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Future trends in ambient air pollution and climate in Germany – Implications for the indoor environment

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ABSTRACT

Until now, only a few studies have addressed the effects of climatic changes on the microclimate in indoor areas. Although global warming presents a problem worldwide, the extent of climate change varies greatly according to region. In this work, climatic changes and their influences on indoor areas in Germany are discussed. Air temperatures, quantity of summer days, sunshine hours and precipitation were extracted from the databases of the German National Meteorological Service. In addition, data on ozone, nitrogen dioxide, PM_{10} and $PM_{2.5}$ were collected from eight monitoring stations in different urban locations. With the aid of single-box models, the effects of these parameters on indoor air were assessed. Furthermore, temperature and humidity measurements were performed in a thermally insulated house during a 14-day period of fine weather. Finally, trends for the indoor temperature through to the year 2040 were estimated.

With the current thermal insulation standards, effects of climate change are associated with an increase in indoor temperatures, which can easily lead to overheating in the event of heatwaves. Nitrogen oxides and ozone are subject to strong daily and seasonal fluctuations. Particle concentrations are declining in Germany. The entry of outdoor air contaminants into the indoor area is essentially dependent on the air exchange. On the whole, measurements and simulations show that climate change substantially influences the indoor air quality. This will have consequences in the medium term as regards measures for thermal insulation and for regulation of the air exchange in private and public buildings.

1. Introduction

After many years of intense discussions, human-induced climate change is now classed as having been accepted both scientifically and socially [1]. Greenhouse gases such as carbon dioxide, nitrous oxide and methane are primarily responsible for global warming. On a regional basis, however, climatic changes will have substantially differing effects, due to the exposure to air pollutants of anthropogenic origin, such as particles (PM_{10} and $PM_{2.5}$) and NO_x [2]. Nitrogen dioxide (NO_2), in turn, forms tropospheric ozone through diverse secondary reactions with the presence of sunlight [2]. Particularly in urban areas with a high population and traffic density, the effects of air pollution and photochemical smog are perceptible [3]. For many years, outdoor air was considered to be “fresh air”; increased concentrations of air pollutants or higher levels of moisture content in indoor spaces were minimized through ventilation. This principle still applies in areas with a high outdoor air quality, but not in general. In situations where the EU-wide limit and guideline values for PM_{10}, $PM_{2.5}$, NO_2 and ozone are exceeded, intensive ventilation is not recommended; this also applies during heatwaves and periods of high humidity.

Germany lies between the 47th and the 55th latitude and therefore in a temperate climatic zone. Its surface area of 357,376 km^2 is inhabited by nearly 83 million people, which corresponds to a population density of 232 per km^2. Since as early as the 1980s, a dense network of air-quality measurement stations has been in existence, so that alterations in air quality are documented on an hourly basis. Temperature data have been available since the 19th century. The possible effects of climate change have been intensively investigated for the geographic and population-structural conditions which prevail in Germany. In a progress report issued by Brasseur et al. [4], the published scientific information is compiled and considered as a whole. The report addresses in detail the physical, ecological and socio-economic aspects. It is therefore surprising that the subject of air quality is mentioned only extremely briefly; statements concerning the possible consequences for the prevailing indoor microclimate are lacking completely. In fact, only a few studies have been carried out to date which deal with the

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influence of climate change and the temporal trend of outdoor air pollution on indoor air quality [5]. Jiang et al. [6] recently studied the impact of low emission zones (LEZ) on air pollution levels. They evaluated air pollutant data for the time period 2002–2012 at different monitoring stations throughout Germany. Effects on indoor air quality were not taken into consideration. The topic is, however, of enormous technical and social importance, as it not only raises questions concerning building regulations and standards [7], retrofitting [8], ventilation concepts [9] and smart technologies [10] but is also of relevance to health [11].

In this publication, the temporal trends of outdoor temperature, precipitation and global radiation are analyzed for Germany. By means of available climate models and literature data, assumptions are made concerning future developments. These data are in turn applied in one-dimensional heat and mass-transfer models. On the basis of the current standards for thermal insulation, the models are then used to compile forecasts for the indoor climate during the summer months up to 2040. Furthermore, the temperature profile in a thermally insulated house was measured over a 14-day fine-weather period and compared with the model.

An analysis of the trends for ozone (O_3), nitrogen dioxide (NO_2) and particles (PM_{10} and $PM_{2.5}$) is also performed. Particular emphasis is placed on urban background stations in the cities of Hamburg, Berlin, Hanover, Leipzig, Cologne, Frankfurt, Stuttgart and Munich. These cities are evenly distributed across Germany (see Fig. 1). More specifically, the indoor/outdoor concentration ratios are calculated by use of single-box models as a function of air exchange rates, penetration factors and deposition factors. The results allow an estimation of current and future influences of the outdoor climate on the microclimate in indoor areas. Accordingly, no indoor sources were taken into account. From the results, possible consequences can be derived for new and existing buildings as well as for future living behavior.

2. Methods

Eight German cities, each with a population > 500,000 and representing different geographic regions, were selected for this study (see Fig. 1). For each city, one air monitoring station representing urban background concentrations was chosen. Climatic data were obtained from the German National Meteorological Service (Deutscher Wetterdienst, DWD), Norddeutscher Klimamonitor and German Federal Environment Agency (Umweltbundesamt). Details of accessed databases are provided in Table SI-1. The outdoor climate data for the calculation of the indoor temperature scenarios up to 2040 are based on a) predicted values from a regional model (REMO) [12] with a spatial resolution of 0.11° (approx. 12 km) and b) the pessimistic RCP 8.5 scenario (Representative Concentration Pathways). This scenario is based on carbon dioxide concentrations of 1370 ppm and a radiative forcing value of 8.5 W/m^2 until 2100 (IPCC, 2013). For the simulations from 2011 to 2040, temperature and relative humidity were used as daily means. WUFI® pro 6 (Fraunhofer Institute for Building Physics (IBP), Holzkirchen, Germany) software was used for the hygrothermal analysis. OriginPro 2018G (OriginLab Corp.) was applied for the statistical treatment of the data.

