bedroom areas between Group A and Group B, indicating that Group B generally has higher concentrations of most chemicals. For formaldehyde, Group B ranges from 11.5 μg/m³ to 20.2 μg/m³, higher than Group A's range of 3.8 μg/m³ to 6.0 μg/m³. For acetaldehyde, Group B ranges from 16.5 μg/m³ to 23.2 μg/m³, while Group A ranges from 3.8 μg/m³ to 10.5 μg/m³. For toluene, Group A ranges from 0.8 μg/m³ to 20.6 μg/m³, higher than Group B's 0.2 μg/m³ to 2.8 μg/m³. However, data for ethylbenzene, m,p-xylene, styrene, p-dichlorobenzene, and 2-ethyl-1-hexanol are incomplete. For α-pinene, Group B ranges from 3.0 μg/m³ to 5.0 μg/m³, higher than Group A's 1.0 μg/m³ to 3.0 μg/m³, with a statistically significant difference. Similarly, d-limonene in Group B ranges from 8.0 μg/m³ to 18.0 μg/m³, higher than Group A's 3.0 μg/m³ to 7.0 μg/m³, also showing a statistically significant difference. For decane and undecane, Group B ranges from 3.0 μg/m³ to 8.0 μg/m³ and 6.0 μg/m³ to 12.0 μg/m³ respectively, higher than Group A's concentrations. Dodecane in Group B ranges from 5.0 μg/m³ to 11.0 μg/m³, while Group A ranges from 2.0 μg/m³ to 6.0 μg/m³, showing a statistically significant difference. Finally, tetradecane in Group B ranges from 3.0 μg/m³ to 5.0 μg/m³, higher than Group A's 1.0 μg/m³ to 3.0 μg/m³. In summary, Group B has higher concentrations for most chemicals, with statistically significant differences observed for 2-ethyl-1-hexanol, α-pinene, d-limonene, α-muurolene, decane and undecane.



(a) Living room



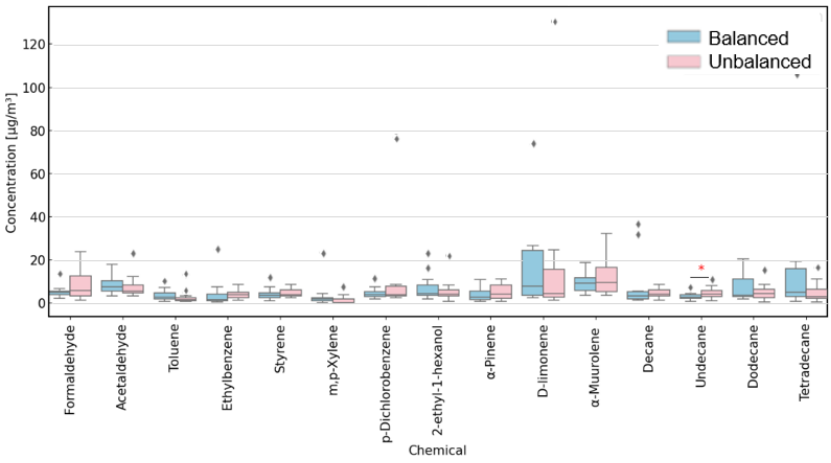
(b) Bedroom

Figure 7. Box plot analysis of VOC concentrations for groups A and B in winter

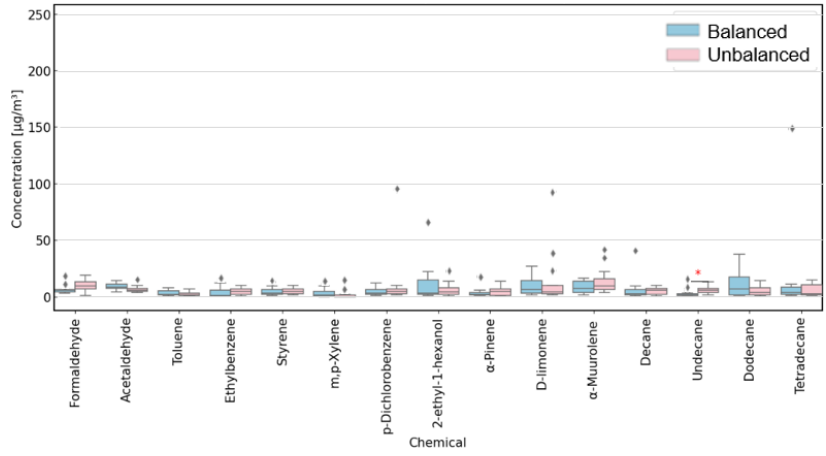
Figure 3-8 shows the comparison of VOC concentrations between Group A and Group B during summer in the living room. Formaldehyde concentrations were slightly higher in Group B (8.9 μg/m³) compared to Group A (5.8 μg/m³). Acetaldehyde concentrations were also higher in Group A (8.5 μg/m³) compared to Group B (7.1 μg/m³). toluene concentrations were 3.4 μg/m³ for Group A and 2.5 μg/m³ for Group B. Ethylbenzene was higher in Group A (3.2 μg/m³) compared to Group B (1.6 μg/m³). Styrene data was not available for Group B but was present in Group A (5.6 μg/m³). The concentration of m,p-xylene was higher in Group A (3.2 μg/m³) than in Group B (1.5 μg/m³. For p-dichlorobenzene, data was not detected in the houses of Group A, but Group B had a significantly higher concentration (85.7 μg/m³). 2-ethyl-1-hexanol concentrations were 14.9 μg/m³ in Group A and 6.7 μg/m³ in Group B. α-pinene concentrations were comparable between Group A (3.6 μg/m³) and Group B (4.8 μg/m³). D-limonene was higher in Group B (26.8 μg/m³) compared to Group A (17.7 μg/m³). α-muurolene concentrations were higher in Group B (13.4 μg/m³) compared to Group A (8.6 μg/m³). Decane concentrations were 10.7 μg/m³ for Group A and 2.7 μg/m³ for Group B. Undecane was significantly higher in Group B (5.4 μg/m³) compared to Group A (2.4 μg/m³). Dodecane concentrations were 13.1 μg/m³ in Group A and 6.5 μg/m³ in Group B. Lastly, tetradecane concentrations were 33.6 μg/m³ for Group A and 5.5 μg/m³ for Group B.

In the bedroom, the VOC concentration comparison shows similar trends. Formaldehyde concentrations were higher in Group B (8.9 μg/m³) compared to Group A (5.8 μg/m³). Acetaldehyde concentrations were 8.5 μg/m³ for Group A and 7.1 μg/m³ for Group B. Toluene levels were 3.4 μg/m³ in Group A and 2.5 μg/m³ in Group B. Ethylbenzene concentrations were higher in Group A (3.2 μg/m³) compared to Group B (1.6 μg/m³). Styrene was

not detected in Group B but was present in Group A (5.6 µg/m³). The concentration of m,p-xylene was higher in Group A (3.2 µg/m³) compared to Group B (1.5 µg/m³). Group B had a significantly higher concentration of p-dichlorobenzene (85.7 µg/m³) compared to Group A, where p-dichlorobenzene was not detected. 2-ethyl-1-hexanol concentrations were 14.9 µg/m³ in Group A and 6.7 µg/m³ in Group B. α-pinene concentrations were 3.6 µg/m³ in Group A and 4.8 µg/m³ in Group B. D-limonene was higher in Group B (26.8 µg/m³) compared to Group A (17.7 µg/m³). α-muurolene concentrations were higher in Group B (13.4 µg/m³) compared to Group A (8.6 µg/m³). Decane concentrations were 10.7 µg/m³ for Group A and 2.7 µg/m³ for Group B. Undecane was significantly higher in Group B (5.4 µg/m³) compared to Group A (2.4 µg/m³). Dodecane concentrations were 13.1 µg/m³ in Group A and 6.5 µg/m³ in Group B. Finally, tetradecane levels were 33.6 µg/m³ for Group A and 5.5 µg/m³ for Group B. In the living room and bedroom, undecane was statistically significantly higher in Group B.



(a) Living room



(b) Bedroom

Figure 3-8. Box plot analysis of summer VOC concentrations in bedroom for groups A and B in summer

Figure 3-9 shows the correlation between air change rate (ACH) and VOCs in the living rooms and bedrooms of groups A and B during winter. A correlation near 1 indicates that as ACH increases, the VOC concentration also increases, while a correlation near -1 signifies a decrease in VOC concentration with increasing ACH. For group A, the results in the living room and bedroom show a tendency towards a negative correlation for formaldehyde and acetaldehyde, approximately -0.2. m,p-xylene, 2-ethyl-1-hexanol, α-muurolene, undecane, and dodecane also exhibited a trend of negative correlation, but very weakly around -0.1. Toluene showed a positive correlation trend around 0.4. In the bedroom, decane and dodecane displayed negative correlations of about -0.3 and -0.4, respectively, while p-dichlorobenzene and undecane showed the highest correlation, over 0.4. The living room of group B exhibited a positive correlation ranging from about 0.3 to 0.7 for formaldehyde, acetaldehyde, toluene, ethylbenzene, m,p-xylene, styrene, decane, and undecane. Additionally, p-dichlorobenzene, 2-ethyl-1-hexanol, and terpenes (α-pinene, d-limonene, α-muurolene) showed a negative correlation trend ranging from about -0.2 to -0.4. Most VOCs in the bedroom had very low correlation coefficients below 0.2, indicating weak correlations.

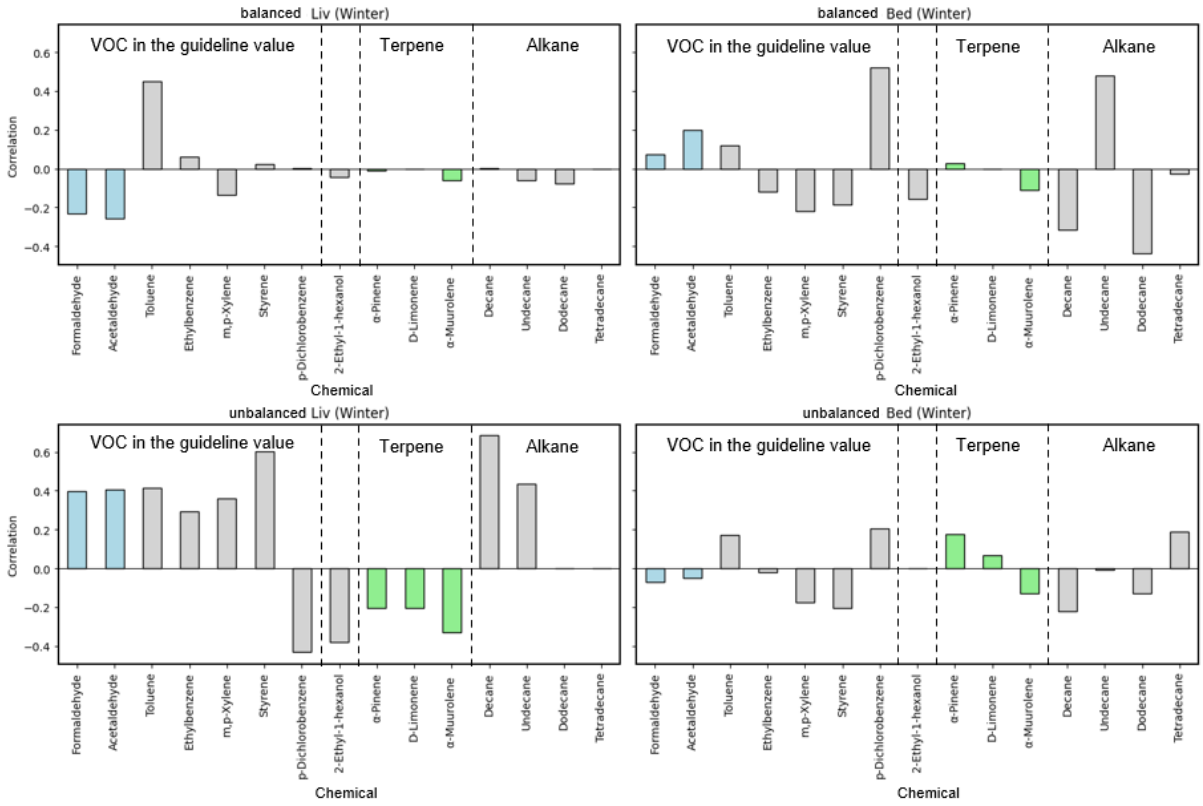


Figure 3-9. Correlations of VOC concentrations with air change rates for groups A and B in winter

Figure 3-10 show the correlation between ACH and VOCs in the living rooms and bedrooms of groups A and B during summer. In the living room of group A, formaldehyde, acetaldehyde, and 2-ethyl-1-hexanol exhibited relatively high negative correlations above -0.5. Terpenes showed a positive correlation trend ranging from about 0.2 to 0.8. In the bedroom, formaldehyde and terpenes displayed negative correlations around -0.2 to -0.3. The living room of group B showed negative correlations for d-limonene, α-muurolene, undecane, dodecane, and tetradecane. In the bedroom, formaldehyde, α-muurolene, and undecane exhibited negative correlations, while acetaldehyde and toluene, along with all other VOCs, showed positive correlations. However, these correlations were weak, and no clear and consistent patterns emerged regarding VOCs and ACH to season, location, and ventilation systems.

