Importance and design of ventilation for an improved sleep quality

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Abstract. Sleep quality, which significantly affects daily life, is also affected by indoor air quality as well as comfort aspects such as noise, temperature, and relative humidity in the sleeping environment. Carbon dioxide concentration is commonly considered as an indicator of indoor air quality, and it has been observed that increasing concentrations increase the number of awakenings resulting in lower sleep quality. In this study, ventilation flow rate is investigated specifically for bedroom and dormitory room dimensions as recommended by different countries to ensure that the carbon dioxide concentration in sleeping spaces is below an upper limit value (1000 ppm). The ventilation flow rate is evaluated by comparison to the methods recommended in ASHRAE standards (Ventilation Rate Procedure–VRP, Indoor Air Quality Procedure–IAQP and Natural Ventilation Procedure–NVP). Although VRP is widely used because a constant flow rate is recommended, which is an advantage due to design simplicity, it has been found to be insufficient for smaller sleeping space volumes and it gives higher values for larger volumes compared to the needed ventilation flow rates recommended by IAQP. Moreover, the fact that the flow rates calculated with VRP and IAQP overlap in a very narrow area shows that the use of VRP values for effective designs is limited. Consequently, we suggest that the system design should be operated with the IAQP and with demand-controlled ventilation, and an approach is offered to designers for direct use in practice.

Keywords. Sleep quality, indoor air quality, ventilation rate
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1. Introduction

Today, people spend most of the day indoors and spend approximately a third of this time sleeping. Sleep quality, which has a significant effect on daily activities, is highly dependent on environmental factors and physical and mental functions of people. A high-quality sleep environment in terms of noise, room temperature, and relative humidity is necessary for good sleep quality [1,2]. CO_2 concentration is considered as an indicator of the ventilation efficiency of the environment and thus the indoor air quality. Higher ventilation rates and so lower CO_2 concentrations provide a reduction in the number of awakenings [2–4] and higher sleep quality [4–6].

Ventilation standards for bedrooms show that countries have different design approaches regarding the fresh air flow that should be provided in bedrooms [7]. These approaches can be classified as (i) consideration of the air flow rates predicted for the unit area and the number of people; (ii) consideration of the air flow rates predicted for the number of people; (iii) consideration of certain values of air change per hour per unit area; (iv) consideration of certain values of air change per hour at the prescribed volume; (v) considering that the CO_2 concentration does not exceed an average value during the sleep period, and (vi) consideration of an upper limit value of the CO_2 concentration in the design.

ASHRAE standards propose three different procedures to determine the ventilation rate: Ventilation Rate Procedure (VRP), Indoor Air Quality Procedure (IAQP), and Natural Ventilation Procedure–(NVP) [8]. As VRP, ASHRAE 62.1-2019 standard [8] provides a calculation of a ventilation rate for any dwelling unit while the total required fresh air flow rate for whole dwelling can be calculated by ASHRAE 62.2-

2019 standard [9].

Pollution concentration is related to volume and dependent on the architectural codes and traditions of the countries, and their social and economic development. Therefore, determining the fresh air flow rate by constant values according to the floor area and/or the number of people, or volume for ventilation (the first four approaches) cannot guarantee that the CO_2 concentration in the bedrooms during the sleeping period will exceed the value that affects the sleep quality, which has negative physical and mental effects. Although VRP is widely used to determine ventilation rate due to providing ease of calculation independent of the intended use of the space, it is not considered appropriate in ventilation design.

On the other hand, NVP should be considered as an auxiliary system by architects and mechanical installation engineers in all building designs, rather than being the main solution. Because natural ventilation can reduce the load of mechanical systems and improve energy savings. However, these systems cannot provide 100% of ventilation safety.

In IAQP, which is the recommended procedure for ventilation design, the required reliable ventilation flow is determined by using the safe pollution level, the volume of the living space, and the anthropometric characteristics of the people using the space as in this study. Monitoring the CO_2 concentration by demand-controlled ventilation provides an adaptation to changes related to volume functions and a possibility to use the advantages of natural ventilation. Although keeping CO_2 concentration below a constant value could be possible in demand-controlled ventilation systems designed by IAQP, it is practically not possible to ensure that the CO_2 concentration during sleep is below an average value considering the parameters affecting the sleep duration. Therefore, the fifth design approach is not considered as appropriate for ventilation design. The proposed design approach is the sixth approach: The ventilation system of the sleeping space should be a demand-controlled system that can provide fresh air at the air flow rate determined according to the maximum number of people and keep the CO_2 concentration in the sleeping space below an upper limit value.