From 25.06.2018 to 10.07.2018, there was a period of fine weather in Northern Germany with no noteworthy precipitation, daily maximum temperatures of between 25 °C and 30 °C, and strong global radiation. During this period, the room temperature and the relative humidity in the living area (75 m^2, ground level) of a single-family house in Braunschweig (latN: 52° 17′, longE: 10° 27′, approx. 60 km west of Hanover) were continuously recorded as 1 h means using a data logger (Rotronic Messgeräte GmbH). The house was built in 1963, was energetically completely refurbished in 2006 in accordance with the specifications of the German Energy Saving Ordinance (EnEV) applicable at that time and is manually ventilated. During the measurement phase, the house was inhabited by 3–4 persons. Windows and doors were opened during the morning and evening hours and closed during the day, with the exception of necessary entries and exits. Shading fixtures were used intensively. The climatic parameters of the surrounding outdoor atmosphere were retrieved from two nearby measurement stations (DENI011 and DENI075). Furthermore, using the available outdoor air data (1 h mean values), a simulation calculation based on heat and moisture transport as well as global radiation was performed.

3. Outdoor and indoor climate

3.1. Climatic trends in Germany

When considering the climate, a strict distinction must be made between global and regional phenomena. For regional considerations, a spatially dense network is necessary, which is available in Germany

City	Station Code (urban background)
Hamburg	DEHH008
Berlin	DEBE10
Hanover	DENI054
Leipzig	DESN059
Cologne	DENW053
Frankfurt/Main	DEHE008
Stuttgart	DEBW013
Munich	DEBY039

Fig. 1. Locations of the selected German cities and monitoring stations with station codes (urban background). More details about the monitoring stations are provided in Tables SI–1.

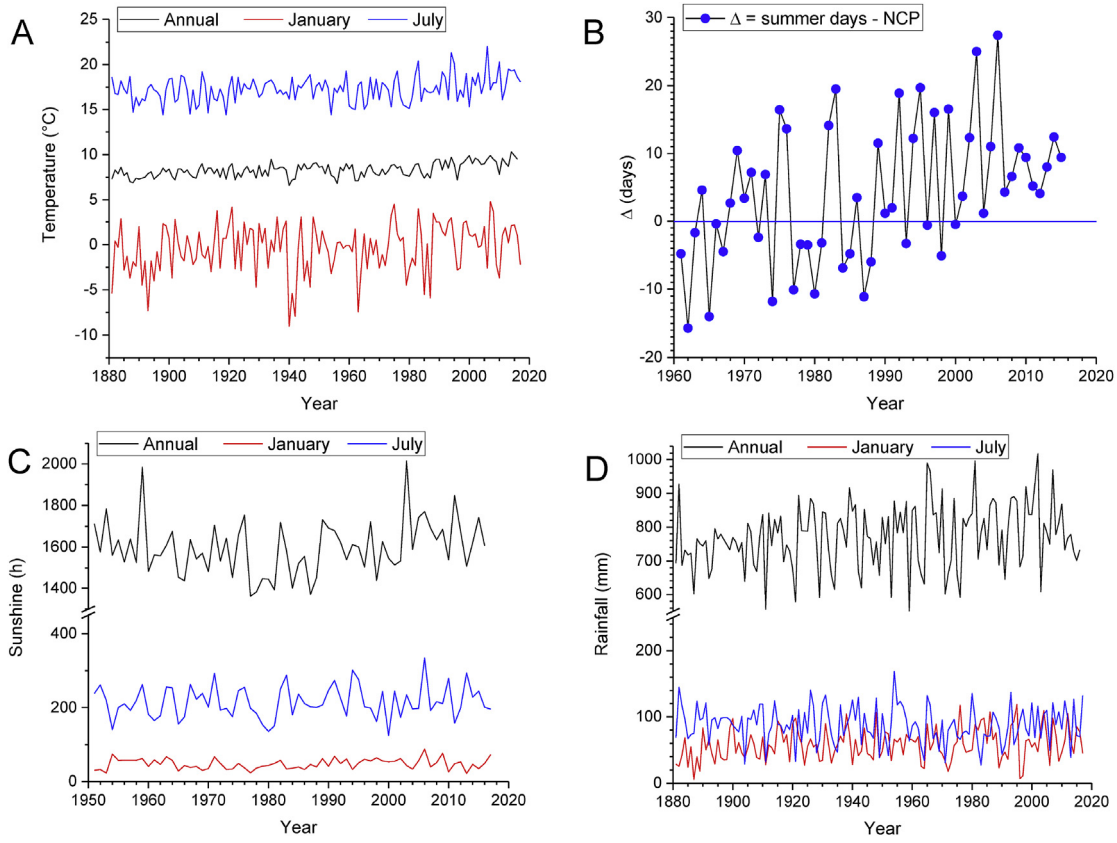

Fig. 2. Trends in Germany for temperature, rainfall, sunshine hours since 1881 (data from Deutscher Wetterdienst, DWD) and summer days since 1951 (data from Norddeutscher Klimamonitor, data set E-OSB 14.0 with a total of 15 stations between List/Sylt and Brocken/Harz). More details about the databases are provided in Table SI-1. NCP is the so-called "normal climate period" and represents the mean of the 1961–1990 data.

with several hundred stations [13]. Starting from 1880, recorded temperatures and precipitation quantities were evaluated through to 2014 [14]. For the temperature, significant increases of 1.3 °C in the annual mean and 1.2 °C in the summer were revealed. The absolute temperatures in Germany for the period from 1880 to 2016 are shown in Fig. 2A as the annual mean and monthly mean (January and July).

The number of summer days (days with temperatures of at least 25 °C) also shows a significant positive trend. In this data set, the respective measured value is correlated against the mean value for the normal climate period (NCP) between 1961 and 1990. Fig. 2B shows the time series for Northern Germany (1961–2015), a region with statistically fewer warm days than the national average. Compared to the NCP between 1961 and 1990, a change of +2.75 summer days per decade (areal average over land areas) was determined. The quantity of sunshine hours has remained relatively constant in Germany since 1951. The long-term annual mean is 1597 h; for the years 1959 and 2003, the value was significantly increased (see Fig. 2C). A major problem is presented by the anticipated accumulation of heat events [15]. During heatwaves in Europe in the years 2003 and 2010, more than 125,000 (70,000 and 55,000 respectively) additional deaths were recorded [16]. For Germany alone, Robine et al. [17] mention 7295 excess deaths. Schär and Jendritzky [18] calculate 900–1300 extra deaths per 10.7 million people for August 2003 in Germany. The increased mortality was accompanied by cases of cardiovascular diseases, circulatory disorders and dehydration. The German Government has therefore compiled recommendations for action concerning the generation of heat action plans for the protection of human health [19].

The annual precipitation quantities in Germany have increased by approx. 10% since 1881. This is due in particular to more frequent rainfall in the winter months. The total precipitation quantity increased by 81 mm in the annual mean and 47 mm in the winter. For the summer months, however, no trend can be observed (see Fig. 2D). Irrespective of the season, more torrential rainfall events and flooding must be expected in Germany. All things considered, the increase in precipitation cannot fully compensate for the temperature-induced intensified evaporation of the water. Assuming the RCP 8.5 scenario, the regional climate model STAR projects a negative water balance for Germany in the long term (see Potsdam Institute for Climate Impact Research, www.klimafolgenonline.com).