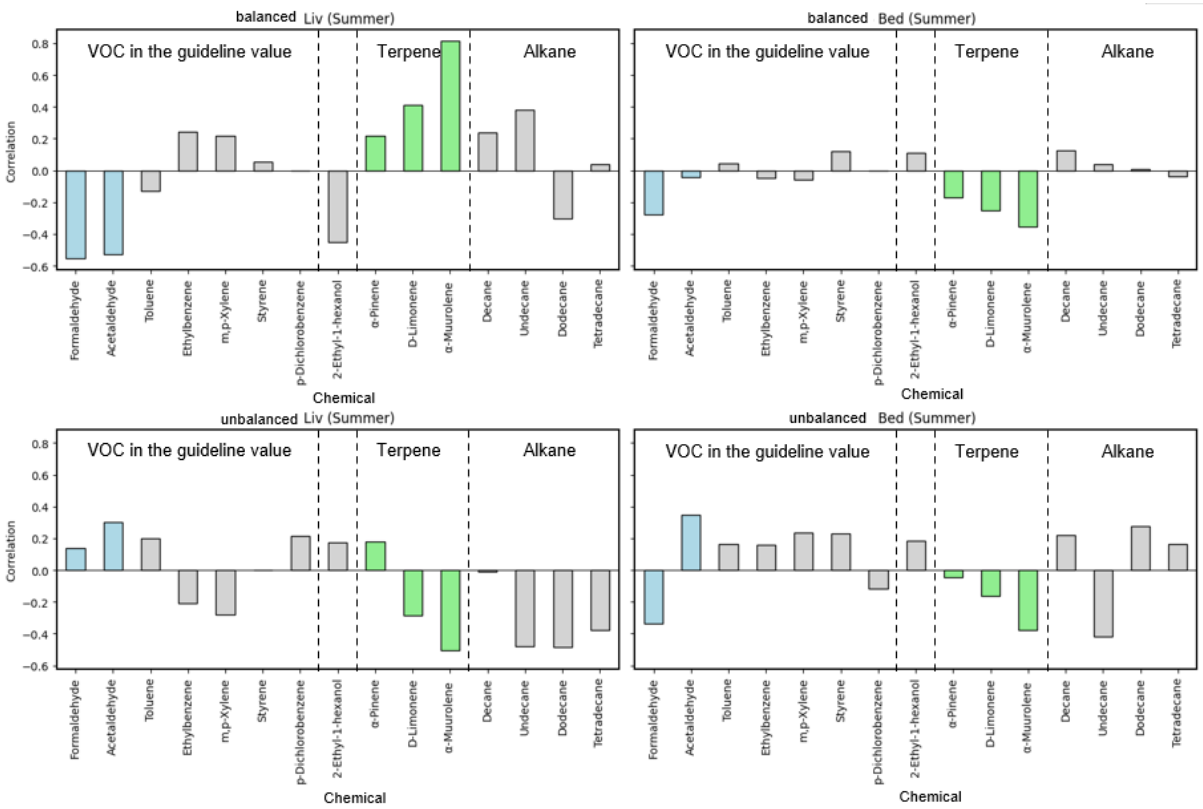
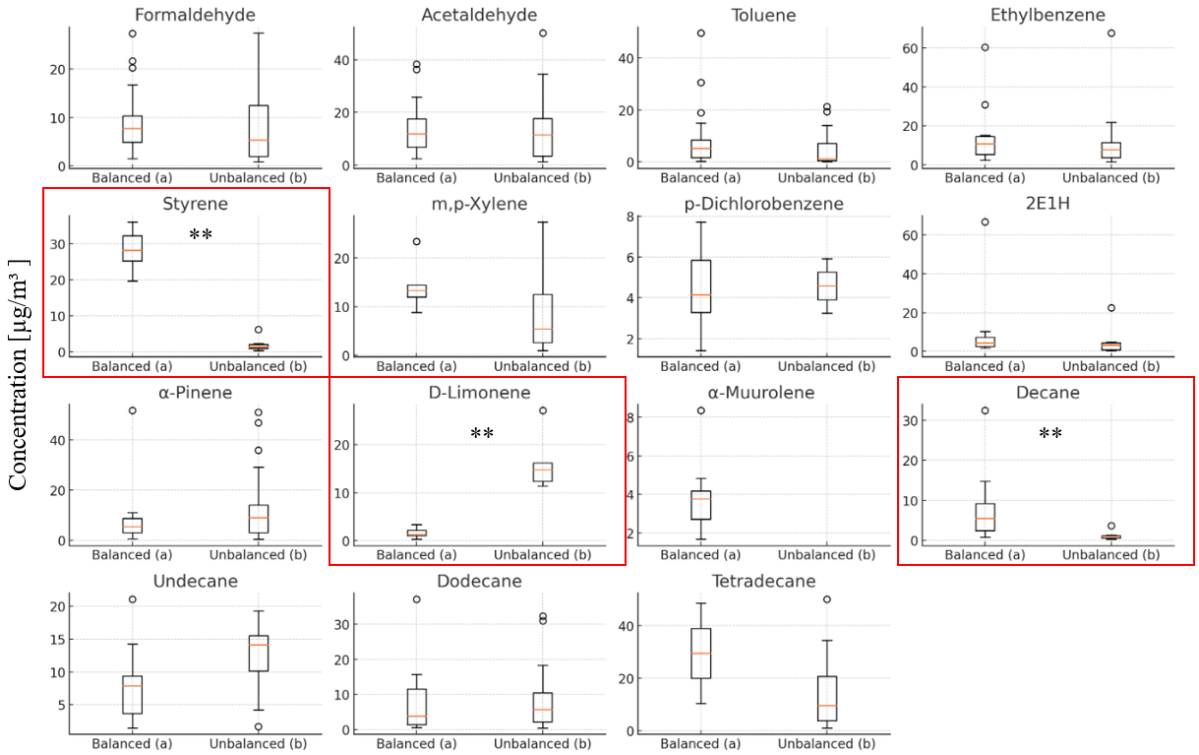


Figure 3-10. Correlations of VOC concentrations with air change rates for groups A and B in summer

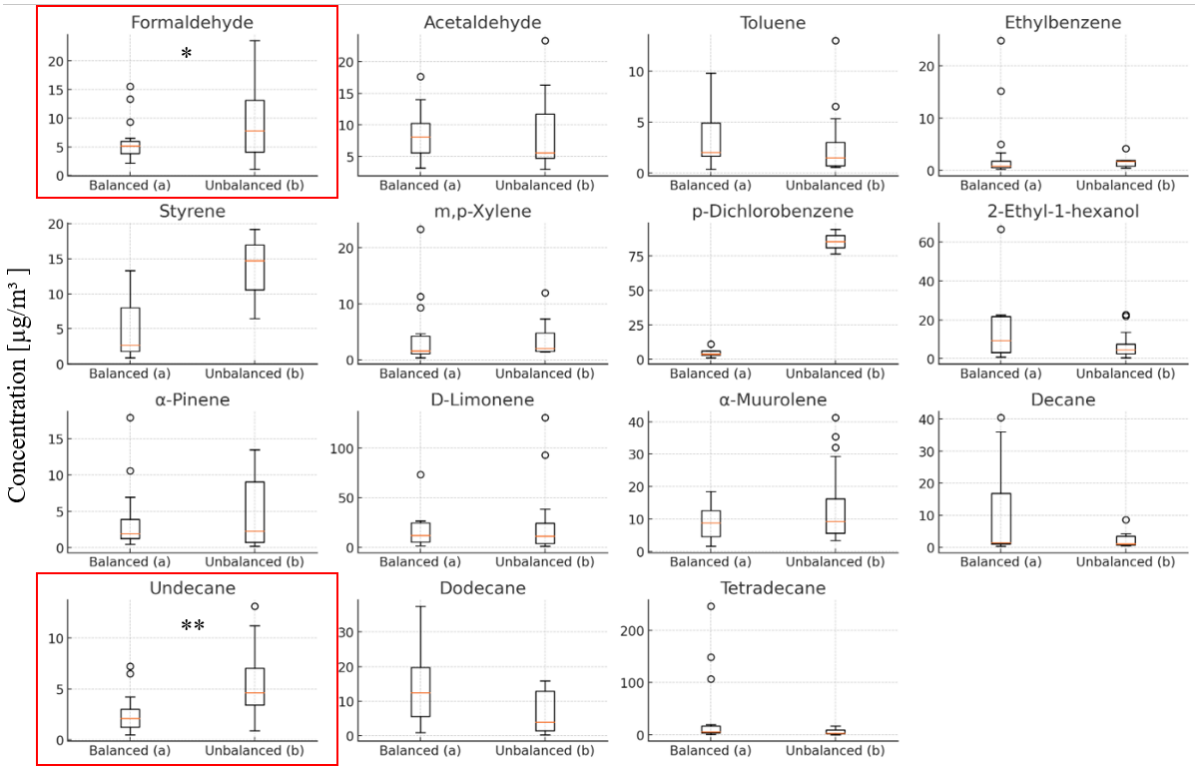
Since the influence of not only negative pressure but also the air change rate may be significant, the concentration of each VOC was converted to the concentration of each VOC under the condition of a ventilation frequency of 0.5 times/h in each house in order to examine differences in ventilation systems by considering the effect of ventilation frequency. The conversion was calculated as follows.

$$VOC_{0.5ACH} = VOC_{measured} \times \frac{0.5ACH}{N_{ACH}}$$

Where, $VOC_{0.5ACH}$ is VOC concentration when converted to 0.5 ACH, $VOC_{measured}$ is actually the VOC concentration measured in the house, N_{ACH} is the ACH value calculated by tracer decision math in each house. Figure 3-11 shows the comparison between balanced ventilation and unbalanced ventilation by converting the measured VOC concentrations to 0.5 ACH. The results are as follows: The average concentrations of formaldehyde in winter for balanced ventilation and unbalanced ventilation were both 9.2 μg/m³, and acetaldehyde concentrations were 13.4 μg/m³ and 12.7 μg/m³, respectively, with no significant difference. On the other hand, p-dichlorobenzene, α-pinene, d-limonene, α-muurolene, decane, undecane, dodecane, and tetradecane tended to be high even when the ventilation frequency for unbalanced ventilation was adjusted to 0.5 h^{-1}. Substances outlined in red indicate statistically significant differences. During the summer, concentrations of 10 substances, except acetaldehyde, toluene, ethylbenzene, 2-ethyl-1-hexanol, and dodecane, tended to be slightly higher for unbalanced ventilation. From the above results, it is thought that the concentration of most of the terpenes and alkanes, which are VOCs generated within the wall structure, tends to be higher in unbalanced ventilation than in balanced ventilation due to the influence of negative pressure.



(a) Winter



(b) Summer

Figure 3-11. Compare VOC concentration by converting all air change rates to 0.5

3.4 Discussion

3.4.1 Influence of indoor temperature and humidity on VOC concentrations

First, this is related content about the indoor temperature and relative humidity measured in the houses. Differences in air conditioning systems between Group A and Group B could explain the significant variance in relative humidity observed during winter in living rooms and bedrooms. Factors such as the presence of humidifiers, heating methods, and ventilation frequency play crucial roles. Frequent ventilation can reduce indoor humidity. Lifestyle habits of the residents also contribute to these differences. Activities like cooking, showering, and plant care increase indoor humidity. The number of occupants and their activity levels also affect humidity, with more people or higher activity levels leading to increased moisture. These combined factors likely account for the observed humidity differences between the two groups.

3.4.2 Seasonal variation and air tightness

In the context of our investigation, the variability in VOC concentrations and air change rates, especially evident during the winter season, can be attributed to several interacting factors [3-13, 3-14]. Modern homes, known for their high airtightness, can easily become negatively pressurized [3-17]. This negative pressure can draw VOCs into living areas from enclosed spaces such as crawl spaces, walls, and floors [3-18, 3-19]. The inconsistency in results between the living rooms and bedrooms of Group B further underscores the complex interplay of factors influencing indoor air quality. This variability could stem from several sources, including unbalanced ventilation which likely contributes to inefficient pollutant removal. The airtightness performance C value of the houses measured in this study has not been investigated, but a certain level is guaranteed because the house manufacturer is the same. However, it is important to note that the number of years a house has been built differs between houses with balanced ventilation and those with unbalanced ventilation. However, since the c-value of the house was not measured, there are limitations.

3.4.3 Comparison of ventilation systems

Our study builds upon previous research that explored the characteristics of VOCs based on variables such as seasonal changes, types of residences, duration of occupancy, and ventilation systems [3-9]. Among these factors, ventilation systems have been shown to have the most significant impact on indoor VOC concentrations, prompting a more detailed investigation into homes with distinctly different ventilation strategies: balanced versus

unbalanced ventilation. The results of our extended analysis confirm that residences employing unbalanced ventilation systems exhibit lower air change rates and consequently higher VOC concentrations compared to those with balanced systems [3-10]. This finding aligns with the hypothesis that inadequate ventilation contributes to poorer indoor air quality by allowing VOCs to accumulate rather than being effectively diluted and removed [3-11, 3-12].

In this study, the measured VOC concentrations did not exceed the guideline values set by the Ministry of Health, Labour and Welfare. It is important to note that all houses, regardless of the type of ventilation used, operated their ventilation systems continuously (24 hours a day), which likely contributed to keeping VOC levels within safe limits. Although not all houses with unbalanced ventilation fell below the standard, those employing unbalanced ventilation in the bedrooms during winter and in both the living rooms and bedrooms during summer, on average, exhibited ventilation frequencies below the Building Standard Law-prescribed rate of 0.5 h-1, showing seasonal variation between winter and summer. Comparing VOC concentrations between winter and summer, some VOCs including formaldehyde were found to have higher levels in the summer, when emission rates are generally higher. However, this trend was not observed for other substances. The year of construction of the houses also appeared to be a factor in the lower emission rates observed. Despite the continuous operation of ventilation systems, the survey revealed a trend where houses with unbalanced ventilation consistently showed higher VOC concentrations compared to those with balanced ventilation. This was particularly evident for α-pinene and d-limonene, which are emitted from wood and household products (cleaning products, air fresheners, and scented candles), [3-20, 3-21]. This indicates that while continuous ventilation helps maintain VOC concentrations within guidelines, the type of ventilation system can influence the overall indoor air quality.

These insights emphasize the need for carefully considered ventilation strategies [3-27]. Effective strategies should not only regulate air exchange rates but also guard against the entry of pollutants from hidden structural areas. Well-designed and maintained ventilation systems are crucial to reducing these risks and ensuring a healthy indoor environment, especially in modern, well-sealed buildings [3-28].

3.4.4 Infiltration of VOCs from the wall

Among the materials used for walls, even waterproof or airtight sheet materials may transmit VOCs, as shown in the research results below. Polyethylene (PE) is the main material for waterproof sheets and airtight tapes and has low permeability to VOCs. However, permeability may increase due to the presence of additives or plasticizers

[3-29]. Because of this, in real-world use situations, VOCs may pass through PE sheets if certain additives are included. Polyvinyl chloride (PVC) also sometimes emits VOCs if it contains plasticizers, so VOCs from other materials are not completely impermeable. According to Griffiths' research, it has been confirmed that VOCs penetrate PVC. A study of the diffusion behavior of VOCs within PVC using molecular dynamics simulations shows that VOCs such as n-hexane penetrate PVC materials. Additionally, VOCs can enter the room through various routes such as tiny cracks. According to Patterson and Davis (2009), gaps and cracks in buildings sometimes allow VOCs to enter indoors [3-30]. Even if these structural defects are small, they can become a pathway for VOCs to enter the indoor environment. The density of wooden house walls is approximately 500-700 kg/m^3, making them highly breathable and porous, which facilitates the entry of VOCs into the interior of a wooden house. Won et al. (2001) showed that VOCs invade through various construction materials such as moisture-proof layers and moisture-proof sheets [3-31]. Although these materials are designed to repel moisture, they cannot completely prevent the spread of VOCs due to their chemical properties or the presence of microscopic cracks. Additionally, the sealing material deteriorates over time, creating small cracks, which may ultimately allow VOCs to enter [3-32, 3-33, 3-34]. Previous studies have reported that it is difficult to maintain airtightness through tape sealing in complex wall joints and that improvements in not only sealing but also in mechanical bonding are necessary to reduce air leakage [3-35]. Another study analyzed the air permeability of wall insulation to evaluate the impact of leak location and pressure differences on the air flow rate and VOC intrusion into the room. As a result, it was reported that the location of the leak and the air change rate had a significant impact on VOC intrusion, and that even with tape sealing, there is the possibility of air, moisture, and VOCs intruding through small gaps [3-36]. To summarize the above-mentioned research results, VOCs generated from walls can easily penetrate into the room through small gaps depending on the pressure difference. In addition, even if waterproof and airtight tape is used at the joints, it is difficult to completely seal the interior, and since the performance deteriorates over time, it is challenging to completely prevent VOCs from entering the room.