In this study, it is recommended to use IAQP to determine the ventilation flow rate to ensure that the CO_2 concentration in the bedroom is below an upper limit value. Considering the difficulty with using this method which is the lack of a designer-friendly and approved calculation tool, this study aims to present an approach that designers can use directly for application by explaining the IAQP specific to bedrooms and dormitories.

2. Research Methods

2.1 CO_2 concentration as a control parameter in IAQP

CO_2 concentration is used as a control parameter which should be below an upper limit for determining ventilation rate by IAQP. The reason behind using the CO_2 concentration is that the source of this pollution is people in an indoor environment, and it is also an indicator of increasing pollutant concentrations since the accumulation of emitted chemicals from other sources occurs along with CO_2 from people in limited-ventilation indoor environments.

A tentative relationship between CO_2 concentration and sleep quality, which needs to be verified, is presented in a comparative study of the ventilation standards in bedrooms by Sekhar et al. [7]. Although the CO_2 concentrations below 750 ppm did not disturb the sleep quality, the range between 750-1150 ppm results in possibly disturbed sleep quality. In addition to disturbing sleep quality in the range of 1150-2600 ppm, reduced next-day cognitive performance was also observed for the concentrations higher than 2600 ppm. Additionally, the CO_2 concentration (higher than the average value of 800-1000 ppm) in the sleeping space shows the following effects [10]: (1) sleep efficiency, (2) waking frequency during the sleep phase, (3) sleep stages, and finally (4) mental performance the next day. Although the range of CO_2 concentration does not disturb sleep quality was given as < 750 ppm in the study [7], this study [10] expressed the range as <800 ppm. Moreover, the CO_2 concentration of 800 ppm is the standard upper limit for bedroom ventilation in Denmark [11].

To date, the upper limit value is 1000 ppm or 700 ppm above the outdoor CO_2 concentration (+700 ppm difference) as given in the “Informative Annex-D” in ASHRAE 62.2-2016. However, discussions continue on the suitability of these values for acceptable indoor air quality and mitigation of impact on human health and performance. Although these values were not given as mandatory upper limits that should be provided by ventilation in the relevant ASHRAE standards (62, 62.1, and 62.2), they were perceived as mandatory limits in practice and ventilation flow rates were used in the design to ensure that these values were not exceeded by the IAQP [12]. It is stated that the reference value of 1000 ppm is not included in ASHRAE 62.1-1999 [13]. Additionally, it was emphasized that the +700 ppm difference value, which is stated as a limit value that people find 80% acceptable in terms of odor quality in ASHRAE 62.1-2016 "Informative Annex-D", is not a limit value, and therefore this information is not included in ASHRAE 62.1-2019 [13].

Considering the mathematical model given in ASHRAE 62.1-2016 "Informative Annex-D", a CO_2 concentration difference limit value varies with each of the ventilation rates per person recommended for the VRP in the ASHRAE 62.1 standards (7.5 l/s of ventilation rate corresponds to the parameters of 1.2 MET of metabolic rate, 0.31 l/s-person of bio-CO_2 emission, +700 ppm difference value for ~350 ppm outdoor CO_2 concentration). However, the +difference values obtained with this mathematical model depend on metabolic rate and anthropometric values as well as ventilation rate. In this case, the results could create a chaotic structure in terms of health safety. Using 1000 ppm of upper limit value instead of 700 ppm above the outdoor CO_2 concentration results in more effective ventilation and thus prevention of indoor accumulation of the various air pollutants and their concentration reaching relatively high levels in indoor air.

Within the scope of these discussions, 1000 ppm of CO_2 concentration value is assumed as an upper limit value to define the ventilation rate in sleeping spaces by IAQP in this study.