3.2. Measurement simulation of the temperature in buildings

Temperature and humidity are important factors of indoor air quality in terms of human comfort and healthiness. Generally, a relative humidity between 30% and 70% provides comfortable conditions in a residential building [20]. When the relative humidity exceeds 70%, people start feeling uncomfortable and the risk of mold growth increases. Vellei et al. [21] state that optimal conditions occur in the range between 40% and 60% relative humidity. Fluctuation of temperature and humidity are linked to certain conditions. The indoor microclimate is affected on the one hand by general building attributes such as location, geometry and material used in the building envelope, and on the other hand by interior environment factors such as ventilation rate, air tightness, air exchange rate, furnishings, number of occupants and many others.

The results of temperature and humidity measurements in the living area of the single-family house are shown in Fig. 3A. Despite the good thermal insulation ($U < 0.35\,W/(m^2 \cdot K)$) and intensive shading measures, a continuous increase in temperature from approx. 20 °C to 24.2 °C can be observed within only a few days from the onset of the fine-weather period, despite windows and doors being closed during the day whenever possible. The relative humidity in the indoor area

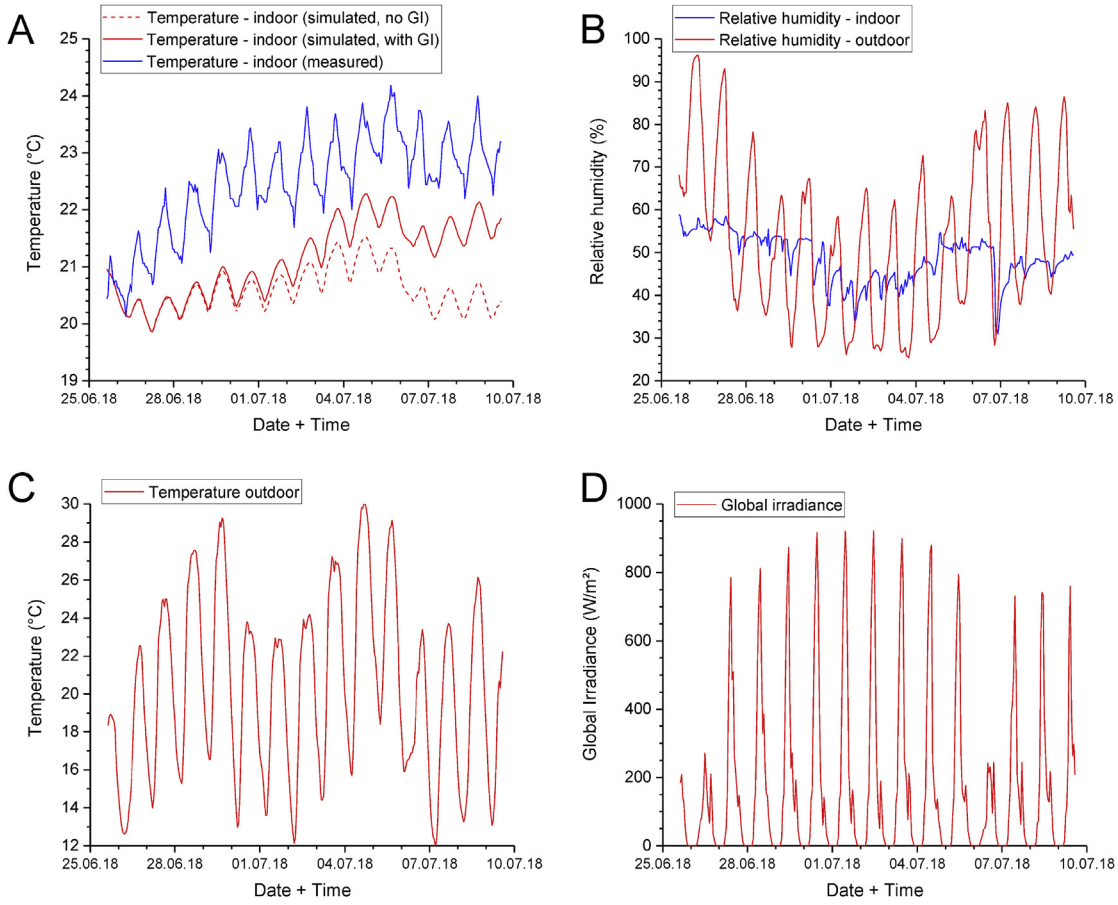

Fig. 3. Measurement data for the indoor air of a residential building in Braunschweig and the surrounding outside air during the period 25.06.2018 to 10.07.2018 (1 h mean, respectively), as well as results of simulations. 3 A: temporal progression of the indoor temperature and simulation calculations with and without consideration of global radiation; 3 B: temporal progression of the relative humidity indoors and outdoors; 3C: temporal progression of the outdoor temperature; 3D: temporal progression of the global radiation. The outdoor air data were retrieved on 11.07.2018 from the measuring station Braunschweig BGSW (DENI011).

fluctuated between 30% and 60% (Fig. 3B). Significantly stronger fluctuations in temperature and humidity were observed in the outdoor areas (see Fig. 3B and C). The temporal profile of the global radiation is shown in Fig. 3D. A temperature of 24 °C is at the upper end of the comfort zone. The WHO is currently developing housing and health guidelines, taking into account the health-related data of people living in dwellings with temperatures above or below 24 °C [22]. During the summer months in Germany, heatwaves with outdoor temperatures in excess of 30 °C occur over periods of days to weeks [23]. In such cases, it is unlikely that a thermally insulated house with manual ventilation will provide protection against overheating. After the end of the measurement phase a heat wave including tropical nights (days with temperatures not below 20 °C) occured in Braunschweig. On 26.07.2018 the recorded maximum temperatures were 33.3 °C outdoors and 26.4 °C in the living area of the single-family house, respectively.

Fig. 3A additionally shows the results of one-dimensional simulation calculations using WUFI® pro 6 ($U = 0.35\ W/(m^2 \cdot K)$, air exchange rate $\lambda = 1.0\ 1/h$) with and without the influence of global radiation and without taking into account the activities of the occupants. The selected air exchange rate of 1.0 1/h takes frequent occupant activities into account. Therefore, λ is higher than the average value of 0.5 1/h, which results from a literature search [24] and which is standard for the conversion of emission rates to concentrations in the European Reference Room [25]. However, the constant value for λ does not consider sudden changes in the air exchange through sporadic occupant activities and atmospheric pressure differences. It is thereby accepted that the simulation is only able to depict the trend, not the absolute temperatures.