3.5 Summary

Our comprehensive study revealed key insights into the impact of ventilation systems on indoor VOC concentrations across different seasons and strategies. Winter and summer analyses demonstrated that Group A, employing balanced ventilation, consistently maintained lower and more stable VOC levels compared to Group B, which uses unbalanced ventilation. This disparity was notably evident in both temperature and relative humidity control, with Group A achieving better regulation across seasons. In winter, Group B exhibited significantly higher and more variable CO_2 and VOC, suggesting poor air exchange and inefficient ventilation. This pattern persisted into summer, where Group B also displayed higher VOC levels, highlighting ongoing challenges in air quality management due to unbalanced ventilation. Statistical analysis confirmed that these differences were significant, emphasizing the effectiveness of balanced ventilation systems in maintaining healthier indoor environments. As a result of comparing the measured airtightness of the house to 0.5 h^{-1}, the unbalanced ventilation was higher than the balanced ventilation in the summer months, and there was a statistically significant difference. The c-value of the measured houses has not been reviewed, but the fact that they are the same housemaker can guarantee some airtightness. Also, VOCs generated from walls can easily penetrate into the room through small gaps depending on the pressure difference. In addition, even if waterproof and airtight tape is used at the joints, it is difficult to completely seal the interior, and since the performance deteriorates over time, it is challenging to completely prevent VOCs from entering the room. Additionally, our findings suggest that VOC concentrations depend not only on ventilation rates but also on the types of VOCs and their emission sources, which can vary between residences. These observations underscore the necessity for tailored ventilation strategies, especially in homes with unbalanced ventilation, that consider both the specific needs of the building and seasonal variations to ensure optimal indoor air quality. Future research will investigate the characteristics of VOC emissions from insulation materials based on existing studies showing that VOCs infiltrate indoors from enclosed spaces under negative pressure conditions in houses using unbalanced ventilation systems.

To sum up the important results, our comprehensive study revealed that balanced ventilation systems significantly reduce and stabilize indoor VOC concentrations across different seasons, compared to unbalanced systems. Group A, with balanced ventilation, consistently achieved lower and more stable VOC and CO_2 concentrations, as well as better temperature and humidity control, in both winter and summer. In contrast, Group B, using unbalanced ventilation, experienced higher and more variable VOC and CO_2 concentrations, indicating poor air exchange and inefficient ventilation. Statistical analysis confirmed the significance of these differences, demonstrating the

superior effectiveness of balanced ventilation systems in maintaining healthier indoor environments. These findings highlight the critical role of tailored ventilation strategies, especially for homes with unbalanced systems, to address seasonal variations and specific building needs for optimal indoor air quality.

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Chapter 4: Assessment of VOC emissions from Insulation materials

4.1 Introduction

In Chapter 3, the differences in VOC concentrations between balanced and unbalanced ventilation housing were examined. In general, the VOC concentration in unbalanced ventilation housing was higher than that in balanced ventilation housing. One of the many causes is that when unbalanced ventilation is used, the room is frequently in a state of negative pressure, making it highly likely that VOCs from confined spaces (walls, floors, roofs) flow into the room. In previous studies, the entire confined space was targeted, and the individual VOCs of building materials were not identified. Most of the research on the VOC emissions of insulation is based on the results of the early 2000s, about 20 years ago, and the types of insulation studied are also limited. Therefore, in Chapter 4, VOC emissions were evaluated for insulating materials, which are one of the layers of confined spaces such as walls and floors, and play a very important role in energy saving among modern building materials. The introduction describes a wide range of content on building materials, including insulating materials. Then, we introduce a previous study that explores how VOCs can be emitted into a room under negative pressure conditions and describe the results. Before dealing with the contents of the insulation, we need to understand the materials that make up the wall and the VOCs that these materials emit.

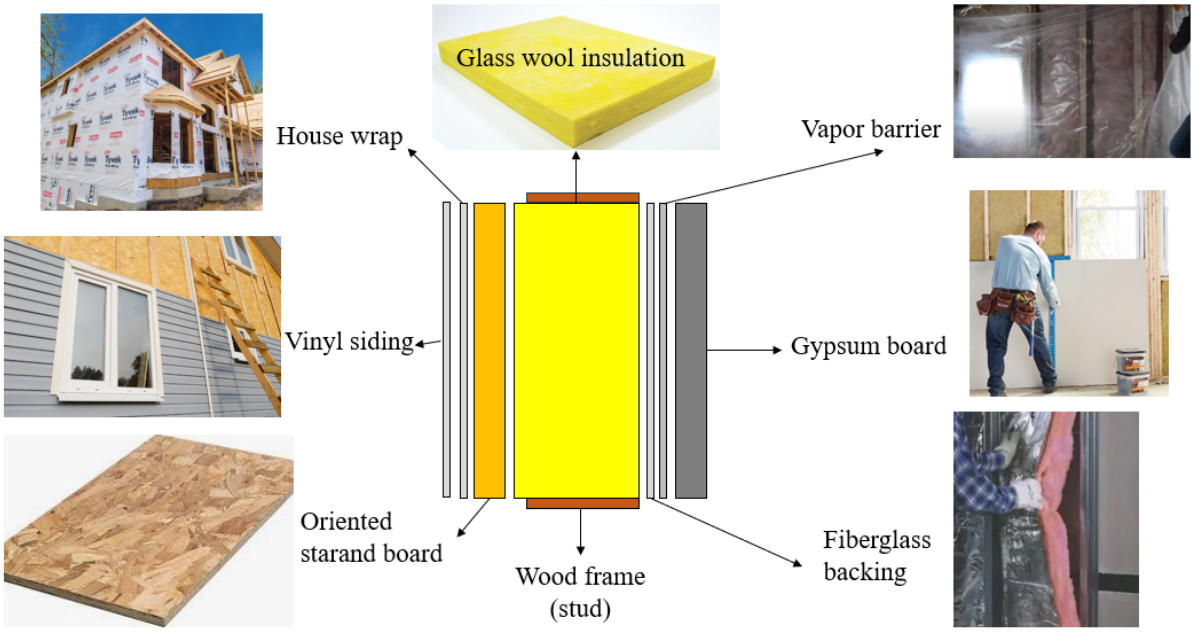


Figure 4-1. Structure of the wall

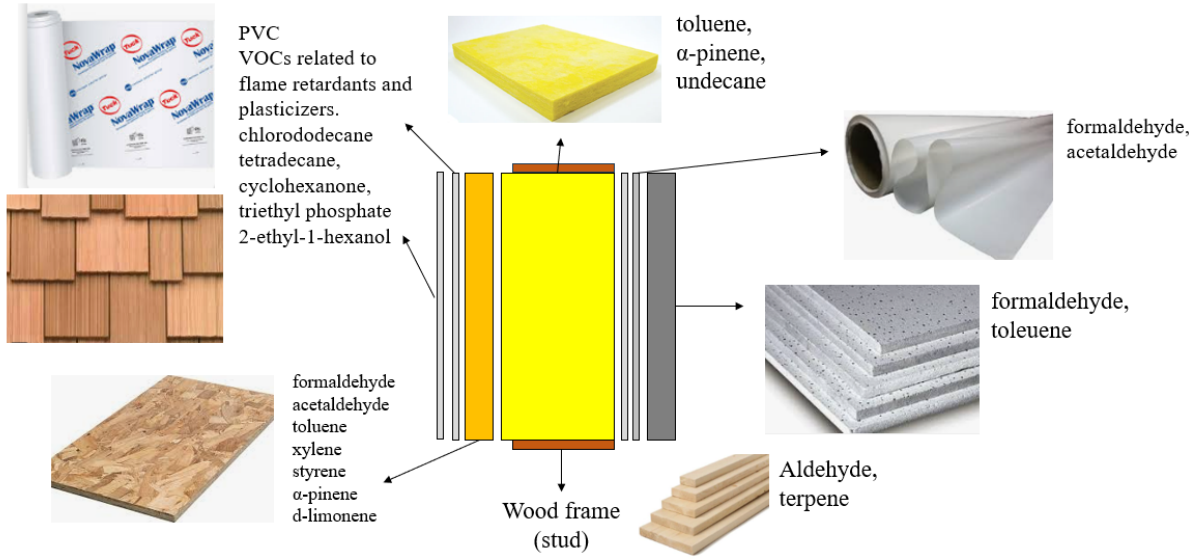


Figure 4-2. Wall of VOC

Gypsum board is a construction material used for interior walls and ceilings [4-1]. VOCs can be emitted during manufacturing and use after installation. For example, formaldehyde mainly originates from plasterboard adhesives and binders [4-2,4-3]. Additionally, toluene is released from solvents and additives used during manufacturing, while styrene is emitted from polystyrene materials, and benzene comes from solvents. In addition to glass wool insulation and drywall, various wall construction materials can emit VOCs. The main materials that emit VOCs from walls include paints and coatings, adhesives and sealants, wallpaper, wood and wooden building materials (stud) [4-4,4-5]. These materials emit VOCs primarily due to the chemicals, adhesives, paints, and finishes used during manufacturing [4-6]. Specifically, the emission of decane, undecane, and styrene was confirmed from paints, adhesives, wallpaper, and other similar materials [4-7]. It was also confirmed that wallpaper releases phthalate esters, particle board and MDF emit formaldehyde and various VOCs, and wood boards release terpenes [4-8].

Existing studies on VOCs occurring in walls are as follows. In a study by Hayashi et al., it was reported that wall materials such as plywood, plastic insulation materials, glass wool, and composite wood were the causes of VOC emissions, and VOC emissions from the wall were measured [4-9]. Other studies have analyzed how multi-layered materials such as walls emit VOCs like ethyl acetate and n-octane. The results showed that materials with higher adsorption and diffusion rates emitted VOCs faster. Additionally, increasing the frequency of ventilation significantly reduced VOC concentrations but had little additional benefit at rates above 2 h^{-1} [4-10]. The goal of another study involving wall materials was to develop a model to describe mass transfer to predict the emission

of VOCs from multilayer building materials with surfaces in contact with air on both sides. The model aims to accurately reproduce the VOC emissions occurring in walls composed of multiple layers of different materials and takes into account the mass transfer resistance through the surface boundary layer. The study demonstrated that the developed model accurately predicts VOC emissions from composite materials and provides a powerful tool for predicting emissions from furniture and multi-layer wall structures [4-11].

In the previous chapters 3, we focused on formaldehyde, styrene, xylene, d-limonene, and undecane as substances that can be emitted from wall materials. In winter, the concentrations of styrene and d-limonene were higher than those of the first species, with a statistically significant difference ($p<0.01$). In summer, the concentrations of formaldehyde, styrene, xylene, d-limonene, and undecane tended to be higher than those of the first type. Among these, there was a statistically significant difference for formaldehyde ($p<0.05$) and undecane ($p<0.01$).

Insulation materials are fundamental to modern construction practices, significantly influencing both energy efficiency and indoor air quality [4-12]. Their primary role is to act as a barrier to heat flow, ensuring that buildings remain thermally stable. This thermal regulation reduces the energy required for heating and cooling, leading to substantial energy savings and a reduction in greenhouse gas emissions. According to the U.S. Department of Energy, proper insulation can reduce heating and cooling costs by up to 20% [4-13]. An important aspect to consider in the use of insulation materials is their interaction with ventilation systems. When the exhaust ventilation system is turned on, negative pressure may be created in the room [4-14]. This negative pressure can cause the emitted of VOCs from confined spaces within walls or floors where insulation is used. VOCs are chemicals that can off-gas from certain insulation materials, especially those containing adhesives or flame retardants [4-15]. VOC emissions can reduce indoor air quality and pose health risks, emphasizing the need for adequate ventilation and the use of low-emission insulation materials [4-16].

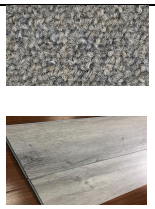
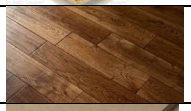
Common types of insulation materials include fiberglass, mineral wool, cellulose, and various types of foam such as polystyrene and polyurethane [4-17]. Each of these materials has unique properties suited for specific applications. Fiberglass insulation, made from fine glass fibers, is praised for its thermal performance and cost-effectiveness [4-18]. However, if not properly installed or if it becomes damaged, it can emitted small fibers into the air that may cause respiratory issues [4-19]. Furthermore, some types of fiberglass insulation are treated with formaldehyde-based resins, which can emit VOCs [4-20]. In response to these concerns, manufacturers have developed formaldehyde-free fiberglass insulation products [4-21]. Mineral wool insulation, which includes rock wool and slag wool, is noted for its fire-resistant properties and effectiveness as a sound insulator. It typically

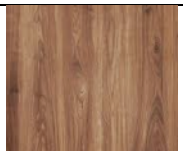
emits fewer VOCs compared to other insulation materials, making it a healthier choice for indoor air quality. Additionally, it is resistant to mold and moisture, reducing the potential for indoor air contamination through biological agents. Cellulose insulation, made from recycled paper products, is treated with fire retardants and offers an eco-friendly alternative [4-22, 4-23]. This type of insulation is known for its good thermal and acoustic properties, as well as its lower environmental impact during production. Cellulose insulation generally emits low levels of VOCs, making it a favorable option for maintaining good indoor air quality [4-24]. Foam insulations, such as polystyrene and polyurethane, offer high R-values, meaning they provide excellent thermal resistance in thin layers. However, these materials can emit significant levels of VOCs, particularly during the curing process [4-25]. Spray polyurethane foam (SPF) is especially noted for its ability to fill gaps and create a strong air barrier, but it must be applied carefully to manage its VOC emissions [4-26].

The selection of insulation materials also depends on factors such as the building's design, climate zone, environmental impact, and specific performance requirements [4-27]. The integration of advanced insulation materials in buildings aligns with global sustainability goals, as highlighted by various building codes and standards, such as those from the International Energy Conservation Code (IECC) and the Leadership in Energy and Environmental Design (LEED) certification [4-28, 4-29]. These standards emphasize the importance of energy efficiency and the use of sustainable building materials to reduce the overall carbon footprint of buildings. Insulation materials are indispensable in modern construction for improving energy efficiency, enhancing indoor comfort, and supporting environmental sustainability [4-30]. The continuous development of innovative insulation technologies promises even greater improvements in building performance. However, attention must be given to the potential emit of VOCs when ventilation systems create negative pressure, highlighting the need for careful material selection and effective ventilation strategies to ensure a healthy indoor environment [4-31].

Research on VOC emissions from building materials, including insulation materials, is diverse. These studies often focus on quantifying emissions from different types of insulation such as glass wool, mineral wool, EPS, XPS, phenolic foam, rigid polyurethane foam, cellulose, and cork. The results highlight variations in VOC emissions based on material composition, usage conditions, and environmental factors. Table 4-1 summarizes key findings from these studies, illustrating the emission rates and the specific VOCs identified.

