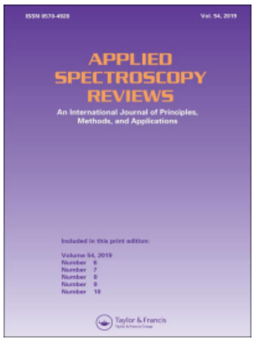

Applied Spectroscopy Reviews

ISSN: (Print) (Online) Journal homepage: https://www.tandfonline.com/loi/laps20

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To cite this article: Andrea Spinazzè, Francesca Borghi, Sabrina Rovelli, Victor G. Mihucz, Benjamin Bergmans, Andrea Cattaneo & Domenico M. Cavallo (2021): Combined and modular approaches for multicomponent monitoring of indoor air pollutants, Applied Spectroscopy Reviews, DOI: 10.1080/05704928.2021.1995405

To link to this article: https://doi.org/10.1080/05704928.2021.1995405

APPLIED SPECTROSCOPY REVIEWS
https://doi.org/10.1080/05704928.2021.1995405

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Combined and modular approaches for multicomponent monitoring of indoor air pollutants

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ABSTRACT
Readers will be introduced to the most common sampling and analytical techniques that are being used indoors for air quality monitoring though the simultaneous determination of several air pollutants (i.e., inorganic and organic gaseous contaminants and airborne particulate matter) and the possible challenges encountered by implementing them. Thus, i) personal exposure equipment; (ii) portable multipollutant monitors; and (iii) fixed monitoring devices have been reviewed. Besides, compiling the most common modular arrangements of instruments, multipollutant analysis approaches through time-integrated and continuous sampling performed during field campaigns organized in several indoor environments in the frame of collaborative research projects are hereby also presented. Our aim was not to compile a comprehensive review on approaches used for multipollutant indoor air quality monitoring but instead to give an overview of potentially useful instrumentation. Discussion on instruments useful for the determination of radicals and bioaerosols as well as use of sensors has been voluntary minimized.

KEYWORDS
active sampling; diffusive samplers; multipollutant exposure; ozone; particulate matter; VOC

1. Introduction

Epidemiological air pollution studies in the last decades have shown a correlation between increased daily air pollution and daily mortality. An increase in long term air contaminants levels reduces also the average life expectancy of urban populations. Mortality increases are clearly associated with fine particulate matter ($PM_{2.5}$), but also other pollutants, especially ozone.[1] Generally, epidemiological studies connect morbidity and mortality statistics to outdoor air pollution levels often measured at urban monitoring sites. However, people tend to spend most of their time indoors such as their homes, workplaces, and transportation vehicles. It is then necessary to develop effective

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Supplemental data for this article is available online at at http://doi/10.1080/05704928.2021.1995405.

policies and control strategies for a healthy indoor environment. The first and basic aspect to consider when planning an indoor monitoring campaign is the definition of the survey objectives. Thus, the sampling and analysis activities should be based on targeted and suitable monitoring strategies. Some possible reasons behind an indoor air monitoring campaign are the following: (i) objective assessment of indoor air quality (IAQ) that allows responding to complaints or problems brought to the attention of company executives, building managers, or competent authorities by the occupants; (ii) identification of the main indoor and outdoor sources of pollution and estimation of their relative contributions; (iii) monitoring as a result of ascertained indoor pollution situations; (iv) assessment of the effectiveness of any restoration/remedy measure adopted; (v) gathering specific information to facilitate the decision-making processes of risk assessors and competent authorities, when assessing the exposure of occupants to toxic substances, in relation to different reference times in a given indoor environment; (vi) IAQ assessment through the verification of the compliance with guide or reference values established by international (e.g., WHO - World Health Organization) or national organizations; (vii) collect useful information about the most important factors (indoor and outdoor sources, human activities, time and spatial trends, etc.) affecting IAQ for an effective risk management.

The assessment of human exposure or air pollution using guideline or limit values specific for occupational environments is well established,[2] although the reference levels to be applied to industrial workers are typically higher than those established for non-industrial (i.e., indoor) work settings. This measurement and assessment strategy is not generally well standardized for nonoccupational indoor environments, for which guideline values were set for some specific pollutants[3] and specific indoor air quality management approaches must be developed. The measured indoor concentration of the target pollutant is usually compared to the corresponding guideline level, considering the correct averaging time. However, combined exposures to multiple stressors are considered in toxicology, where in the simplest case the total estimated risk can be attributed to the sum of risks deriving from each pollutant in the mixture.[4,5] However, when health effects or discomfort complaints have multiple causes, there is no guarantee that the final effect would be attributed to the proper causes, because only the total effect of multiple stressors is seen or reported. When guideline values are set for a specific contaminant, the scientific evidence used as background information may include data on effects caused together with coexisting pollutants to some extent.[6] However, several air pollutants simultaneously present in the study place can contribute to the health stress in synergetic or additive ways. Therefore, there is an increasing interest to evaluate simultaneous and cumulative exposures; for these reasons novel techniques and approaches are needed to study the effects of these multiple stressor exposures. Particular care is needed to protect human health in situations where the concentration of each single pollutant is below the guideline, but the combination of these pollutants poses somewhat a possible health risk.[7–11]

The evaluation of bioaerosol (i.e., airborne particles of biological origin which may influence the health of humans by infectious, sensitizing, or toxic action) indoor concentrations or exposure is a complex task, considering the great diversity of bioaerosols, the limitations of the measurement methods available and the lack of occupational exposure limits. Further, bioaerosols active sampling with proper samplers usually lasts only a few minutes indoors and guideline values or limits for indoor air are not feasible. Passive sampling is mainly used to collect long-term information on bioaerosol contamination; however, to our knowledge, these solutions are not used in monitoring stations for a combined chemical and biological air monitoring. Besides, a specific online monitoring for indoor bioaerosols is not currently achievable. Further discussion on equipment used for bioaerosol analysis has been excluded from the present study, anyway.

The development of sensor technologies for air quality monitoring has experienced a considerable increase in the past decades and, to date, there are several commercially available multipollutant systems based on low-cost sensors.[12] Sensor-based measurements may be more appropriate indoors than outdoors since indoor environments are less variable to changes in ambient temperature. Among advantages offered by sensors, lower space, noise, price, and power requirements should be mentioned. However, these advantages are often accompanied by reduced accuracy, sensitivity and selectivity.[12,13] Nevertheless, the data so obtained are not comparable with those collected by multipollutant systems, used for large scale IAQ studies, especially when data are expressed as IAQ index (e.g., good-bad quality) and not as air concentrations. Therefore, sensor-based approaches will not either be discussed in the present manuscript. Although analytical techniques to determine radicals [i.e., OH, NO_3, HO_2 and RO_2 (organic peroxy radicals)] concentrations have evolved considerably recently,[13] radical-based analyses are still not very widespread, since radical species exist at typically part per trillion concentration or below and have lifetimes of the order of seconds. Therefore, such challenging measurements will not either be discussed in the present manuscript. However, radical concentrations can be estimated through determination of the oxidative potential (OP) of indoor $PM_{2.5}$.[14,15]

Thus, the objective of this work was to review instrumental approaches used for multipollutant determination indoors from personal exposure equipment to portable multipollutant monitoring units and fixed monitoring stations. Besides reviewing the most common modular arrangements of instruments, compiling information on multipollutant analysis approaches through time-integrated and continuous sampling performed during field campaigns organized in several indoor environments in the frame of collaborative research projects across Europe has also been aimed. Our aim was not to compile a comprehensive review on approaches used for multipollutant indoor air quality monitoring but instead to give an overview of potentially useful instrumentation to help planning a field campaign for the monitoring of indoor air quality.

2. Investigated indoor pollutants

Air pollution is caused by different substances exerting adverse impact on human health even in a synergistic way.[5,16] The major air pollutants are gaseous (inorganic and organic) contaminants as well as airborne particulate matter (PM). Since major gaseous pollutants are strongly linked together *via* transformations occurring through gas-phase reactions,[17] we briefly discuss their presence in indoor environment together and separately from PM.

2.1. Major indoor gaseous pollutants

Among inorganic gaseous pollutants ozone (O_3), nitrogen oxides (NO_x) and the related compounds [e.g., nitrous acid (HONO) and nitric acid (HNO_3)] should be mentioned. Also, ammonia (NH_3) concentration can achieve twice as larger values indoors than outdoors (e.g., 1-5 ppb).[18] Carbon dioxide is also important due to its relatively high concentrations in air and low water-solubility. Carbon monoxide evolves from incomplete combustion of fossil fuel.

