Journal Pre-proof

A critical evaluation of the dynamic nature of indoor-outdoor air quality ratios

Samuel Stamp, Esfand Burman, Lia Chatzidiakou, Elizabeth Cooper, Yan Wang, Dejan Mumovic

PII: S1352-2310(22)00020-6

DOI: https://doi.org/10.1016/j.atmosenv.2022.118955

Reference: AEA 118955

To appear in: Atmospheric Environment

Received Date: 19 November 2021

Revised Date: 10 January 2022

Accepted Date: 11 January 2022

Please cite this article as: Stamp, S., Burman, E., Chatzidiakou, L., Cooper, E., Wang, Y., Mumovic, D., A critical evaluation of the dynamic nature of indoor-outdoor air quality ratios, *Atmospheric Environment* (2022), doi: https://doi.org/10.1016/j.atmosenv.2022.118955.

This is a PDF file of an article that has undergone enhancements after acceptance, such as the addition of a cover page and metadata, and formatting for readability, but it is not yet the definitive version of record. This version will undergo additional copyediting, typesetting and review before it is published in its final form, but we are providing this version to give early visibility of the article. Please note that, during the production process, errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

© 2022 Published by Elsevier Ltd.



Author Credit Statement

DM and SS contributed to the conceptualization of the paper. SS conducted the formal analysis and original draft. All authors contributed to the methodology, data collection, review and editing of the paper.

ournal Propo

1 A critical evaluation of the dynamic nature of indoor-outdoor air quality ratios

2 Samuel Stamp^a, Esfand Burman^a, Lia Chatzidiakou^b, Elizabeth Cooper^a, Yan Wang^a, Dejan Mumovic^a

^a Institute for Environmental Design and Engineering, University College London (UCL), Central House, 14
 Upper Woburn Place, London, WC1H ONN, UK

6 Department of Chemistry, University of Cambridge, Cambridge, CB2 1EW, UK

8 Abstract

7

9 Long term, continuous indoor and outdoor pollutant monitoring was evaluated from a case study 10 hospital, school, office and 18 apartments in the UK. Data was examined in order to explore the 11 dynamic behaviour of indoor-outdoor ratios (I/O) for both particulate matter and nitrogen dioxide. 12 Traditionally I/O ratios have been determined as single aggregate values or static parameters, from 13 passive sampling or short periods of continuous monitoring. Whilst widely reported, I/O ratios are 14 seen as too variable to be of wider use. However, this work reveals the dynamic nature of I/O ratios, 15 with strong diurnal and seasonal variation observed for both particulate matter and nitrogen 16 dioxide. Higher I/O ratios tended to be seen during core or occupied hours, associated with 17 increased human activity and higher ventilation rates. This means that static I/O ratios determined 18 by passive sampling techniques, rather than continuous measurements filtered to core hours, may 19 underestimate I/O ratios associated with occupant exposure. Further, the I/O ratio is shown to be 20 strongly influenced by occupant activity and window opening behaviour. As such, it may represent a 21 personal variable as much as one associated with a building. It is argued that traditionally reported 22 static I/O ratios simplify these dynamic behaviour and modes of operation into a single aggregate 23 value, losing key information in the process. Further, without contextual information on the 24 operation and use of a building during measurements a reported I/O ratio may be hard to interpret 25 or compare to wider studies. Finally, it is argued that the I/O ratio, whilst a limited metric, when 26 evaluated dynamically provides a useful building operation parameter, describing the relationship 27 the building has with the outdoor environment. This can help better define ventilation strategies, 28 schedules, the influence of occupant behaviour and significance of indoor sources.

29

30 1. Introduction

31 Although the most important environmental predictor driving health effects is considered to be

- 32 individual-level exposure, most epidemiological studies concerning air pollution utilise outdoor
- 33 concentrations at the nearest central monitoring station or modelled ambient concentrations often
- 34 at postcode level (Hoek et al., 2013). Models for chronic health impacts and mortality tend to use
- 35 outdoor annual mean concentrations (Lelieveld et al., 2015; Pedersen et al., 2013), without
- 36 accounting for any diurnal /seasonal variations or modifying effects of building envelopes on indoor
- 37 exposure. However, people spend most of their time indoors and within indoor microenvironments.
- 38 The use of outdoor concentrations as proxies of exposure therefore implicitly assumes the
- 39 attenuation of ambient concentrations by buildings is the same for all participants (Cohen et al.,
- 40 2009), with exposure estimates that do not take into account time activity patterns, indoor
- 41 generated pollutants or factors influencing infiltration of pollutants indoors. Evidence from the built
- 42 environment indicates that building characteristics, ventilation, penetration efficiency and indoor
- 43 activities have a significant impact on indoor concentrations (Blondeau et al., 2005; Kim et al., 2019;
- 44 Majd et al., 2019; Stranger et al., 2008; Wichmann et al., 2010). The influence of building

- 45 characteristics has even been picked up within population health studies, for example, PM₁₀
- 46 associated hospital admissions were lower in US cities with higher proportions of air conditioning

47 (Janssen et al., 2002).

- 48 The modifying impact building envelopes have on outdoor concentrations penetrating in the indoor
- 49 environment has not been adequately addressed in many health studies and therefore remains a
- 50 potential source of exposure error and a cause of the large bandwidth seen in mortality estimates (Ji
- 51 and Zhao, 2015; Sarnat et al., 2007).

52 In light of these concerns, some epidemiological studies and large scale building stock models have

53 instead attempted to incorporate metrics to quantify the modifying effect of a building upon

54 outdoor concentrations (Chen et al., 2012; Ji and Zhao, 2015; Taylor et al., 2019). To this purpose, an

- 55 infiltration factor (Finf), the fraction of outdoor pollutants remaining airborne after penetrating
- 56 indoors, may be used. Under assumptions of ideal and instantaneous mixing, uniform air exchange 57
- rates and approximated steady state (i.e. analysed over a minimum of 24hours), indoor
- 58 concentrations can be described by a first-order differential equation with the analytical solution:
- 59 (Wallace and Williams, 2005):

$$60 C_{in} = F_{inf}C_{out} + \frac{S}{W(a+b)}$$

61 Where the infiltration factor can further be defined as:

$$F_{inf} = \left(\frac{P \cdot a}{(a+k)}\right) \qquad 2$$

63 Here, C_{in} and C_{out} are the indoor and outdoor mass concentration ($\mu g/m^3$), P is the penetration factor

64 across the building envelope, a is the air exchange rate (h^{-1}), k is the total decay rate of particles (h^{-1}) 65 that depends on the aerodynamic diameter, S is the source strength $(\mu g/h^{-1})$ and V is the volume of 66 the building (m³).

67 Estimates of Finf can be generated through a variety of methods. Modelling studies may base estimates 68 of P, a and k upon previous literature (e.g. Chen et al., 2012; Fabian et al., 2012; Fazli et al., 2021; Li 69 and Friedrich, 2019), sometimes supported by building surveys and questionnaires, in order to 70 determine the likely relationship between indoor and outdoor concentrations (Cohen et al., 2009). 71 Alternatively, Finf may be based upon direct measurements of pollutants with no internal sources (e.g. 72 particulate sulfate (Wilson et al., 2000)), via linear regression or a range of dynamic solutions to the 73 mass balance equation (Diapouli et al., 2013a; Wang et al., 2016). However, in reviewing these 74 techniques Diapouli et al (2013) note the determination of *P* and *k* remains challenging, with reported 75 values within the literature varying significantly. Similarly, approaches must consider how to 76 accurately filter out or account for internally generated contributions across analytical periods. Finally, 77 the air exchange rate, a, is considered a critical exposure factor that may potentially modify health 78 effect estimates reported in epidemiological studies (Long and Sarnat, 2004). However, studies show 79 significant variation in air exchange rates, both in time and between buildings, countries and 80 occupants (Dimitroulopoulou, 2012; Dimitroulopoulou and Bartzis, 2014; Øie et al., 1998). Further, measurements of representative air exchange rates are subject to significant uncertainties, 81 82 particularly regarding occupancy, multizonal airflows, sensor accuracies, temporal variations and 83 analytical methods (Batterman, 2017; Johnston and Stafford, 2016; Kabirikopaei and Lau, 2020). As a 84 result of these challenges, the determination of more accurate and relevant methods of determining 85 the relationship between indoor and outdoor air is thought to be a key future task in air pollution 86 exposure assessment (Sarnat et al., 2007).