Table 4-1. Research on VOC emission testing of building materials

Author	Year	Building materials	Condition	Substance	Picture
Renata Wiglusz et al [4-32]	2002	Laminated flooring Particle Board 2High Density Fiber	Temp.: 23+-0.3 , 29+-0.5 , 50+-0.5℃ RH : 45+-2%, 40+-2%, 10+-0.5% ACH : 0.5+-0.01 Duration :1-7Day	VOC, HCHO	
Peder Wolkoff [4-33]	1998	nylon carpet, Pvc Flooring , wooden floor polish, sealant, Water-based wall paint on gypsum board	Temp. : 23, 35, 60℃ RH: 0 or 50 Wind speed : 5cm/s	VOC	
YinpingZhang et al [4-34]	2007	Particle Board, vinyl floor sheet, Medium Density and High Density board.	Temp. : 18, 30, 40, 50℃ RH : 60+-8%	HCHO	
Chen Zhou et al [4-35]	2017	Newly built house in china (xian, hangzhou)	Temp. : 7.5~35.2℃ (Outdoor -3~33.8 RH : 26.3~90% (Outdoor 19~97)	VOC, HCHO	
Ib Andersen et al [4-36]	1975	Particle Board	Temp : 14~31℃	HCHO	
Myers [4-37]	1985	ParticleBoard	Temp. : 23~40℃	HCHO	
Lin et al. [4-38]	2009	Wooden flooring	Temp. : 15-30 ℃ Duration : 8.3 day	VOC, HCHO	
J. Xiong and Y.Zhang [4-39]	2010	MDB	Temp. : 25.2, 33.3, 41.4, 50.6℃ RH : 50.0+-1.0 % Wind speed : 5.0+-0.1l/min	HCHO	
L. Fang, et al [4-40]	1998	PVC flooring , Carpet, Floor varnish , Wall paint, Sealant	Temp : 18, 23, 28℃ RH : 30, 50, 70% (dry air 10, 30%) warm and humid air 29, 90%	VOC, TVOC	 

					
Shan Zhou et al [4-41]	2019	Artificial board	Temp. : 18, 23, 28°C +-1 (RH60%) RH:45, 60, 75+-5% (Temp. 23°C) ACH: 1.0	HCHO , TVOC (Only formaldehyde emission increases with temperature)	
Z. Liu et al [4-42]	2014	Poly methyl pentene film	Temp.: 10, 23, 30 RH : 15 50 70 (Detail conditions 23°C to relative humidity 15,50,70 (relative humidity 50 to temperature 10,23,30)	HCHO , VOC	
Young-kyu et al [4-43]	2012	Laminated flooring (HDF)	Temp. : 25,35,45+-1 RH : 55+-10 ACH : 90	TVOC, 5VOCs, HCHO (Emission increases with temperature rise)	

The following research on insulation and VOCs, titled "Evaluation of environmental impact on the formaldehyde emission and flame-retardant performance of thermal insulation materials" by Wi et al., quantitatively assesses the pollutant emissions and gas hazards of 18 different insulation materials commonly used in construction. Using the 20-L small chamber method and KS F 2271 fire testing, the study measures emissions of formaldehyde, TVOC, and other VOCs under combustion conditions. The results show that expanded polystyrene (EPS) materials emitted the highest levels of pollutants, particularly TVOC and HCHO, due to incomplete polymerization and the presence of flame retardants. In contrast, inorganic materials like glass wool (GW) and mineral wool (MW) exhibited minimal emissions. The gas hazard analysis, using rat behavioral tests, indicated that certain polyurethane insulation materials emit significant toxic gases during combustion, reducing evacuation times. The study underscores the need for stricter regulations on insulation materials to mitigate health risks associated with indoor air pollution and fire hazards [4-44].

4.1.1 Impact of negative pressure on VOC emissions

An important aspect to consider in the use of insulation materials is their interaction with ventilation systems. When the exhaust ventilation system is turned on, negative pressure may be created in the room. In Leivo's study, the research investigates the pressure differences between indoor and outdoor environments, as well as staircases,

in 26 multi-family buildings in Finland, comprising 152 apartments. The study measures these differences before and after renovations aimed at improving energy efficiency. Results show that buildings with mechanical exhaust systems had average indoor-to-outdoor pressure differences of -7.8 Pa before renovation, which increased to -19.1 Pa post-renovation. Similarly, pressure differences between indoor spaces and staircases averaged -18.6 Pa before and -9.0 Pa after renovation. The increase in negative pressure post-renovation is attributed to improved building airtightness without corresponding adjustments in ventilation systems. The research results show that modern houses have high airtightness and that the exhaust system operates to create sufficiently negative pressure inside the room due to the pressure difference between the inside and the outside [4-45]. This negative pressure can cause the emit of VOCs from confined spaces within walls or floors where insulation is used. VOCs are chemicals that can off-gas from certain insulation materials, especially those containing adhesives or flame retardants. VOC emissions can reduce indoor air quality and pose health risks, emphasizing the need for adequate ventilation and the use of low-emission insulation materials. There are studies on $PM_{2.5}$, PM_{10}, and MVOC that are not VOCs generated in closed spaces, but studies and reports on VOCs entering indoors are very lacking. Hayashi's study examined the impact of hidden pollution sources on indoor air quality by altering the ventilation system in a test house from air-supply to air-exhaust and measuring formaldehyde levels before and after the change [4-46]. They used the tracer-gas method to assess emission rates from concealed spaces in three different house structures: a wooden post-and-beam test house, a prefabricated house with a wooden panel structure, and a steel-prefabricated house. The results showed that switching to an exhaust-ventilation system significantly increased indoor formaldehyde levels, with infiltration ratios varying by location within the house. Beam spaces and partitions exhibited higher infiltration ratios compared to crawl spaces, and these ratios increased with greater pressure differences between indoor and outdoor environments. The study highlighted the need for improved ventilation systems that minimize pollutant infiltration from concealed spaces, emphasizing the importance of designing houses to limit the use of harmful chemicals in these areas and implementing barriers to prevent pollutant migration. In conclusion, to achieve acceptable indoor air quality, it is crucial to control emissions from concealed spaces and manage ventilation systems effectively, as infiltration from these hidden areas significantly contributes to indoor pollutant levels.

In other study focuses on the impact of basements on indoor air quality in 74 residences in Detroit, Michigan, by examining the levels and movement of VOCs and air exchange rates between basements and living areas. The research found that basements often contain significant VOC emission sources, such as solvents, household

cleaners, and gasoline-powered equipment, which can migrate to the living spaces. The study revealed that AERs in basements were generally higher than in living spaces, with significant seasonal variations; for example, AERs were highest in basements during the winter. The findings highlight that while the high AERs in basements help dilute VOC concentrations, the presence of many VOC sources in basements remains problematic. Therefore, advisories against storing and using strong VOC-emitting items in basements are recommended to improve IAQ. We report that basements, while not entirely enclosed spaces, significantly influence indoor VOC levels [4-47].

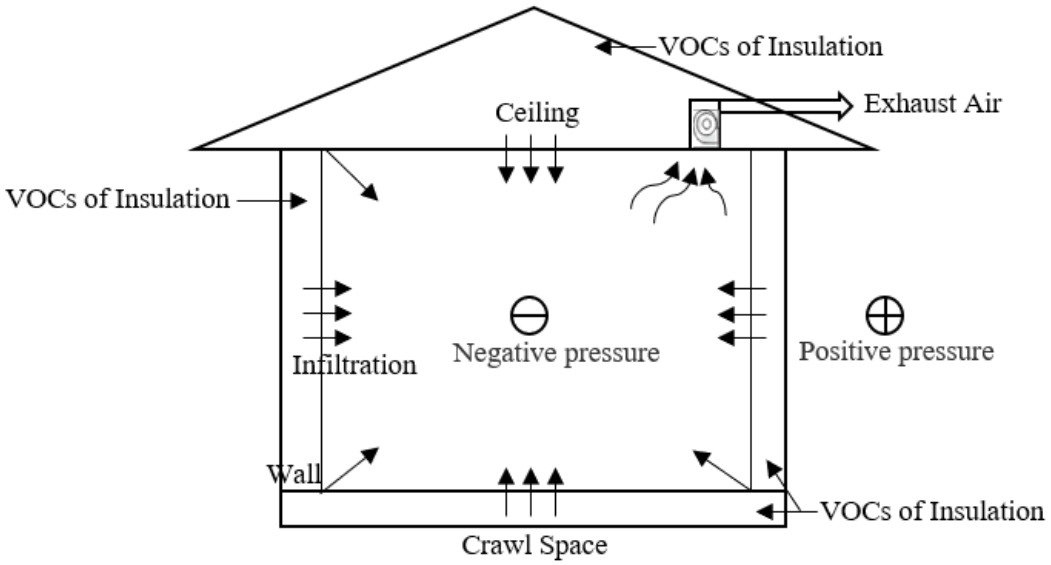


Figure 4-3. VOC schematic diagram introduced into the room under negative pressure

4.1.2 Characteristics of insulation materials and VOC emissions

In research on insulation materials and VOC emissions, this study, conducted by researchers from Yonsei University, examines the impact of VOCs emitted from building insulation materials on IAQ. Insulation materials are essential in construction for improving thermal efficiency and reducing energy loss, but they can also emit harmful pollutants. The researchers focused on 8 common insulation materials: Glass wool, Mineral wool, expanded polystyrene (EPS), extruded polystyrene (XPS), phenolic foam (PF), rigid polyurethane foam (PUR), Cellulose and Cork. Using a test bed designed to simulate real-world conditions, the study measured pollutant emissions over a 90-day period. The small chamber method was employed to provide controlled conditions for

accurate quantification of emissions. The results revealed significant differences in VOC emissions among the materials. PF and EPS, in particular, showed higher levels of TVOCs, indicating a higher potential for indoor air pollution. The study highlighted the need for stringent legal standards to regulate emissions from insulation materials. Currently, regulations tend to focus on final finishing materials, such as flooring and wallpapers, and often neglect insulation materials. This oversight is problematic given the substantial contribution of insulation materials to indoor pollution. The findings stress the importance of improved ventilation systems that can effectively manage pollutant infiltration from hidden spaces within buildings. Furthermore, the research underscored the necessity of designing buildings to minimize the use of harmful chemicals and implementing barriers to prevent pollutant migration. By enhancing the understanding of long-term VOC emissions from insulation materials, the study advocates for the establishment of legal standards to protect indoor environments and ensure the health and safety of occupants. The study reports that a comprehensive evaluation of pollutant emissions emphasizes the importance of proactive measures in building design and material selection to achieve better IAQ. Figure 4-2 shows the eight types of insulation materials used in this study. The characteristics of each insulation material are explained in (a) to (h) below.

(a) Glass wool

Glass wool, also known as fiberglass, is one of the most widely used insulation materials globally [4-48]. It is renowned for its thermal performance, acoustic properties, and cost-effectiveness. Glass wool is made from recycled glass and sand, which are melted and spun into fine fibers, forming a wool-like material. The use of glass wool is extensive worldwide due to its versatility and affordability. It is commonly used in residential, commercial, and industrial buildings for thermal and acoustic insulation. The global market for glass wool has been growing steadily, driven by increasing energy efficiency regulations and the need for sustainable building practices. According to market reports, the glass wool insulation market is expected to continue its growth, with significant demand in regions such as North America, Europe, and Asia-Pacific. Glass wool is considered environmentally friendly to a certain extent, as it is made from recycled glass, which helps reduce landfill waste and the need for raw materials. However, the production process of glass wool involves high energy consumption and the emission of greenhouse gases. Additionally, glass wool fibers can pose health risks if inhaled, necessitating proper handling and installation procedures. To mitigate these issues, manufacturers are continuously improving production processes to reduce energy consumption and emissions. Glass wool insulation is generally more affordable than other types of insulation materials like rigid foam or spray foam. The cost can vary depending on the thickness,

density, and specific product used, but it typically ranges from $0.40 to $1.00 per square foot for standard applications. This affordability makes glass wool a popular choice for both new construction and retrofit projects. Glass wool is valued for its excellent thermal and acoustic insulation properties. It is available in various forms, including batts, rolls, and loose-fill, providing flexibility for different installation requirements. Glass wool typically has an R-value of 2.2 to 4.3 per inch, depending on the density and thickness. The R-value is a measure of thermal resistance, with higher values indicating better insulating properties. For example, a glass wool batt with a thickness of 3.5 inches may have an R-value of approximately 13. The thermal conductivity of glass wool ranges from 0.030 to 0.045 W/m·K. Lower thermal conductivity values indicate better insulation performance, as the material is less capable of conducting heat. Glass wool is also highly effective in soundproofing, reducing noise transmission through walls, floors, and ceilings. Its fibrous structure helps absorb sound waves, making it a preferred choice in buildings requiring good acoustic performance, such as offices, schools, and theaters. Glass wool is non-combustible and can withstand temperatures up to 1,000°C (1,832°F) without melting. This property enhances the fire safety of buildings, providing valuable time for evacuation in case of a fire. While glass wool itself is not hydrophobic, it is often treated with water-repellent additives to improve its resistance to moisture. Proper installation and the use of vapor barriers are essential to prevent water ingress, which can reduce insulation performance and lead to mold growth. In conclusion, glass wool is a versatile, cost-effective, and widely used insulation material with excellent thermal and acoustic properties. Its environmental impact is mitigated by the use of recycled materials, though energy consumption and health risks during installation remain concerns. Overall, glass wool remains a popular choice for various insulation applications due to its performance, affordability, and adaptability to different building requirements. Additionally, studies have indicated that glass wool insulation can emit VOCs due to the binders and resins used during its manufacturing process. These emissions can contribute to indoor air quality issues, particularly in tightly sealed buildings. Research has shown that the levels of VOC emissions from glass wool are generally low and decrease over time, but it is still crucial to ensure proper ventilation during and after installation to minimize potential health risks. Some studies have reported the presence of formaldehyde and other VOCs, though at levels often within acceptable limits.

(b) Mineral wool

Mineral wool, often referred to as rock wool or slag wool, is another widely used insulation material known for its excellent thermal and acoustic properties [4-49]. It is made from natural rock materials such as basalt or diabase, or from industrial by-products like slag from steel mills, which are melted and spun into fibers. The global usage

of mineral wool is extensive due to its versatility, non-combustibility, and excellent performance in both thermal and acoustic insulation. It is widely used in residential, commercial, and industrial buildings, especially in regions with stringent fire safety regulations. The demand for mineral wool has been increasing globally, driven by its superior fire resistance and energy-saving capabilities. Europe, North America, and Asia-Pacific are the primary markets, with extensive applications in both new constructions and retrofits. According to market analysis, the mineral wool insulation market is expected to grow steadily, fueled by increasing awareness of energy efficiency and sustainable building practices. Mineral wool is considered environmentally friendly as it is made from abundant natural resources and recycled materials. Its production has a relatively low environmental impact compared to other insulation materials. However, the manufacturing process involves high energy consumption and emissions, including carbon dioxide and other pollutants. Additionally, fine fibers emitted during installation can pose health risks if inhaled, necessitating proper protective measures. Manufacturers are continually working to improve the sustainability of production processes, aiming to reduce energy consumption and emissions. Mineral wool insulation is generally competitively priced compared to other high-performance insulation materials. The cost can vary depending on the form and density of the product, typically ranging from $0.60 to $1.20 per square foot. Its affordability, combined with its performance characteristics, makes it a popular choice for various insulation applications. Mineral wool is highly valued for its thermal and acoustic insulation properties. It is available in several forms, including batts, boards, and loose-fill, providing flexibility for different installation requirements. Mineral wool typically has an R-value of 3.0 to 3.3 per inch, depending on the density and thickness. For example, a mineral wool batt with a thickness of 3.5 inches may have an R-value of approximately 12. The R-value is a measure of thermal resistance, with higher values indicating better insulating properties. The thermal conductivity of mineral wool ranges from 0.035 to 0.040 W/m·K. Lower thermal conductivity values indicate better insulation performance, as the material is less capable of conducting heat. Mineral wool is highly effective in soundproofing, reducing noise transmission through walls, floors, and ceilings. Its dense, fibrous structure helps absorb sound waves, making it an ideal choice for buildings that require good acoustic performance, such as residential buildings, offices, and theaters. Mineral wool is non-combustible and can withstand temperatures up to 1,200°C (2,192°F) without melting or releasing toxic fumes. This property significantly enhances the fire safety of buildings, providing crucial protection and valuable time for evacuation in case of a fire. Mineral wool is hydrophobic and does not absorb water. This characteristic helps prevent mold growth and maintains its insulating properties even in humid conditions. Proper installation and the use of vapor barriers further enhance its performance in moisture-prone areas. Mineral wool insulation has been found to emit low levels of VOCs, mainly

due to the binders used during its production. Studies have indicated that while the VOC emissions from mineral wool are generally low and decrease over time, it is essential to ensure proper ventilation during and after installation to minimize potential health risks. Some research has reported the presence of formaldehyde and other VOCs, but typically at levels within acceptable limits. Continuous advancements in manufacturing processes aim to reduce these emissions further, enhancing the safety and environmental friendliness of mineral wool insulation. In conclusion, mineral wool is a versatile, cost-effective, and widely used insulation material with excellent thermal and acoustic properties. Its environmental impact is mitigated by the use of natural and recycled materials, though energy consumption and health risks during installation remain concerns. Overall, mineral wool remains a popular choice for various insulation applications due to its performance, affordability, and adaptability to different building requirements. The study reports that a comprehensive evaluation of pollutant emissions emphasizes the importance of proactive measures in building design and material selection to achieve better IAQ.