2.2 CO_2 emissions for the design of ventilation systems in sleeping spaces

The main source of CO_2 emissions in sleeping spaces (residential bedrooms, dormitories, hotel rooms, etc.) is emissions from sleeping people. The CO_2 emission of people depends on their metabolic rate (MR), anthropometric characteristics, respiratory coefficients depending on their dietary habits (carbohydrate, fat, protein), temperature and altitude. Therefore, it would be appropriate to take the maximum possible values of the CO_2 emissions of the people who sleep in these spaces to design a system providing adequate service within the physical life of the space. Here, the 95-percentile values were used for the weight (W) and height (H) of the men and women staying in the sleeping spaces [14]. The respiratory coefficient (RC) which is defined by average dietary habits and the MR are assumed as 0.83 and 1 MET, respectively. Since it is inversely proportional to temperature, the lowest value of temperature (18 °C) was used as the design temperature of the sleeping space. Tab. 1 presents the calculated CO_2 emission values by the model of Kalema and Viot [15] as a function of these parameters. The values given in Tab. 1 are used to calculate the total CO_2 emissions for different sleeping spaces, as indicated in Tab. 2.

Tab. 1 - CO_2 emission rates in terms of the safe design of ventilation systems in sleeping spaces.

	95-percentile		**MR (MET)**	**RC**	**CO_2 emission rate (g/s)**
	W (kg)	**H (m)**			
Woman	95.3	1.7985	1	0.83	0.0093
Man	96.8	1.7740			0.0095

Tab. 2 – The total emission values in sleeping spaces.

Sleeping Space	**CO_2 emission rate (g/s)**
Residential bedroom - Double	0.0188 (=0.0095+0.0093)
Residential bedroom - Single	0.0095
Dormitory rooms	Number of beds × 0.0095
Hotel room - Single	0.0095
Hotel room - Double	0.0188 (=0.0095+0.0093)
Hotel room - Multiple	Number of beds × 0.0095

2.3 Sleeping spaces

Within the scope of indoor air quality, the pollutant concentration is related to the volume of the sleeping space which depends on the countries' architectural codes, traditions, socio-economic status. Tab. 3 presents the minimum sizes of bedrooms and dormitory rooms. Most of the dimensions in the Tab. 3 indicate the general practice instead of the minimum requirement. Although the ceiling height is also an important parameter for ventilation, some of the sources only gave a value of area per person. Tab. 3 shows that the minimum volume per person in dormitory rooms all over the world is 10 m^3/person. The highest value is calculated as 21.6 m^3/person corresponding to the area of 9 m^2/person given by TS 11498 for single-person dormitories [16–18].

Tab. 3 – The total emission values in sleeping spaces.

Country	**Area (m^2/person) / Volume (m^3/person)**	**Dormitories**	**Ref.**
Turkey	**9** / 21.6	Single-room Min. Ceiling height = 2.4 m	[16–18]
	5 / 12.5	Multiple-room Min. Ceiling height = 2.4 m	
	8 / 19.2	Single-room Min. Ceiling height = 2.4 m	[19]
	4.17 / 10	Multiple-room (3-6 people) Min. Ceiling height = 2.4 m Min. Volume = 10 m^3/person	
	6 / 13.2	Single-room Min. Ceiling height = 2.2 m	
	4.55 / 10	Multiple-room	

		Min. Ceiling height = 2.2 m Min. Volume = 10 m^3/person	
	2.98 / -	8-people Room floor area = 23.8 m^2	[18]
USA	12.1 / -		[20]
	8.6 / -		[21]
	4.6 / -		[22]
China	4.5 / 12.5	4-people Ceiling height = 2.8 m	[23]
	3.73 / 10	6-people Ceiling height = 2.8 m	
Australia	10-12 / -		[24]
	20 / -		

Country	**Area (m^2/person) / Volume (m^3/person)**	**Bedrooms**	**Ref.**
Turkey	**9** / 21.6 – 2.34	Ceiling height = 2.4 – 2.6 m	[25]
UK	8 / -	Single-room	[26]
	6 / -	Double-room	
	7.5 / -	Single-room Min. ceiling height = 2.3 m	[27]
	5.75 / -	Double-room Min. ceiling height = 2.3 m	
	16-25 / -		[28]

3. CO_2 Concentration in Sleeping Environments

In many countries, the variation of the CO_2 concentration in sleeping spaces (bedrooms) in buildings with different purposes (residential, residence, dormitory, etc.) has been measured in different seasons and the effect of this variation on sleep quality has been tried to be determined by objective or subjective methods. Most of these studies show that the CO_2 concentration increases during sleep and in some cases reaches serious values (7500 ppm) for the absence of ventilation in the sleeping spaces and the conditions of the open window and/or door during the measurements.