An alternative, simpler metric is to define the ratio of indoor pollutant concentrations in relation to outdoor concentrations. This indoor-outdoor (I/O) ratio allows a more direct field measurement and avoids uncertainties in determining individual parameters in equation 2. The I/O ratio has been measured for particulate matter and nitrogen dioxide since the 1980s (Monn et al., 1997) and can be defined by:

3

4

92
$$I/O = \frac{C_{in}}{C_{out}}$$

93 Or, importantly when utilising continuous, real-time measurements:

$$I/O_{i,j} = \frac{\sum_{i}^{j} c_{in}}{\sum_{i}^{j} c_{out}} = \frac{\sum_{i}^{j} c_{in}/n}{\sum_{i}^{j} c_{out}/n}$$

Where i and j are the beginning and end of an analytical period and *n* is the number of measurements
over that period. More fully, combining equation 4 and equation 1:

97
$$I/O = \frac{C_{in}}{C_{out}} = \left(\frac{P \cdot a}{(a+k)}\right) + \frac{S}{V \cdot (a+k)C_{out}}$$

As can be seen in equation 5, the I/O ratio is a function of a building's penetration factor (of each specific pollutant), air change rate, loss rate due to chemical sinks or deposition and the presence of any indoor sources. It can also be considered that in the absence of indoor sources (S = 0) the indooroutdoor ratio can be considered equivalent to the infiltration factor (I/O = F_{inf}).

The I/O metric therefore not only describes the building as a pollutant modifier but also incorporates the operation of and activity within a building, including indoor sources. Estimates of I/O ratios have been established for a range of pollutants, buildings and countries, with significant review articles covering studies on nitrogen dioxide (Hu and Zhao, 2020; Kalimeri et al., 2019; Salonen et al., 2019),

particulate matter (Chen and Zhao, 2011; Kalimeri et al., 2019) and ozone (Kalimeri et al., 2019;

107 Weschler, 2000). Importantly, significant ranges in I/O estimates exist both across study samples,

108 between studies and within buildings across time (Wallace and Williams, 2005). This latter point is

109 important, with further studies indicating seasonal and shorter-term temporal variations in I/O

110 ratios measured within a single building or zone (Allen Ryan W. et al., 2012; Cyrys et al., 2004).

111 The focus and scope of this paper is restricted to understanding I/O ratios within the context of

112 human health and buildings designed and operated for occupied spaces. As such, some conclusions

113 will not apply to other areas where I/O ratios have also been adopted, such as in cultural heritage

114 where they are used to detail relationships between outdoor and various indoor environments, such

as showcases and closed depositories (e.g. Grau-Bové and Strlič, 2013; Lazaridis et al., 2015).

116 1.1 Measuring the I/O ratio

94

117 Importantly, the definition of any measured I/O ratio is then further formed by the nature of the 118 measurement itself. Many of the largest studies of indoor air quality have at least partly used some form of diffusive sampling over 5 to 14 day periods (e.g. Allen Ryan W. et al., 2012; Mandin et al., 119 120 2017; Schneider et al., 2001; Shaw et al., 2020; SINPHONIE, 2014). Some of these data sets have then 121 been used to determine distributions of I/O ratios for different building types (Kalimeri et al., 2019). 122 However, fundamentally this means the measured I/O incorporates significant unoccupied periods 123 during which activities and building operation and indoor sources (S) are likely to significantly differ. 124 For example, a 5-day passive measurement would typically represent just a third of core occupied 125 school hours (e.g. 8am – 4pm) and two-thirds unoccupied periods. Similarly, out-of-hours sampling of internal VOCs has been found to either over or under-estimate concentrations, depending on thepollutant species (WHO Europe, 2020).

128 This bias in sampling period is further compounded by the non-static nature of buildings. Air exchange 129 rates in both mechanically and naturally ventilated buildings are likely to increase during occupied 130 hours, meaning these periods may experience a higher ingress of outdoor air pollution to the indoor 131 environment than during unoccupied periods. More specifically, this can be considered an increase in 132 the F_{inf} component, as a result of the higher air exchange rate across occupied periods. Even in a 24hour operation, sealed envelope hospital, demand-controlled ventilation has been found to result 133 134 in higher I/O ratios for nitrogen dioxide across the core daytime hours (Stamp et al., 2020). The result 135 is that any aggregated passive measurement is likely to underestimate the I/O experienced across this 136 period. The potential bias in sampling period may be given further significance by the fact that the 137 daytime periods with the highest I/O ratio may also coincide with the highest, traffic-related, outdoor 138 concentrations.

- 139 Shorter-term aggregate sampling, taking place over a few hours or days and eliciting a single measured
- 140 concentration for this time period, can avoid this out-of-hours sampling period bias (e.g. (Viana et al.,
- 141 2014). However, even with measurements across multiple seasons (e.g. Li and Lin, 2003), these may
- only represent small snap-shots of a complex and highly variable parameter.

143 The alternative is to use continuous or real-time measurements (e.g. laser scattering, optical particle 144 counters, electrochemical sensors). This approach allows careful filtering and selection of appropriate 145 analysis periods. Until recently, with large, expensive and impractical equipment, continuous 146 measurements have often been limited to periods of a few days to weeks (e.g. (Branco et al., 2014; 147 Huang et al., 2015; Jones et al., 2000). However, improvements in sensing technologies can enable 148 more affordable measurements at improved accuracies, in multiple locations, allowing I/O ratios to 149 be assessed and examined at high resolutions across significantly longer periods (Chatzidiakou et al., 150 2019).

151 *1.2 Aims*

In summary, I/O ratios have been measured across numerous studies since the 1980s and continue to be widely reported for a range of pollutants in many IAQ studies. Whilst the I/O ratio has been directly used in a few health studies (Borrego et al., 2006; Setton et al., 2008), more commonly building modifiers are omitted or else infiltration factors preferred (Fazli et al., 2021). Therefore, whilst I/O ratios remain widely measured and reported, the large variation in measured values and range of influencing factors mean that they are unlikely to be useful to epidemiologists (Poupard et al., 2005).

This paper aims to use several long-term, continuous data sets from a range of UK building types to evaluate the variation seen in I/O ratios under greater detail. This includes variation due to diurnal and seasonal effects, measurement methods, building operation modes, occupant behaviour and measurement uncertainties. The aim of these investigations is to understand the dynamic nature of I/O ratios and the influential factors behind this variation. From this, a critique of the validity and relevance of I/O ratios can be made, particularly when they are evaluated as a static metric.

A further hypothesis is that the information held within a dynamically assessed I/O ratio might prove more insightful as a building operation parameter. Whilst individual, static I/O ratios may prove useful within a larger population of results, a dynamic I/O may indicate, on an individual building basis, how the influence of outdoor air, ventilation practices and indoor sources of pollutants impact indoor air quality – particularly when longer data sets can be acquired. Subsequently this may lead to improved evidence on how building design and operation might mitigate these effects.

170 2. Research Methods

171 Data from long-term, simultaneous outdoor and indoor air quality measurements has been re-

analysed focusing on determining I/O ratios from continuous monitoring. This includes measurements

173 from a hospital, a school, an office and in 18 low-energy apartments over a 6 - 12 month period.

174 Further details of the case study buildings and monitoring campaigns can be found in previous

175 publications (Cooper et al., 2021; Stamp et al., 2020). A summary of the case studies can be found in

176 Table 1.

Case Study	Monitored	Monitored	Ventilation	Year of	Air	Location
	Locations	Duration	strategy	Completion	Permeability	
Hospital	1 (3 sampled	8 Months	Mechanical	2015	$< 5 \text{ m}^{3} \cdot \text{h}^{-1}/\text{m}^{2}$	City
	indoor zones)	(Jan – Aug)	Ventilation,		at 50 Pa	Centre, UK
			fully sealed			
			(F9 filters)			
School	1 (3 sampled	6 Months	Mechanical	2014	< 5 m ³ ·h ⁻¹ /m ²	South
	indoor zones)	(Jan-June)	ventilation,		at 50 Pa	London,
			openable			UK
			windows			
			(F7 filters)			
Office	1 (3 sampled	8 Months	Naturally	2014	4.7 m³/h per	Town-
	indoor zones)	(Jan – Aug)	ventilated		m² at 50 Pa	Centre, UK
Apartments	11 (1 zone –	Between 6	Background	2015	2–3 m³/(h.m²)	East
(Site A)	living room)	- 12	MVHR (G3		at 50Pa	London,
		Months	filters),			UK
		(June –	openable			
		May)	windows			
Apartments	7 (1 zone –	Between 6	natural	2007	< 5 m ³ ·h ⁻¹ /m ²	East
(Site B)	living room)	- 12	ventilation		at 50 Pa	London,
		Months	and trickle-			UK
		(June –	ventilators			
		May)				

177 Table 1: Details of monitoring campaigns used in analysis.

178

The analysis of I/O ratio focuses on measurements of particulate matter (PM_{2.5} and PM₁₀) and nitrogen 179 180 dioxide (NO₂). Complimentary measurements of carbon dioxide (CO₂, as a proxy for ventilation rates) 181 and TVOCs (as a proxy for internally generated pollutants) have been made simultaneously in all 182 locations, alongside temperature, humidity and locally installed weather stations. Within the case 183 study apartments, window-opening and occupant presence have additionally been monitored 184 directly, allowing more explicit evaluation of occupant behaviours on I/O ratios. Details of the monitoring equipment is given in Table 2. Sensors have been co-located and evaluated against 185 186 reference instruments, with appropriate linear corrections applied to improve accuracy or precision 187 between the deployed sensors. Specific details can be found in the supplementary material of Stamp 188 et al., (Stamp et al., 2020), with Chatzidiakou et al., (2019) providing a broader look this process and 189 a more detailed evaluation of sensor performance. Particulate matter data has additionally been 190 filtered to avoid bias at high humidity (>88%), because exposure of the particles to relative humidity 191 (RH) results in hygroscopic growth of particles and leads to mass overestimation (Crilley et al., 2018). 192 The optical particle counters limited ability to capture particles at the lower end of particle sizes (range 193 $0.38 = 17 \,\mu\text{m}$) may also impact the results of the study. For example, parts of the urban aerosol fine 194 mode fraction $(0.2 - 0.3 \,\mu\text{m})$ may not be sufficiently observed. This may further limit the equivalence 195 of I/O ratios (see section 4.6 for further discussion).