In further simulation calculations, the possible regional effects of climate change on indoor temperatures up to the year 2040 were investigated using the example of the eight selected cities. Solely the non-heating period was thereby considered. Despite the fact that the temperature distribution in the indoor area is influenced by numerous factors, a one-dimensional model for the heat and moisture transport was applied here, which calculates the indoor temperature in close proximity to the wall (distance of up to 1 m). Because the effect of outdoor temperature and humidity on indoor conditions forms the focus, other effective factors were not considered in the simulation.

Properties of external walls have been extracted from TABULA [26]. In this project, a set of models is described for residential buildings in Germany, with their energy-related properties in certain construction periods and specific building size. External walls of buildings constructed during the periods 1979–1983 and 1995–2001 have been taken into consideration as the first and third epochs in which the Energy Saving Ordinance (EnEV) in Germany applied. Heat transfer coefficients (U-values) of external walls constructed between 1979 and 1983 were around $0.8\ W/(m^2 \cdot K)$; this value decreased to $0.5\ W/(m^2 \cdot K)$ in the 1995–2001 period.

Climate models are tainted with major uncertainties, especially as regards regional modeling. The model selected here is one-dimensional and solely considers the heat and moisture transition through an external wall on the basis of the RCP 8.5 scenario (high greenhouse gases emission pathway) with a constant air exchange $\lambda = 0.5\ 1/h$ and $U = 0.5\ W\ (m^2\ K)$ (high insulation properties). For each city and each decade between 2011 and 2040, the number of days with an indoor temperature of more than 18 °C (daily mean) was calculated. This

temperature lies at the lower limit of the thermal comfort range in indoor areas [27]. As the model only considers daily mean values, the absolute temperatures are underestimated (see Figure SI-1). Nevertheless, important trends for the indoor area can be recognized. The model clearly identifies strong regional differences, but also shows that the development of the outdoor climate under the RCP 8.5 scenario will have an impact on the microclimate in indoor areas in the future (see Figure SI-2). Furthermore, the model predicts a significant increase in relative humidity (figure not shown).

4. Pollutants

4.1. Ozone

The World Health Organization [28] determined a health-related guideline value of 100 µg/m^3 as the 8 h mean value for ozone. In the European Union, a concentration of 120 µg/m^3 (8 h mean value) is defined as the reference value and a concentration of 240 µg/m^3 (1 h mean value) as the alarm threshold. The reference value may be exceeded on a maximum of 25 days per year, averaged over three years. A specific guideline value for indoor air was defined by Health Canada [29] at 40 µg/m^3 (as 8 h mean).

The determination of human exposure to ozone is difficult, as the ozone concentration is subject to significant daily fluctuations in dependence on the precursor substances and solar radiation. A simplified reaction profile is shown in Fig. 4. Ultimately, the photochemical reaction of NO_2 always leads to ozone formation. The NO_2 concentration, however, depends not only on the volume of traffic but also on further precursor substances, such as hydrocarbons [30] and NO. For Central Europe, Derwent and Hjellbrekke [31] were able to verify a clear dependence of ozone formation on the degree of latitude.

Measuring stations are differentiated into traffic/industrial, urban background and rural background. According to evaluations by the German Federal Environment Agency, the annual mean values for ozone rose slightly until the turn of the millennium and have remained stable since then. The annual mean values for the urban background have lain within a range from 40 µg/m^3 to 50 µg/m^3 since 2001. For the evaluation of eight large German cities shown in Fig. 5A, the span is 29 µg/m^3 to 53 µg/m^3 (see Tables SI–2). The annual mean values for the rural background are consistently higher and lie within a range of 55 µg/m^3 to 65 µg/m^3 (see Figure SI-3). The number of days on which

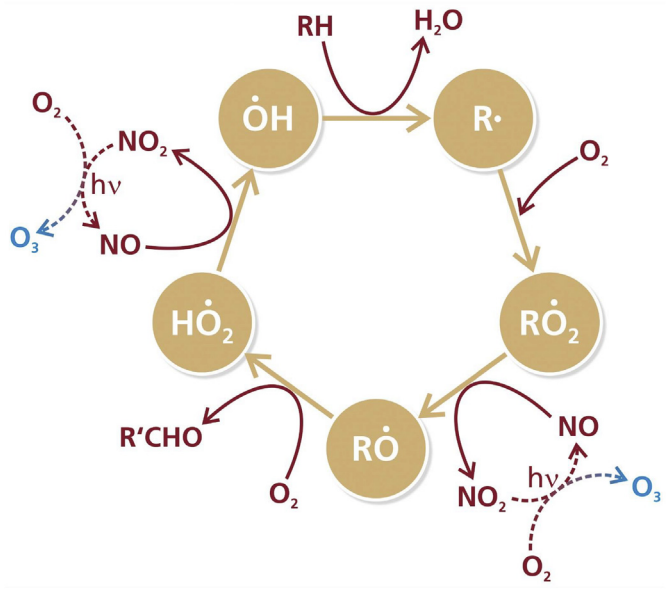

Fig. 4. Catalytic cycle of atmospheric ozone formation in polluted air. The figure was adapted from Hites et al. [68], page 66.