(c) Expanded polystyrene

Expanded polystyrene (EPS) is a widely used insulation material known for its lightweight, moisture resistance, and excellent thermal insulating properties [4-50]. Made from expanded polystyrene beads that are fused together, EPS is utilized globally in residential, commercial, and industrial buildings, particularly in applications requiring effective thermal insulation at a low cost. The global demand for EPS has been increasing due to its cost-effectiveness and versatility, with significant markets in North America, Europe, and Asia-Pacific. Environmentally, EPS is advantageous as it is recyclable and made from 98% air and only 2% plastic, reducing its overall material footprint. However, the production of EPS involves the use of pentane as a blowing agent, which can contribute to VOC emissions and has raised environmental concerns. Additionally, EPS is non-biodegradable, which can contribute to long-term waste issues if not properly recycled. Cost-wise, EPS is among the most affordable insulation materials, typically costing between $0.25 and $0.70 per square foot, making it a popular choice for budget-conscious projects. In terms of characteristics, EPS typically has an R-value of 3.6 to 4.2 per inch, indicating good thermal resistance, and a thermal conductivity of about 0.035 to 0.040 W/m·K. It is also highly effective in soundproofing, though to a lesser extent than materials specifically designed for acoustic insulation. EPS is fire-retardant, but it can melt and emit toxic fumes when exposed to high temperatures, necessitating the use of additional fire protection measures in certain applications. It is hydrophobic and does not absorb water, which helps prevent mold growth and maintains its insulating properties in humid conditions. Regarding VOC emissions, EPS can emit small amounts of styrene and other VOCs, especially shortly after

installation, but these levels generally decrease over time and are typically within safe limits. Ensuring proper ventilation during and after installation is crucial to minimize any potential health risks. In conclusion, EPS is a versatile, cost-effective, and widely used insulation material with excellent thermal properties, though considerations for its environmental impact and VOC emissions are important for ensuring safe and sustainable use. The study reports that a comprehensive evaluation of pollutant emissions emphasizes the importance of proactive measures in building design and material selection to achieve better IAQ.

(d) Extruded polystyrene

Extruded polystyrene (XPS) is a widely used insulation material known for its high compressive strength, moisture resistance, and excellent thermal insulating properties [4-51]. Made from polystyrene resin that is melted and then extruded into rigid boards, XPS is utilized globally in residential, commercial, and industrial buildings, particularly for below-grade applications like foundations, basements, and under slab insulation. The global demand for XPS has been increasing due to its durability and superior performance in harsh conditions, with significant markets in North America, Europe, and Asia-Pacific. Environmentally, XPS has both advantages and challenges. It is recyclable and can contribute to energy savings in buildings, but its production involves blowing agents such as hydrofluorocarbons (HFCs), which have high global warming potential. Additionally, XPS is non-biodegradable, raising concerns about long-term waste management. Cost-wise, XPS is more expensive than EPS but offers better performance in specific applications, typically costing between $0.50 and $1.30 per square foot. In terms of characteristics, XPS typically has an R-value of 4.5 to 5.0 per inch, indicating excellent thermal resistance, and a thermal conductivity of about 0.028 to 0.034 W/m·K. It is also highly effective in soundproofing, though primarily valued for its thermal insulation capabilities. XPS is fire-retardant but can emit toxic fumes when exposed to high temperatures, necessitating the use of fire barriers in certain applications. Its closed-cell structure makes it highly resistant to water absorption, which helps prevent mold growth and maintains its insulating properties even in wet conditions. Regarding VOC emissions, XPS can emit small amounts of blowing agents and other chemicals, especially shortly after installation, but these levels generally decrease over time and are typically within safe limits. Ensuring proper ventilation during and after installation is crucial to minimize any potential health risks. In conclusion, XPS is a durable, effective, and widely used insulation material with excellent thermal properties, though considerations for its environmental impact and VOC emissions are important for ensuring safe and sustainable use.

(e) Phenolic

Phenolic foam insulation is a high-performance material known for its excellent thermal properties, fire resistance, and low smoke emission [4-52, 4-53]. Made from phenolic resin, it is produced by a chemical reaction between phenol and formaldehyde, which creates a highly cross-linked polymer structure that is then foamed and cured. Phenolic foam is used globally in residential, commercial, and industrial buildings, particularly in applications where high fire safety standards are required, such as in walls, roofs, and HVAC ductwork. The demand for phenolic foam has been increasing due to its superior fire resistance and thermal efficiency, with significant markets in North America, Europe, and Asia-Pacific. Environmentally, phenolic foam has several advantages, including its low thermal conductivity, which contributes to energy savings in buildings. However, the production process involves formaldehyde, which raises concerns about potential emissions and health risks. Phenolic foam is non-biodegradable, but efforts are being made to recycle and reuse the material to mitigate waste issues. Cost-wise, phenolic foam is more expensive than traditional insulation materials like EPS and XPS, typically costing between $1.00 and $2.50 per square foot, but its superior performance justifies the higher price in many applications. In terms of characteristics, phenolic foam typically has an R-value of 4.0 to 4.5 per inch, indicating excellent thermal resistance, and a thermal conductivity of about 0.020 to 0.025 W/m·K, which is lower than many other insulation materials, making it highly efficient. It is also highly effective in soundproofing, although it is primarily valued for its thermal insulation and fire-resistant properties. Phenolic foam is non-combustible and can withstand temperatures up to 1,200°C (2,192°F) without igniting, melting, or releasing significant amounts of toxic fumes. This property significantly enhances the fire safety of buildings, providing crucial protection and valuable time for evacuation in case of a fire. Phenolic foam is also resistant to moisture and does not absorb water, which helps prevent mold growth and maintains its insulating properties even in humid conditions. Proper installation and the use of vapor barriers further enhance its performance in moisture-prone areas. Regarding VOC emissions, phenolic foam can emit small amounts of formaldehyde and other VOCs, especially shortly after installation, but these levels generally decrease over time and are typically within safe limits. Ensuring proper ventilation during and after installation is crucial to minimize any potential health risks. In conclusion, phenolic foam is a high-performance insulation material with excellent thermal properties and fire resistance. Its environmental impact is mitigated by its energy-saving capabilities, though considerations for its production process and VOC emissions are important for ensuring safe and sustainable use. Overall, phenolic foam remains a popular choice for various insulation applications due to its superior performance, fire safety, and durability.

(f) Rigid polyurethane

Rigid polyurethane foam, often referred to as hard urethane insulation, is a highly efficient insulation material known for its superior thermal properties, structural strength, and versatility [4-54]. It is produced through a chemical reaction between a polyol and a diisocyanate, which creates a rigid foam structure. This insulation is extensively used worldwide in residential, commercial, and industrial buildings, particularly for applications that require high thermal performance and load-bearing capacity, such as in walls, roofs, and refrigerated storage facilities. The global demand for rigid polyurethane foam has been growing steadily, driven by increasing energy efficiency standards and the need for high-performance insulation in various climatic conditions, with significant markets in North America, Europe, and Asia-Pacific. Environmentally, rigid polyurethane foam has the advantage of significantly reducing energy consumption in buildings due to its low thermal conductivity, thereby contributing to lower greenhouse gas emissions over the building's lifetime. However, the production process involves the use of blowing agents, which historically included chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) that have high global warming potentials. Modern formulations have largely transitioned to more environmentally friendly blowing agents, but concerns about emissions during production and end-of-life disposal remain. The material is non-biodegradable, posing long-term waste management challenges, though recycling and reuse practices are being developed. Cost-wise, rigid polyurethane foam is more expensive than traditional insulation materials like fiberglass or EPS, typically costing between $1.00 and $3.00 per square foot, depending on the specific product and application. Despite the higher initial cost, its exceptional insulating properties often result in long-term cost savings through reduced energy bills. In terms of characteristics, rigid polyurethane foam typically has an R-value of 6.0 to 7.0 per inch, indicating outstanding thermal resistance, and a thermal conductivity of about 0.020 to 0.024 W/m·K. This makes it one of the most effective insulation materials available. It also provides good acoustic insulation, although its primary advantage lies in thermal performance. Rigid polyurethane foam is fire-retardant but can emit toxic fumes when exposed to high temperatures, necessitating the use of additional fire protection measures in certain applications. Its closed-cell structure makes it highly resistant to water absorption, which helps prevent mold growth and maintains its insulating properties even in wet conditions. Regarding VOC emissions, rigid polyurethane foam can emit small amounts of isocyanates and other chemicals, particularly during installation [4-55]. Proper ventilation and protective measures are crucial during application to minimize potential health risks. Over time, the emissions typically decrease to safe levels. Ensuring proper installation and following safety guidelines are essential to mitigate any adverse effects. In conclusion, rigid polyurethane foam is a high-performance insulation material with excellent thermal properties, structural strength, and moisture resistance. Its environmental impact is balanced by its energy-saving capabilities, though

considerations for its production process and VOC emissions are important for ensuring safe and sustainable use.

(g) Cellulose

Cellulose insulation is a sustainable and environmentally friendly insulation material made primarily from recycled paper products, such as newsprint, which are treated with fire retardants like borates to enhance their performance [4-56]. Known for its excellent thermal and acoustic insulating properties, cellulose is widely used in residential and commercial buildings. The demand for cellulose insulation is growing globally, particularly in regions like North America and Europe, due to increasing awareness of sustainable building practices and the drive for energy efficiency. Environmentally, cellulose insulation is highly advantageous as it is made from up to 85% recycled materials, significantly reducing landfill waste and the demand for new raw materials. Its production process requires less energy compared to many other insulation materials, contributing to a lower overall environmental footprint. Additionally, cellulose insulation has a relatively low embodied energy, which means that the total energy required to produce and transport the material is less compared to traditional insulation materials like fiberglass or foam. However, cellulose insulation must be installed carefully to avoid settling over time, which can reduce its effectiveness. Cost-wise, cellulose insulation is competitively priced, typically costing between $0.30 and $1.00 per square foot, depending on the installation method and application. This affordability, combined with its environmental benefits, makes it a popular choice for both new construction and retrofit projects. In terms of characteristics, cellulose insulation typically has an R-value of 3.2 to 3.8 per inch, indicating good thermal resistance, and a thermal conductivity of about 0.034 to 0.040 W/m·K. These properties make it effective at reducing heat transfer and improving energy efficiency in buildings. Cellulose is also highly effective at soundproofing, absorbing sound waves and reducing noise transmission through walls and ceilings. Its dense structure helps in creating a quieter indoor environment. Fire resistance is another key feature of cellulose insulation. When treated with fire retardants like borates, cellulose insulation can achieve a Class 1 fire rating, meaning it has a low flame spread and smoke development. This treatment also helps in deterring pests and mold growth, contributing to a healthier indoor environment. Moisture resistance is a consideration for cellulose insulation. While it can absorb moisture, it also has the ability to emit it, which can help in preventing mold growth if properly installed. Ensuring proper vapor barriers and ventilation is crucial to maintaining its performance in humid conditions. Regarding VOC emissions, cellulose insulation is relatively low in VOCs compared to other insulation materials. The use of natural and recycled materials, along with the non-toxic fire retardants, generally results in lower emissions. However, it is still important to ensure good ventilation during and after installation to

mitigate any potential health risks from the initial off-gassing. In conclusion, cellulose insulation is a sustainable, cost-effective, and widely used insulation material with excellent thermal and acoustic properties. Its environmental impact is significantly reduced due to its high recycled content and low energy production process, though considerations for proper installation and moisture management are important for ensuring long-term performance.

(h) Cork

Cork insulation is a natural and sustainable insulation material derived from the bark of cork oak trees. Known for its excellent thermal and acoustic insulating properties, cork is widely used in residential, commercial, and industrial buildings [4-57]. The global demand for cork insulation is growing, particularly in Europe and North America, driven by the increasing emphasis on sustainable building materials and eco-friendly construction practices. Cork is harvested without harming the trees, which continue to grow and absorb carbon dioxide, making it an environmentally friendly option with a low carbon footprint. Environmentally, cork insulation is highly advantageous due to its renewable and biodegradable nature. The production process is energy-efficient and involves minimal waste, as cork bark can be harvested every nine years without damaging the trees. Additionally, cork has natural fire-resistant properties and does not emit toxic fumes when burned, contributing to safer indoor environments. However, cork insulation can be more expensive than other insulation materials due to the time and labor involved in harvesting and processing the cork bark. Cost-wise, cork insulation typically costs between $1.50 and $3.00 per square foot, depending on the thickness and form (e.g., boards or granules). Despite the higher initial cost, its durability and sustainability often justify the investment, particularly in green building projects. In terms of characteristics, cork insulation typically has an R-value of 3.6 to 4.2 per inch, indicating good thermal resistance, and a thermal conductivity of about 0.037 to 0.040 W/m·K. This makes cork effective at reducing heat transfer and improving energy efficiency in buildings. Cork is also highly effective at soundproofing due to its cellular structure, which absorbs sound waves and reduces noise transmission. Its natural resilience and elasticity further enhance its acoustic properties. Fire resistance is another key feature of cork insulation. Cork's natural fire-retardant properties prevent it from igniting easily and ensure that it does not emit toxic fumes when exposed to high temperatures. This makes it a safer option for insulation, particularly in areas with stringent fire safety regulations. Moisture resistance is also a significant advantage of cork insulation. Cork is naturally hydrophobic, meaning it repels water and resists mold and mildew growth. This property helps maintain its insulating performance even in damp conditions and contributes to a healthier indoor environment. Proper installation and

the use of vapor barriers can further enhance its moisture resistance.

Regarding VOC emissions, cork insulation is relatively low in VOCs, especially compared to synthetic insulation materials. Its natural composition and the minimal use of additives during processing result in lower emissions. However, it is still essential to ensure proper ventilation during and after installation to minimize any potential health risks from initial off-gassing. Cork insulation, being a natural product derived from the bark of cork oak trees, can emit VOCs, including terpenes, which are organic compounds that contribute to the characteristic smell of cork. These emissions are due to the natural substances present in the cork itself and can impact indoor air quality. Studies have shown that while cork insulation has many environmental benefits, including being renewable and biodegradable, it can still emit terpenes, especially shortly after installation. This makes it important to ensure good ventilation during and after the installation of cork insulation to minimize any potential health risks. According to the study by Horn et al. (1998), the emissions of acetic acid and furfural from cork products are primarily associated with the degradation of hemicellulose [4-58]. These emissions occur due to the heat treatment applied during the manufacturing process. In conclusion, cork insulation is a sustainable, effective, and environmentally friendly insulation material with excellent thermal and acoustic properties. Its low environmental impact, combined with its fire resistance and moisture resistance, makes it a highly desirable choice for various insulation applications. While the initial cost may be higher, the long-term benefits in terms of energy efficiency, durability, and sustainability often justify the investment. The list of VOCs likely to be emitted from all insulation materials is shown in Table 4-2 [4-59, 4-60, 4-61, 4-62, 4-63].