Among organic gaseous pollutants, several volatile organic compounds (VOCs) characterized by a wide boiling point range (i.e., $50\,°C - 260\,°C$) such as alkanes, aromatics, terpenes, alcohols, aldehydes, ketones, esters, and halogenated compounds, have been identified indoors.[18] It has been estimated that a large number of (between 50 and 300) VOCs can be found in nonindustrial indoor air environments.[19,20] Among them, mono- and sesquiterpenes can widely be present in indoor environments but they can be easily oxidized (e.g., by O_3) because of containing unsaturated isoprene units [$CH_2{=}C(CH_3){-}CH{=}CH_2$].[21,22] Besides biogenic origin (emission by vegetation), monoterpenes such as α-pinene and d-limonene are commonly found in room fresheners, cleaning products, wood-based furniture coatings, etc.[17]. Aldehyde emission indoors is traced back to pressed wood products, urea-formaldehyde foam insulation, combustion and environmental tobacco smoke, textiles and glues.[19,23] An important aldehyde pollutant indoor is acrolein, a strong skin, eye and nasal irritant as a by-product of burnt edible oil.[13] However, its analysis is difficult containing a highly reactive C-C double bond.[24] Monocarboxylic acids such as acetic acid, formic acid[25] as well *n*-alkenoic acids (e.g., linoleic and oleic acids) are the most abundant indoors.[26] Recently, occurrence of several amino acids, fatty acids, triglycerides, unsaturated oils (e.g., squalene), low molecular weight VOCs (e.g., isoprene and lactic acid), NH_3, siloxanes emitted by human skin have been described indoors.[27–33] However, monitoring of such emissions requires *in situ* mass spectrometric techniques such as direct analysis real-time mass spectrometry.[34] Ozone (O_3) is one of the most important tropospheric gases and it is considered as the major oxidant indoors.[21] Exposure to O_3 may cause asthma as well as decreased lung function.[35] Most indoor ozone originates outdoors and enters with ventilation air, indoor emission can also contribute to increase indoor ozone concentrations;[21] for example, the indoor air purifiers can also generate $O_{3.}$[36] The O_3 concentrations indoors are usually lower than outdoor. However, these lower concentration levels indoors should not be overlooked because most people spend indoors large fractions of their time. Another O_3 source indoors is the corona discharge produced by photocopiers and laser printers, especially in workplaces.[37–39] Nevertheless, this source of subordinate importance compared to infiltration from outdoors. Indoor O_3 concentration can be reduced by gas-phase reactions. For example, O_3 reacts with isoprene units containing terpenes originating indoors from the personal care products, cleaning products as well as from timber, paints, polishes, etc. through ozonolysis.[37] Some of the ozonolysis products such as carbonyls (e.g., formaldehyde, acrolein) and hydroperoxides are known or supposed to adversely affect human health. Oxidation than ozonolysis between ozone and skin oil leads to the formation of the secondary organic aerosol.[40] The O_3 concentration can be decreased indoors also by filtering through charcoal or chemically impregnated filters in buildings equipped with mechanical ventilation systems.[41–43]

Another important oxidant is NO_2 that disproportionates in the presence of water to HONO and HNO_3. Nitrous acid is an important constituent of indoor air because it is the photolabile precursor to OH radicals.[44] Like O_3, NO_2 is also formed in the troposphere through a set of complex reactions[45] The reduction in the NO_2 level is accompanied by an increase in the O_3 concentration of air[46] due to the following reactions leading to generation of photochemical smog: i) $NO_2 + h\nu\ (\lambda < 420\,\text{nm}) \rightarrow NO + O$; ii) $O + O_2$ + energy-absorbing third body $\rightarrow O_3$; iii) $O_3 + NO \rightarrow NO_2 + O_2$.[47] The major sources of NO_2 and NO_x are motorized road traffic (e.g., diesel vehicles equipped with oxidation catalysts implemented for reducing PM emission) and heating combustion.[48] Typical NO_2 urban concentration levels indoors are between 20 and 40 ppbv depending on indoor sources.[39,49,50] In the absence of such indoor sources like gas appliances, smoking and kerosene heaters, a strong indoor/outdoor (I/O) relationship for NO_2 has been reported.[51]

2.2. Airborne fine particulate matter

Nowadays, particulates with an aerodynamic diameter less than 2.5 µm ($PM_{2.5}$) are primarily collected and analyzed, since these inhaled particles penetrate to the alveolar region. Usually, the PM analyses are aimed at the determination of elemental carbon (EC), organic carbon (OC), trace elements and inorganic (oxy)ions. The organic macromolecules in PM may reach 50% of PM2.5 mass,[52] and can be of both primary (e.g., biomass burning) and secondary origin.[53–55] It may be useful to extend the analyses to the determination of other pollutants, such as VOCs (benzene, toluene, formaldehyde, chlorinated compounds, etc.), since the occurrence of VOCs indoors is prevailing.[56,57] Although trace elements constitute only a small portion by mass of PM, some of them (e.g., redox cycling ones) may cause oxidative stress *via* reactive oxygen species (ROS) production. The ROS deplete antioxidants from the respiratory tract lining fluid (RTLF) compartment.[58,59] Therefore, determination of the water-soluble fraction of PM is crucial since many elements may readily pass the air-lung fluid interface[60] in water-soluble form. Thus, besides total element determination after MW-assisted acid digestion, appropriate amounts of filter samples should be sonicated at room temperature, then filtered and acidified prior to analysis.[61] Linkage to toxicological characterization of PM through oxidative potential measurements is now preferred.[62] The oxidative potential (OP) captures the redox active components and sources that can be related to health end points, and it can be used as predictor of adverse health impacts associated with an oxidative stress mechanism. Thus, the OP is a multipollutant parameter that integrates the composition effects into just one measurement requiring a set of instrumental measurements. The OP values are either based on time rate-of-change, expressed as nmol or % depletion per min. They may be also volume-based values (nmol or % depletion per min per m^3 of air) or mass-based data (nmol or % depletion per min per µg of particulate matter, PM); sometimes referred as intrinsic OP. Such a protocol variability strongly affects the magnitude of the measured responses, with the consequence that not reliable and conclusive results are reported within every study, which can make comparisons across studies difficult. Therefore, care should be taken when comparing OP results from different studies, as they often use varying operative protocols, since a unique standard method is still lacking.

3. Sampling and analytical methods for monitoring major indoor air pollutants

Monitoring of air pollutants have been made indoors and outdoors for many years now. Indoor air pollutants can be collected either diffusively or actively (by sampling pumps) and using stationary monitors or human volunteers wearing samplers (personal exposure monitoring).

Sampling strategies and analytical methods used for determination of main gaseous pollutants indoors are presented in Table 1. The built up of PM samplers as well as the main analytical techniques for physico-chemical characterization of PM have been summarized in Tables 2 and 3, respectively. Thus, in the case of PM, this means instruments that determine the concentration of particles in number, mass, surface area, size distributions, shape, etc. The PM_{10} and $PM_{2.5}$ samplers to be used in sampling activities should comply with the characteristics of the UNI

EN 16000-34 and 16000-37 standards.[73,81] A classic approach for the characterization of PM is determination of the mass concentration and the chemical characterization involving determination of metals, metalloids, major inorganic ions, organic compounds (e.g., polycyclic aromatic hydrocarbons) and EC/OC. There are several challenges for the characterization of indoor PM. One is the low amount of PM mass

Table 1. Sampling strategies and analytical methods used for determination of main gaseous pollutants.

	Time-integrated			
Pollutant	Sampler	Quantification	Continuous	Ref.
CO	–	–	Dispersive IR or electrochemical	[63–65]
			Cavity Ring-Down Spectroscopy	[66]
CO_2	–	–	Non-dispersive IR spectroscopy	[64]
	–	–	Cavity Ring-Down Spectroscopy	[66]
NO_2	Chemiadsorption as nitrite ion and conversion into a reddish-purple azo dye with an adequate reagent	UV–Vis spectrophotometry	Chemiluminescence	[67]
		IC		[68]
O_3	Diffusive *via* selective acid-catalyzed ozonolysis and subsequent condensation a yellow colored azide	UV–Vis spectrophotometry	Calibrated UV-based online instruments with a certain time resolution response	[67]
VOCs	Passive or active sampling using sorbents	GC-MS	Photoionisation PTR-MS	[66]
carbonyls	Passive or active sampling using DNPH on silica gel	HPLC-UV	IR spectroscopy Cavity Ring-Down Spectroscopy	[69,70]

Abbreviations: GC-MS = gas chromatography mass spectrometry; HPLC-UV = high performance liquid chromatography ultraviolet detection; IC = ion chromatography; IR = infrared; UV-Vis = ultraviolet visible; PTR-MS = proton-transfer-reaction mass spectrometry

Table 2. Built up of a PM sampling system.