- 196 Measurements were aggregated to hourly intervals to estimate I/O ratios to reduce noise and reduce
- 197 the effect of lags between indoor and outdoor concentrations. All analysis was performed in R

198 software (R Core Team, 2021).

199 Table 2: Details of monitoring equipment used in case study buildings.

Parameter	Sensor	Range	Resolution	Accuracy
Temperature	Thermistor	-30.0 to 65.0°C	0.1°C	±0.2°C at 20°C ±0.4°C for -5 to 40°C ±1.0°C for -20 to 65°C
Relative Humidity	Capacitive	0.0 to 100.0%	0.10%	±2% RH (0 to 90% RH) ±4% RH (0 to 100% RH)
CO ₂	Non-dispersive infra red (E+E Electronik)	0-5000ppm	1ppm	<±50ppm, +3%
Particulate Matter (PM _{2.5} & PM ₁₀)	Optical Particle Counter (Alphasense OPC-N2) Size segregated particles in the range (0.38 to 17 µm)	0 to 500 μg/m ³	0.01 μg/m3	-
Airflow	-	0.00 to 500 ml/s	0.01 ml/s	-
Nitrogen Dioxide (NO ₂)	Electrochemical (Alphasense NO2-A43F)	0.00 to 3.00 ppm	0.1 ppb	-
Total Volatile Organic Compounds (TVOCs)	Photoionization detector (Alphasense PID-AH2)	0.00 to 50.00 ppm	10ppb	-
Occupancy	Passive Infrared Sensors (PIR) (HOBO UX90-05)	0-1 (Unoccupied – Occupied)	-	-
Window Status	Reed Contact Switches (Eltek GS34)	0-1 (Open – Closed)	-	-

200

201

202 **3. Results**

Results from re-analysed I/O ratios are explored within this section. This includes diurnal variations
(section 3.1), seasonal variations (3.2), differences associated with passive and continuous
measurement methods (3.3), variations between apartments (3.4) and variations with occupant
behaviour (3.5).

207 3.1 Diurnal Variations in I/O ratios

The diurnal behaviour of I/O ratios can be seen for the monitored hospital, school, office and 18 apartments in Figure 1.

210 In the mechanically ventilated hospital and hybrid school, the I/O ratio for nitrogen dioxide is seen to

211 increase by a factor of two during core operation hours. Both buildings adopted CO₂ based demand-

212 controlled ventilation strategies, that would increase ventilation rates during core hours to maintain

213 low indoor CO₂ concentrations. However, by increasing the air change rate to maintain low indoor CO₂

 $214 \qquad \text{concentrations, there was an unintended ingress of NO_2 from the outside air.}$

215 For particulate matter, within the fully mechanical hospital (F9 filters) and hybrid school (F7 filters),

the impact of filtration is clear, with low I/O ratios for both PM_{2.5} and PM₁₀. However, within all non-

- 217 domestic buildings, the I/O ratio of particulate matter is again seen to increase across core hours.
- Again, this may be associated with increased ingress from outdoors, but, given particle filtration in

Journal Pre-proof

- 219 mechanically ventilated buildings, the more significant contribution to this is likely to by indoor 220 generated and re-suspended particles during the occupied periods, particularly for PM₁₀.
- 221 With more varied occupancy schedules, apartments show altogether different diurnal patterns. The
- $\label{eq:loss} 222 \qquad I/O\ ratio\ for\ NO_2\ again\ peaks\ during\ the\ middle\ of\ the\ day,\ but\ peaks\ in\ particulate\ matter\ occur\ during$
- the evening, associated with increased occupant activity. A strong peak is seen around 19:00-20:00,
- with I/O ratios reaching above 1.5 for $PM_{2.5}$ and above 2 for PM_{10} . These indicate the strong influence
- of internal sources, e.g. cooking, across these periods.
- 226 Such diurnal variations are seen in other studies, with indoor concentrations and I/O ratios varying
- significantly across the day (Challoner and Gill, 2014; Jones et al., 2000), and between weekdays and
- weekends (Branco et al., 2014) or peaking during core hours in schools (Branco et al., 2019) and
- hospitals (Cyrys et al., 2004).





Figure 1: Top row - Aggregated or 'typical' I/O ratios for NO₂, PM_{2.5} and PM₁₀. Evaluated over 6-9 month periods in a school, hospital, office (3 internal zones in each -Mon-Fri only) and 18 apartments (living rooms). Bottom row – corresponding outdoor concentrations for each site and pollutant.

235 3.2 Seasonal variations in I/O ratios

- 236 Figure 2 shows the I/O ratios for NO₂, PM_{2.5} and PM₁₀ in the heating (Oct-Mar) and non-heating
- 237 seasons (Apr-Sep). Increases in the I/O across the non-heating season can be observed in all cases
- 238 for NO_2 and $PM_{2.5}$. Such differences are not seen for PM_{10} , where potentially higher indoor
- 239 contributions and the impact of re-suspension may negate any seasonal effect.
- 240 Similar seasonal variations have been picked up in a number of previous studies. Trends may be
- 241 dependent on the significance of internal sources, but typically higher I/O ratios have been observed
- during the summer under higher ventilation rates (Cyrys et al., 2004; Hu and Zhao, 2020;
- 243 Martuzevicius et al., 2008; Stamp et al., 2021). This trend corresponds to findings of a stronger
- association between both ambient NO₂ and PM_{2.5} concentrations and mortality during warm
- seasons, when along with spending more time outside, increased ventilation rates resulted in higher
- ingress of ambient NO₂ to the indoors (Rojas-Bracho et al., 2002; Samoli Evangelia et al., 2013).
- 247 The seasonality seen here has two main implications. Firstly, it re-enforces the need to capture
- 248 seasonal variation within I/O measurements. Secondly, it indicates the influence of changing building
- 249 operation and occupant behaviours between the two seasons.







Figure 2: Seasonal variations in I/O ratios for the case study hospital, school and office (all working hours only), as well as
 the 18 apartments.

255 3.3 Comparing I/O ratios defined by diffusive and continuous sampling

For a metric to be comparable and provide useful insights, it must be consistent across potential measurement methods. However, there is a risk that I/O ratios measured by either passive or continuous sampling yield different results. The diurnal variations already seen in section 3.1 suggest that out-of-hours sampling period bias may significantly impact passive sampling methods, where a static I/O ratio is measured across full 24-hour periods.

In Figure 3, continuously measured I/O ratios for the non-domestic case studies have been re-analysed into a series of hypothetical 5-day passive measurements and compared to hourly continuous estimates utilising occupied hours only (i.e. the full data set has been analysed with either a range of average 5-day indoor and outdoor concentrations across this period, representing passive measurements of the I/O ratio, or as continuous measured and calculated hourly I/O ratios, across core hours only).

In some cases, for example NO₂ within the naturally ventilated office, there is little observable difference between the passive and continuous approaches, as I/O ratios are not seen to drastically vary across the day (Figure 1). However, when building operation is significantly different during core hours (e.g. hospital – NO₂) or when human activity increases across core hours (e.g. office – particulate matter) significant differences are seen. The overall difference between I/O ratios defined in core hours, outside core hours and across all weekday hours can be seen in Table 3.

273 When bias occurs, it can have a significant impact, particularly as core building hours may coincide with peaks in external traffic-related pollutants. For example, annual mean ambient NO₂ 274 275 concentrations, across all hours at the school site were 19 ppb. Applying the median I/O ratio of 0.43, 276 calculated across all hours (i.e. as if from passive measurements) would give a mean internal 277 concentration of 8.2ppb. However, applying the typical daily profile of the I/O ratio (as seen in Figure 278 1), across outdoor concentrations during core hours (23.8ppb), would give a concentration during core 279 occupied hours of 12.2ppb (49% higher). Similarly, for PM_{2.5} the two approaches lead to mean internal 280 concentrations of 3.2 and 6.3 μ g/m³ respectively (97% higher).