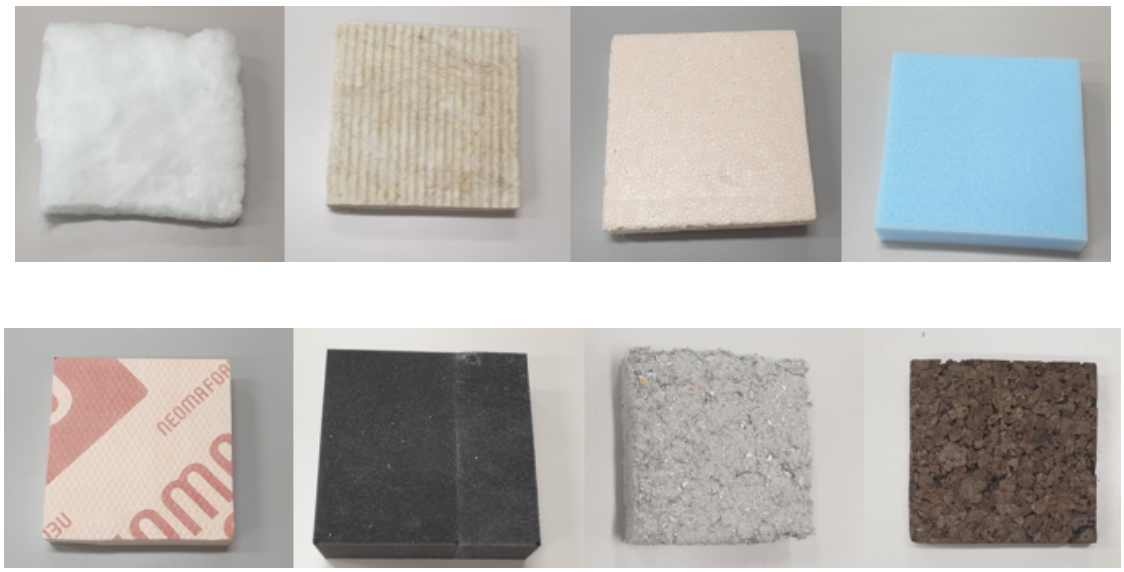


Figure 4-4. 8 insulation materials used in the measurement

Table 4-2. List of VOCs likely to be emitted from insulation materials

Insulation type	Possibly emitted VOCs
Glass wool	formaldehyde, acetaldehyde phenol, styrene, glycols
Mineral wool	formaldehyde, phenol, ammonia, acetaldehyde, hydrocarbons, naphthalene
Expanded polystyrene	benzene, toluene, xylene, styrene, pentane
Extruded polystyrene	benzene, toluene, styrene, pentane, HFCs
Phenolic	formaldehyde, acetaldehyde phenol, methanol
Rigid polyurethane	isocyanates, amine catalysts, blowing agents, formaldehyde, CFCs
Cellulose	formaldehyde, acetic acid, methanol, boric Acid
Cork	formaldehyde, acetic Acid, phenols, tannins, terpen

4.2 VOC emissions from different insulation materials

4.2.1 Small chamber method

The small chamber method is a widely used technique for assessing the emission rates of VOCs from building materials, consumer products, and furnishings. This method involves placing the test sample in a controlled environment, typically a small-scale environmental chamber, which allows for precise regulation of temperature, humidity, and airflow. The fundamental principle behind the small chamber method is to simulate indoor conditions in a reproducible manner, enabling the measurement of VOC emissions over time. The process begins by preparing the test specimen according to standardized dimensions and conditions, ensuring that it represents the typical usage scenario. The chamber itself is usually made of inert materials, such as stainless steel or glass, to prevent any interaction between the chamber walls and the emitted compounds. The chamber's size can vary, but it is generally small enough (typically between 20 to 200 liters) to allow for a high concentration of emissions to be detected within a relatively short period. Once the specimen is placed inside the chamber, the environmental conditions are stabilized. The temperature is usually set to standard indoor conditions, around 23°C, and the relative humidity is controlled, typically at 50%. The air exchange rate is another critical parameter, often set to about 0.5 to 1 ACH, ensuring that fresh air is continuously supplied while the emitted VOCs are simultaneously removed. Air samples are collected from the chamber's exhaust outlet at specified intervals using sorbent tubes or other sampling devices. These samples are then analyzed using gas chromatography-mass spectrometry (GC/MS) or similar analytical techniques to identify and quantify the VOCs present. The concentration of VOCs in the chamber air is measured, and from these data, the emission rates are calculated. The emission rate, often expressed in micrograms per square meter per hour ($\mu g/m^2h$), is determined by considering the concentration of the VOCs in the chamber, the volume of the chamber, the area of the emitting surface, and the air exchange rate. This calculation allows for the determination of how much of a particular VOC is emitted from the material into the air over a given period. The small chamber method is highly valued for its ability to provide reproducible and controlled conditions, making it possible to compare the emission characteristics of different materials and products. It is also essential for regulatory compliance, as many standards and guidelines specify acceptable emission levels for indoor air quality.

Figure 4-5. Small chamber method（20L）

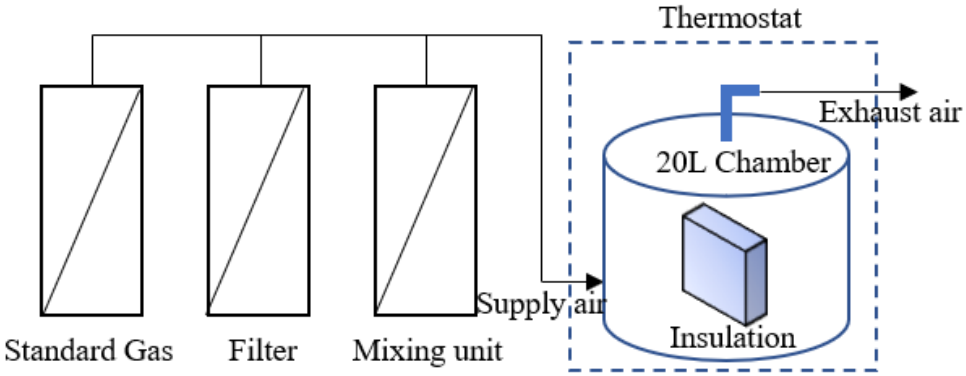


Figure 4-6. Schematic diagram of 20L small chamber method

4.2.2 Measurement conditions

In this research, I adhered to the JIS A 1902-4:2015 standard, which outlines methods for measuring VOCs, formaldehyde, and other carbonyl compound emissions from insulation materials. I began by collecting

representative samples from various batch locations to ensure accurate and comprehensive testing. These samples were stored in airtight, inert containers to prevent any contamination or alteration of their properties. Proper handling and labeling were critical to maintaining the integrity of the samples throughout the process.

For sample preparation, I cut the materials into specific dimensions of 135 mm x 135 mm x 30 mm. The edges of these specimens were sealed with aluminum tape to ensure that only the intended surfaces emitted VOCs during testing. This step was essential to prevent any side emissions that could skew the results. After preparation, the specimens were conditioned in a controlled environment at 23°C and 50% relative humidity for at least 24 hours to stabilize their properties before testing.

The testing was conducted in a precisely controlled environmental chamber maintained at 23°C ± 1°C and 50% ± 5% relative humidity, with an air exchange rate of 0.5 ± 0.05 air changes per hour (total supply flow rate 0.166 L/min). These conditions are crucial for simulating typical indoor environments and obtaining reliable emission data. The chamber's inert material construction prevented any interaction with the VOCs being measured, ensuring the accuracy of the test results. The air in the chamber was collected using a Tenax TA tube. The flow rate of the air suction pump (MINIPUMP Σ MP 30 made by SIBATA) was 0.166 L/min, which was collected for 30 minutes.

Air samples were collected from the chamber at predetermined intervals—specifically at 3, 7, 14, and 28 days—to capture the emission profile over time (In this study, only 1, 3, and 7 days were measured.). These samples were analyzed using gas chromatography-mass spectrometry (GC/MS), a sophisticated analytical technique that provides precise identification and quantification of the emitted VOCs. The GC/MS system was meticulously calibrated with standard VOC mixtures to maintain accuracy. The emission rates were calculated based on the VOC concentrations in the chamber air, the volume of the chamber, the surface area of the test specimen, and the air exchange rate. These calculations were essential for determining the emission factors, expressed in micrograms per square meter per hour (μg/m²h). After sampling, the Tenax TA tube was analyzed by GC/MS, and the emission rate was calculated using the result of the obtained VOC concentration. The calculation method of the emission rate EFa is as follows.

$$EFa = C_t \times \frac{n}{L} = C_t \times \frac{Q/v}{S/v} = C_t \times \frac{Q}{S} \ [\mu g/m^2h]$$

C_t : concentration[μg/m³], n : Air change rate[h^{-1}], L : sample loading factor [m^2/m^3], Q : ventilation rate [m^3/h], S : Exposed area [m^2] , v : Chamber volume [m^3]

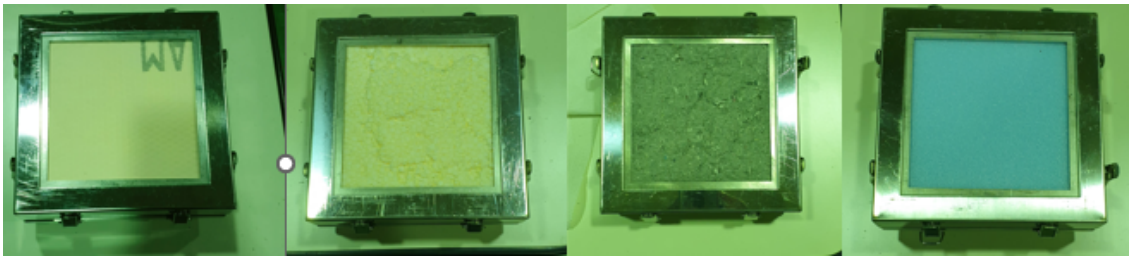


Figure 4-7. Example of customized to seal box

4.3 Results

4.3.1 Emission rate of VOC

This Figure 4-6 shows the emission rate of toluene, a VOC, emitted from various insulation materials over different time periods (1 day, 3 days, 5 days and 7 days), as determined by the small chamber method. For glass wool, the emission rate starts at 20.6 μg/m²h on the 1st day, rises to 40.7 μg/m²h on the 3rd day, then drops to 15.1 μg/m²h on the 5th day, with undetected toluene on the 7th day. The 3rd-day value exceeds the guideline of 38 μg/m²h. Mineral wool shows an increase from 20.4 μg/m²h on the 1st day to 44.7 μg/m²h on the 3rd day, with undetected toluene on the 5th and 7th days; the 3rd-day value also exceeds the guideline. Expanded polystyrene has the highest emission rate on the 1st day at 216.7 μg/m²h, which decreases over time to 144.9 μg/m²h on the 3rd day, 130.3 μg/m²h on the 5th day, and 84.0 μg/m²h on the 7th day; all values exceed the guideline. Extruded polystyrene starts at 36.4 μg/m²h on the 1st day, drops to 14.7 μg/m²h on the 3rd day, with undetected toluene on the 5th day, and further decreases to 13.7 μg/m²h on the 7th day; values are within the guideline except for the 1st day. Phenolic only has data for the 7th day at 89.9 μg/m²h, which exceeds the guideline, with undetected toluene on the earlier days. Rigid polyurethane shows a decrease from 82.4 μg/m²h on the 1st day to 40.5 μg/m²h on the 3rd day, with undetected toluene on the 5th and 7th days; both of these values exceed the guideline. Cellulose has a emission rate of 21.4 μg/m²h on the 1st day, with undetected toluene on the following days; the value is within the guideline. Cork has undetected toluene on all days, indicating that toluene emission rates were not detected.

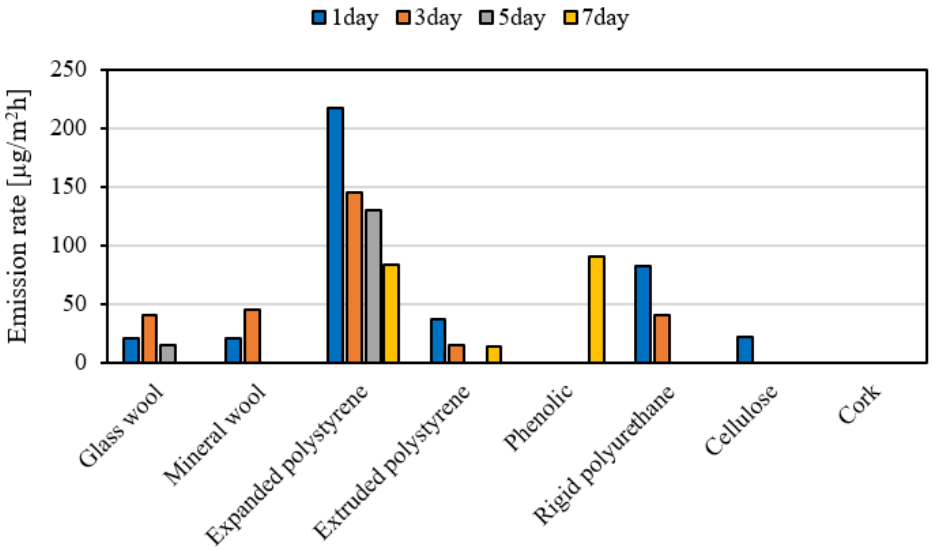


Figure 4-8. Toluene emission rates from insulation materials

Figure 4-7 shows the emission rates of ethylbenzene, a VOC, emitted from various insulation materials over different time periods (1 day, 3 days, 5 days and 7 days), as determined by the small chamber method. The guideline value for safe ethylbenzene emission rates is less than 550 µg/m²h. For glass wool, the emission rate starts at 16.8 µg/m²h on the 1st day, increases to 23.7 µg/m²h on the 3rd day, and slightly decreases to 20.8 µg/m²h on the 5th day, with undetected ethylbenzene on the 7th day. All values are well within the safety guidelines. Mineral wool shows an emission rate of 49.9 µg/m²h on the 1st day, with undetected ethylbenzene on the 3rd, 5th, and 7th days. The initial value is within the guideline. For expanded polystyrene, the emission rate is 137.8 µg/m²h on the 1st day, which decreases to 90.3 µg/m²h on the 3rd day, 87.6 µg/m²h on the 5th day, and 89.4 µg/m²h on the 7th day. All values are within the guidelines. Extruded polystyrene starts with an emission rate of 13.8 µg/m²h on the 1st day, with undetected ethylbenzene on the 3rd and 5th days, and then 13.8 µg/m²h on the 7th day. The values are within the safety guidelines. Phenolic has an emission rate of 77.5 µg/m²h on the 1st day, which decreases to 55.0 µg/m²h on the 3rd day, 23.5 µg/m²h on the 5th day, and 29.3 µg/m²h on the 7th day. All values are within the guidelines. Rigid polyurethane shows an emission rate of 82.2 µg/m²h on the 1st day and 32.1 µg/m²h on the 3rd day, with undetected data on the 5th and 7th days. The recorded values are within the guidelines. For cellulose, the emission rate of ethylbenzene is undetected on the 1st, 3rd, 5th, and 7th days, indicating that ethylbenzene emission rates were not detected. Cork also has undetected ethylbenzene on all days, indicating that ethylbenzene emission rates were not detected.