Components	Notes	Type	Application	Ref.
pre-separator	for size selection	impactors	air quality standards	[71]
		cyclones	low-flow rates	
filter media	porosity, net mass, hygroscopicity and sensitivity to electrostatic charges should be evaluated	cellulose esters; PC membranes	Microscopy	[72]
		PTFE	Gravimetry	[73]
		PTFE/cellulose	multielemental analysis	[74–77]
		QFFs	OC/EC	[78]
air flow meter	classical gold standard	soap bubble flowmeter		–
	current alternatives	NIST-traceable calibrators		–
tubing	suitable material to prevent kinks	flexible tygon		–
sampling pump	membrane pumps	Low flow battery operated	Personal sampling	[79]
	rotary-vane vacuum pumps	High flows	Fixed sampling	[80]

Abbreviations: QFF = quartz fiber filter; EC = elemental carbon; OC = organic carbon; PTFE = polytetrafluoroethylene

Table 3. Summary of multipollutant analyses in indoor particulate matter. (For oxidative potential analyses see Table 4).

Analyses	Analytical technique	Notes	Ref.
Mass concentrations Real-time	Optical light scattering	Particles from 0.3 μm to > 20 μm can be detected	[81]
Number concentrations Real-time	CPC	Particles from 4 nm to > 1 μm can be detected	[81]
Mass concentrations Time-integrated	Gravimetry	Microbalance in climatic chamber; preferably 1 μg resolution	[73]
Size distribution Real-time	DMPS	Particles from a few nm to about 1 μm can be detected	[81]
EC/OC Time-integrated	TOT	1) Combustion of OC in He; ii) For EC, combustion in He/O_2; iii) Charring correction *via* transmission of a He-Ne laser beam through the sample; iv) Reduction of the evolving CO_2 to CH_4; v) quantification with FID	[14,82]
Elements Time-integrated	ICP XRF	Differences in sampling media, sample destructivity and costs between ICP and XRF	[83]
Major ions Time-integrated	IC	Either anion or cation exchange column packaging	[14,84–89]
VOCs Time-integrated	GC-MS, LC-MS	GC-MS of GC-FID after thermal desorption are widely used	[90–92]

Abbreviations: CPC = condensation particle counter; DMPS = differential mobility particle sizer; EC = elemental carbon; FID = flame ionizatno detector; GC-FID = gas chromatography – flame ionization detector; GC- MS = gas chromatography – mass spectrometry; IC = ion chromatography; ICP = inductively coupled plasma mass spectrometry; LC-MS = liquid chromatography – mass spectrometry; OC = organic carbon; TOT = thermal optical transmittance; XRF = X-ray fluorescence

collected on the filters due to the relatively short sampling time and the low flow rates used to reduce noise annoyance for occupants. In short, a single pollutant monitoring or determination (e.g., elemental, major inorganic ions, EC/OC) is not typically enough for a comprehensive assessment of IAQ and to study the key exposure determinants for a sound risk assessment and management.

Several epidemiological studies proved that knowledge of the mass concentration and chemical composition of PM is necessary but not always sufficient to explain the

Table 4. Possibilities of determining oxidative stress induced by atmospheric aerosols.

	OP measurement (symbol)	Observation	
Acellular (cell-free)	Electron spin resonance (OP^{ESR})	suitable only for OH·	
	Dithiothreitol assay (OP^{DTT})	antioxidant depletion	DNA protection
	Ascorbate assay (OP^{AA})		RTLF constituents
	Glutathione assay (OP^{GSH})		
	ROS measurements		
	Fluorescent-based techniques on PM	for radical determination	
Cellular	A549 (human lung carcinoma cell line)	[20–22]	
	NR8383 (rat alveolar macrophage)	[23–25]	

AA = ascorbic acid; DTT = dithiothreitol; GSH = reduced glutathione; OP = oxidative potential; PM = particulate matter; RTLF = respiratory tract lining fluid.

adverse health effects of PM. The OP of PM can be estimated by acellular and *in vitro* cellular assays. The acellular assays have the advantages of low price, speed, practicality, and high data throughput, when compared to cellular assays.[93–99] Among them, the most commonly used are based on dithiothreitol as a proxy of cellular reductants (DTT assay, OP^{DTT}),[100,101] or endogen antioxidant species, such as the AA (OP^{AA}) and GSH (OP^{GSH}) assays[102]

The most common acellular methods include i) electron paramagnetic resonance (EPR) measurements with 5,5-dimethylpyrroline N-oxide, which traps OH· formed in the presence of H_2O_2,[103,104] ii) fiber optic spectrometric measurements on the fluorescence after sampling[105] and iii) determination of antioxidant depletion. The EPR test has a relatively small sample requirement, it is relatively cost effective, and it can be used in large-scale air pollution studies.[59] Its big drawback is that it only provides information about OH·. In the case of *in vitro* cellular assays, ROS produced in living cells (e.g., macrophages) and exposed to PM samples are determined by fluorescent probes.[106,107] The methods used to determine oxidative stress are summarized in Table 4.

Dithiothreitol is a surrogate for the cellular oxidant NADPH, which reduces oxygen to the superoxide anion. The DTT assay is based on the ability of redox active compounds associated with PM to transfer electrons from the DTT to oxygen. This generates superoxide that subsequently converts to H_2O_2 and oxygen.[100,108] The non-reacted DTT is determined with the thiol reagent 5,5′-dithiobis-2-nitrobenzoic acid (DTNB). This reaction is proportional to the concentration of the redox active species. The DTT consumption over time constitutes the rate of the reaction. However, the rate is also a limitation of the assay, since incubation times of up to 45 minutes are needed to achieve significant consumption compared to the blanks. The DTT assay is mainly sensitive to the organic compounds accumulated in the fine PM fraction.[94,109] In contrast, OP^{AA} and OP^{GSH} are mostly responsive to metals, mainly those related to non-exhaust traffic emissions (Cu, Zn, Cr, Fe, Ni, Mn, Sn, Cd, and Pb), that are mainly accumulated in the coarse PM. However, DTT can be used only at low temperatures in an inert atmosphere and loses its nucleophilicity as the pH decreases.

The chemical OP^{AA} protocol is very similar to the OP^{DTT} protocol. After controlled incubation of the antioxidant AA in the aqueous extract, the measurement of AA depletion over time is directly followed by the decrease of UV-VIS absorbance of the AA at 265 nm.[110,111] Modified AA procedures have also been used, performed in a synthetic surrogate (i.e., RTLF), containing endogenous antioxidants at physiological levels, which is more indicative of realistic lung conditions, i.e., GSH, UA, and citrate, in addition to AA.[112] In some OP protocols, the remaining concentrations of antioxidants in RTLF is quantified at specific time intervals, using reversed-phase high-performance liquid chromatography (HPLC) with electrochemical detection and/or enzyme-linked assay for total GSH and GSH.[14,59,98,113,114] In general, GSH determination requires the reduction of disulfide bonds between GSH and other thiols or proteins. Almost all methods are based on products of GSH derivatization. Such analytical methods use colorimetric reagents, e.g., Ellman's reagent (DTNB)[115,116] or Sanger's reagent (1-fluoro-2,4-dinitrobenzene, DNFB),[117,118] and then HPLC is applied. Glutathione may also be determined as an *o*-phthalaldehyde derivative[119] by HPLC with fluorescence detection.[120,121] Direct, selective, and sensitive methods for quantitative determination of GSH are based on its electrochemical properties.[122] However, LC-MS-based methods for determination of GSH have not gained much popularity.