3.1 CO_2 balance in sleeping environments

The CO_2 balance in any sleeping space (bedroom, dormitory, etc.) is expressed considering the amount of CO_2 produced in the volume, entering and leaving the volume if there is no other CO_2 source in the volume (Fig. 1). Components entering the sleeping space are the CO_2 rates emissions of sleeping people in the volume, entering by ventilation and infiltration. The outputs are the CO_2 rates which release through infiltration and ventilation. Considering these values, the CO_2 rate in the sleeping volume is given in equation (1). The parameters in equation (1) are explained in Tab. 4.

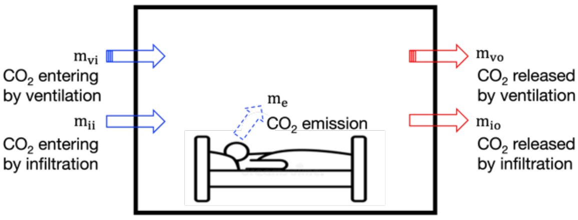

Fig. 1 – CO_2 balance in the sleeping space

$$\frac{dm(t)}{dt} = M_e + m_{vi} + m_{ii} - m_{vo} - m_{io} \qquad (1)$$

Tab. 4 – Parameters of the CO_2 balance in the sleeping space

$m(t) = VC(t)$	The amount of CO_2 in the sleeping volume (mg)
V	Sleeping volume (m^3)
t	Time (h)
$M_e = N_b m_e$	Time-dependent CO_2 concentration in the sleeping volume (mg/m^3)
N_b	Number of people in the sleeping space (-)
m_e	CO_2 emission of the sleeping person (mg/s)
$m_{ii} = Q_i \mathrm{C_d}$	CO_2 concentration entering the volume by infiltration (mg/s)
Q_i	Infiltration rate (m^3/s)
$\mathrm{C_d}$	Outdoor CO_2 concentration (mg/m^3)
$m_{io} = Q_i C(t)$	CO_2 concentration released by infiltration (mg/s)
$m_{vi} = Q_v \mathrm{C_d}$	CO_2 concentration entering the volume by ventilation (mg/s)
Q_v	Ventilation rate (m^3/s)
$m_{vo} = Q_v C(t)$	CO_2 concentration released by ventilation (mg/s)

By substituting the terms given in equation (1) and Tab. 4, the time-dependent change in CO_2 concentration is:

$$\frac{dC(t)}{dt} = \frac{N_b m_e}{V} + \frac{Q_v + Q_i}{V}[\mathrm{C_d} - C(t)] \qquad (2)$$

3.2 Minimum sleeping volume without ventilation and infiltration

Since the highest CO_2 concentration value should be in the case where ventilation and infiltration are not taken into account, equation (2) is written as:

$$\frac{dC(t)}{dt} = \frac{N_b m_e}{V} \quad (3)$$

When the boundary conditions for $C(0)=C_d$ at $t=0$ and $C=C(t)$ at $t=t$ are employed in equation (3), the equation required to define the maximum CO_2 concentration value is obtained as

$$C_{\max}(t) = \mathrm{C_d} + \frac{N_b m_e}{V} t \quad (4)$$

The maximum CO_2 concentration in equation (4) is rewritten in ppm unit instead of mg/m^3 by equation (5).

$$C_{\max}(t) = \mathrm{C_d} + \frac{N_b m_e}{V} t \left[\left(\frac{\mathrm{V_m}}{\mathrm{M_{CO_2}}} \right) \left(\frac{273+T}{273} \right) \left(\frac{1013}{P} \right) \right] \quad (5)$$

Here, the molecular weight of CO_2 is M_{CO2} = 44.01 g/mol, and the molar volume of air is V_m = 24.47 l/mol. T and P are the temperature [°C] and pressure [hPa] of the air.