- 281
- 282

283

285 Table 3: I/O ratios during core and outside of core hours. Note: ALL is still only working days, not including weekends.

	Pollutant	Core Hours - median	Outside Hours - median	All Hours - median
	NO ₂	0.40	0.41	0.40
Office	PM _{2.5}	0.71	0.50	0.58
	PM ₁₀	1.01	0.57	0.69
	NO ₂	0.71	0.48	0.57
Hospital	PM _{2.5}	0.09	0.05	0.07
	PM ₁₀	0.25	0.09	0.16
School	NO ₂	0.61	0.39	0.43
	PM _{2.5}	0.37	0.29	0.31
	PM ₁₀	0.63	0.39	0.45





295 Figure 3: Comparison of I/O ratio measured by hypothetical 5-day passive sampling (black) and continuous sampling based 296 upon core hours only (red), across full 6-8month monitoring campaigns.

297 3.4 Variations in between and within apartments

The variable and dynamic nature of I/O ratios can be further explored by looking at data across 6-12 298 299 months in cumulative distribution plots from 18 individual apartments (Figure 4). Initially, it should 300 be noted that there is both a significant range in the I/O ratio defined within each individual apartment over the measurement period, but also between apartments and between the two 301 302 different developments as a whole. Differences may be observed in the distribution of I/O ratios at 303 values less than 1, generally more reflective of the relationship between indoor and outdoor air, and 304 in the proportion of time spent with an I/O ratio greater than 1, a reflection of the magnitude,

305 frequency and decay rate of indoor sources.

306 Two cases are highlighted in the left image of Figure 4. Case A reveals significant periods in which

307 the I/O ratio is greater than 1, with 24% of total recorded time spent above this threshold. In

contrast, case B exceeds an I/O ratio above 1 for just 2.7% of the total monitored period. This is a 308 309 clear response to the strength, duration and frequency of indoor sources experienced in each

310 apartment.

311 Cumulative frequency plots can be seen for I/O ratios for NO_2 in Figure 5. Here, apartment C is

highlighted as having a significantly lower I/O ratio when the I/O ratio is less than 1. Apartment C 312

313 had a window open in the living room just 19% of the time, compared to an average of 46% of time

314 across all apartments, indicating a lower rate of ingress from the external environment.

Previous studies have typically defined buildings with I/O ratios below 0.8 as having few internal 315

sources and above 1.2 as likely having significant indoor sources (Deng et al., 2017). Here it can be 316

317 seen that using static I/O ratios in such definitions may be a simplification of a range of modes in

318 each apartment. This may include various use of mechanical ventilation, configurations of openable

- windows and internal partitions and activities within the apartments. The measurement of I/O ratios 319
- 320 over extended periods of time therefore can be useful in understanding the variation of I/O ratio
- 321 under different modes of operation and the significance of internal sources.



Figure 4: Cumulative frequency plots for I/O (PM_{2.5}) for individual flats within two developments. Individual apartments are
 represented with grey lines, mean of all apartments in blue/red.

325

322





Figure 5: Cumulative frequency plots for I/O (NO₂) for individual flats within two developments. Individual apartments are
 represented with grey lines, mean of all apartments in blue/red. Fewer flats recorded NO₂ across the full period than
 particulate matter (N=7).

330 3.5 Variations in I/O ratios due to occupant behaviour – window use

The role of occupant behaviour as a driving force of varying I/O ratios can be more directly investigated through window monitoring. Measurements in the 18 apartments were accompanied by recordings of both window use (open/shut) and occupancy. This allows a more direct examination of occupant actions upon the I/O ratio and on indoor air quality more generally.

Table 4 shows the median I/O ratio under a range of conditions between the apartments. Firstly, I/O ratios show significant increases between periods in which windows are closed or open, increasing from 0.38 to 0.63 for PM_{2.5}, 0.37 to 0.74 for PM₁₀ and 0.42 to 0.69 for NO₂. This indicates the strong
 influence of ventilation practices and occupant behaviours within apartments.

339 Additionally, passive infrared (PIR) and CO_2 sensors can be used to help determine occupancy. The 340 most significant difference between occupied and unoccupied periods can again be see for $PM_{10}(0.35-$ 341 0.60). Given the I/O ratio only increases at this higher particle size, it is thought this largely relates to 342 particle resuspension during occupied periods. Further, infiltration factors may be obtained by 343 selecting periods in which both windows are closed and the apartment has been unoccupied for at 344 least the proceeding 4 hours (to remove lingering indoor sources). These values are again significantly lower than within other periods and may prove useful in understanding the proportion of pollutant 345 346 ingress that occurs via uncontrolled infiltration through the fabric.

347

Table 4: I/O in apartments	under different	conditions.
----------------------------	-----------------	-------------

		Median I/O	<u>x</u>
	PM _{2.5}	PM10	NO ₂
All Data	0.47	0.49	0.51
Windows Open	0.63	0.74	0.69
Windows Closed	0.38	0.37	0.42
Occupied	0.49	0.60	0.52
Unoccupied	0.47	0.35	0.43
Windows Closed –	0.19	0.11	0.13
Unoccupied for 4 hr			

348

The impact of window actions can be seen across short timescales in Figure 6 and Figure 7. Here, the average indoor concentration or I/O ratio is shown for CO₂, PM_{2.5}, NO₂ and TVOCs as a function of the duration a window has been open (Figure 6) or shut (Figure 7). Whilst opening a window leads to reductions in both CO₂ and TVOCs, the I/O for both PM_{2.5} and NO₂ is seen to increase in response, indicating a higher proportion of outdoor pollutants enter the indoor environment.

When a window is closed, the opposite effect can be observed. Increasing CO₂ and TVOC concentrations are accompanied by reducing I/O ratios in both NO₂ and PM_{2.5}. Across shorter measurements in both open and closed states, both Cyrys et al., (2004) and Yin et al., (2019) noted similar increases in I/O ratios for particulate matter when widows were open. This is further evidence not only in the dynamic nature of I/O ratios but also on the direct influence of occupant behaviour, leading to seasonal effects seen elsewhere.

360

361

- 363
- 364
- 365





from whole monitoring periods within the 18 apartments from moment of window opening/closing and the subsequent 3
 hours. Data from full monitored periods (June-May).



373

374 Figure 7: Impact of shutting a window upon CO2, PM2.5, NO2, TVOC, I/O ratio (PM2.5) and I/O ratio (NO2). Aggregated

response from whole monitoring periods within the 18 apartments from moment of window opening/closing and the
 subsequent 3 hours. Data from full monitored periods (June-May).

377 4. Discussion

378 4.1 Static I/O ratios

379 Previous studies have helped to establish the relationship between I/O ratios and ventilation types

380 (Ho et al., 2004; Hu and Zhao, 2020), building locations (Nunes et al., 2016), airtightness

- 381 (Chatzidiakou et al., 2015; Poupard et al., 2005) and therefore identifying the potential for higher
- 382 exposure in such settings. In particular, cheaper, non-intrusive passive sampling has allowed larger
- 383 samples of buildings to be assessed, helping to determine values or distributions of I/O ratios to be
- 384 used within modelling or epidemiological studies.
- 385 However, as analysis here has shown, passive sampling over several full 24-hour periods is likely to
- lead to a sampling period bias in many buildings. In most cases, this will have the effect of
- 387 underestimating the I/O ratio which is associated with core operation hours and most strongly
- 388 associated with exposure. Significantly, static measurements of I/O ratios using passive sampling

- 389 may also dampen the difference between buildings with significant operational differences. For
- example, if we considered two identical schools, one with a 5 L/s.p ventilation rate the other
- adopting 10 L/s.p. Across occupied hours we would expect a significant increase in penetration of
- 392 pollution from outdoors into the building with higher ventilation rate (assuming that the ventilation
- air is 100% outdoor air and not filtered). However, through static measurements this difference is
- largely diminished through the inclusion of 16 unoccupied hours alongside the 8 occupied hours.
- This means that such passively measured values should not be used in further modelling or epidemiological studies and may not accurately reflect a building's operation or exposure of
- occupants. Equally, in the absence of contextual details (e.g. ventilation rate, hours of operation,
- 398 window status, weather conditions) the use of such values is prone to mis-interpretation.