4. Technical solutions

4.1. General considerations

Human exposure to air pollutants can be assessed using direct measurements, biological monitoring, or estimated using indirect methods.[123] These latter apply to models predicting exposure from fixed sampling locations indoors or outdoors.[123] In the case of direct measurements, personal sampling is the preferable way for assessing human exposure.[123,124] Studies using this type of approach have the disadvantage of being time consuming and expensive and not always recommendable when it is necessary to determine concentrations of several co-pollutants since wearing several instruments interferes with normal daily activities.[123] Therefore, methodologies to assess personal exposure to multiple airborne pollutants have been developed.[125–127] The association between personal exposure and measurements from fixed stations depends mainly on some factors as the distance of sampling equipment with respect to the subjects' breathing zone[128] and the distance from point sources. The association may also vary as a function of the sampling period. Short-time fluctuations in concentrations occurring in the proximity of point source emissions are undetectable when sampling is carried out for a longer time period.[129] Urban background monitoring stations are not suitable to characterize exposures significantly influenced by individual point sources.[130]

4.2. Portable multipollutant monitoring units

As reported in the literature,[131] portable instruments, due to their intrinsic advantages, have been widely used in exposure assessment studies, significantly improving the assessment of human exposure to airborne pollutants, since these kinds of instruments are able to measure the exposure at individual level (defined as the exposure constantly measured in proximity - within 3 m - to the subjects) or at personal level (which is preferable for assessing human exposure, as it is representative of the contaminant concentration in the breathing zone).[16,123] Accordingly, a personal monitoring approach may assess the subjects' exposure based on measuring the concentration of a pollutant ideally within the subject breathing zone for a defined time.[132] As the subjects move in

Table 5. Basic experimental designs adapted from [133].

Aim	Methodology	Advantages	Drawbacks
Use of individual air pollution monitors for direct determination of exposure	Each subject in the study population would wear/carry an individual air pollution monitor. The same individuals would also be subjected to continuous/periodic evaluation of health responses.	n.d.	Because of economic constraints, only relatively small populations could be studied following this direct approach.
Use of individual air pollution monitors to adjust results from fixed stations	By monitoring exposure of subjects with individual air pollution monitors in areas also monitored with fixed stations.	If one or several relatively constant relations were found in various areas, fixed-station data would then be corrected for use in estimating population exposures.	n.d.
Use of representative sampling to determine subgroup exposure	A carefully selected sample of the study population would be asked to wear/carry individual air pollution monitors. The sample would be stratified, grouping those expected to have similar exposures.	The measured exposure of each subgroup in the sample could be used as representative of the entire group.	n.d.
Use of individual air pollution monitors to calibrate personal activity models	In such an experiment, a carefully selected sample of the study population would be asked to wear individual air pollution monitors, and their measured exposure would be compared with the estimated exposure of the activity model.	Activity models have not been applied in air pollution epidemiology except in a very limited way, and they could be best calibrated or verified through experiments using individual air pollution monitors.	n.d.

changing environments, their personal/individual exposure to airborne pollutants vary accordingly and, to record these variation portable devices are needed.[132] In this regard, the standard ISO 16000-34[81] mentions a series of portable samplers and instruments usually worn by people during indoor activities to monitor their personal exposure, even though no guidelines are clearly specified for these kinds of devices.

For a reliable evaluation of air pollution health effects, it is necessary to know the (i) actual personal exposures and the (ii) distribution of those exposures in a population: the assessment of personal exposure can be useful in several experimental designs, as reported by the literature.[133] Four basic experimental designs are reported in Table 5.

The use of portable instrumentation is mainly used in the outdoor environment, particularly when the study design foresees the exposure assessment of moving subjects, where some typical characteristics of the fixed instrumentation must necessarily be passed (e.g., high dimensions and weight, need for continuous power). For this reason, exposure assessment studies, based on the evaluation of personal exposure in indoor environments only, using portable instruments, are scarce. More often, the assessment of indoor personal exposure is conducted *via* portable monitors whether the study design includes movement in multiple microenvironments by the monitored subjects. A non-exhaustive list of exposure assessment studies, conducted *via* portable and multi-pollutant instruments to highlight examples is reported by way of example in Table 6.

Table 6. Brief description of the methods used in some exposure assessment studies.

Monitored pollutants and used instrumentation	Instrumentation case	Instrument inlets	Indoor environment and subjects monitored	Monitoring period and location	Reference
$PM_{2.5}$ and PM_{10}: PEMs O_3; NO_2; SO_2: passive badge samplers Select VOCs: sorbent tubes	Backpack The entire sampler weighed approximately 5 lb	Sampling inlets were placed on the shoulder strap of a backpack to correspond to the breathing zone of each subject	n.d.; Older adults	Summer 1998 and Winter 1999; Baltimore	[134]
$PM_{2.5}$ and PM_{10}: Impactor OC and EC: Inertial impactor Sulfate and Nitrate: Inertial impactor O_3; SO_2; NO_2: Passive diffusion sampler	n.d.	n.d.	n.d.; n.d. (experimental characterization)	n.d.; n.d. (experimental characterization)	[125]
UFP (ultrafine particles); FP (fine particles); CP (coarse particle): (i) TSI P-Trak 8525 Ultrafine Particle Counter (UPC)[a]; (ii) Lighthouse Handheld 3016 Optical Particle Counter (OPC)[b] CO: Langan CO measurers T15v[c] O_3: Aeroqual Series 500	Soundproof aluminum case - trolley Outer dimensions: 90 × 45 × 31cm; weight: 25 kg	Top-level of the unit (approximately 90 cm from the ground), except for O_3 and CO monitors, that were fastened on the outer wall of the unit (80 cm from the ground)	Office; n.d.(141)	April and October; Milan	[16]
$PM_{2.5}$ and PM_{10}: CPEM[d] NO_2 and O_3: Ogawa passive dosimeters[e]	Monitoring vest The use of a vest fixed the participant burden at 454 g	Breathing zone	n.d.; Adults having documented degrees of asthma severity	February 2008 - April 2009	[135]
PM and UFP: (i) Lighthouse Handheld 3016 Optical Particle Counter (OPC)[b]; (ii) P-Trak Ultrafine Particle Counter 8525 condensation particle counter (CPC)[a] CO: Langan CO measurers T15v[c]	Backpack	The sampling inlets were not placed in strict correspondence with the breathing zone: the study results refer to the so-called "individual" measurement	Office; n.d.	July 2008, October - November 2008, February - March 2009; Milan	[136]
$PM_{2.5}$: MicroPEM Personal $PM_{2.5}$ samplers - v3.2[f] BC: MicroAeth BC Monitor model AE-51[g]	n.d.	Personal level will represent the breathing zone exposure of a person if the unit is worn nominally within 30 cm of the oral/nasal cavity	n.d.; Graduate students	December 2014 - March 2015; Shanghai	[137]

(*continued*)

Table 6. Continued.

Monitored pollutants and used instrumentation	Instrumentation case	Instrument inlets	Indoor environment and subjects monitored	Monitoring period and location	Reference
PM (PM_1, $PM_{2.5}$, PM_4, PM_{10}, and TSP): Aerocet 831 Met One[h] NO_2: CairClip NO_2, Cairpol[i]	Backpack	Breathing zone	Pregnants	October 2017-April 2018; Milan	[138]
PM: SKC five-stage Sioutas Cascade Impactor[j] BC: MicroAeth BC Monitor model AE-51[g]	Trolley	Air inlet tube placed in the breathing zone	n.d.; Children	May 2 - June 22, 2018; Lisbon	[139]
UFP: DiSCmini Miniature Diffusion Size Classifier[k] PM (PM_1, $PM_{2.5}$, PM_4, PM_{10}, and TSP): Aerocet 831 Met One[h] $PM_{2.5}$: AirBeam, HabitatMap Inc.[l] $PM_{2.5}$: GK2.05 sampler[m] NO_2: CairClip NO_2, Cairpol[i]	Backpack	All the inlets were placed in the breathing zone of the operator, with 30 cm-radius hemispheres extending in front of the face	Commuters	March 11-15 and 18-22, 2019; July 8-12 and 15-19 and 25 2019; Milan	[140]
BC: MicroAeth BC Monitor model AE-51[g] UFP: DiSCmini Miniature Diffusion Size Classifier[k]	Backpack	Inlet tubing fixed at the breathing zone	Household microenvironment, study-related microenvironment, leisure locations; School students	July - August 2016; Chengdu	[141]

Abbreviations and explanations: n.d.: not defined.
aTSI P-Trak 8525 Ultrafine Particle Counter; TSI Inc., Shoreview, Minn.;
bLighthouse Handheld 3016 Optical Particle Counter; Light-house Worldwide Solutions, Fremont, Calif.;
cCO: Langan CO measurers T15v; Inc. San Fran-cisco, Calif..
[d]CPEM; RTI International, Research Triangle Park, NC;
eOgawa passive dosimeters; Pompano Beach, FL.;
fPersonal $PM_{2.5}$ samplers - v3.2 MicroPEM, RTI, USA;
gBC: MicroAeth BC Monitor model AE-51; Magee Scientific, USA;
hAerocet 831-Met One Instrument Inc., Grant Pass, Oregon, USA;
iCairClip NO2, Cairpol; La Roche Blanche, France;
jSKC five-stage Sioutas Cascade Impactor - SKC Inc., U.S. Patent No. 6,786,105;
kUFP: DiSCmini Miniature Diffusion Size Classifier; Matter Aerosol AG, Wohlen AG, Swiss;
lAirBeam, HabitatMap Inc., Brooklyn, NY, USA;
mGK2.05 sampler; BGI Inc., Waltham, MA, USA.