The required time to reach the 1000 ppm value defined as the upper limit for any volume of CO_2 concentration is expressed by

$$t = \frac{(1000 - \mathrm{C_d})V}{\left(\frac{\mathrm{V_m}}{\mathrm{M_{CO_2}}} \right) M_e \phi_T \phi_P} \quad (6)$$

where $\phi_T = \frac{273+T}{273}$ and $\phi_P = \frac{1013}{P}$.

Additionally, the volume in which ventilation is not required during sleep is given as a function of time in equation (7).

$$V_f = \frac{N_b m_e}{C(t) - \mathrm{C_d}} t \quad (7)$$

The National Sleep Foundation (NSF) recommends between 7 and 9 hours of sleep for adults aged 18-25 years [29,30]. The V_f was found to be 311.4 m^3/person if the CO_2 concentration in the room did not exceed the C(t)=1000 ppm value after 9 hours of sleep. The variation of this value according to sleep duration is presented in Fig. 2. The V_f values obtained for the 7-9 hours of sleeping period recommended for an adult human are inaccessible for conventional sleeping spaces, therefore it should be noted as a necessity to ventilate any sleeping space smaller than the V_f value.

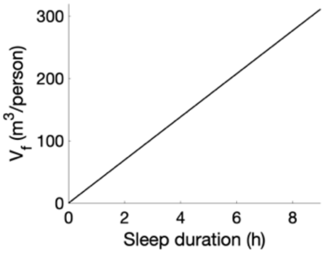

Fig. 2 – The variation of smallest sleeping space volume which does not require ventilation with the sleeping duration (($C(t))_{max}$=1000 ppm).

Fig. 3 shows the time-dependent variation of CO_2 concentration for different volumes per person. Since the sleeping volume changes in the range of 10-21.6 m^3/person in dormitories (Tab. 3), the minimum value of the sleeping volume is defined as 10 m^3/person in this study. As expected, the increasing slope of the CO_2 concentration decreases for higher volumes, and the time to reach the 1000 ppm of CO_2 concentration also increases as the volume increases.

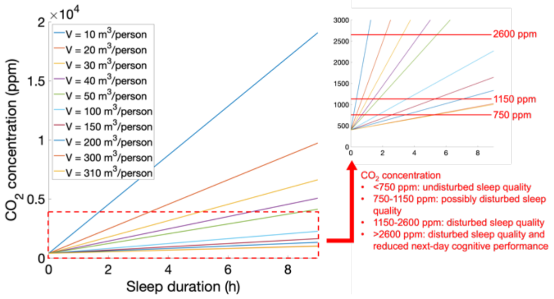

Fig. 3 – The time-dependent variation of CO_2 concentration for different volumes and the typical effects of CO_2 concentration on people [7].

3.3 Safe ventilation rates for sleeping environments

In order not to affect sleep quality in any sleeping place, the differential solution of the equation (2) is required to define CO_2 concentration in the sleeping space sourced by sleeping people's emissions. The equation (2) is rearranged by neglecting the infiltration as

$$\frac{dC(t)}{dt} = \frac{N_b m_e}{V} + \frac{Q_v}{V} [\mathrm{C_d} - C(t)] \quad (8)$$

Equation (8) is a first-order linear ordinary differential equation, and its solution is given by

$$C(t) = c_1 e^{-\frac{tQ_v}{V}} + \mathrm{C_d} + \frac{N_b m_e}{Q_v} \quad (9)$$

When the boundary conditions for $C(0)=C_d$ at $t=0$ and $C=C(t)$ at $t=t$ are employed in equation (9), the equation (9) is re-written as

$$C(t) = \mathrm{C_d} + \frac{N_b m_e}{Q_v} \left(1 - e^{-\frac{tQ_v}{V}} \right) \quad (10)$$

The equation (10) allows calculating the safe ventilation rate (Q_v) in the sleeping space for the volume per person by applying the upper limit value of ($C(t))_{max}$=1000 ppm. The variation of the safe ventilation rate with the volume per person is presented in Fig. 4 for 9 hours of sleep duration, and the ventilation rate decreases as the volume per person increases. However, this reduction in the ventilation rate, which is obtained from the volume of 100 m^3/person, has no implications in real-life conditions. Because the sleeping volume ranges between 10 and 25 m³/person in real life, and the ventilation flow rate remains almost constant (9.60 l/s-person) in this volume range.