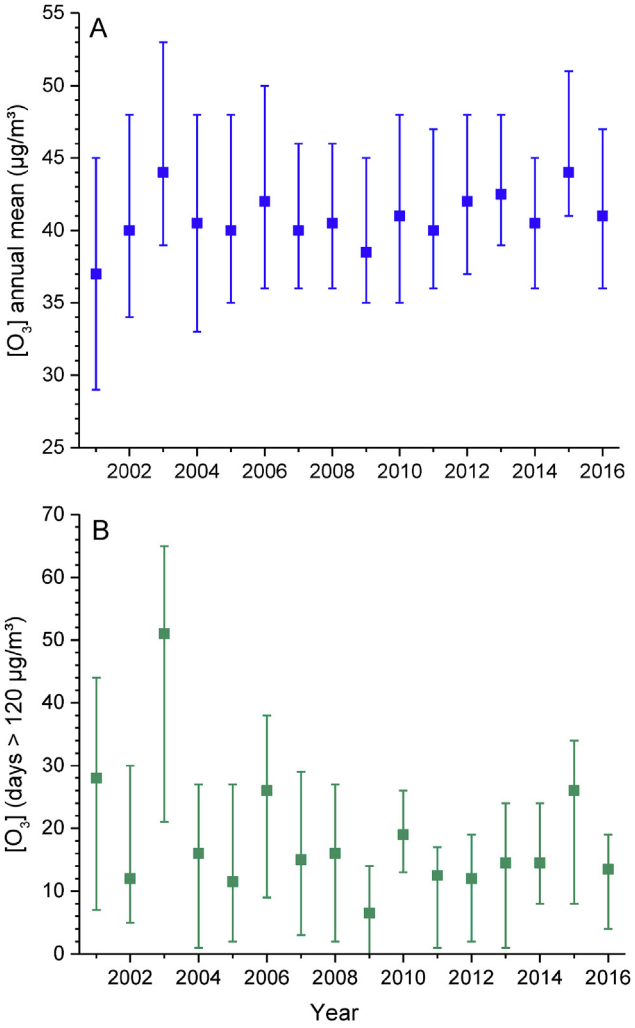

Fig. 5. Annual mean of ozone concentrations A) and number of days with exceedances of 120 µg/m^3 (8 h mean) B) for the 8 urban background monitoring stations shown in Fig. 1. The boxes (■) represent the median value and the whiskers represent minima and maxima.

the reference value of 120 µg/m^3 is exceeded varies greatly (see Fig. 5B). On average, a value of 10–30 days must be assumed for the eight observed German cities (urban background). A significant exception is the summer year of 2003 with, in some instances, more than 60 breaches of the 120 µg/m^3 threshold. Melkonyan and Wagner [32] have investigated the ozone formation potential for Germany under consideration of the NO_x concentrations and the temperature and conclude that with a temperature increase of 3 K, the number of days with concentrations > 120 µg/m^3 will increase by 87% in the rural background and by 135% in areas close to industrial locations. Model calculations by Karlsson et al. [33] for the northern European region assume a considerable decrease in ozone peak concentrations on summer days, with a simultaneous increase in small and medium concentrations on summer nights and winter days.

The entry of ozone in indoor air can be estimated by means of the mass balance equation (1), which assumes a well-mixed single-box compartment. The model does not consider the influence of the building envelope, air flow pattern or mechanical ventilation systems [40]. The restrictions associated with these simplifications are addressed by Lai et al. [34]. $[O_3]_{out}$ and $[O_3]_{in}$ are the ozone concentrations in outdoor and indoor air (µg/m^3), λ is the air exchange rate (1/h) and k_{O3} is the deposition rate of ozone in the indoor area. Measured k_{O3} values lie frequently within the range of 2.5 1/h to 3.5 1/h [35].

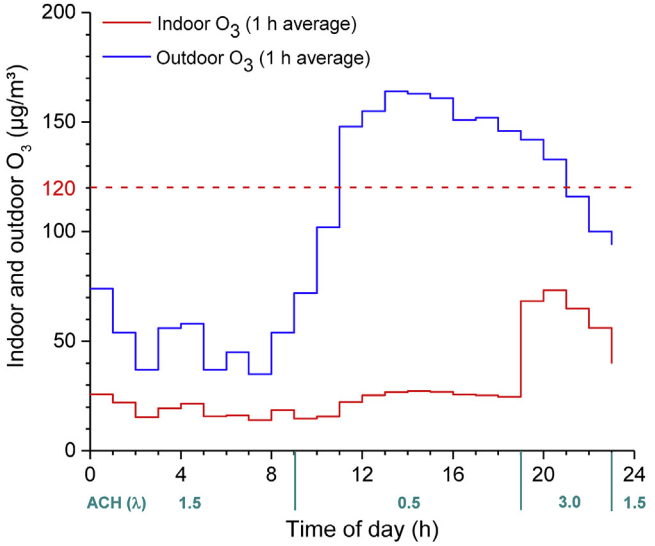

Fig. 6. Diurnal changes of ozone concentrations (1 h mean) in outdoor and indoor air. The outdoor data were taken from the monitoring station Munich, Lothstraβe for the 22.06.2017. The indoor data were calculated from equation (1) with k_m = 2.5 1/h.

$$\frac{d[O_3]_{in}}{dt} = \lambda \cdot [O_3]_{out} - \lambda \cdot [O_3]_{in} - k_{O3} \cdot [O_3]_{in} \quad (1)$$

At equilibrium with d $[O_3]_{in}$/dt = 0 equation (1) passes into equation (2).

$$\frac{[O_3]_{in}}{[O_3]_{out}} = \frac{\lambda}{\lambda + k_{O3}} \quad (2)$$

A calculated indoor/outdoor scenario for a summer day with peak concentrations of > 160 µg/m³ is shown in Fig. 6. The outdoor air data for ozone are the 1 h mean values measured at the station in Lothstraβe, Munich on 22.06.2017. For the night hours (00:00–08:00), an air exchange of λ = 1.5 1/h was assumed through window ventilation. During the day (08:00–19:00), windows and doors were kept closed (λ = 0.5 1/h). In the evening hours (19:00–23:00), intensive ventilation occurred (λ = 3.0 1/h). From 23:00 onwards, night ventilation once again took place (λ = 1.5 1/h). The air exchange rates were estimated on the basis of experimental data [36]. The calculation of the indoor concentration was performed using equations (1) and (2) under assumption of a deposition rate of k_m = 2.5 1/h. Possible ozone sources in the indoor area [37] were not taken into account. At night and during the day, the ozone concentration in the indoor area remains relatively constant with values of between 15 µg/m³ and 27 µg/m³. In the evening hours and during intensive ventilation, the indoor concentration increases to values of between 55 µg/m³ and 77 µg/m³. The model calculation shows that even on a day with extreme ozone concentrations in the outdoor air, with reasonable ventilation behavior, the WHO guideline value of 100 µg/m³ ozone is barely exceeded. Furthermore, the assumed deposition rate of k_{O3} = 2.5 1/h is conservatively estimated. Salonen et al. [38] present the results of more detailed calculations with variable values for k_{O3} and λ.

From equation (2), an I/O ratio of 0.55 can be calculated for λ = 3.0 1/h. Conversely, appropriate ventilation behavior is, however, also important, in order to provide effective protection against high ozone concentrations [39,40]. The example selected here refers to living space with manual ventilation. The model does not, however, take into account the concentration gradients between open windows and doors and the center of the room. Modern residential units often have mechanical ventilation systems with corresponding air pollutant filter elements [9].