Most of the studies involve the use of an instruments case, which can be a trolley,[16,139] a backpack,[136,138,140,141] or, less commonly, a monitoring vest.[135] The choice of the case type can be made according to different factors, such as (i) the number and characteristics (in terms of dimensions and weight) of the instrumentation used; (ii) the impact of the case on the normal activities of the monitored subjects; (iii) study design ; (iv) use of extra materials such as additional batteries, as reported by Cattaneo *et al.*[16]

Moreover, the instruments used in such studies (i.e., evaluation of personal exposure assessment *via* multipollutant approach) normally are (i) portable, (ii) small in size, so as not to interfere with the subject usual behaviors and habits throughout the day, (iii) flexible, (iv) robust, (v) user-friendly, (vi) lightweight and (vi) battery operated (or passive).[51,132,142]

Overall, the number of instrumental systems commercially available for the real-time measurement of indoor air pollutants increased considerably. In many cases, a given constituent can be determined by a variety of instrumental techniques. The choice of the best approach can be a complex decision, which is influenced by several factors, including cost, system portability, selectivity and sensitivity, and the quantity and concentration of potential interferences. Direct-reading instruments could provide substantial benefits (including lower efforts at lower cost) when applied in IAQ monitoring if compared to traditional exposure assessment methods, which rely on time-integrated sampling devices, (i.e., by means of sampling pumps or diffusion methods), sampling substrates (e.g., sampling filters, adsorbent substrates) and a subsequent analytical phase (e.g., gravimetric determinations, chemical characterization). More in detail one of the advantages of direct-reading techniques is to provide new insights on exposure dynamics due to their ability in collecting data at greater spatio-temporal resolutions.[143] Further, direct-reading devices can report and process the data as soon as they are collected and while the instrument is still deployed (i.e., real time analysis). Then, due to these features (i.e., reduced cost, ease of deployment, direct reading capabilities together with the wireless network ability and the possibility of integrating them with other exposure estimation methods) new ways of collecting and sharing information become possible.[144,145] For these reasons, not only the need for accurate evaluation of exposure to airborne pollutants is confirmed and reiterated, but a step forward is required as regards the methods, the techniques, and the technologies to be used for this purpose. Despite their advantages, it’s important to underline that these kinds of technologies should be deeply evaluated before use, especially in terms of measurement precision and accuracy.[146–149] Then, despite expected advantages, the use of direct-reading methods can also present drawbacks and difficulties.[131,150] Direct-reading methods are generally less reliable (in terms of accuracy, sensitivity, precision and specificity to the chemical/variable of interest) if compared to high-end devices.[151–153] Overall, these kind of sensors are being successfully used complimentary to reference monitoring,[154] but they are not yet validated as alternative techniques for (or to replace) reference instruments (especially for purposes of mandatory monitoring).[153] Although some studies are available[131,150,155] definitive and comprehensive evaluations concerning the agreement between sensor systems and reference instrumentation are not available, and neither performance evaluations of direct-reading techniques in different exposure scenarios. Further, unlike what happens for reference-grade instrumentations that are subjected to comprehensive regulatory standards and processes for evaluation and certification, only few standards exist for direct-reading techniques.[156] Further, biases in the acquisition and interpretation of the data obtained with direct-reading techniques can derive from different sources of measurement error and interference, which arise once operating in the field and which cannot be completely covered in the development and calibration phases carried out in the laboratory.[151] For these reasons, direct-reading techniques should be operated applying rigorous quality assurance and quality control protocols.[144,157]

The most recent studies also associate the instrumentation for measuring exposure to the various selected pollutants with the use of a GPS (Global Positioning System) and/or an electronic TAD (Time-Activity Diary), to be filled in by the monitored subject. This latter can be of particular interest if used in indoor environments, as it allows to evaluate the contribution of the different activities carried out in a particular environment to the total exposure.[136,137] Indeed, TADs and questionnaires are essentials in personal exposure assessment studies to cover data on human behavior and activities[132,158] and several important studies (e.g. the TEAM[159,160] and EXPOLIS[126] studies) have integrated them into their experimental design to gather data about the participants' whereabouts and activities. More in detail, activity information may be collected in a predetermined time interval (e.g., 10 min intervals, as reported by Yang *et al.*161] or 15-30 min interval as reported by Wheeler *et al.*[162] or may be collected using open-ended diaries.[163] In other cases[134], subjects were given a choice of completing two types of TADs during the project by a (i) closed-form diary (in which subjects recorded their activities and location in closed, time-interval spaces) or in a (ii) open-form, activity-based diary. In this latter case each page of the diary corresponded to a specific activity with space available on the page to indicate location and any special conditions likely to affect exposure).

The activities may also be organized according to the classifications defined *a priori* or they can be detailed by the monitored subject. For example, studies focused on the assessment of indoor exposure commonly require the compilation of specific TADs, with indications concerning (i) cooking activities; (ii) time of cleaning; (iii) proximity to any smokers; (iv) possible effects modifiers (e.g., windows/doors opened).[162,164] Obviously, the required level of detail and the chosen classifications depend on the specific aim of the study and on the monitored pollutants. The use of a TAD is therefore of fundamental importance also in indoor monitoring studies since, as reported in the literature, the indoor sources of airborne pollutants are diverse. For example, Gonzalez-Martín *et al.*[165] have reported in their study how different indoor environments are characterized by different pollutant sources.

As said and as reported in Table 7, indoor exposure results from interactions between different factors, such as (i) building characteristics, (ii) furnishings and (iii) individuals acting, as well as (iv) outdoor environment:[132] these characteristics can therefore be qualified/quantified *via* compilation of TADs and questionnaires.

Even when carrying out personal monitoring, attention must be paid to the fact that the measurements remain within the specificities of the instruments. Moreover, the instrument should be correctly calibrated and checked before use as recommended by

Table 7. Source of indoor air pollution (adapted from [165]).

Indoor environment	Pollutants	Sources
Office	O_3	Electronic devices
	VOCs	Office supplies
Kitchen	CO, PM and VOCs	Cooking and heating devices
	VOCs	Cleaning products
Living room	HCHO and BTEX	Furniture, carpets and air fresheners
	CO, PM and VOCs	Fireplaces and burning stoves
Bathroom	VOCs	Cleaning agents, cosmetics and personal care products
Bedroom	VOCs	Furniture and personal care products
	Naphthalene	Insect repellers
Construction materials	HCHO and fibers	Insultating materials

the manufacturer. The clocks of the instrumentation, even more when using a multipollutant system, should also be synchronized with each other before the start of monitoring.

As reported in the bibliography, the time at which the measurement is performed depends on the potential sources and its inputs,[81] as well as the aim of the study. It would also be important to start the monitoring before specific activities related to pollutants sources start (e.g., vacuum cleaning) and finish it some time later (e.g., 1 h), to monitor background concentrations, those re-entrained during the activity, and those emitted at its term.

4.3. Fixed multipollutant monitoring stations

The general principles for IAQ monitoring typically requires different measurement techniques and strategies; some information are available for example in a recent WHO publication[166] and in technical standards.[167] The classical experimental approach for a multi-pollutant monitoring in indoor settings is based on active, passive, or continuous sampling at a fixed location. IAQ problems have multifactorial causes, and multipollutant measurement systems are generally used to account for the different chemical, biological and physical agents and quantify their contamination levels in indoor air. Basic microclimatic parameters such as T and RH should also be monitored because of their influence (e.g., uptake rates of passive samplers) or interference with air sampling (e.g., electrochemical sensors).