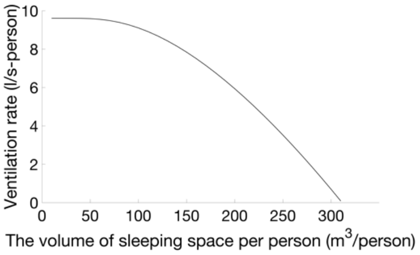

Fig. 4 – The change of safe ventilation rate with sleeping volume per person (C(9 h)=1000 ppm).

Although the need for ventilation decreases as the volume of the sleeping area increases, Fig. 5 shows the necessity of ventilation for the volumes of 10-30 m³/person from the first hour of the sleep period in case of the CO₂ concentration to not exceed the upper limit of 1000 ppm.

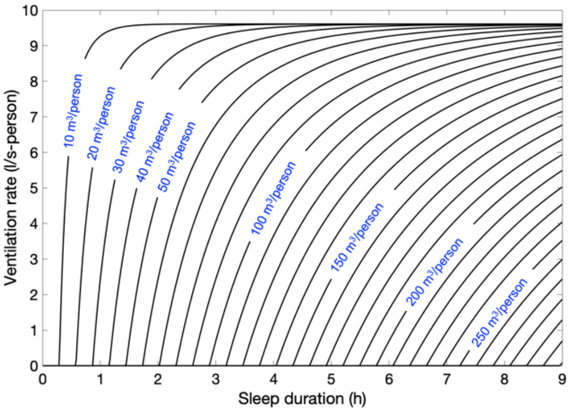

Fig. 5 – The variation of the ventilation rate with sleep duration for different sleeping volumes per person.

3.4 Comparison of IAQP and VRP in determining the safe flow rate to be used in the design of ventilation

Sekhar et al. [7] investigated the ventilation rate standards of 15 countries, from the USA to China for bedrooms. In the table they presented to compile all standards of different countries, specific ventilation design flow rates for bedrooms are defined in the European Union and in five countries, and in some of 9 countries, separate values are given for kitchens and bathrooms, and the same design flow rates are recommended for all remaining residential spaces. This study aims to investigate the effect of these recommended flow rates on the CO₂ concentration in the sleeping space and the difference from the values recommended by IAQP. The proposed design flow rates within the scope of VRP and the models given in ASHRAE Standard 62.1 and 62.2 to be used in the comparison of IAQP and VRP are presented in Tab. 5. In ASHRAE Standard 62.2, A_{floor} is the residential floor area (m²) and N_{br} is the number of bedrooms (N_{br}>1) in the residential. N_b represents the number of people in the bedroom and A_b represents the area of the bedroom (m²) in ASHRAE Standard 62.1.

Tab. 5 – The ventilation rates obtained from [7] and proposed by VRP.

Country	Flow rates
ASHRAE 62.2	$Q_{total} = 0.15A_{floor} + 3.5(N_{br} + 1)$ (l/s)
Belgium	1 (l/s m²)
Sweden	0.35 (l/s m²)
Netherlands	0.90 (l/s m²)
UK	0.30 (l/s m²)
Country	**Flow rates**
ASHRAE 62.1	$Q_b = 2.5N_b + 0.3A_b$ (l/s)
CEN – Category 1	2.90 (h^{-1})
CEN – Category 2	2.00 (h^{-1})
CEN – Category 3	1.20 (h^{-1})
Norway	7.20 (l/s person)
Denmark	0.30 (l/s m²)
China	1.00 (h^{-1})
India	3.00 (h^{-1})

The effect of the ventilation rates for the bedroom determined by VRP on the CO₂ concentration in the sleeping space is shown in Fig. 6 for 9 hours of sleep duration. The volume of the sleeping space was calculated considering the minimum ceiling height as 2.4 m corresponding to the minimum volume value in the standard TS 11498 [16–18]. It is observed that the ventilation rates calculated by VRP for the bedroom decrease the CO₂ concentration below 1000 ppm as the volume of sleeping space per person increases. In addition, the CO₂ concentration for 10 m³/person, which is the lowest volume of sleeping space in the standards, is above 1000 ppm for the values recommended in all standards.