4.2. Nitrogen dioxide

Potential health hazards caused by nitrogen oxides (NO_x), especially nitrogen dioxide (NO_2), are being intensively discussed in Germany, particularly as regards the release of NO_x from diesel engines. This concerns not only the exposure to the substances themselves but also their role in atmospheric photochemistry [2] (see also Fig. 4). On the basis of an evaluation by the World Health Organization [28], the EU has set a threshold value for NO_2 of 40 µg/m³ as an annual mean. For short-term exposure, the WHO states a guideline value of 200 µg/m³ (1 h). The WHO has also published guidelines for indoor areas of 40 µg/m³ (annual) and 200 µg/m³ (1 h) [41]. The NO_2 guideline values derived for indoor areas in Germany are 350 µg/m³ (0.5 h) and 60 µg/m³ (1 week) [42].

According to an evaluation performed by the German Federal Environmental Agency, the annual mean of 40 µg/m³ was exceeded in 142 from a total of 517 nationwide monitoring stations in 2015. These were predominantly traffic stations. However, the German Federal Environment Agency also states that between 2002 and 2011, the total NO_x emission released by transport reduced by approximately 50% (2002: 1018 kt, 2011: 538 kt). Jiang et al. [6] found that concentrations of NO, NO_2 and NO_x at monitoring stations with urban background were reduced on average by 13.8% (NO), 5.0% (NO_2) and 8.6% (NO_x) after the introduction of LEZ. In indoor areas, the quantity of potential emission sources for NO_x has decreased sharply over the years. In the past, high NO_x concentrations in indoor spaces were primarily caused by the operation of gas stoves [43] and gas-fired heaters. Today, burning candles are often a cause of NO_x emissions [44].

NO_2 is effectively degraded on surfaces. This has been confirmed in investigations by Spicer et al. [45], who experimentally determined the material-dependent degradation rates k_{NO2} up to 8.5 1/h. Taking into account the materials used in indoor areas and the available surfaces, Spicer et al. [45] state an average degradation rate of 0.8 1/h. During later investigations in an uninhabited house, Spicer et al. [46] determined degradation rates of between 1.04 1/h and 1.45 1/h. Deposition velocities for NO_2 were investigated by Grøntoft and Raychaudhuri [47], subject to the air humidity and drawing on literature data. The deposition velocity can be converted into the deposition rate as per $k_{NO2} = v_{NO2} \cdot S/V$, whereby S denotes the room surface and V the room volume. If the European Reference Room [25] is taken as the basis with S/V = 1.97 m²/m³, k_{NO2} values are obtained which are similar to those of Spicer et al. [45]. For the further calculations, a value of k_{NO2} = 1.0 1/h was assumed for the NO_2 degradation in indoor areas.

For a number of years, hardly any breaches of the EU threshold value of 40 µg/m³ have been registered in the annual mean for urban and rural-background monitoring stations. However, the NO_2 concentration in areas with high traffic volumes is subject to pronounced daily and seasonal fluctuations [48]. This can be observed using the example of 1 h mean values for the monitoring station Stuttgart Bad Cannstatt in 2017 (see Figure SI-4). With an annual mean of 30 µg/m³, the 1 h mean values vary within the course of the year between 3 µg/m³ and 108 µg/m³. In winter, the NO_2 concentrations are on average higher, caused by a reduction in the photochemical degradation due to the diminished global radiation.

Without taking indoor sources into account, NO_2 room air concentrations can be calculated analogous to ozone using equations (1) and (2). Fig. 7 shows the comparison for outdoors and indoors of calculated 1 h mean values based on the station Stuttgart Bad Cannstatt in 2017. For the respective winter and summer periods, average air exchange rates of 0.5 1/h and 1.0 1/h were assumed. It can be seen that through infiltration, neither the guideline values of the World Health Organization [28] nor the guideline values of Englert [42] will be exceeded. The illustration, however, provides no information concerning short-term emission peaks due to strong indoor sources.

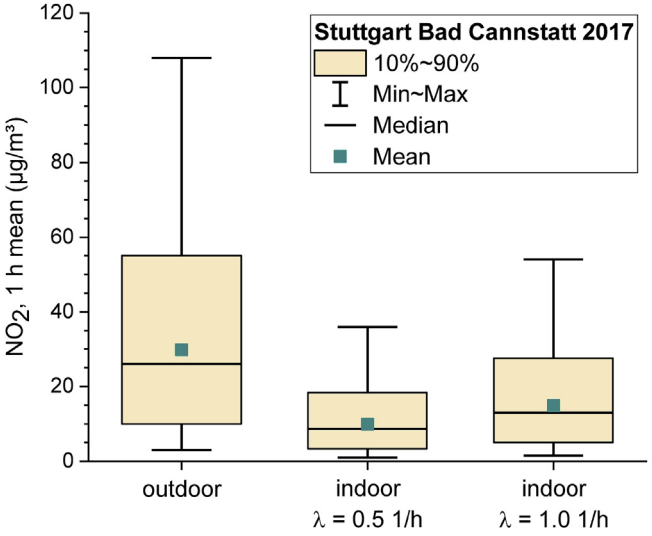

Fig. 7. Boxplot of measured NO_2 outdoor concentrations (1 h mean) for Stuttgart Bad Cannstatt in 2017. The boxplots of indoor concentrations represent simulated data using equation (2) with k_{NO2} = 1.0 1/h and two different ventilation scenarios (λ = 0.5 1/h and λ = 1.0 1/h).

4.3. Particles

The infiltration of outdoor particles into the indoor environment can be expressed by the mass balance equation (3),

$$\frac{d[PM]_{in}}{dt} = \lambda \cdot P \cdot [PM]_{out} - \lambda \cdot [PM]_{in} - k_p \cdot [PM]_{in} \quad (3)$$

where $[PM]_{in}$ and $[PM]_{out}$ are indoor and outdoor PM concentrations, respectively, λ is the air exchange rate (1/h), P is the particle penetration factor through the building envelope or openings, and k_p is the particle deposition rate in the indoor environment (1/h). Here, no indoor source is considered. As in equation (1), neither the concentration gradients nor the artificial ventilation are taken into consideration.

The infiltration factor (F_{in}) is defined as the equilibrium fraction of outdoor particles that penetrate the building and remain suspended indoors [49]. When indoor PM reaches equilibrium, F_{in} is obtained by equation (4).

$$F_{in} = \frac{PM_{in}}{PM_{out}} = \frac{\lambda P}{\lambda + k_p} \quad (4)$$

In the assessment of the results of existing studies, the problem turned out to be the selection of proper parameters λ, P and k_p. The air exchange rate was assumed to be 1.0 1/h in warm seasons and 0.5 1/h in cold seasons. We used the particle indoor deposition rates of 0.29 (for PM_{10}) and 0.17 (for $PM_{2.5}$), and particle penetration factors in cold seasons of 0.76 (PM_{10}) and 0.85 ($PM_{2.5}$) [50]. In the warm seasons, when windows are frequently opened (tilted), a particle penetration factor of 1 was assumed [51]. Taylor et al. [52] suppose that in overheated rooms, the exposure to $PM_{2.5}$ increases due to the frequent opening of windows and doors.