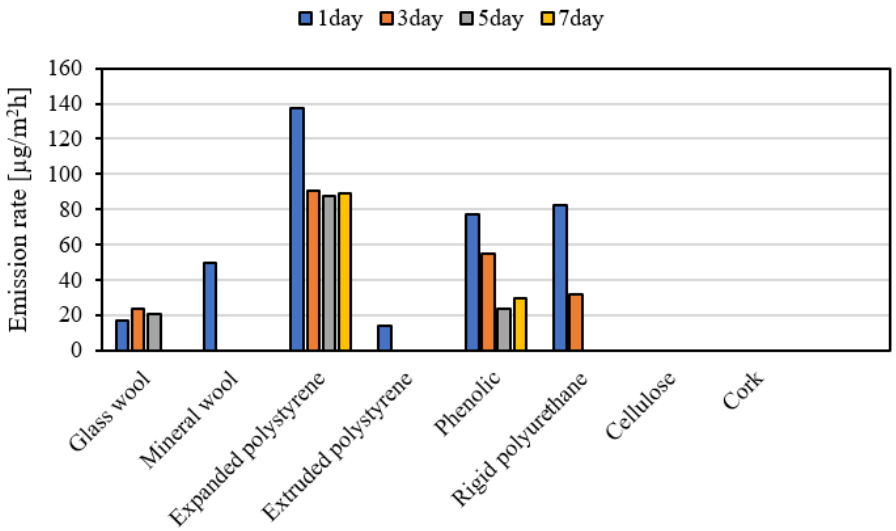


Figure 4-9. Ethylbenzene emission rates from insulation materials

The following figure shows the emission rates of styrene, a VOC, emitted by various insulation materials over different time periods (1 day, 3 days, 5 days and 7 days), as determined by the small chamber method. The

guideline value for safe styrene emission rates is less than 32 μg/m²h. For glass wool, the emission rate starts at 7.9 μg/m²h on the 1st day, increases to 48.5 μg/m²h on the 3rd day, drops to 31.0 μg/m²h on the 5th day, and further decreases to 19.0 μg/m²h on the 7th day. The 3rd-day value exceeds the guideline. Mineral wool shows an emission rate of 41.8 μg/m²h on the 1st day, with not detected styrene for the 3rd, 5th, and 7th days; the 1st-day value exceeds the guideline. Expanded polystyrene has the highest emission rates, starting at 307.3 μg/m²h on the 1st day, decreasing to 290.1 μg/m²h on the 3rd day, 298.6 μg/m²h on the 5th day, and 266.6 μg/m²h on the 7th day, all of which significantly exceed the guideline. Extruded polystyrene shows an emission rate of 259.8 μg/m²h on the 1st day, dropping to 185.6 μg/m²h on the 3rd day, 147.4 μg/m²h on the 5th day, and slightly rising to 157.1 μg/m²h on the 7th day, all of which exceed the guideline. Phenolic, rigid polyurethane, cellulose, and cork all have not detected styrene for the 1st, 3rd, 5th, and 7th days, indicating no recorded emission of styrene from these materials over the observed time periods.

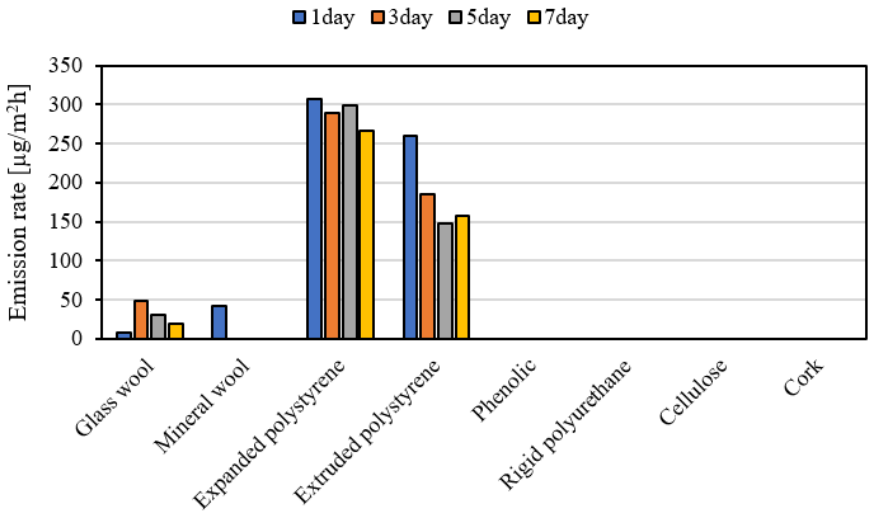


Figure 4-10. Styrene emission rates from insulation materials

The following Figure 4-8 shows the emission rates of terpane compounds emitted from cork insulation over different time periods (1 day, 3 days, 5 days and 7 days), as determined by the small chamber method. For α-Pinene, the emission rate starts at 13.9 μg/m²h on the 1st day, slightly increases to 14.9 μg/m²h on the 3rd day, drops to 13.8 μg/m²h on the 5th day, and further decreases to 12.7 μg/m²h on the 7th day. α-Muurolene shows an emission rate of 98.6 μg/m²h on the 1st day, slightly decreasing to 93.9 μg/m²h on the 3rd day, 91.6 μg/m²h on the 5th day, and 76.4 μg/m²h on the 7th day. Cubenol starts at 14.6 μg/m²h on the 1st day, increases to 21.5 μg/m²h

on the 3rd day, drops to 15.6 μg/m²h on the 5th day, and rises again to 23.5 μg/m²h on the 7th day. α-Cadinol shows an emission rate of 92.5 μg/m²h on the 1st day, decreasing to 86.2 μg/m²h on the 3rd day, 72.9 μg/m²h on the 5th day, and 50.2 μg/m²h on the 7th day. Notably, substances such as toluene, ethylbenzene, styrene, and xylene were not detected in the emissions from cork insulation.

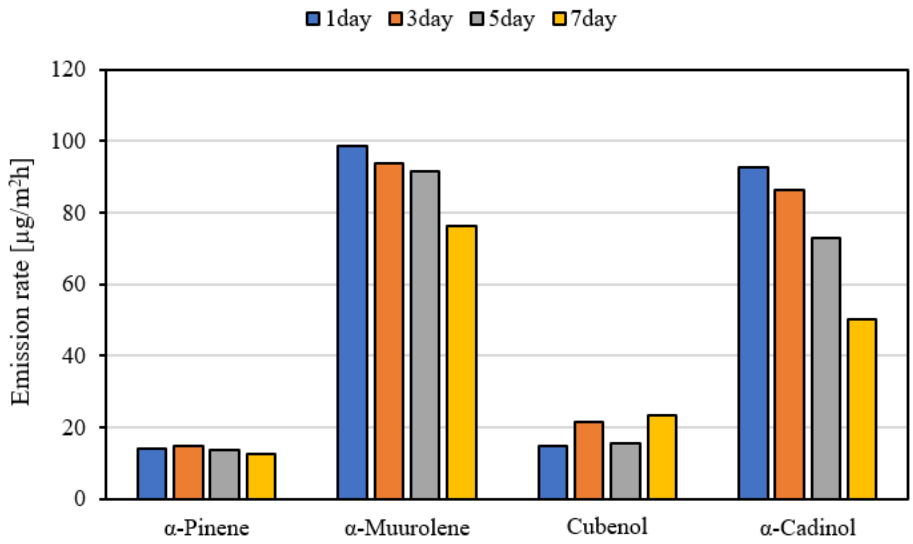


Figure 4-11. Terpene emission rates from cork insulation

4.3.2 Emission rate of formaldehyde and acetaldehyde

The following Figure 4-9 shows the emission rates of formaldehyde emitted by various insulation materials over different time periods (1 day, 3 days, and 7 days) as determined by the small chamber method. For glass wool, the emission rate starts at 8.8 μg/m²h on the 1st day, decreases to 1.7 μg/m²h on the 3rd day, and further drops to 1.1 μg/m²h on the 7th day. Mineral wool shows an emission rate of 7.6 μg/m²h on the 1st day, which decreases to 1.8 μg/m²h on the 3rd day and 0.9 μg/m²h on the 7th day. Expanded polystyrene has an emission rate of 7.7 μg/m²h on the 1st day, dropping to 1.1 μg/m²h on the 3rd day and 0.6 μg/m²h on the 7th day. Extruded polystyrene starts with an emission rate of 0.6 μg/m²h on the 1st day, which slightly increases to 0.8 μg/m²h on the 3rd day and remains at 0.6 μg/m²h on the 7th day. Phenolic shows an emission rate of 4.0 μg/m²h on the 1st day, decreasing to 3.6 μg/m²h on the 3rd day and 2.1 μg/m²h on the 7th day. Rigid polyurethane emits formaldehyde at a rate of 1.6 μg/m²h on the 1st day, which decreases to 0.8 μg/m²h on the 3rd day and 0.5 μg/m²h on the 7th day. Cellulose shows an emission rate of 4.2 μg/m²h on the 1st day, dropping to 0.8 μg/m²h on the 3rd

day and 0.7 μg/m²h on the 7th day. Cork has an emission rate of 7.0 μg/m²h on the 1st day, which decreases to 1.4 μg/m²h on the 3rd day and 1.2 μg/m²h on the 7th day.

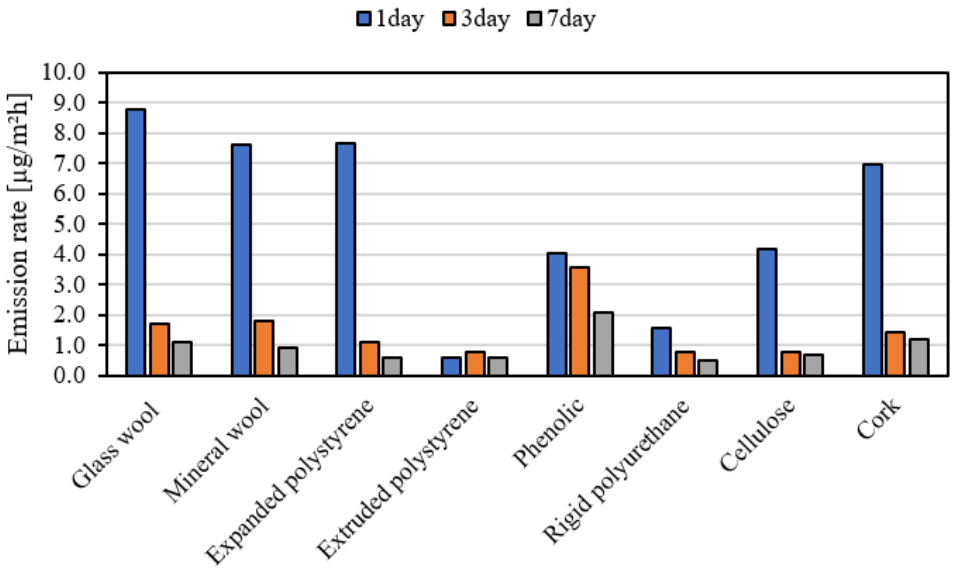


Figure 4-12. Formaldehyde emission rates from insulation materials

The following Figure 4-10 shows the emission rates of acetaldehyde emitted by various insulation materials over different time periods (1 day, 3 days, and 7 days) as determined by the small chamber method. For glass wool, the emission rate starts at 11.4 μg/m²h on the 1st day, decreases to 3.3 μg/m²h on the 3rd day, and further drops to 2.6 μg/m²h on the 7th day. Mineral wool shows an emission rate of 14.4 μg/m²h on the 1st day, which decreases to 1.7 μg/m²h on the 3rd day and 1.4 μg/m²h on the 7th day. Expanded polystyrene has an emission rate of 19.5 μg/m²h on the 1st day, dropping to 2.2 μg/m²h on the 3rd day and 1.8 μg/m²h on the 7th day. Extruded polystyrene starts with an emission rate of 9.0 μg/m²h on the 1st day, which decreases to 0.9 μg/m²h on the 3rd day and 0.4 μg/m²h on the 7th day. Phenolic shows an emission rate of 3.3 μg/m²h on the 1st day, decreasing to 0.7 μg/m²h on the 3rd day and 0.8 μg/m²h on the 7th day. Rigid polyurethane emits acetaldehyde at a rate of 0.7 μg/m²h on the 1st day, which increases to 5.9 μg/m²h on the 3rd day and then decreases to 1.2 μg/m²h on the 7th day. Cellulose shows an emission rate of 0.7 μg/m²h on the 1st day, increasing to 1.0 μg/m²h on the 3rd day and dropping to 0.7 μg/m²h on the 7th day. Cork has the highest initial emission rate of 24.9 μg/m²h on the 1st day, which decreases to 2.5 μg/m²h on the 3rd day and 2.3 μg/m²h on the 7th day.

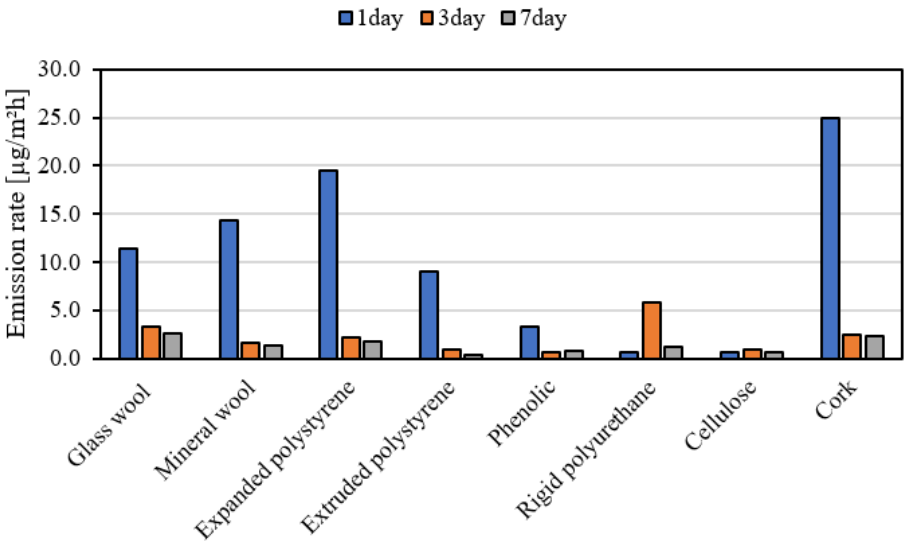


Figure 4-13. Acetaldehyde emission rates from insulation materials

4.4 Comprehensive review of the characteristics of eight insulation materials

Various studies have investigated the emissions of VOCs and formaldehyde from building materials, including glass wool. MansourElie found that different types of wool can absorb VOCs, including formaldehyde, to varying degrees [4-64]. Xiong developed a model to predict VOC and formaldehyde emissions from building materials [4-65], which both Maskell and Brown found could be useful in minimizing emissions [4-66, 4-67]. Highlighting the potential impact of bio-based insulation materials on indoor air quality, Maskell specifically mentioned their ability to reduce VOCs, and Sidheswaran and Hodgson further investigated formaldehyde emissions from building materials [4-68, 4-69]. Overall, these studies found that while glass wool can emit VOCs and formaldehyde, it can also absorb some of these compounds and affect indoor air quality. This suggests that the impact may be influenced by factors such as wool type and the presence of other building materials.

Mineral wool insulation emits VOCs and formaldehyde, which can pose potential health risks [4-70]. To solve this problem, low formaldehyde binders for mineral wool insulation are being developed [4-71]. Wool, including mineral wool, has been found to absorb VOCs, including formaldehyde, which may help mitigate emissions [4-72].

Expanded polystyrene (EPS) insulation can emit VOCs and formaldehyde, which can affect indoor air quality. Adding powdered graphite to EPS can reduce VOC emissions, especially styrene [4-73]. However, emissions of formaldehyde and other VOCs from EPS can still be significant [4-74]. EPS insulation has been found to contain high levels of the hazardous substance benzene [4-75]. Ignition of EPS can emit potentially toxic emissions, including formaldehyde and total VOCs [4-76]. Products containing EPS, such as pillows and cushions, can also contribute to indoor air pollution by emitting styrene and other chemicals [4-77]. VOCs emitted from EPS-based food containers vary with temperature and compounds such as benzaldehyde, styrene, ethylbenzene, and tetradecane are identified [4-78].