399 4.2 Dynamic I/O ratios

400 Assessing I/O ratios on a dynamic basis will allow data to be filtered and sampling period bias to be

401 avoided, with the estimated I/O values more closely linked to exposure during occupation. These

402 may therefore provide more suitable values for modelling and epidemiological studies. Sensor costs,

403 performance and intrusiveness have limited the length and scale of such measurements to date,

- 404 although technological improvements in air quality sensors may overcome these challenges and405 capture longer term behaviours.
- 406 Beyond the determination of the I/O for use in wider studies, it may also be argued that a
- 407 dynamically assessed I/O ratio may be useful as a building performance metric. This may provide
- 408 useful information on an individual building basis. For example, dynamic I/O ratios demonstrated
- 409 here have shown the impact on indoor air quality of opening windows, CO₂ demand-controlled
- 410 ventilation (with and without filtration), ingress from infiltration, cases with significant indoor
- sources and the influence of particulate resuspension during occupancy. This might lead to suitable
- 412 interventions, for example scheduling ventilation rates around external pollution, control based on
- 413 external pollutants, optimising window opening or improving extraction to reduce internal peaks.

414 4.3 Limitations in I/O as a metric

The broader limitations of a I/O, whether analysed statically or dynamically, can also be considered. In their review, Chen and Zhao, (2011) concluded that the considerable variation of I/O ratios meant that they were 'hardly helpful for understanding the indoor-outdoor relationship', with the penetration factor a more relevant metric. However, given the influence of ventilation and window opening on exposure during occupied hours, it would seem understanding ventilation and occupant behaviours remains crucial.

421

422 4.4 I/O ratio as a physical or personal variable

423 The I/O ratio has been described as a building modifier, representing the impact the building fabric 424 and services might have on outdoor pollutants reaching the indoor environment. However, results 425 here indicate that two key influential factors are related to the occupant(s) rather than the building 426 itself. Both indoor generation and window opening, are shown to be influential variables for the I/O 427 ratio. However, both vary significantly between buildings or apartments and may be linked to a much 428 wider range of social-physical factors. Within dwellings, particularly where the occupant has 429 significant control over ventilation, the I/O ratio may therefore provide a much stronger description 430 of the occupants than the building itself.

431 4.5 Uncertainty in I/O

432 Uncertainty in estimated I/O ratios is a result of the uncertainty in both indoor and outdoor433 measurements. Error propagation rules would define the uncertainty in an I/O ratio as:

434
$$\frac{\delta(I/O)}{I/O} = \left[\left(\frac{\delta C_{in}}{C_{in}} \right)^2 + \left(\frac{\delta C_{out}}{C_{out}} \right)^2 \right]^{1/2}$$

435 Where, $\delta(I/O)$, δC_{in} and δC_{out} are the uncertainties in the I/O ratio, measured indoor and measured 436 outdoor pollutant concentrations.

437 The EU Directive 2008/50/EC defines acceptable levels of uncertainty for indicative ambient air quality 438 measurements of particulate matter (50%) and sulphur dioxide, nitrogen dioxide and carbon 439 monoxide (25%) and Ozone (30%). If both indoor and outdoor measurements incorporated 440 uncertainties at these limits, this would translate to overall significant uncertainties in I/O estimates 441 for particulate matter (70%), sulphur dioxide, nitrogen dioxide and carbon monoxide (35%) and Ozone 442 (42%). Clearly, even with good quality instrumentation and stable conditions, any estimated I/O ratio 443 will contain significant measurement uncertainty, particularly at low concentrations. This supports the 444 argument that the dynamic I/O ratio works better as an informative metric, rather than an absolute 445 value.

446

447 4.6 Lack of equivalency between measurements.

448 Measurement uncertainties are further compounded by the lack of equivalency between 449 measurement systems, particularly with lower cost portable systems (Karagulian et al., 2019). Such 450 inequivalent indoor and outdoor measurements will only serve to increase the uncertainty in I/O 451 estimates. Even if identical measurement systems are used in both indoor and outdoor 452 measurements, their performance may alter with the varying composition of pollutants and their 453 respective environments, with humidity and temperature bias impacting measurements of many 454 pollutants (Crilley et al., 2018). An example within this study is the limitations of low-cost sensors to 455 capture fine-mode aerosols in the urban environment. Where compositions of outdoor and indoor air 456 differ, these limitations in measurements may lead to further bias.

Estimates of I/O ratios have also been determined with C_{out} based upon local central monitoring stations (e.g. Braniš et al., 2009; Tang et al., 2018). Whilst this may improve the accuracy of outdoor measurements, issues around equivalency remain and such approaches may introduce further spatial uncertainty between the central monitoring station and at the site of a case study building.

461 Finally, there may be significant definitional uncertainty in estimated I/O ratios. Definitional

462 uncertainty is defined by JCGM 100 as 'Component of measurement uncertainty resulting from the

463 finite amount of detail in the definition of a measurand'. In this case, it applies to the uncertainty in

the definition of the I/O ratio and in particular the location of an external measurement. For

- 465 example, an I/O ratio of an apartment defined by an external measurement at ground level will not
- 466 be equivalent to an I/O ratio defined by an external measurement at the height of the apartment.
- 467 Whilst urban background and roadside measurements are generally well defined, what external
- 468 measurement point is most appropriate or indeed informative is less clear, particularly with mixed
- 469 mode ventilation strategies, and may be a function of ventilation intake locations, natural ventilation
- 470 openings, local spatial effects and practical constraints.
- 471

472 *4.7 Comparisons to F_{inf}*

473 Infiltration factors have been cited as less variable and therefore more useful in health studies than
474 I/O ratios (Poupard et al., 2005). There is however an important note of terminology.

475 Within the built environment, infiltration is typically defined as uncontrolled air movement between 476 indoor and outdoor environments (e.g. via cracks and gaps in the building fabric). On the other hand, 477 ventilation describes the controlled provision of outdoor air either via mechanical systems or via 478 openable windows etc. The infiltration factor is perhaps therefore a misnomer. The air exchange rate 479 of a building is tied to both infiltration and ventilation rates. Particularly in modern, low-energy, 480 airtight buildings, it is the ventilation component that is likely to dominate, particularly during 481 occupied hours. The infiltration factor is therefore closely linked to ventilation practices and as such, 482 to the operation of a building and actions of its occupants. Given the strong associated with ventilation 483 practices, F_{inf} is again expected to be a function of both a building and its occupants, varying in similar 484 ways to the I/O ratio investigated here.

485

486 **5. Conclusions**

487 The study has analysed long term indoor and outdoor pollutant monitoring in a hospital, school, office 488 and 18 apartments. The results indicated strong diurnal and seasonal variation in I/O ratios for 489 particulate matter and nitrogen dioxide. Diurnal variation was shown to reveal the potential for 490 significant bias if static I/O ratios are measured through passive sampling. As a result, static I/O 491 measurements may not reasonably account for, and are likely to underestimate, the expected 492 exposure levels in associated indoor environments. This is likely to result in many of the I/O ratios and 493 infiltration factors previously estimated to underestimate actual indoor concentrations during 494 occupied periods.

495 The results presented indicate how I/O ratios are strongly influenced by ventilation rates, window 496 opening actions and by the occupancy patterns and activities within a building. Therefore I/O ratios 497 should not be thought of as a physical building parameter, but rather a metric that describes both the 498 building and the behaviour of the occupants that occupy and manage it. Whilst academic literature 499 has focused on measuring and reporting I/O ratios over the past four decades, the dynamic nature 500 and behavioural component of I/O ratios means that drawing together and making use of reported 501 I/O ratios is not straightforward and it is perhaps unsurprising that reported values demonstrate too 502 much variation to often be useful. Static I/O ratios hide this dynamic behaviour and as a result, such 503 comparisons or compiled data sets may mean little without contextual information, both on the 504 measurement made and the state of the building during the measurement (e.g. ventilation rate, 505 window opening, activities, occupancy).

However, it is argued that the I/O ratio, when evaluated dynamically from continuous measurements,
provides a useful building operation parameter. Dynamic I/O ratios may help describe the relationship
the building has with the outdoor environment and importantly the role of building operation and
occupant behaviour. This can help better understand ventilation strategies, schedules, the influence
of occupant behaviour and the significance of indoor sources. The use of a dynamic I/O ratio may
therefore provide effective information for designing and operating buildings to improve indoor air
quality.