Fig. 8 shows the annual PM_{10} concentrations from 2001 to 2016, and $PM_{2.5}$ concentrations from 2008 to 2016 at urban background stations. For PM_{10} the overall trend is clearly decreasing. In most cities, PM_{10} has shown a consistent decreasing trend throughout the last 16 years, except for Frankfurt Ost, where PM_{10} decreased until 2008, and has stabilized since then. Monitoring of $PM_{2.5}$ started around 2008 and was only available in four of the eight stations. However, $PM_{2.5}$ also showed a decreasing trend. The high PM_{10} events (daily mean > 50 µg/m³) also occurred less frequently in recent years (see SI). The median PM_{10} and $PM_{2.5}$ concentrations in 2016 were 18 and 11 µg/m³, respectively; close to the WHO guidelines (20 µg/m³ for PM_{10} and 10 µg/m³ for $PM_{2.5}$).

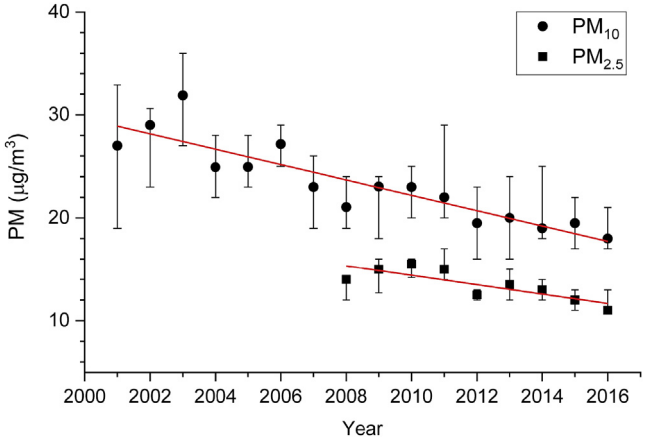

Fig. 8. Median of PM_{10} (●) and $PM_{2.5}$ (■) annual mean concentrations of eight German cities ($PM_{2.5}$ was only available in the cities of Hamburg, Hanover, Munich and Stuttgart). The upper and lower error bars represent the minimum and maximum of the annual mean values among cities. Linear regression models are applied for determining the annual trend.

Unlike O_3, the impact of climate change on ambient PM_{10} and $PM_{2.5}$ is a very complex issue. One important reason is that PM is a mixture of particles with different sources, physical properties and formation mechanisms, and these components can respond differently to the change of climate parameters. The major PM components include organic matter, sulfate, nitrate, ammonium, elemental carbon, mineral dust and sea salt [53]. For instance, sulfate concentration will increase with higher temperature due to faster SO_2 oxidation, but particulate nitrate and semi-volatile organic compound concentrations will reduce by prompting evaporation [54,55]. Increased air humidity could increase the water content of PM and enhance the absorption of gaseous pollutants, thus increasing PM concentration. Precipitation would scavenge PM from the atmosphere; however, it is the frequency of precipitation, rather than the intensity, which plays the major role [56].

If the future emission scenarios are considered, ambient PM are expected to decrease significantly. Lacressonnière et al. [57] modeled a reduction of 1.8–2.9 µg/m³ PM_{10} concentration in Europe under a +2 °C climate and future emission scenario. In Germany, particularly due to a gradual phase-out of diesel-powered passenger cars, there will be much room to lower PM concentrations. As an estimate, the mid-term annual mean PM_{10} and $PM_{2.5}$ concentrations are likely to reach 14 and 8 µg/m³, respectively.

Table 1 shows the PM_{10} and $PM_{2.5}$ infiltration factors, as well as calculated indoor PM_{10} and $PM_{2.5}$ concentrations in warm and cold seasons, respectively. In warm seasons, due to higher air exchange rates and greater particle penetration factors, the infiltration factors are much higher than in cold seasons. The indoor PM is thus higher in warm seasons than in cold seasons. $PM_{2.5}$ infiltrates overall more efficiently than PM_{10}. In the future, the indoor PM_{10} and $PM_{2.5}$

Table 1
PM_{10} and $PM_{2.5}$ infiltration factors and calculated indoor PM_{10} and $PM_{2.5}$ concentrations (µg/m³) caused by infiltration from outdoors.

		Outdoor	Indoor	
			warm season	cold season
PM_{10}	Infiltration factor		0.78	0.48
	Present concentration	18	14.0	8.6
	Future concentration	14	10.9	6.7
$PM_{2.5}$	Infiltration factor		0.85	0.63
	Present concentration	12	10.3	7.6
	Future concentration	8	6.8	5.0

concentrations are expected to decrease.

Of course, climate change could also influence living behavior or habits, which may directly influence the air exchange rate and particle penetration factor and, subsequently, the indoor PM. For instance, as the climate model indicates, there will be more hot days in Germany. People are more likely to keep the windows open during the night, thus favoring PM infiltration. This may have a significant impact for those who live next to major streets, where PM levels are higher than the urban background.

5. Discussion

In a highly industrialized country, daily life generally comprises complex dynamic processes. The people, whether adults or children, spend the day in different places, and a considerable proportion of time is spent on transportation. Mazaheri et al. [58] recorded more than 90 activities for Australian schoolchildren and divided them into broad categories such as home, school, transportation and others. Based on these aspects, investigations can only refer to indoor and outdoor concentrations, but not to the actual exposure.

The indoor and outdoor conditions under which people in Germany live and work are, moreover, very different. The influence of the Gulf Stream ensures a mild climate, hot spells (as in 2003 and 2010) and cold spells are comparatively rare. Traditionally, living accommodation and workplaces in Germany are manually ventilated, but air-conditioning systems have been customary in large office buildings for decades. In contrast, artificial ventilation in private residential environments, usually in conjunction with heating systems, has only recently become an issue. For the case of Slovakian homes Földváry et al. [59] have shown that occupants of thermally insulated houses without artificial ventilation systems are often dissatisfied with the quality of the indoor air. Havarinen-Shaughnessey et al. [60] did not find associations between retrofit status of Finnish and Lithuanian houses and occupant satisfaction in summer temperature.