The answer to the question on “where” the monitoring campaign should be carried out is crucial in this type of approach: the selection of indoor sampling locations is generally based on specific criteria, which are strictly dependent on the purposes of the study, while at the same time responding to specific requirements (e.g., security concerns, practical logistics). Moreover, some general aspects must always be considered carefully,[168] viz (i) in the presence of outdoor emission sources, a significant portion of the indoor air pollution may derive from outdoor mobile or stationary emissions and air exchange rates play a major role in determining the relative influence of the outdoor pollution on the indoor concentrations; (ii) other determinants, besides ventilation, may affect the indoor/outdoor relationship (e.g., climatic conditions, seasonal patterns, indoor activities); iii) the concentration levels of indoor pollutants may vary among rooms and within the same room, because of possible differences in ventilation efficiencies or the presence of strong emission sources.

When measurements are carried out at fixed indoor locations, not every room in a flat or a building can be controlled, mainly because of practical constrains. If the primary aim of the fieldwork is the IAQ assessment or the exposure assessment of occupants (as pupils in schools, workers in offices or people in residential buildings), the monitored rooms should be selected to best represent the investigated scenario (e.g., when personal sampling is inconvenient for the involved subjects, area samples should be taken in such a way that the investigated area(s) is representative of the personal exposure of the subjects). In general, the number of investigated room(s) per building should be a compromise between a comprehensive IAQ characterization for the whole building and budget and feasibility issues (e.g., availability of monitoring devices).[64,67] Then, the location of sampling devices inside each room should comply with the following guidelines, in agreement with ISO 16000-1:[169] (i) the center of the room is generally considered the most suitable location but, if this is not possible, ii) the equipment should be placed at least 1 m from the nearest wall and on a height of about 1.1–1.5 m (sitting or standing height) above the floor,[67] which approximately corresponds to the breathing zone of humans; however, especially in the case of long-term measurements, a somewhat higher location (2 m or more, e.g., on the room ceiling) may be appropriate to avoid annoyance for or interferences by occupants; iii) places in the sun, nearby heating systems, ventilation channels and clearly identified indoor emission sources should be avoided. Moreover, the sampling location should not disturb the normal activities carried out inside the room and it must comply with any safety regulation. Special care must be taken when the investigated environment is characterized by a certain variability of air concentration levels.

Contrarily, if the goal is the study of specific sources of indoor air contamination, the priority decision would fall on the room(s) where the source is present. By way of example, if the emissions depend on specific activities carried out by occupants, measurements should be performed in the environment where these activities take place. Moreover, in this special case, it may be appropriate to choose a fixed site location close to the specific emission source instead of the center of the room not to underestimate occupants' exposure.

The choice of the sampling point in indoor environments is of even greater importance than outdoors.[170] If it is a building, it is generally not necessary to investigate all the microenvironments, but the most representative areas must be identified in relation to the objective of monitoring (e.g., generally the environments in which people stay for longer periods).[170] The position of the sampler within an environment strongly influences the measurement and therefore the results.[171] If the investigated area includes several microenvironments, it is necessary to make a virtual subdivision of the building and establish one or more sampling sites within it.[170] If the emission levels depend on particular activities plan by the occupants, it is useful to carry out measurements in the area in which this activity is carried out as well as in background positions (presumably not exposed/influenced by the activities of the occupants).[170] At the same time, concomitant outdoor sampling in the vicinity of the indoor sampling point should be carried out. In the presence of anomalous concentration levels, it may be useful to study the emissive characteristics of the source(s) using instrumentation with high temporal resolution.[170]

Finally, as stated before, since the outdoor pollution can significantly affect the indoor environment, it may be useful to make additional measurements at a fixed site station in the outdoor air close to the investigated room(s), to allow the quantification of the outdoor contribution on the indoor concentrations, especially in the proximity of potential outdoor sources (e.g., traffic).[64] Generally, the outdoor site should be placed in a safe location sheltered from rain and direct sunlight and at a reasonable distance from walls or other objects or systems (e.g., the air intake). If possible, outdoor monitoring should be carried out on the same side of the building and at the same height as that which the indoor sampling take place;[98] in case of air-conditioned buildings, it may be useful to perform the outdoor sampling at the inlet of the HVAC system.[98] Positioning under a low roof or on another floor can be acceptable if necessary for security concerns or practical constrains (e.g., access to electrical supply, presence of a balcony).[168]

At the selected fixed (indoor and outdoor) locations, the multi-pollutants devices are typically put together in a single solution which may result in different types of structural assembly. The samplers may be fixed to a wire attached to the ceiling,[67] placed in a metallic rack[67,168] or mounted on a tripod.[172]

The arrangement of microenvironmental monitors (MEMs) in a suitcase composed of sound-absorbent containers made of medium-density fiberboard may be another strategic solution,[16,63] especially when the experimental design requires the use of bulky devices (in terms of size and weight) which could hardly be placed on a simple tripod. For example, in the framework of the EXPOLIS study, the MEMs suitcase contained a $PM_{2.5}$ EPA-WINS impactor above the box, two filter holders inside the box connected to the $PM_{2.5}$ impactor by a Y-joint, a charger below the filter holders with tubing and a pump outside the box. The pump was placed inside the lower part of the box and the doors were closed during the runs.[63] In the outdoor location, a Graseby-Andersen PM_{10} inlet was used to avoid wind and rain effects and the pump was enclosed in a weatherproof case.[63] As regards the monitoring techniques, several sampling and detection methods exist. The applied strategies may include active sampling, diffusive sampling and/or continuous monitoring for both particles and gas/vapor phase. The selection of the most suitable methods for each chemical and physical parameter is generally performed following the reference standards ISO 16000 series. If no reference method is available for a specific pollutant, methods published by national or international governments (e.g., OSHA, NIOSH, HSE) and described in ISO, ASTM, CEN guidelines may be considered, as well as methods published in peer reviewed journals, with a backup validation data, or methods under development. Furthermore, among the various available methods, the most suitable choice is also dependent on sampling time, selectivity, sensitivity, detection limits, acceptable uncertainty, budget and available resources. In the past 30 years, several instrumental approaches have been proposed for multipollutant determination aiming at monitoring of indoor air quality. A summary of field campaigns conducted in the frame of collaborative projects across Europe and the US has been summarized in Table 8. Temperature and relative humidity were monitored using data logging sensors. Over the years and with the development of analytical instrumentation, more and more components have been quantified simultaneously during the field campaigns of these collaborative projects (Table 8). The most investigated

Table 8. Summary of multicomponent analyses performed during field campaigns in the frame of collaborative research projects.

Project acronym (reference)	AUDIT [190]	EXPOLIS [63,184]	RIOPA [168]	THADE	AIRMEX [186,187]	OFFICAIR [67]	SINPHONIE [65]
Project years	1992–1994	1996–1997	1999–2001	2002–2003	2003–2008	2010–2013	2010–2012
Study location	Office buildings in Europe	Six European cities	100 dwellings in each of three cities in US with different air pollution sources and weather conditions	Dwellings across Europe	Public buildings and kindergartens across Europe	Office buildings across Europe	Schools across Europe
Particulate matter							
PM$_{10}$	**YES**	**NO**	**NO**	**NO**	**NO**	**NO**	**YES**
PM$_{2.5}$	**NO**	**YES**	**YES**	**YES**	**NO**	**YES**	**YES**
UFP	**NO**	**NO**	**NO**	**NO**	**NO**	**YES**	**NO**
Inorganic gaseous pollutants							
NO$_2$	**NO**	**YES**	**NO**	**YES**	**NO**	**YES**	**YES**
O$_3$	**NO**	**NO**	**NO**	**NO**	**NO**	**YES**	**YES**
CO	**YES**	**YES**	**NO**	**YES**	**NO**	**NO**	**YES**
CO$_2$	**YES**	**NO**	**NO**	**YES**	**NO**	**YES**	**YES**
Organic gaseous pollutants							
VOC	**YES**^{a}	**YES**	**YES**	**NO**	**YES**	**YES**	**YES**
Alkanes	NO	YES	NO	NO	YES	YES	NO
Alcohols	NO	YES	NO	NO	YES	YES	NO
Aromatics	NO	YES	YES	NO	YES	YES	YES
Cycloalkanes	NO	YES	NO	NO	YES	NO	NO
Dienes	NO		YES	NO	NO	NO	NO
Halogenated	NO	YES	YES	NO	NO	YES	YES
Ethers	NO	YES	YES	NO	YES	YES	NO
Terpenes	NO	YES	YES	NO	YES	YES	YES
Carbonyls	**NO**	**YES**	**YES**	**YES**	**YES**	**YES**	**YES**
Sum of studied classes	4	5	3	5	2	7	8

a= only total VOCs.