- 513
- 514
- 515

516 Acknowledgements

517 Funding for this work was provided by the European Institute of Technology-Digital (EIT-Digital), 518 activity number 19144. This work was part of the EIT project "Quasimodo" and the study described

- 519 was carried out in partnership with Philips, IMEC and Forum Virium Helsinki.
- 520

521 References

- Allen Ryan W., Adar Sara D., Avol Ed, Cohen Martin, Curl Cynthia L., Larson Timothy, Liu L.-J. Sally,
 Sheppard Lianne, Kaufman Joel D., 2012. Modeling the Residential Infiltration of Outdoor
 PM2.5 in the Multi-Ethnic Study of Atherosclerosis and Air Pollution (MESA Air). Environ.
 Health Perspect. 120, 824–830. https://doi.org/10.1289/ehp.1104447
- Batterman, S., 2017. Review and Extension of CO2-Based Methods to Determine Ventilation Rates
 with Application to School Classrooms. Int. J. Environ. Res. Public. Health 14, 145.
 https://doi.org/10.3390/ijerph14020145
- Blondeau, P., Iordache, V., Poupard, O., Genin, D., Allard, F., 2005. Relationship between outdoor
 and indoor air quality in eight French schools. Indoor Air 15, 2–12.
- Borrego, C., Tchepel, O., Costa, A.M., Martins, H., Ferreira, J., Miranda, A.I., 2006. Traffic-related
 particulate air pollution exposure in urban areas. Atmos. Environ. 40, 7205–7214.
 https://doi.org/10.1016/j.atmosenv.2006.06.020
- Branco, P.T.B.S., Alvim-Ferraz, M.C.M., Martins, F.G., Sousa, S.I.V., 2019. Quantifying indoor air
 quality determinants in urban and rural nursery and primary schools. Environ. Res. 176,
 108534. https://doi.org/10.1016/j.envres.2019.108534
- Branco, P.T.B.S., Alvim-Ferraz, M.C.M., Martins, F.G., Sousa, S.I.V., 2014. Indoor air quality in urban nurseries at Porto city: Particulate matter assessment. Atmos. Environ. 84, 133–143.
 https://doi.org/10.1016/j.atmosenv.2013.11.035
- Braniš, M., Šafránek, J., Hytychová, A., 2009. Exposure of children to airborne particulate matter of
 different size fractions during indoor physical education at school. Build. Environ. 44, 1246–
 1252. https://doi.org/10.1016/j.buildenv.2008.09.010
- 543 Challoner, A., Gill, L., 2014. Indoor/outdoor air pollution relationships in ten commercial buildings:
 544 PM2.5 and NO2. Build. Environ. 80, 159–173.
- 545 https://doi.org/10.1016/j.buildenv.2014.05.032
- 546 Chao, C.Y.H., Tung, T.C., 2001. An empirical model for outdoor contaminant transmission into
 547 residential buildings and experimental verification. Atmos. Environ. 35, 1585–1596.
 548 https://doi.org/10.1016/S1352-2310(00)00458-1
- Chatzidiakou, L., Krause, A., Han, Y., Chen, W., Yan, L., Popoola, O.A.M., Kellaway, M., Wu, Y., Liu, J.,
 Hu, M., Barratt, B., Kelly, F.J., Zhu, T., Jones, R.L., 2020. Using low-cost sensor technologies
 and advanced computational methods to improve dose estimations in health panel studies:
 results of the AIRLESS project. J. Expo. Sci. Environ. Epidemiol. 30, 981–989.
 https://doi.org/10.1038/s41370-020-0259-6
- Chatzidiakou, L., Krause, A., Popoola, O.A.M., Antonio, A.D., Kellaway, M., Han, Y., Squires, F.A.,
 Wang, T., Zhang, H., Wang, Q., Fan, Y., Chen, S., Hu, M., Quint, J.K., Barratt, B., Kelly, F.J.,
 Zhu, T., Jones, R.L., 2019. Characterising low-cost sensors in highly portable platforms to
 quantify personal exposure in diverse environments. Atmospheric Meas. Tech. 12, 4643–
 4657. https://doi.org/10.5194/amt-12-4643-2019
- Chatzidiakou, L., Mumovic, D., Summerfield, A.J., Tàubel, M., Hyvärinen, A., 2015. Indoor air quality
 in London schools. Part 2: long-term integrated assessment. Intell. Build. Int. 7, 130–146.
 https://doi.org/10.1080/17508975.2014.918871
- 562 Chen, C., Zhao, B., 2011. Review of relationship between indoor and outdoor particles: I/O ratio,
 563 infiltration factor and penetration factor. Atmos. Environ. 45, 275–288.
 564 https://doi.org/10.1016/j.atmosenv.2010.09.048

565	Chen, C., Zhao, B., Weschler, C.J., 2012. Indoor Exposure to "Outdoor PM10": Assessing Its Influence
566	on the Relationship Between PM10 and Short-term Mortality in U.S. Cities. Epidemiology 23,
567	870–878. https://doi.org/10.1097/EDE.0b013e31826b800e
568	Cohen, M.A., Adar, S.D., Allen, R.W., Avol, E., Curl, C.L., Gould, T., Hardie, D., Ho, A., Kinney, P.,
569	Larson, T.V., Sampson, P., Sheppard, L., Stukovsky, K.D., Swan, S.S., Liu, LJ.S., Kaufman, J.D.,
570	2009. Approach to Estimating Participant Pollutant Exposures in the Multi-Ethnic Study of
571	Atherosclerosis and Air Pollution (MESA Air). Environ. Sci. Technol. 43, 4687–4693.
572	https://doi.org/10.1021/es8030837
573	Cooper, E., Wang, Y., Stamp, S., Burman, E., Mumovic, D., 2021. Use of portable air purifiers in
574	homes: Operating behaviour, effect on indoor PM2.5 and perceived indoor air quality. Build.
575	Environ. 191, 107621. https://doi.org/10.1016/j.buildenv.2021.107621
576	Crilley, L.R., Shaw, M., Pound, R., Kramer, L.J., Price, R., Young, S., Lewis, A.C., Pope, F.D., 2018.
577	Evaluation of a low-cost optical particle counter (Alphasense OPC-N2) for ambient air
578	monitoring. Atmospheric Meas. Tech. 11, 709–720. https://doi.org/10.5194/amt-11-709-
579	2018
580	Cvrvs, J., Pitz, M., Bischof, W., Wichmann, HE., Heinrich, J., 2004, Relationship between indoor and
581	outdoor levels of fine particle mass, particle number concentrations and black smoke under
582	different ventilation conditions. J. Expo. Sci. Environ. Epidemiol. 14, 275–283.
583	https://doi.org/10.1038/si.jea.7500317
584	Deng G. Li Z. Wang Zhichao Gao L. Xu Z. Li L. Wang Zhivong 2017 Indoor/outdoor
585	relationship of PM2.5 concentration in typical buildings with and without air cleaning in
586	Beijing, Indoor Built Environ, 26, 60–68, https://doi.org/10.1177/1420326X15604349
587	Diapouli, E., Chaloulakou, A., Koutrakis, P., 2013a. Estimating the concentration of indoor particles of
588	outdoor origin: A review 1 Air Waste Manag. Assoc. 63, 1113–1129.
589	https://doi.org/10.1080/10962247.2013.791649
590	Diapouli, E., Chaloulakou, A., Koutrakis, P., 2013b. Estimating the concentration of indoor particles of
591	outdoor origin: A review 1 Air Waste Manag. Assoc. 63, 1113–1129.
592	https://doi.org/10.1080/10962247.2013.791649
593	Dimitroulonoulou C 2012 Ventilation in European dwellings: A review Build Environ
594	International Workshop on Ventilation Comfort and Health in Transport Vehicles 47 109–
595	125. https://doi.org/10.1016/i.buildeny.2011.07.016
596	Dimitroulonoulou C Bartzis I 2014 Ventilation rates in European office huildings: A review
597	Ventilation rates in European office buildings: A review Indoor Built Environ 23 5–25
598	https://doi.org/10.1177/1420326X13481786
599	Fabian P. Adamkiewicz, G. Lewy LL 2012 Simulating indoor concentrations of NO2 and PM2.5 in
600	multifamily housing for use in health-based intervention modeling. Indoor Air 22, 12–23
601	https://doi.org/10.1111/i.1600-0668.2011.00742.v
602	Fazli T Dong X Fu IS Stephens B 2021 Predicting IIS Residential Building Energy Use and
603	Indoor Pollutant Exposures in the Mid-21st Century Environ Sci Technol 55 3219–3228
604	https://doi.org/10.1021/acs.est.0c06308
605	Grau-Boyé I. Strlič M. 2013 Fine particulate matter in indoor cultural heritage: a literature review
606	Herit Sci 1 & https://doi.org/10.1186/2050-7445-1-8
607	How K E Coo LL Harrison P.M. Lee S.C. Bay K.K. 2004 Indeer/outdoor relationships of organic
608	carbon (OC) and elemental carbon (EC) in PM2.5 in roadside environment of Hong Kong
600	Atmos Environ 28, 6227–6225 https://doi.org/10.1016/j.atmosony.2004.08.007
610	Hoek C Krishnan R.M. Boelen R. Beters A. Ostro R. Brunekreef R. Kaufman I.D. 2013 Long
611	term air pollution exposure and cardio, recoiratory mortality; a roviou, Environ, Health 12
612	12, https://doi.org/10.1186/1476-0608-12.42
612	43. IIIIps.//WUI.UIg/WUI.TOU/14/0-003A-12-43 Hu V Zhao R 2020 Relationship between indeer and outdeer NO2: A review Ruild Environ
614	106909. https://doi.org/10.1016/j.buildenv.2020.106909