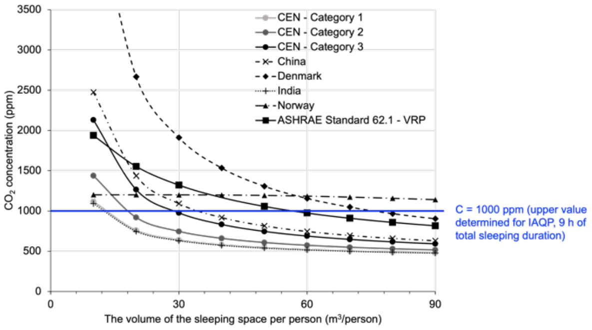

Fig. 6 – The effect of ventilation rates calculated by VRP on the CO₂ concentration for different volumes of sleeping space per person.

Fig. 7 shows the variation of ventilation rates with the volume per person after 9 hours of the sleep. In the calculations, the minimum residential floor area is assumed as 28.5 m^2 and the lowest area that the sleeping space was defined as 9 m^2 [25]. The ventilation flow rates for the whole dwelling were calculated as the ratio of the sleeping space area to the total floor area of the dwelling (0.32).

Although the flow rates calculated by VRP for low volumes of sleeping spaces are insufficient, the flow rates calculated by VRP exceed the safe ventilation flow rate calculated with IAQP for higher volumes. Intersecting the ventilation rates defined by VRP and IAQP in a narrow area limits the utilization of the VRP. In the cases where VRP values are insufficient or too high, it is recommended to demand-controlled ventilation by IAQP considering the system design.

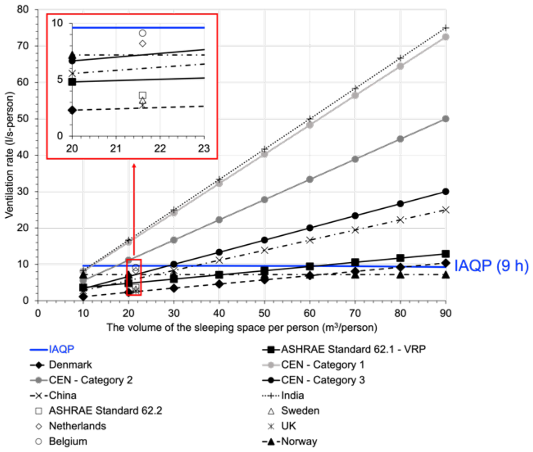

Fig. 7 – The ventilation rates calculated by VRP and IAQP for different volumes of sleeping space per person.

4. Conclusions

Within the scope of different approaches for the required ventilation rate for sleeping spaces, the applicability of these procedures proposed by ASHRAE standards has been evaluated in this study. In order to ensure that the CO_2 concentration in the sleeping space is below an upper limit value (1000 ppm), the proposed IAQP is explained specifically for bedrooms and dormitory rooms, and an approach that designers can use directly for implementation is presented. Ventilation rate diagrams have been proposed for different sleeping space volumes and sleep times by assuming the upper limit value of the CO_2 concentration as 1000 ppm. In addition, the model in which the upper limit value of CO_2 concentration, space volume, and sleep duration are variable has been developed to calculate a reliable ventilation flow.

While the mathematical model given in ASHRAE 62.1-2016 “Informative Annex-D” indicates the appropriateness of 7.5 l/s person ventilation flow per person to reach a safe 1000 ppm value, the safe ventilation rate recommended by the IAQP method, which accepts the upper limit of the CO_2 concentration as 1000 ppm, has been calculated as 9.60 l/s person, up to a sleeping area volume of approximately 70 m^3/person.

The proposed design criteria for sleeping spaces within the scope of this study can be summarized as follows:

- Upper limit value of CO_2 concentration: 1000 ppm
- The procedure for calculating ventilation rate should be IAQP
- The number of people in the bedroom:
  - Main bedroom: 2
  - Other bedrooms: 1 or 2 depending on the size of the room
  - Dormitory rooms: possible maximum number of people
- Maximum CO_2 emission per sleeping person: 9.5 mg/s
- System control: Demand-controlled ventilation

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Data Statement

The datasets generated during and/or analysed during the current study are not publicly available because they are to be the subject of further studies but will be available from the corresponding author on reasonable request.