Abbreviations: AIRMEX European Indoor Air Monitoring and Exposure Assessment Project; AUDIT European Audit Project to Optimize Indoor Air Quality and Energy Consumption in Office Buildings; EXPOLIS Acronym created from exposure and polis (city, gr.); OFFICAIR On the reduction of health effects from combined exposure to indoor pollutants in Modern Offices; PEOPLE Population exposure to Air Pollutants in Europe; RIOPA: Relationships of Indoor, Outdoor, and Personal Air; SINPHONIE Schools Indoor Pollution and Health: Observatory Network in Europe; THADE: Toward Healthy Air in Dwellings in Europe;

environments during these field campaigns have also been summarized in Table 8. Briefly, they included kindergarten, schools, offices and dwellings. Table 9 presents the detailed information on the pollutants determined during these and international studies as well as some national studies conducted in the last decades focusing on the assessment of airborne multi-pollutants concentrations in different kinds of indoor settings. Information about the investigated pollutants, the type of structural arrangement used at the fixed monitoring locations as well as the selected sampling and analytical methods are reported. The tabulated information on PM, inorganic and organic pollutants can be found as supplementary data information (Tables S1–S4, supplementary material).

4.4. Advantages and drawbacks of multipollutant monitoring for indoor air quality

A recent WHO publication[166] presents and discusses the advantages and drawbacks of some methods for sampling and analysis that have been used to characterize chemical pollution of indoor air, selected among (i) those recommended by the International Organization for Standardization (ISO), (ii) the most commonly used for sampling and analysis of indoor air pollutants in public settings. The document presents both passive (also called diffusive) or active sampling method. As told, these are discontinuous methods, thus pollutants are collected on a filter or sorbent for further analysis in a laboratory. Laboratory analysis usually consists of extraction, clean-up (when necessary) and the analysis itself using analytical equipment. The extraction can be carried out using thermal desorption or solvent desorption. Analysis can be performed by means of different analytical systems depending on the pollutant to be analyzed (such as gas chromatography (GC) and liquid chromatography (LC) coupled to different detectors and an ultraviolet-visible (UV-Vis) spectrophotometer).[166] More in detail, putting together multiple monitoring devices and systems in a single solution for multi-pollutant monitoring offers a series of advantages for a comprehensive assessment and management of indoor air quality but presents, at the same time, some technical limitations and criticalities.

Among the possible advantages of multipollutant monitoring are: i) to allow a comprehensive health risk assessment accounting for combined exposure to multiple air pollutants;[173] ii) to explain or estimate air contamination trends and relative source contributions accounting for indoor air reactions;[174] iii) to allow the correction of sensor cross-interferences using simultaneously collected data of co-pollutants.[175] By way of example, amperometric O_3 sensors are known to suffer NO_2 interference and NO_2 sensors are highly sensitive to O_3 and simple or multivariate regression methods and artificial neural networks can be applied to field data for improving their accuracy.[176] Another interference was studied by Collins et al..[44] During an indoor study, NO and NO_2 were determined as 60 second running average concentrations by chemiluminescence with an uncertainty of ±10% for both pollutants. The instrument was equipped with a blue light converter for selective observation of NO_2. According to the authors, interference on NO_2 measurements from $RO_2\bullet$ formed within the blue light converter[177] might have arisen when photo labile organic compounds (e.g., glyoxal) were

Table 9. Summary of some national and international studies conducted in the last decades and focused on the assessment of airborne multi-pollutants concentrations in different kinds of indoor settings (e.g., residential buildings, schools, offices).

IAQ study and/ or (reference)	Investigated indoor environment	Type of fixed site arrangement	Investigated pollutants	Sampling methods	Analysis or detector
OFFICAIR Project[67]	Offices	Wire attached to the ceiling or metallic rack; indoor and outdoor	$PM_{2.5}$	Pumped sampling – sampling medium quartz filter with impactor for $PM_{2.5}$ (Harvard type impactor, with quasi noiseless pumping unit)	Gravimetric analysis Elemental characterization by inductively coupled plasma sector filed mass spectrometry (ICP-SF-MS) after vapor-phase microwave (MW)-assisted *aqua regia* digestion Water-soluble fraction analysis by sonication-assisted extraction, filtration and IC analysis EC/OC determination by a Sunset thermal-optical-transmittance analyzer (TOT) Determination of the oxidative potential of $PM_{2.5}$ through antioxidant depletion
			Ultrafine particles (UFP, $PM_{0.1}$)	Continuous monitoring	Condensation particle counter (CPC)
			NO_2	Gradko® circular diffusive sampler for NO_2	UV–Vis spectrophotometry
			O_3	Radial symmetry diffusive sampler	UV–Vis spectrophotometry
			CO_2	Continuous monitoring (transportable instrument)	NDIR (non dispersive infrared) detector
			VOCs	Radiello® passive sampler: diffusive body code 120-2 and cartridge code 145	Thermal desorption, gas chromatography mass spectrometry (TD-GC-MS)
			Carbonyls	Radiello® DNPH passive sampler: diffusive body code 120-1 and cartridge code 165	High performance liquid chromatography coupled with UV detection

EXPOLIS Study[63,184]	Homes	Microenvironmental monitors (MEMs) suitcase; indoor and outdoor	$PM_{2.5}$	EPA-WINS Impactor equipped with 47-mm Gelman Teflo filters	Gravimetric analysis Elemental analysis by energy-dispersive X-ray fluorescence (ED-XRF)
			VOCs	Active sampling with glass tube containing two stacks of active charcoal (only in Basel)	CS_2 desorption with GC-MSD analysis
				Active sampling on stainless-steel tubes packed with Tenax TA	Thermal desorption with GC-MSD/FID analysis
			Carbonyls	Active sampling with glass tube containing two stacks of active charcoal (only in Basel)	CS_2 desorption with GC-MSD analysis
				Active sampling on stainless-steel tubes packed with Tenax TA	Thermal desorption with GC-MSD/FID analysis
RIOPA Study[168]	Homes	Metallic rack; indoor and outdoor	$PM_{2.5}$	Harvard type Impactor equipped with Teflon filters	Gravimetric analysis
			VOCs	Organic Vapor Monitors (Model No. OVM3500) for passive VOCs sampling on charcoal pad	Extraction by ultrasonication into a high purity 2:1 acetone/CS_2 solvent and GC-MS analysis
			Carbonyls	Active method with DNPH-coated cartridge	Extraction with acetonitrile and HPLC/UV analysis
				Passive Aldehydes and Ketones Sampler (PAKS)	Extraction with acetonitrile and HPLC analysis with fluorescence detection
AIRMEX Study[186,187]	Public buildings, schools/kindergartners, private homes	Wire attached to the ceiling or metallic rack; indoor and outdoor	VOCs	Radial type diffusion passive samplers (Radiello®, charcoal/carbograph type)	Chemical desorption with CS_2 followed by GC-FID analysis
			Carbonyls	Radial type diffusion passive samplers (Radiello® DNPH-covered)	Extraction with acetonitrile and HPLC/UV analysis
AUDIT Project[190]	Offices	Type of arrangements not specified; indoor and outdoor	PM10	Not specified	Not specified
			CO	Continuous monitoring	Dispersive IR or direct reading colorimetric detector
			CO_2	Six different instruments for either continuous or spot CO_2 measurements	NDIR (non dispersive infrared) detector
			Total VOC	Direct measurement	Infrared absorption using photoacoustical detection

(*continued*)

Table 9. Continued.