Unquestionable is the global increase in average air temperature and warm days caused by climate change (see Fig. 2). It is expected that this will also be accompanied by an increase in the average absolute humidity. At 20 °C, one cubic meter of air can absorb 18.3 g of water; at 25 °C, it is 24.8 g. The results of the comparatively simple model used here, which does not directly take into account solar radiation, hot spells, heating or ventilation activities, predict a significant increase in heat transfer into the indoor area over the next few decades. Furthermore, the occurrence of hot spells is expected to increase in the future [4]. The heat action plans of the Bund/Länder Ad-hoc Arbeitsgruppe Gesundheitliche Anpassung an die Folgen des Klimawandels [19] include measures for indoor areas. These are divided into a) short-term measures such as providing shade, occupying cooler rooms, adapting the ventilation behavior, avoiding the operation of heat-emitting devices; (b) mid-term measures such as the installation of sun protection and insulation, placement of vegetation, installation of technical cooling techniques, and (c) active cooling of indoor areas, whereby this is only recommended if other measures do not lead to success or are exhausted. Fundamentally, good thermal insulation can only protect against the overheating of indoor areas if intelligent ventilation behavior is deployed. A substantial amount of effort is required in order to passively cool a thermally insulated living area which has become too hot inside.

The heat action plans also address potential health risks arising through increased tropospheric ozone pollution. For Germany, the average annual concentrations have remained constant since the turn of the millennium; the differences between the eight monitoring stations considered here are also slight. The number of days with breaches of the information threshold of 180 μg/m^3 and the alarm threshold of 240 μg/m^3 has declined significantly. It is expected that this trend, constant annual mean and simultaneous decline in peak concentrations, will continue in the coming years. The major influencing factors will be future global radiation, NO_2 emissions and hydrocarbon emissions (see Fig. 4). In indoor areas, ozone is rapidly degraded. Therefore, even on summer days with peak concentrations in the outdoor air, it is unlikely that high ozone levels in the range of the WHO guideline value of 100 μg/m^3 will occur in indoor areas, provided indoor sources are negligible. With customary ventilation behavior, the highest indoor concentrations are then attained during the evening hours. The WHO guideline value, however, takes into account solely the inhalative effect of ozone. Dermal exposure, which leads to reactions with organic components in the surface of the skin, such as squalene [61], remains unconsidered. Especially in environments with high occupant density, such as school classrooms and offices, ozone removal by humans becomes relevant [38]. On the basis of Canadian data [62], an increase in the tropospheric ozone background concentration is anticipated in the future. In their work, the authors also note the formation of possible secondary products and particles through ozone-initiated reactions and refer to air purifiers installed in air-conditioning systems as an effective option for reducing ozone pollution in indoor areas.

The NO_2 pollution of the indoor and outdoor atmosphere can only be considered and evaluated under regional aspects. By taking into account the average of all the monitoring stations nationwide, a decreasing trend can be determined (see Figure SI-5). If, however, the eight selected stations in urban settings are compared (see Tables SI–3), this decreasing trend is barely obvious, particularly for the period 2007–2016. At traffic stations, breaches of the annual mean of 40 μg/m^3 remain frequent. As combustion engines constitute the main emission source for NO_x, the high traffic density in these regions is therefore responsible. Whether a reduction can be expected here in the foreseeable future depends in particular on future environmental legislation and the application of NO_x-reducing technologies. High NO_2 values, with simultaneous low ozone values, occur during the winter months; in summer, the reverse is true. In indoor areas, NO_2 is effectively degraded, but not, however, quite as quickly as ozone. Under normal ventilation conditions (0.5 1/h – 1.0 1/h), NO_2 concentrations are expected to be low in the absence of other emission sources, as demonstrated by the example of Stuttgart Bad Cannstatt (2017) (see Fig. 7).

The pollution of outdoor air with particulate matter (PM_{10} and $PM_{2.5}$) has decreased considerably in recent years. On average, the pollution is significantly below the EU annual limit of 40 μg/m^3. Nevertheless, at stations with heavy traffic, the EU daily limit of 50 μg/m^3 is still exceeded, e.g. at the Stuttgart Am Neckartor traffic station (station code DEBW118), where breaches were measured on a total of 45 days in 2017. The EU annual limit value for $PM_{2.5}$ of 25 μg/m^3 was not exceeded at any station in 2017. The simulation of the infiltration of PM_{10} and $PM_{2.5}$ into indoor areas (see Table 1) shows that outdoor air is not a significant source of particulate pollution in indoor areas and, with the continuing decreasing trend, will also remain insignificant in the future. As regards exposure, the sources in the indoor area are of far greater importance here [63].

6. Conclusion

In this work, the possible impacts of climate change on indoor air quality were investigated with regard to the geographical circumstances in Germany. For the transport of heat and moisture through exterior walls, a simple one-dimensional model was applied. For the estimation of I/O ratios, single-box models were used under the assumption of complete mixing. These models allow a rational assessment of trends. However, they are suitable neither for the calculation of spatial concentration differences nor for the prediction of exposure scenarios. The prediction of energy consumption, indoor air pollution and potential health burden requires sophisticated modeling and coupled models under consideration of multiple sources and facilities [64].

In the future, higher background concentrations of ozone and decreasing concentrations of NO_2, PM_{10} and $PM_{2.5}$ can be expected in outdoor air. With intelligent manual ventilation behavior, adjusted to

suit the conditions of the day, it is unlikely that concentrations which are critical in terms of health aspects will be attained in indoor areas through infiltration. Residential spaces with artificial ventilation generally have appropriate filtering devices. The current and future role of volatile organic compounds (VOCs) is the subject of current discussions. McDonald et al. [30] postulate that VOCs from consumer products, which exfiltrate from indoor areas into the outdoor areas, may contribute to tropospheric photo smog.

With regard to indoor areas, however, the greatest challenges in the coming years will arise in the regulation of temperature and humidity [65]. This applies both to the long-term temperature trend and to sudden incidents such as heatwaves and torrential rain. By means of a simple experiment, it could be shown that without additional measures, thermally insulated and manually ventilated houses will not provide protection against overheating of the indoor area in the medium term. Improved concepts and preventive measures are required here in order to obviate unpleasantly high or even health-endangering temperatures and an indoor climate which also facilitates microbial growth. For the UK, Vardoulakis et al. [66] identified four major categories: a) indoor temperature; b) indoor air quality; c) indoor allergens and infections; d) flood damage and water contamination.

Finally, it must be kept in mind that the statements and conclusions made in this publication might be applicable to Germany and Central Europe, but not to other regions of the world. In many regions and metropoles, people suffer due to hot and humid climates and/or extreme air pollution, which demand different measures and action plans [67].

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.buildenv.2018.07.050.

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