Extruded polystyrene insulation can emit a variety of VOCs, including acetic acid, pentanal, hexanal, and styrene [4-74]. Adding powdered graphite to insulation can reduce these emissions, especially styrene emissions [4-73]. However, insulation can still emit harmful substances such as benzene, which can exceed health limits [4-75]. When ignited, polystyrene foam can emit potentially toxic emissions, including formaldehyde and total VOCs [4-76].

For cellulose, study presents the primary VOC emission fluxes, ozone (O_3) reaction probabilities (γ), and yields

of O_3 reaction byproducts for eight widely used insulation materials. The primary VOC fluxes, measured in a continuous flow reactor with proton transfer reaction-time of flight-mass spectrometry, ranged from 3 μmol/m^2h for polystyrene with thermal backing to 61 μmol/m^2 h for cellulose. The estimated total VOC mass emission rates varied between approximately 0.3 and 3.3 mg/m^2h. The main primary VOC emissions from cellulose were tentatively identified as compounds likely resulting from the chemical and thermal decomposition of cellulose [4-79].

A number of studies have been conducted on the emissions of VOCs and formaldehyde from various building materials, including cork insulation. Kort found that expanded cork insulation does not emit harmful substances at levels that could affect human health [4-75]. Similarly, Maskell highlighted the potential of bio-based insulation, including cork, to improve indoor air quality [4-66]. Fuczek highlighted the influence of production parameters and flame retardants on VOC emissions from wood fiber insulation, suggesting that similar factors may also affect cork insulation [4-59]. Silvestre and Yu discussed the environmental impacts and emission parameters of insulation materials, respectively, providing a broader context for understanding cork insulation [4-80, 4-81].

In this study, expanded polystyrene had the highest toluene emission rate, exceeding the guideline of 38 μg/m²h. Rigid polyurethane also exceeded the guideline at 40.5 μg/m²h on the 7th day. Similar to the results of previous studies, the emission of toluene was confirmed, and the emission rate was somewhat different. Expanded polystyrene had the highest emission rate of ethylbenzene at approximately 140 μg/m²h, but did not exceed the guideline of 550 μg/m²h. Phenolic, rigid polyurethane, and glass wool were also found to emit emissions. p-xylene was not detected in all insulation materials. For styrene, expanded polystyrene was the highest on the 7th day at about 250 μg/m²h, and extruded polystyrene was also at about 150 μg/m²h on the 7th day.

Expanded polystyrene insulation and extruded polystyrene insulation had the highest styrene emissions. This can be attributed to their chemical composition and manufacturing process. Polystyrene, the primary component of both EPS and XPS, is a polymer derived from the monomer styrene. During production, residual styrene monomers can remain trapped within the polymer matrix. Over time, these residual monomers can off-gas, leading to detectable styrene emissions. Additionally, the extrusion process used to create XPS can introduce more styrene monomers due to the higher temperatures and pressures involved, potentially increasing the emission levels compared to other insulation materials.

Terpene substances can be emitted from cork insulation due to the natural chemical composition of cork. Cork is

derived from the bark of the cork oak tree (Quercus suber), which contains high levels of terpenes, such as limonene and pinene [4-82]. These compounds are a part of the tree's defense mechanism against pests and pathogens. When cork is processed into insulation material, the natural terpenes present in the bark can be emitted into the surrounding environment. The emit of these VOCs is influenced by factors such as temperature, humidity, and the age of the cork material. Scientific studies have shown that natural cork emits a variety of terpenes, contributing to its characteristic scent and potential VOC emissions in indoor environments.

As in previous studies, formaldehyde and acetaldehyde were also detected in all eight insulation materials, but the amount emitted by the 7th day was so low that it did not pose a problem.

4.5 Summary

In Chapter 4, additional research was conducted to supplement the results in Chapter 3. As a result of comparing balanced ventilation and unbalanced ventilation houses in Chapter 3, the VOC concentration in unbalanced ventilation houses was high. There are various reasons for this, but one significant factor is the improved airtight performance of the house. If an unbalanced ventilation device is used, the room can easily become negatively pressurized, causing VOC emissions from the walls, floor, and roof to flow into the room. Therefore, VOC emissions were measured by selecting insulation as one of the building materials. The results are as follows:

a) Among the substances subject to the guidelines, toluene was confirmed to be emitted from all seven insulation materials except cork insulation. However, except for expanded polystyrene, emissions were very low. Expanded polystyrene emitted 220 μg/m²h on the 1st day, which decreased to approximately 70 μg/m²h on the 7th day, but it was still slightly higher than the guideline value of 38 μg/m²h.

b) For ethylbenzene, it was detected in six insulation materials excluding cellulose and cork. Among them, expanded polystyrene had the highest level (80 μg/m²h on day 7), followed by phenolic and rigid polyurethane. However, it did not exceed the guideline value of 550 μg/m²h.

c) Styrene was detected in glass wool, expanded polystyrene, and extruded polystyrene. Glass wool showed very low emissions, but expanded polystyrene and extruded polystyrene had very high emissions of approximately 250 μg/m²h and 160 μg/m²h, respectively. The guideline value of 32 μg/m²h was exceeded.

d) No VOCs other than terpenes were detected in cork insulation. Among the detected terpenes, α-Muurolene and α-Cadinol were approximately 70 μg/m²h and 50 μg/m²h, respectively, on the 7th day. α-Pinene and Cubenol were less than 20 μg/m²h.

e) Formaldehyde and acetaldehyde were detected in eight insulation materials. Formaldehyde emissions were measured to be over 4 μg/m²h for five insulation materials, including glass wool, on the first day, but were very low at 1 μg/m²h on the 7th day. The amount of acetaldehyde emitted was over 10 μg/m²h for 5 insulation materials, including glass wool, on the 1st day, but was measured as low at less than 2 μg/m²h on the 7th day.

The findings from Chapter 4 supplement the results of Chapter 3 by providing detailed insights into VOC emissions from various insulation materials in houses with unbalanced ventilation systems. It was observed that unbalanced ventilation contributes to higher VOC concentrations due to the negative pressurization of the room,

causing emissions from building materials to infiltrate indoor air. Specifically, expanded polystyrene insulation exhibited the highest emissions of toluene, ethylbenzene, and styrene, often exceeding guideline values, particularly in the initial days. Although emissions of formaldehyde and acetaldehyde were initially high across multiple materials, they significantly decreased over time. Therefore, houses that use unbalanced ventilation should be more concerned with ventilation and managed. The selection of low-emission insulation and the use of a balanced ventilation system have the advantage of maintaining healthier indoor air quality by mitigating the adverse effects of VOC emissions from building materials.

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Chapter 5. Conclusions

5.1 Outcome

The novelty of this study is to reveal the characteristics of VOC concentration in various regions and types of houses targeting houses actually occupied by residents in Japan, and to evaluate the VOC concentration of balanced ventilation (central air conditioning) and unbalanced ventilation equipment as a factor. One point of focus in the study is that it was hypothesized that a stable amount of ventilation was secured in houses with balanced ventilation facilities, while in houses with only unbalanced ventilation, the concentration of VOCs increased due to the attraction of VOCs indoors caused by a lack of ventilation or negative pressure. Ventilation facilities have previously been mandated as a measure against sick building syndrome; however, the reality is that there has been very little research on the effects of ventilation methods (balanced and unbalanced), and there is also very little research on VOC concentration in houses using central air conditioning systems. Additionally, this study is meaningful as it examines not only how ventilation determines indoor VOC concentration but also the effect of ventilation equipment on indoor air quality from the walls in residential homes. Moreover, we measured and evaluated the VOCs of various types of insulation materials generated from insulation materials in the walls due to negative pressure. The results from Chapters 2 to 4 can be summarized as follows.

The results of the research in Chapter 2 are as follows. This study measured VOCs and carbonyls in the living rooms and bedrooms of 116 Japanese houses in winter and 66 houses in summer. Most substances were present at very low concentrations, but 12% of houses exceeded guideline values for formaldehyde and acetaldehyde. Houses with higher acetaldehyde concentrations were characterized by daily alcohol consumption. Living room concentrations were slightly higher than bedroom concentrations, with no significant seasonal differences. VOC concentrations were highest in apartments and higher in built-for-sale houses than in custom-made houses. In winter, statistically significant differences ($P<0.01$) in xylene concentrations were found between apartments and private rental houses, and between other housing types ($P<0.05$). In summer, significant differences ($P<0.01$) in acetaldehyde concentrations were observed between custom-made and private rental houses, and between other housing types ($P<0.05$). VOC concentrations did not generally vary with residence period, except for acetaldehyde. Houses with unbalanced ventilation had higher pollutant concentrations than those with balanced ventilation, especially in winter. In winter, decane concentrations in homes using kerosene fan heaters were 57.8% to 136.4% higher than in homes using other heating devices.

The research results of Chapter 3 are as follows. The survey measured 33 houses, with 48.5% using balanced

ventilation and 51.5% using unbalanced ventilation. In homes with unbalanced ventilation, 70.6% had air inlets in the living room, with 52.9% always open. During summer, 93.8% of Group A and 76.5% of Group B used ventilation systems 24 hours a day; similar usage was observed in winter. Heating devices in winter varied, with balanced ventilation homes primarily using central air conditioning, while unbalanced ventilation homes used a mix of air conditioners, electric heaters, and other devices. Group A had significantly higher average temperatures in winter and summer compared to Group B, indicating the impact of air conditioning systems. Regarding relative humidity in winter, Group A's living rooms had a mean of 37.4% and bedrooms of 42.9%, significantly lower than Group B's living rooms (47.0%) and bedrooms (53.9%), highlighting better humidity control by Group A. In summer, humidity control was similar between the groups. Group A's CO_2 concentrations were significantly lower in winter, with Group B's CO_2 concentrations remaining slightly higher and more variable in summer. Group A consistently maintained higher air change rates in both seasons, exceeding the required rate of 0.5 h^{-1}, while Group B's rates were lower, especially in bedrooms. The higher CO_2 concentrations in Group B during winter were influenced by the use of oil stoves and fan heaters. Seasonal behaviors, such as window opening in summer and heating in winter, contributed to the observed differences between the groups. In winter, VOC detection rates for formaldehyde and acetaldehyde were high in both groups, with Group B having generally higher concentrations of most chemicals, especially α-pinene, d-limonene, undecane, and dodecane, showing statistically significant differences. During summer, VOC detection rates increased, and Group B still showed higher VOC concentrations, but with less variability. A negative correlation between air change rate (ACH) and VOC concentrations was observed in Group A, while Group B showed a mix of positive and negative correlations. The results indicate that Group A's balanced ventilation system more effectively controls VOC concentrations than Group B's unbalanced system. The correlations between ACH and VOCs are weak and vary by season, location, and ventilation system, with no consistent patterns.

In Chapter 4, additional research was conducted to supplement the results from Chapter 3, focusing on VOC emissions in houses with unbalanced ventilation. Measurements of insulation materials showed the following: Toluene was emitted from all but cork insulation, with expanded polystyrene exceeding the guideline value. Ethylbenzene was detected in six materials, highest in expanded polystyrene, but below the guideline value. p-xylene was not detected in all insulation materials. Styrene emissions were high in expanded and extruded polystyrene, exceeding the guideline value. Cork insulation emitted only natural chemicals called terpenes, with α-Muurolene and α-Cadinol being the most common. Formaldehyde and acetaldehyde were detected in eight

materials, with emissions decreasing significantly by the 7th day. Overall, it was found that VOC emissions from insulation materials indicate that under negative pressure conditions, VOCs from enclosed spaces are likely to infiltrate the indoor environment.

The study revealed that while VOC and carbonyl concentrations in most Japanese houses were low, 12% exceeded guidelines for formaldehyde and acetaldehyde, particularly in homes with frequent alcohol consumption. Living rooms had higher VOC levels than bedrooms, regardless of season. Apartments and built-for-sale houses exhibited the highest concentrations, with significant differences observed between housing types for xylene and acetaldehyde levels. Unbalanced ventilation systems were associated with higher pollutant concentrations compared to balanced systems, especially in winter. Kerosene fan heaters significantly increased decane concentrations in winter.

The study provides valuable data on indoor VOC and carbonyl concentrations in Japanese houses during winter and summer. It highlights the importance of housing type, ventilation system, and heating method in affecting indoor air quality, suggesting that tailored strategies are necessary to mitigate VOC exposure. Balanced ventilation systems were shown to significantly reduce and stabilize VOC and CO_2 concentrations, achieving better temperature and humidity control compared to unbalanced systems. The findings emphasize the need for selecting low-emission insulation materials and employing balanced ventilation to maintain healthier indoor environments.

5.2 Limitations and future perspective

Although 182 homes (in Chapter 2) and 34 homes (in Chapter 3) constitute a relatively solid sample, it may still be insufficient to represent the variety of living conditions and housing types across Japan. Moreover, VOC concentrations can vary depending on local environmental factors, so the study results may be limited if the homes sampled are not evenly distributed across multiple geographic areas. Since VOC sampling was conducted only once in winter and summer, I believe more accurate results can be obtained if sampling is continuously measured and monitored on a weekly or, at minimum, monthly basis. We also identified the VOC concentration in the living room and bedroom of the house according to the ventilation system. The daily usage time of the ventilation system and other air purifiers was obtained through a questionnaire survey. Window opening was also investigated. I believe a better analysis could have been made if the frequency and duration of window opening had been measured in the same house during both winter and summer, and if the presence or absence of interior renovations during the measurement period had also been identified. There is also a limitation in not measuring the c-value of the house.

Future studies should address the aforementioned shortcomings by expanding the sample size and diversity of housing types, conducting periodic VOC monitoring, investigating all activities related to ventilation, and providing detailed identification of VOC pollutant sources such as renovations, introduction of new furniture, and use of household products, along with assessing health impacts.

Achievement

(a) Published papers

1. Park S., Kagi N., Umishio W., Hasegawa, K., Mitamura, T., Tamura, J. (2024). VOC concentrations in houses in Japan: correlations with housing characteristics and types of ventilation. Journal of Asian Architecture and Building Engineering, 1-17.

2. Park S., Kagi, N., Umishio W., Shinohara N, Kim H. Influence of Mechanical Ventilation Systems on Indoor VOC Concentrations in Residential Buildings (submitted on June 21, 2024)

(b) Conference presentation

1. Sang-in Park, Naoki Kagi, Wataru Umishio, Kenichi Hasegawa, Jo Tamura, Teruaki Mitamura. Study on Housing Characteristics and Volatile Organic Compound Concentration, Proceedings of the 40th Air Purification and Contamination Control Research Conference, pp. 223-225, April 2023.

2. Sang-in Park, Naoki Kagi, Wataru Umishio, Kenichi Hasegawa, Jo Tamura, Teruaki Mitamura. VOC Concentrations in Houses in Japan: Correlations with Housing Characteristics and Types of Ventilation, The Asian Symposium on Contamination Control, pp. 253-260, August 2023.

3. Sang-in Park, Naoki Kagi, Wataru Umishio, Naohide Shinohara, Hun Kim. Effects of Differences in Residential Mechanical Ventilation Equipment on Volatile Organic Compound Concentration, Proceedings of the 41st Air Purification and Contamination Control Research Conference, pp. 199-201, April 2024.

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