615 Huang, L., Pu, Z., Li, M., Sundell, J., 2015. Characterizing the Indoor-Outdoor Relationship of Fine 616 Particulate Matter in Non-Heating Season for Urban Residences in Beijing. PloS One 10, 617 e0138559. https://doi.org/10.1371/journal.pone.0138559 618 Janssen, N.A.H., Schwartz, J., Zanobetti, A., Suh, H.H., 2002. Air conditioning and source-specific 619 particles as modifiers of the effect of PM(10) on hospital admissions for heart and lung 620 disease. Environ. Health Perspect. 110, 43–49. Jeremy A. Sarnat, *, Christopher M. Long, ‡, Petros Koutrakis, †, Brent A. Coull, §, Joel Schwartz, † 621 and, Suh⁺, H.H., 2002. Using Sulfur as a Tracer of Outdoor Fine Particulate Matter [WWW 622 623 Document]. https://doi.org/10.1021/es025796b Ji, W., Zhao, B., 2015. Estimating Mortality Derived from Indoor Exposure to Particles of Outdoor 624 625 Origin. PLOS ONE 10, e0124238. https://doi.org/10.1371/journal.pone.0124238 Johnston, D., Stafford, A., 2016. Estimating the background ventilation rates in new-build UK 626 627 dwellings – Is n50/20 appropriate?: Indoor Built Environ. 628 https://doi.org/10.1177/1420326X15626234 629 Jones, N.C., Thornton, C.A., Mark, D., Harrison, R.M., 2000. Indoor/outdoor relationships of 630 particulate matter in domestic homes with roadside, urban and rural locations. Atmos. 631 Environ. 34, 2603–2612. https://doi.org/10.1016/S1352-2310(99)00489-6 632 Kabirikopaei, A., Lau, J., 2020. Uncertainty analysis of various CO2-Based tracer-gas methods for 633 estimating seasonal ventilation rates in classrooms with different mechanical systems. Build. 634 Environ. 179, 107003. https://doi.org/10.1016/j.buildenv.2020.107003 635 Kalimeri, K.K., Bartzis, J.G., Sakellaris, I.A., de Oliveira Fernandes, E., 2019. Investigation of the PM2.5, NO2 and O3 I/O ratios for office and school microenvironments. Environ. Res. 179, 636 637 108791. https://doi.org/10.1016/j.envres.2019.108791 638 Karagulian, F., Gerboles, M., Barbiere, M., Kotsev, A., Lagler, F., Borowiak, A., European Commission, 639 Joint Research Centre, 2019. Review of sensors for air quality monitoring. 640 Kim, J., Hong, T., Lee, M., Jeong, K., 2019. Analyzing the real-time indoor environmental quality 641 factors considering the influence of the building occupants' behaviors and the ventilation. 642 Build. Environ. 156, 99–109. https://doi.org/10.1016/j.buildenv.2019.04.003 643 Lazaridis, M., Katsivela, E., Kopanakis, I., Raisi, L., Panagiaris, G., 2015. Indoor/outdoor particulate 644 matter concentrations and microbial load in cultural heritage collections. Herit. Sci. 3, 34. 645 https://doi.org/10.1186/s40494-015-0063-0 646 Lelieveld, J., Evans, J.S., Fnais, M., Giannadaki, D., Pozzer, A., 2015. The contribution of outdoor air 647 pollution sources to premature mortality on a global scale. Nature 525, 367–371. 648 https://doi.org/10.1038/nature15371 649 Levy, J.I., 1998. Impact of Residential Nitrogen Dioxide Exposure on Personal Exposure: An 650 International Study. J. Air Waste Manag. Assoc. 48, 553–560. 651 https://doi.org/10.1080/10473289.1998.10463704 652 Li, C.-S., Lin, C.-H., 2003. Carbon profile of residential indoor PM1 and PM2.5 in the subtropical 653 region. Atmos. Environ. 37, 881-888. https://doi.org/10.1016/S1352-2310(02)00998-6 654 Li, N., Friedrich, R., 2019. Methodology for Estimating the Lifelong Exposure to PM2.5 and NO2—The 655 Application to European Population Subgroups. Atmosphere 10, 507. 656 https://doi.org/10.3390/atmos10090507 Long, C.M., Sarnat, J.A., 2004. Indoor-Outdoor Relationships and Infiltration Behavior of Elemental 657 658 Components of Outdoor PM2.5 for Boston-Area Homes. Aerosol Sci. Technol. 38, 91–104. 659 https://doi.org/10.1080/027868290502281 660 Majd, E., McCormack, M., Davis, M., Curriero, F., Berman, J., Connolly, F., Leaf, P., Rule, A., Green, T., 661 Clemons-Erby, D., Gummerson, C., Koehler, K., 2019. Indoor air quality in inner-city schools and its associations with building characteristics and environmental factors. Environ. Res. 662 663 170, 83–91. https://doi.org/10.1016/j.envres.2018.12.012 664 Mandin, C., Trantallidi, M., Cattaneo, A., Canha, N., Mihucz, V.G., Szigeti, T., Mabilia, R., Perreca, E., Spinazzè, A., Fossati, S., De Kluizenaar, Y., Cornelissen, E., Sakellaris, I., Saraga, D., Hänninen, 665

666	O., De Oliveira Fernandes, E., Ventura, G., Wolkoff, P., Carrer, P., Bartzis, J., 2017.
667	Assessment of indoor air quality in office buildings across Europe – The OFFICAIR study. Sci.
668	Total Environ. 579, 169–178. https://doi.org/10.1016/j.scitotenv.2016.10.238
669	Martuzevicius, D., Grinshpun, S.A., Lee, T., Hu, S., Biswas, P., Reponen, T., LeMasters, G., 2008.
670	Traffic-related PM2.5 aerosol in residential houses located near major highways: Indoor
671	versus outdoor concentrations. Atmos. Environ. 42, 6575–6585.
672	https://doi.org/10.1016/j.atmosenv.2008.05.009
673	Mohammed, M.O.A., Song, WW., Ma, WL., Li, WL., Ambuchi, J.J., Thabit, M., Li, YF., 2015.
674	Trends in indoor–outdoor PM2.5 research: A systematic review of studies conducted during
675	the last decade (2003–2013). Atmospheric Pollut. Res. 6, 893–903.
676	https://doi.org/10.5094/APR.2015.099
677	Monn, C., Fuchs, A., Hogger, D., Junker, M., Kogelschatz, D., Roth, N., Wanner, H.U., 1997.
678	Particulate matter less than 10 mu m (PM10) and fine particles less than 2.5 mu m (PM2.5):
679	relationships between indoor, outdoor and personal concentrations. Sci. Total Environ. 208,
680	15–21. https://doi.org/10.1016/S0048-9697(97)00271-4
681	Nunes, R.A.O., Branco, P.T.B.S., Alvim-Ferraz, M.C.M., Martins, F.G., Sousa, S.I.V., 2016. Gaseous
682	pollutants on rural and urban nursery schools in Northern Portugal. Environ. Pollut., Special
683	Issue: Urban Health and Wellbeing 208, 2–15. https://doi.org/10.1016/j.envpol.2015.07.018
684	Øie, L., Stymne, H., Boman, CA., Hellstrand, V., 1998. The Ventilation Rate of 344 Oslo Residences.
685	Indoor Air 8, 190–196. https://doi.org/10.1111/j.1600-0668.1998.t01-1-00006.x
686	Pedersen, M., Giorgis-Allemand, L., Bernard, C., Aguilera, I., Andersen, AM.N., Ballester, F., Beelen,
687	R.M.J., Chatzi, L., Cirach, M., Danileviciute, A., Dedele, A., van Eijsden, M., Estarlich, M.,
688	Fernandez-Somoano, A., Fernandez, M.F., Forastiere, F., Gehring, U., Grazuleviciene, R.,
689	Gruzieva, O., Heude, B., Hoek, G., de Hoogh, K., van den Hooven, E.H., Haberg, S.E., Jaddoe,
690	V.W.V., Kluemper, C., Korek, M., Kraemer, U., Lerchundi, A., Lepeule, J., Nafstad, P., Nystad,
691	W., Patelarou, E., Porta, D., Postma, D., Raaschou-Nielsen, O., Rudnai, P., Sunyer, J.,
692	Stephanou, E., Sorensen, M., Thiering, E., Tuffnell, D., Varro, M.J., Vrijkotte, T.G.M., Wijga,
693	A., Wilhelm, M., Wright, J., Nieuwenhuijsen, M.J., Pershagen, G., Brunekreef, B., Kogevinas,
694	M., Slama, R., 2013. Ambient air pollution and low birthweight: a European cohort study
695	(ESCAPE). Lancet Respir. Med. 1, 695–704. https://doi.org/10.1016/S2213-2600(13)70192-9
696	Poupard, O., Blondeau, P., Iordache, V., Allard, F., 2005. Statistical analysis of parameters influencing
697	the relationship between outdoor and indoor air quality in schools. Atmos. Environ. 39,
698	2071–2080. https://doi.org/10.1016/j.atmosenv.2004.12.016
699	R Core Team, 2021. R: A Language and Environment for Statistical Computing. R Foundation for
700	Statistical Computing, Vienna, Austria.
701	Rojas-Bracho, L., Suh, H.H., Oyola, P., Koutrakis, P., 2002. Measurements of children's exposures to
702	particles and nitrogen dioxide in Santiago, Chile. Sci. Total Environ. 287, 249–264.
703	https://doi.org/10.1016/S0048-9697(01)00987-1
704	Salonen, H., Salthammer, T., Morawska, L., 2019. Human exposure to NO2 in school and office
705	indoor environments. Environ. Int. 130, 104887.
706	https://doi.org/10.1016/j.envint.2019.05.081
707	Samoli Evangelia, Stafoggia Massimo, Rodopoulou Sophia, Ostro Bart, Declercq Christophe,
708	Alessandrini Ester, Díaz Julio, Karanasiou Angeliki, Kelessis Apostolos G., Le Tertre Alain,
709	Pandolfi Paolo, Randi Giorgia, Scarinzi Cecilia, Zauli-Sajani Stefano, Katsouyanni Klea,
710	Forastiere Francesco, null null, 2013. Associations between Fine and Coarse Particles and
711	Mortality in Mediterranean Cities: Results from the MED-PARTICLES Project. Environ. Health
712	Perspect. 121, 932–938. https://doi.org/10.1289/ehp.1206124
713	Sarnat, J.A., Wilson, W.E., Strand, M., Brook, J., Wyzga, R., Lumley, T., 2007. Panel discussion review:
714	session 1 — exposure assessment and related errors in air pollution epidemiologic studies. J.
715	Expo. Sci. Environ. Epidemiol. 17, S75–S82. https://doi.org/10.1038/sj.jes.7500621