IAQ study and/or (reference)	Investigated indoor environment	Type of fixed site arrangement	Investigated pollutants	Sampling methods	Analysis or detector
[64]	Residetial buildings	Microenvironmental monitors (MEMs) suitcase; indoor and outdoor	PM_{10}	GK2.69 cyclone connected to a personal sampling pump with a flow rate of 1.6 l/min	Gravimetric analysis
			$PM_{2.5}$	GK2.05 cyclone connected to a personal sampling pump with a flow rate of 4.0 l/min	Gravimetric analysis
			Size-segregated PM	Personal cascade impactor sampler (PCIS) connected to a sampling pump with a flow rate of 9 l/min	Gravimetric analysis
			NO_2	Radial symmetry diffusive sampler	UV–Vis spectrophotometry
			O_3	Radial symmetry diffusive sampler	UV–Vis spectrophotometry
			CO	Continuous monitoring	Passive electrochemical sensor
			CO_2	Continuous monitoring	NDIR (non dispersive infrared) dectector
[172]	Schools	Tripod or specific suitcases; indoor and outdoor	$PM_{2.5}$	Continuous monitoring	Nephelometer, at a flow rate of 3 l/min with a size-selective inlet for $PM_{2.5}$
			Size-segregated PM	Continuous monitoring	Optical particle counter (OPC)
			NO_2	Radial symmetry diffusive sampler	UV–Vis spectrophotometry
			O_3	Radial symmetry diffusive sampler	UV–Vis spectrophotometry
			CO_2	Continuous monitoring	NDIR (non dispersive infrared) detector
			CO	Continuous monitoring	Passive electrochemical sensor
P-TEAM Study[188,189]	Homes	Microenvironmental monitors (MEMs) suitcase; indoor	PM_{10}	Marple/Turnen impactor connected to a 10 l/min pump	Gravimetric analysis

present.[44] Moreover, air introduced to an O_3 instrument was passed through a Teflon filter to limit interference from aerosol particle scattering.[44] When VOCs are released into the atmosphere, they react with nitrogen oxides (NO_x) to create ozone molecules.[178]

When a number of air sampling systems and real-time analyzers for the monitoring of some key indoor air pollutants are assembled in a single monitoring unit, it is first necessary to provide an adequate power supply to each active sampler/analyzer in order to perform a survey based on a sampling duration commensurate with i) the time of occupation of the specific indoor environment by people (typically 8 hours/day for occupational environments e.g., offices and up to 24 hours/day for residential environments); and ii) the reference time with respect to which indoor guideline values or estimated thresholds for protecting indoor occupants against adverse health effects are expressed.

The most classic experimental set-up for indoor air monitoring is based on active or passive air sampling in a fixed location, which requires a main power supply to be used for achieving of the aforementioned objectives without major problems, except for the possible presence of cables and extensions in some critical areas of the investigated room (e.g., those characterized by frequent people movement). Conversely, when a micro-environmental or personal monitoring approach is required for exposure assessment purposes or to be easily moved across various premises, some mobile monitoring options typically consisting of a shoulder bag, backpack, trolley suitcase, etc. should be used. In this case all the active samplers/analyzers are battery powered and it may happen that the autonomous power provided by internal batteries is not enough to ensure a correct sampling duration. If that is the case, the portable monitoring system should be necessarily equipped with an ancillary battery supply with suitable charge supply and an ad hoc electronic board should be also developed for specific voltage control.[16,179] This solution will thus guarantee a long-term mobile monitoring, but at the expense of lightness and compactness, which could lead to a hindrance of usual activities.

When active sampling is required, which is for instance the only choice when good quality data about indoor air concentrations of PM are needed, some criticalities can typically arise. First, air is forced to enter the sampling train through sampling inlets, and this can alter the aerosol sampling efficiency if sampling inlets are too close each other and at relevant air velocities and sampling flow rates. Then, the operation of sampling pumps involves noise and heat emissions, depending also on the instrumental type and on operative flow rates. Moreover, the use of active sampling increases the sampling system size and weight. That is the reason why, in an EU-wide study, volatile organic compounds were sampled using a sampling tube fed by the vacuum created by a pump for PM monitoring *via* a T-joint system. The sampling flows were controlled by an experimentally designed flow-restrictor made of stainless-steel capillary tubes placed before and after the sampling tubes and adjusted with a valve.[180]

Multipollutant stations specifically developed for indoor air monitoring and containing sampling pumps are generally well-sealed and insulated with sound-absorbing materials to reduce noise emissions at levels that would typically ensure acoustic wellness for people (e.g., The WHO sleep disturbance noise guideline corresponds to a level of 42 dBA outdoors[181]). Although the use of sound-absorbing materials is fundamental in this regard, this typically involves a parallel thermal insulation and consequent potential increase in temperature inside the monitoring station, such as to cause important interferences on the measurement of a series of IAPs monitored *via* real-time monitoring instruments based on electrochemical sensors e.g., for CO and NO_2. In such a case, it would be first of key importance to verify that the maximum allowable temperatures indicated by the manufacturers are not exceeded, and then the instrumental baseline should be corrected for temperature based on field- or lab- experimental curves.[64,182] Moreover, the emission of chemicals by some peculiar instruments, as well as of water or heat, should be controlled or abated not to cause important changes in the indoor air composition or microclimate (see for instance the abatement system used for 2-propanol emission control in[16]).

A further possible criticality of multiparametric monitoring systems for indoor pollutants can be the presence of mechanical vibrations directly generated by the functioning of pumps used in active sampling systems and, in the case of mobile monitoring units, by rapid displacements, jolts and mechanical stress caused by moving, walking or produced by contact of possible wheels with the ground. As an example, tilting with respect to the horizontal position can generate data loss in the case of ultrafine particle counters or low frequency vibrations can cause a disturbance to the instrument signal and it is therefore useful to remedy these problems by equipping the multiparameter system of suitable cushioning systems and balancing or anti-tilting devices to mechanically keep the sensitive instruments as much as possible disjoined from the rest of the system.[16]

5. Conclusions

Surveys aiming at monitoring of indoor air quality in dwellings require unobtrusive, energy-saving, and cost-efficient instrumentation such as portable, accurate on-line trace gas analyzers or diffusive samplers followed by off-line analysis. Nevertheless, such measurements can provide either spatial or temporal resolution but rarely both.[183] Costly and large benchtop instruments for real-time monitoring, such as proton transfer reaction-mass and negative ion chemical ionization mass spectrometers have been successfully used for monitoring of organic contaminants mainly in laboratory and chamber studies to understand chemical processes. However, these instruments placed in the study area influence their immediate environment by heating air, representing new surface area, and reducing volume for study. Some attempts have been made to solve this problem such as placing this type of equipment in a detached room next to the studied dwelling or by analyzing removable building materials in laboratory. Moreover, in order to plan the monitoring activity by identifying the appropriate techniques for the collection and chemical characterization of multiple pollutants present in indoor environments, it is first necessary to define the correct sampling duration. For example, to compare the concentration obtained with a reference guide value, the monitoring duration must comply with the reference time associated with these values. By way of example, this is 24 h for PM_{10} and $PM_{2.5}$. If the sampling duration is less than that provided by the guideline/reference value and it is chosen based on the work activities, residence time of occupants, or activation of internal sources, the measurement provides only an indicative value. In return, that will be useful to identify and estimate the possible contributions to the contamination levels of a given environment, induced by the activity of the individual sources in operation at the time of detection.[170] Also, long-term measurements including periods of occupation of indoor environments by people and periods in which these environments are unoccupied should be considered only as indicative.

Therefore, the field campaign measurements must be accompanied by a questionnaire to be filled by occupants, facility management, etc. to acquire information about the presence/absence of occupants and what type of activities they carry out within the premises, on the operation of mechanical ventilation, heating systems, etc. Thus, the parameters and conditions of use of the indoor environments for which the measurements can be considered representative must be specified. To summarize, measurements should be planned in relation to the monitoring objectives, considering the activities, the source(s), possible temporal variations (hourly, daily, monthly, seasonal), and microclimatic variables as well.

Multipollutant monitoring devices should then comply with the need of obtaining reliable results while being functional to reach the monitoring goals (e.g., source identification/apportionment, IAQ assessment, IAQ real-time management, risk assessment arising from exposures to multiple chemicals).

Acknowledgments

This publication is based upon work from COST Action CA17136 supported by COST (European Cooperation in Science and Technology) (www.cost.eu) for which the authors are grateful. The support and original concept designed by Nicola Carslaw (University of York) is hereby, acknowledged.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work is based upon work from COST Action INDAIRPOLLNET, supported by COST (European Cooperation in Science and Technology).

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