716 Schneider, P., Gebefugi, I., Richter, K., Wolke, G., Schnelle, J., Wichmann, H.E., Heinrich, J., 2001. 717 Indoor and outdoor BTX levels in German cities. Sci. Total Environ. 267, 41–51. 718 https://doi.org/10.1016/S0048-9697(00)00766-X Setton, E.M., Keller, C.P., Cloutier-Fisher, D., Hystad, P.W., 2008. Spatial variations in estimated 719 720 chronic exposure to traffic-related air pollution in working populations: A simulation. Int. J. 721 Health Geogr. 7, 39. https://doi.org/10.1186/1476-072X-7-39 722 Shaw, C., Boulic, M., Longley, I., Mitchell, T., Pierse, N., Howden-Chapman, P., 2020. The association 723 between indoor and outdoor NO2 levels: A case study in 50 residences in an urban 724 neighbourhood in New Zealand. Sustain. Cities Soc. 56, 102093. 725 https://doi.org/10.1016/j.scs.2020.102093 726 SINPHONIE, 2014. SINPHONIE – Schools Indoor Pollution and Health Observatory Network in Europe 727 - Final Report [WWW Document]. EU Sci. Hub - Eur. Comm. URL 728 https://ec.europa.eu/jrc/en/publication/eur-scientific-and-technical-research-729 reports/sinphonie-schools-indoor-pollution-and-health-observatory-network-europe-final-730 report (accessed 2.18.20). 731 Stamp, S., Burman, E., Shrubsole, C., Chatzidiakou, L., Mumovic, D., Davies, M., 2021. Seasonal 732 variations and the influence of ventilation rates on IAQ: A case study of five low-energy 733 London apartments. Indoor Built Environ. 1420326X211017175. 734 https://doi.org/10.1177/1420326X211017175 735 Stamp, S., Burman, E., Shrubsole, C., Chatzidiakou, L., Mumovic, D., Davies, M., 2020. Long-term, 736 continuous air quality monitoring in a cross-sectional study of three UK non-domestic 737 buildings. Build. Environ. 107071. https://doi.org/10.1016/j.buildenv.2020.107071 738 Stranger, M., Potgieter-Vermaak, S.S., Van Grieken, R., 2008. Characterization of indoor air quality in 739 primary schools in Antwerp, Belgium. Indoor Air 18, 454–463. 740 https://doi.org/10.1111/j.1600-0668.2008.00545.x 741 Tang, C.H., Garshick, E., Grady, S., Coull, B., Schwartz, J., Koutrakis, P., 2018. Development of a 742 modeling approach to estimate indoor-to-outdoor sulfur ratios and predict indoor PM 2.5 743 and black carbon concentrations for Eastern Massachusetts households. J. Expo. Sci. Environ. 744 Epidemiol. 28, 125–130. https://doi.org/10.1038/jes.2017.11 745 Taylor, J., Shrubsole, C., Symonds, P., Mackenzie, I., Davies, M., 2019. Application of an indoor air 746 pollution metamodel to a spatially-distributed housing stock. Sci. Total Environ. 667, 390-747 399. https://doi.org/10.1016/j.scitotenv.2019.02.341 748 Tung, T.C.W., Chao, C.Y.H., Burnett, J., 1999. A methodology to investigate the particulate 749 penetration coefficient through building shell. Atmos. Environ. 33, 881-893. 750 https://doi.org/10.1016/S1352-2310(98)00299-4 751 Viana, M., Rivas, I., Querol, X., Alastuey, A., Sunyer, J., Alvarez-Pedrerol, M., Bouso, L., Sioutas, C., 752 2014. Indoor/outdoor relationships and mass closure of quasi-ultrafine, accumulation and 753 coarse particles in Barcelona schools. Atmospheric Chem. Phys. 14, 4459–4472. 754 https://doi.org/10.5194/acp-14-4459-2014 755 Wallace, L., Williams, R., 2005. Use of personal-indoor-outdoor sulfur concentrations to estimate the 756 infiltration factor and outdoor exposure factor for individual homes and persons. Environ. 757 Sci. Technol. 39, 1707–1714. https://doi.org/10.1021/es049547u 758 Wang, F., Meng, D., Li, X., Tan, J., 2016. Indoor-outdoor relationships of PM2.5 in four residential 759 dwellings in winter in the Yangtze River Delta, China. Environ. Pollut. 215, 280–289. 760 https://doi.org/10.1016/j.envpol.2016.05.023 761 Weschler, C.J., 2000. Ozone in Indoor Environments: Concentration and Chemistry. Indoor Air 10, 762 269-288. https://doi.org/10.1034/j.1600-0668.2000.010004269.x Wheeler, A.J., Wallace, L.A., Kearney, J., Van Ryswyk, K., You, H., Kulka, R., Brook, J.R., Xu, X., 2011. 763 764 Personal, Indoor, and Outdoor Concentrations of Fine and Ultrafine Particles Using 765 Continuous Monitors in Multiple Residences. Aerosol Sci. Technol. 45, 1078–1089. 766 https://doi.org/10.1080/02786826.2011.580798

- WHO Europe, 2020. Methods for sampling and analysis of chemical pollutants in indoor air. WorldHealth Organisation.
- Wichmann, J., Lind, T., Nilsson, M.A.-M., Bellander, T., 2010. PM2.5, soot and NO2 indoor–outdoor
 relationships at homes, pre-schools and schools in Stockholm, Sweden. Atmos. Environ. 44,
 4536–4544. https://doi.org/10.1016/j.atmosenv.2010.08.023
- Wilson, W.E., Mage, D.T., Grant, L.D., 2000. Estimating Separately Personal Exposure to Ambient and
 Nonambient Particulate Matter for Epidemiology and Risk Assessment: Why and How. J. Air
 Waste Manag. Assoc. 50, 1167–1183. https://doi.org/10.1080/10473289.2000.10464164
- Yin, H., Liu, C., Zhang, L., Li, A., Ma, Z., 2019. Measurement and evaluation of indoor air quality in naturally ventilated residential buildings. Indoor Built Environ. 28, 1307–1323.
- 777 https://doi.org/10.1177/1420326X19833118 778

ounderer

Highlights: A critical evaluation of the dynamic nature of indoor-outdoor air quality ratios

- I/O ratios are shown to have significant dynamic variation, varying diurnally, seasonally as well as with building operation and occupant behaviour.
- Whilst static I/O ratios are widely reported, they hide this dynamic behaviour, limiting their interpretation and comparability..
- I/O ratios measured passively across integer 24 hour periods will be meaningless in many buildings.
- I/O ratios are shown to be strongly related to occupant behaviours and therefore represent a personal variable as much as a physical building metric.
- Dynamically assessed I/O ratios may prove useful as building performance metrics, indicating the relationship between indoor-outdoor air and the role of building operation.

John al Preven

Declaration of interests

 \boxtimes The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

□The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: