

Review

The Application of In Situ Methods to Monitor VOC Concentrations in Urban Areas—A Bibliometric Analysis and Measuring Solution Review

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Abstract: Urbanisation development affects urban vegetation both directly and indirectly. Since this process usually involves a dramatic change in land use, it is seen as likely to cause ecological pressure on local ecosystems. All forms of human activity, including urbanisation of areas close to residential buildings, significantly impact air quality. This study aims to identify and characterise different measurement solutions of VOCs, allowing the quantification of total and selective compounds in a direct at source (in situ) manner. Portable devices for direct testing can generally be divided into detectors, chromatographs, and electronic noses. They differ in parameters such as operating principle, sensitivity, measurement range, response time, and selectivity. Direct research allows us to obtain measurement results in a short time, which is essential from the point of view of immediate reaction in the case of high concentrations of tested compounds and the possibility of ensuring the well-being of people. The paper also attempts to compare solutions and devices available on the market and assess their application.

Keywords: BTEX; health; portable sensors; urbanisation; VOCs; well-being



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1. Introduction

1.1. The Main Contents in This Review

This manuscript contains the results of the initial literature search of the terms “urban areas” and “VOCs” in five academic databases, as well as a bibliometric analysis of the records from the Scopus database. As part of this analysis, overlay a visualisation of the Scopus results with LinLog normalisation was made, as well as a network graph between the most cited papers. Moreover, the paper reviews the solutions and devices of various in situ methods to monitor VOC concentrations in urban areas available on the market with an evaluation of their application, their strengths, and weaknesses.

1.2. VOCs in Urban Areas

The intensive development of cities around the world is inseparable from municipal and industrial infrastructure [1]. The process of urbanisation is characterised by many different aspects, among which are demographic, economic, and sustainability aspects [2–6].

Urbanisation development affects urban vegetation both directly and indirectly. Since this process usually involves a dramatic change in land use, it is seen as likely to cause ecological pressure on local ecosystems [7–9]. All forms of human activity, including urbanisation of areas close to residential buildings, significantly impact air quality. An increasing number of pollutants, including volatile organic compounds (VOCs), originates, among other things, from municipal (landfills, sewage treatment plants, mechanical-biological waste treatment plants) and industrial (refineries, breweries, distilleries, paper, wood, or meat-processing plants) facilities, but also from motor vehicle exhausts [10–13]. The listed sources are of anthropogenic origin. However, some sources of VOC emissions are also

natural, represented mainly by green vegetation [14,15]. The term VOC should be understood as a broad group of organic chemical compounds, including natural compounds such as terpenes and alcohols, carbonyl compounds such as ketones, aldehydes, ethers, and aromatic hydrocarbons, and acids, characterised by vapour pressure at 0.01 kPa at 20 °C and boiling point from 50–100 °C to 240–260 °C [16–20]. The temperature ranges given are based on the type of VOC. WHO has classified VOCs, found mainly indoors, into three groups based on the volatility of their compounds, such as very volatile (gaseous) organic compounds (VOCs, example compounds: propane, butane, methyl chloride), volatile organic compounds (VOCs, example compound: formaldehyde, d-Limonene, toluene, acetone, ethanol (ethyl alcohol), 2-propanol (isopropyl alcohol, hexanal)), and semi-volatile organic compounds (SVOCs, example compounds: pesticides (DDT, chlordane, plasticisers, fire retardants)) [21]. In addition, VOCs are poorly soluble in water, and in the atmospheric zone, they participate in photochemical reactions and thus may contribute to photochemical smog pollution [22]. Some volatile organic compounds consume ozone in the stratosphere and cause ozone holes. Others, e. g., benzene, toluene, and formaldehyde are toxic and harmful to human health, due to carcinogenic, teratogenic, and mutagenic effects [23,24].

The compounds of most interest to scientists are aromatic compounds such as benzene, toluene, xylene, and ethylbenzene, commonly known as BTEX.

The World Health Organization (WHO), in its published guidelines for air quality in Europe [25], establishes maximum concentration values for selected substances in the air: 260 g/m³ within 1 week for toluene, 100 g/m³ within 30 min for formaldehyde, and 250 g/m³ within one year for tetrachloroethylene. In addition, the WHO guidelines also indicate guide values for benzene corresponding to concentration levels associated with an excess mortality risk of 1/10,000, 1/100,000, and 1/1,000,000, equal to 17, 1.7 and 0.17 g/m³, respectively. Almost all plants can synthesise a variety of VOCs, numbering up to hundreds of thousands, and subsequently emitting these compounds into the atmosphere. VOCs synthesised and emitted by plants include simple forms, such as methanol (CH₃OH), and very complex ones, such as (1S,3aR,4S,8aS) decahydro-4,8,8-trimethyl-9-methylene-1,4-methanoazulene, or longifolene, a tricyclic member of the C₁₅H₂₄ family of sesquiterpenes. Biogenic volatile organic compounds, BVOCs, play an essential role as signals and protectants against biotic factors, as well as abiotic stress [26,27]. Most BVOCs are synthesised by one of the three main metabolic pathways, i.e., isoprenoid, lipxygenase, or chiacid [28–32]. BVOCs are emitted by both above- and below-ground plants, with the most significant emission occurring through the leaves. Increased emission of the mentioned compounds also occurs when plants are damaged [33,34].

A wide range of plants, mainly flowers, is characterised by the emission of VOCs, inducing a positive olfactory sensation. For this reason, they are deliberately planted in urban areas (urbanised areas). In addition to visual considerations, their task is also to improve the well-being of residents through smell [35,36].

Plant odours are generally experienced differently in herbaceous waste, which usually undergo a composting process [37]. Characterisation of VOCs emitted by green waste during composting was carried out in work by Kumar et al. [38]. The researchers detected 100 types of VOCs during the study, including those belonging to the BTEX group, in the emitted process gases.

The implementation of sustainable urban development should involve the minimisation of environmental pollution [39]. Therefore, environmental monitoring is crucial [40]. Some VOCs are also classified as pollutants, mainly due to their negative impact on human health (e.g., skin and respiratory diseases) and comfort of living (e.g., odour nuisance). Correct assessment of air quality is not an easy task due to the dynamics of changes. Therefore, performing frequent measurements at the source seems to be a suitable method to conduct VOC monitoring [41–43].

Large quantities of VOCs are emitted into the troposphere from anthropogenic and biogenic sources—for example, large quantities of oxygenated VOCs are emitted from vegetation, while non-methane organic compounds are emitted by combustion sources

(vehicle and fossil-fueled power plant emissions), fuel storage and transport, solvent usage, emissions from industrial operations, landfills, and hazardous waste facilities [44]. As Atkinson wrote, advances in our knowledge of the kinetics, products, and mechanisms of the tropospheric reactions of VOCs does not only result in more accurate chemical mechanisms for use in urban and regional airshed models, but also in more accurate estimates of VOC reactivities [44].

This study aims to identify and characterise different measurement solutions of VOCs, allowing the quantification of total and selective compounds in a direct-at-source (in situ) manner. Direct research allows us to obtain measurement results in a short time, which is vital from the point of view of immediate reaction in the case of high concentrations of tested compounds and the possibility of ensuring the well-being of people. The paper also attempts to compare solutions and devices available on the market and assess their application.

2. Bibliometric Analysis

The initial literature search conducted on 1 April 2022 was based on a current search for the terms “Urban areas” and “VOCs” in titles, abstracts, and keywords in five academic databases: Web of Science, Scopus, Science Direct, PubMed, and SAGE. The data range was set to “published all years to the present”. The document type was set to “scientific articles and review articles”, and the language was set to “English”. The results are presented in Figure 1. Of the five databases used, Scopus recorded the highest number of publications of a specific type related to the occurrence of VOCs in urban areas, with 875 articles. Ultimately, records from the Scopus database were selected to continue the review.

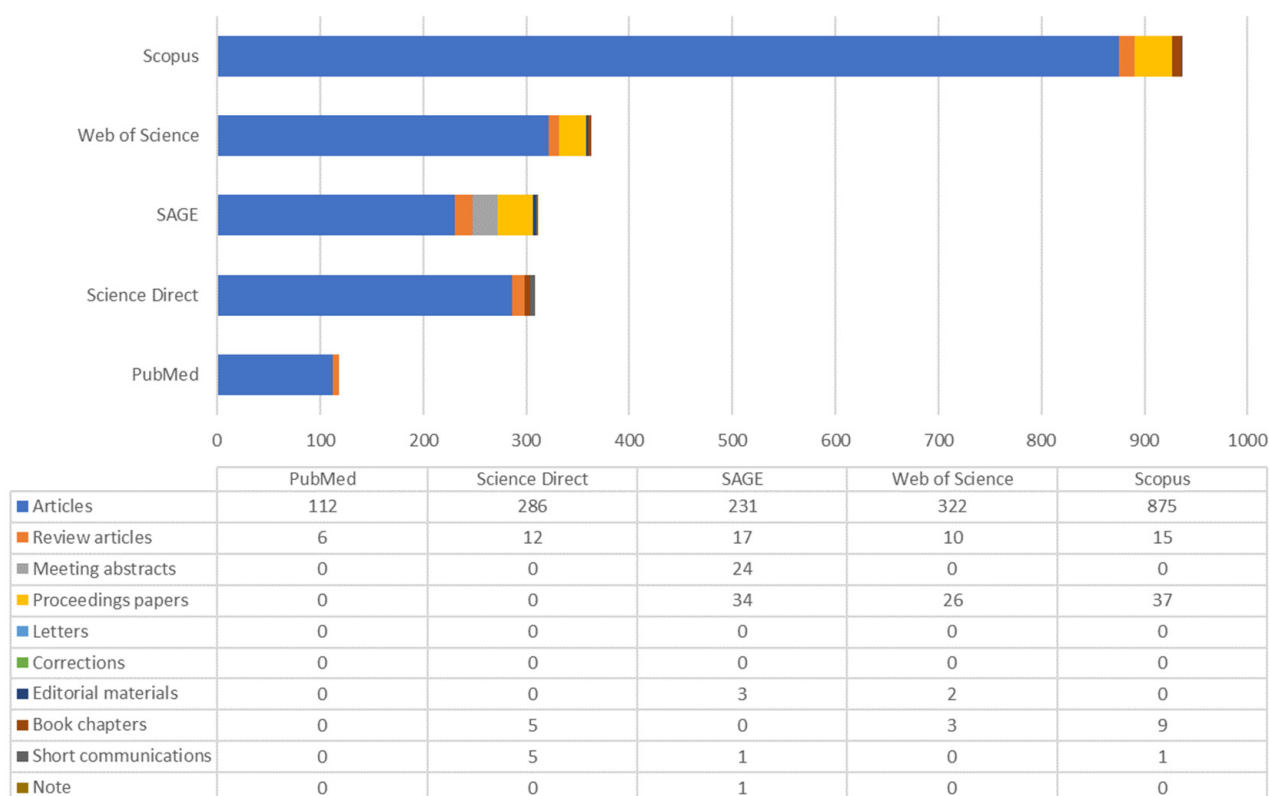


Figure 1. Results of bibliographic search for the terms “urban areas and “VOCs” in five academic databases.

The temporal distribution of the collected bibliographic records on VOCs in urban areas was examined. The earliest related work was published in 1989. The number of publications published each year is shown in Figure 2.

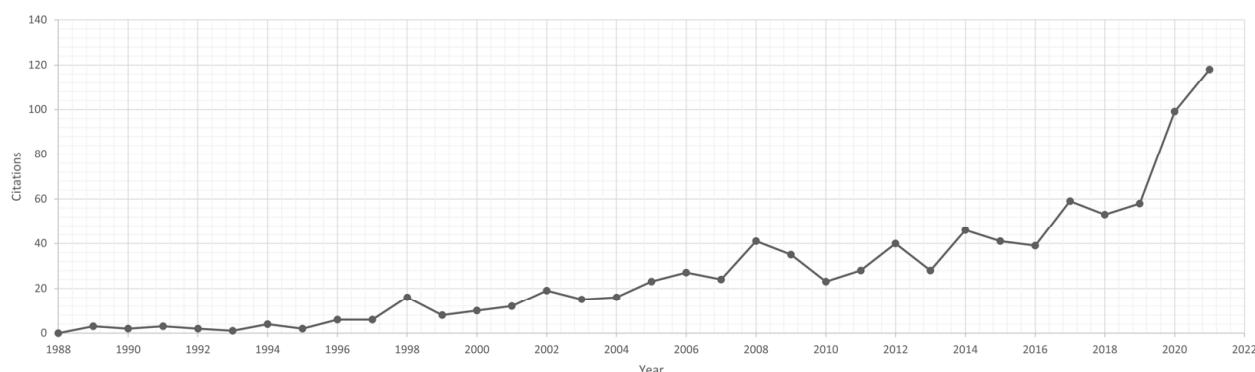


Figure 2. The number of publications about VOCs in urban areas published in each year.

Figure 3 contains an overlay visualisation of the Scopus results with LinLog/modularity normalisation, prepared using VOSviewer software to analyse and visualise the relationships among the authors, countries, journals, co-citations, and terms [45,46]. The energy-based (force-based) technique has advantages such as good layout quality, ease of implementation, and code flexibility. It is based on the assumption that there is an attraction force between the connected vertices while all vertices repulse each other [47]. The LinLog model is an energy algorithm to provide an efficient way to minimise the energy in our force-directed cluster graph. The lowest energy is achieved by minimising the ratio between the average edge length to average distance to all nodes. The LinLog model provides the minimal ration of the arithmetic mean of the edges to the geometrical mean of node distances. Therefore, the nodes were assigned in a network to clusters. A cluster is a set of closely related nodes. Each node in a network is assigned to exactly one cluster, while the number of clusters is determined by a resolution parameter—the higher the value of this parameter, the larger the number of clusters.

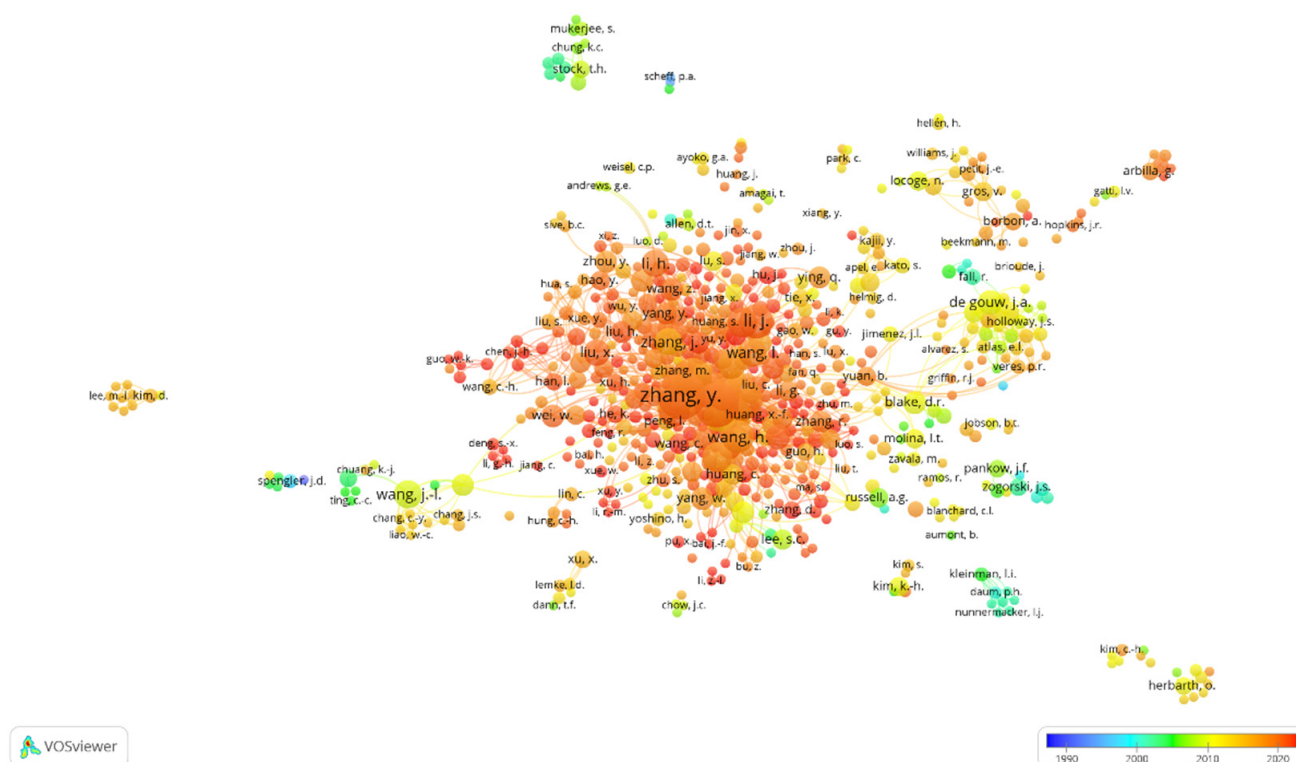


Figure 3. Overlay visualisation with publication year.

The set is divided into 20 clusters—the largest contains 86 items, the smallest 6 items. Colours mark individual time periods.

Among the articles on volatile organic carbons in urban areas, the number of articles published in 2020–2021 was much higher than in previous years, reaching 118 in 2020. The bibliographic analysis showed that more and more attention is being paid to this topic. Figure 4 provides a summary of documents by author, subject area, and country/region.

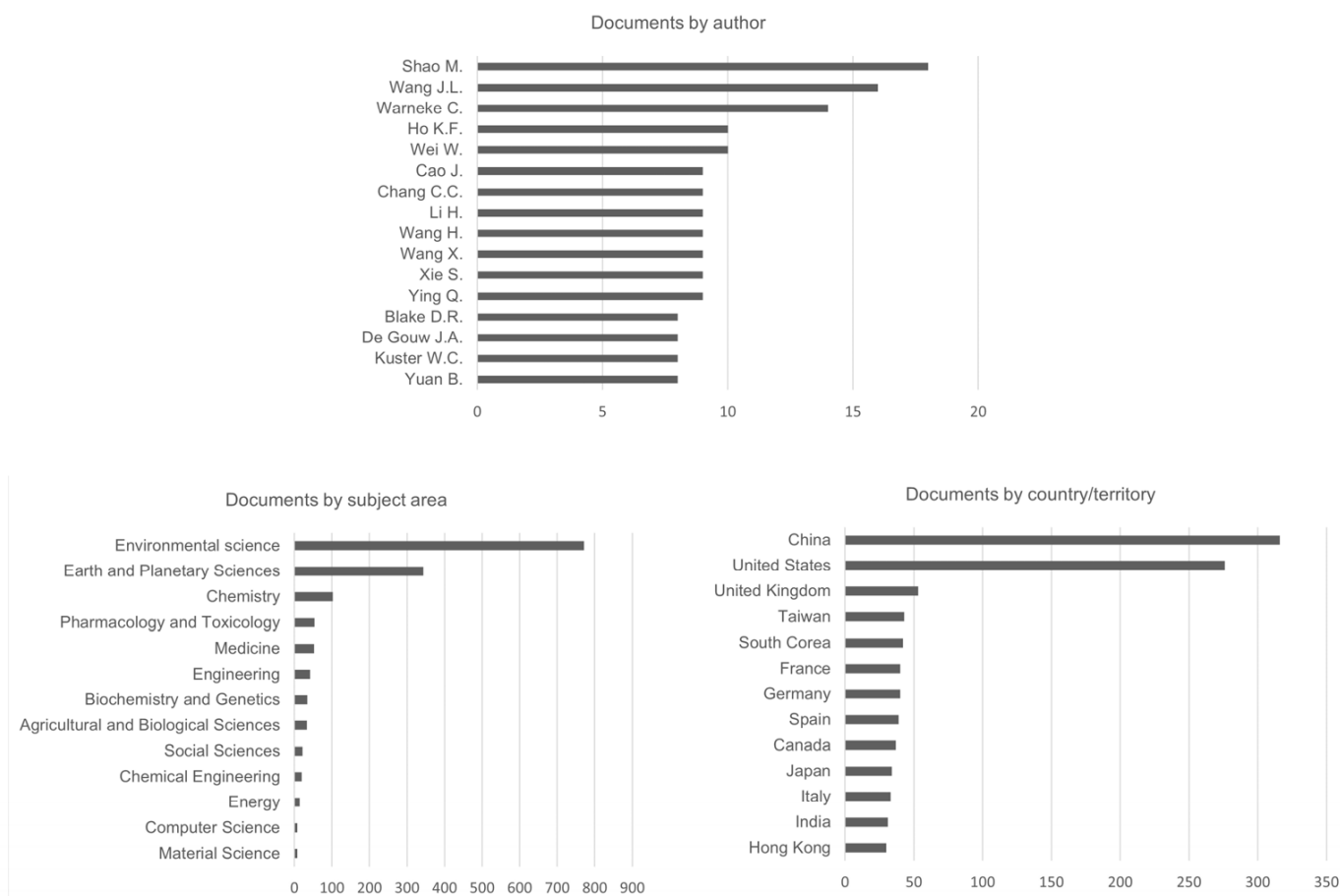


Figure 4. Summary of documents by author, subject area, and country/territory.

The most articles on VOCs in urban areas were written by Shao M. (18 articles), Wang J.L. (16), Warneke C. (14), Ho K.F. (10), and Wei W. (10). As many as 771 articles fell into the subject area of environmental science, compared with 343 in earth and planetary science, 102 in chemistry, and 53 in pharmacology and toxicology. A total of 316 articles were affiliated with China, 276 with the United States, and 53 with the United Kingdom. Among the articles that are most cited are:

- Atkinson R. (2000) [44]—cited by 2182 authors.
- Carter W.P.L. (1994) [48]—cited by 1101 authors.
- Khalili N.R., Scheff P.A., Holsen T.M. (1995) [49]—cited by 1093 authors.
- Hoek G et al. (2008) [50]—cited by 920 authors.
- Volkamer R. et al. (2006) [51]—cited by 766 authors.
- Li K. et al. (2019) [52]—cited by 613 authors.
- Watson J.G. et al. (2001) [53]—cited by 442 authors.

The connections between Atkinson [44] and highly cited articles are presented in Figure 5.

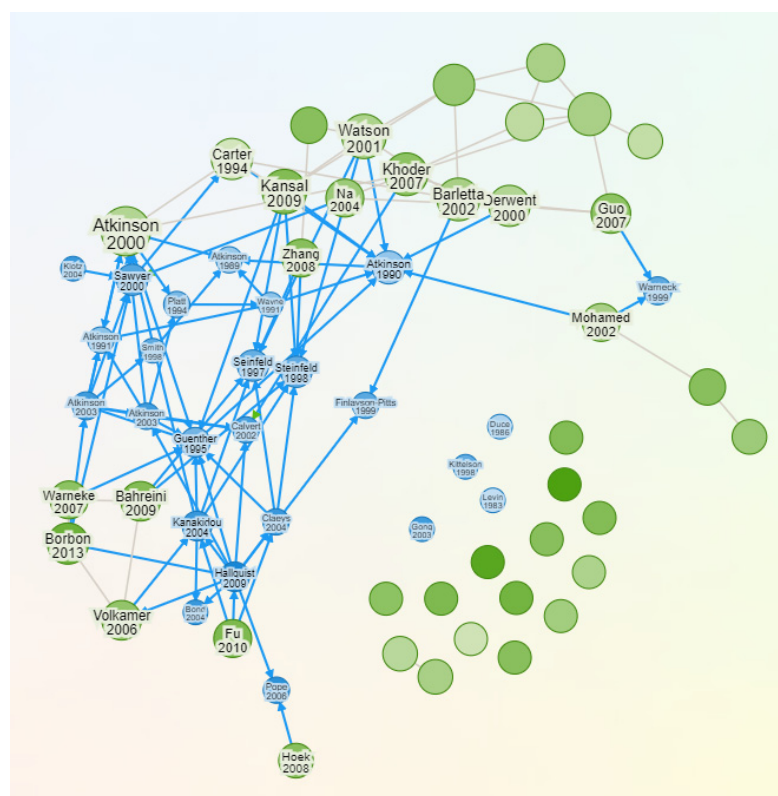


Figure 5. Connections between Atkinson [44] and highly cited articles [48–53].

A citation network graph contains green nodes representing the most cited papers using the phrases “urban areas” and “VOCs” in the Scopus database, and blue nodes representing what in this case are citations of Atkinson [44]. Each line connecting the nodes represents a citation relationship between the papers. As of the nodes themselves, intensity of the colours is based on recency, and the size of the blue nodes is based on the number of connections they have with the green ones. Since some of the green nodes are not citations of that publication, there are no connections between them.

3. Characteristics of Various Methods for VOC Determination

Over the years, there have been rapid developments in the scientific field, including devices measuring VOCs [54–60]. Portable devices for direct testing can generally be divided into detectors, chromatographs, and electronic noses. They differ in operating principle, sensitivity, measurement range, response time, and selectivity [61–64]. The individual measurement solutions are discussed in more details later in this paper.

3.1. Portable Gas Detectors

Portable gas detectors are devices equipped with more sensors to quantify one or more gases [1,58]. There are several primary and commonly used types of sensors available on the market: photoionisation (PID), electrochemical (amperometric, EC), metal oxide semiconductor (MOS), nondispersive infrared (NDIR), and thermal (PELLISTOR). They can immediately detect a disturbance in a process, the environment, or health conditions [65]. The emphasis is on new column heating techniques requiring no convection oven [8–11,66–69], microscale inline preconcentrators and inlet devices [12,70], arrays of partially selective sensors [13–18,71–76], and methods of micromachining to produce small and efficient columns [10,19,20,68,77–79]. Table 1 presents the characteristics of the different types of sensors.

Table 1. Characteristics of basic types of sensors.

Sensor Type	Operation Principle	Quantification	Advantages	Limitations	References
PID	Sensor operation is based on the ionisation phenomenon; photons of high energy, usually in the UV range, are used; ionisation of molecules occurs after excitation of molecules by UV light; the photon energy is typically 10.6 eV	0–20,000 ppm depending on the manufacturer	Low level of quantification, as low as one ppb	Lack of possibility to detect compounds with high ionisation energy (e.g., acetylene—11.40 eV, chloroform—11.37 eV, chloromethane—11.26 eV; usually lacks selectivity—ionisation of all compounds with energy lower or equal to the lamp energy	[80–86]
EC	An electrochemical reaction results from the transfer of charge from the electrode to the electrolyte. The electrolyte may be in a solid, gel, liquid, or gaseous state. Electrochemical cells are equipped with two, three, or four electrodes depending on the manufacturer. The gas to be measured diffuses into the measuring electrode via the diaphragm sensor. According to Nernst's law for electrochemical reactions, the transfer of electrons induces a current whose measure is proportional to the gas concentration.	0–100 ppm depending on the manufacturer	Low purchase cost, energy-saving, compact; response time is approx. 120 s dependent on the temperature	Not much selectivity; lower limit of quantification typically at 0.1 or 2 ppm.	[87–89]
MOS	The principle of the sensor is based on the receptor and transducer function. The function of the receptor is based on the recognition of a given gas at the gas–solid interface by an electron change on the surface of the metal oxides. The role of the transducer is to transfer the surface phenomenon to a change in electrical resistance in the sensor. There are two main types of semiconducting metallic oxides: n, the primary carrier is an electron, and p, in which the leading carrier is a hole.	0–5000 ppm depending on the manufacturer	Small size, low purchase cost, low energy consumption, good sensitivity.	Low selectivity.	[90–94]

Table 1. Cont.

Sensor Type	Operation Principle	Quantification	Advantages	Limitations	References
NDIR	The principle of operation is based on the absorption of infrared radiation waves (IR) of a specific length by individual gases. If there is a particular gas in the radiation area, some of the radiation is absorbed. The sensor consists of a source of IR radiation and a detector that measures its intensity. The decrease in intensity of the IR radiation corresponds to the concentration of the gas.	0–100%	Possibility to work in conditions without access to oxygen.	Low selectivity; rapid temperature changes may cause erroneous measurement results. The presence of corrosive gases can damage the sensor.	[73,95]
PELLISTOR	The PELLISTOR consists of two spirals of thin platinum wire embedded in aluminium beads and connected to a Wheatstone bridge. One of the beads is impregnated with a unique catalyst to promote the oxidation of combustible gases, while the other is modified to inhibit oxidation. Platinum spirals are passed through an electric current and heated to a temperature that oxidises the combustible gases on the catalyst. The temperature rise is a measure of the concentration of flammable substances.	0–100% LEL (lower explosion limit) depending on the manufacturer	Low cost of purchase; fast response time; stability	Sensor activity requires 10% oxygen content in the tested gases	[78,96,97]

There are several companies manufacturing gas detectors, for example, Alpha-Sense (Great Notley, UK), Dräger (Lübeck, Germany), Ion Science (Cambridge, UK), Mocon-Baseline (Lyons, CO, USA), and RAE System (San Jose, CA, USA). Manufactured detectors and dedicated sensors differ mainly in purchase cost, detection range, and ability to work with sensors of other gases.

3.2. Portable Gas Chromatographs

Portable gas chromatographs, otherwise known as micro-GC or portable GC, are exciting analytical devices from the point of view of environmental monitoring, the chemical industry, medicine, as well as system failure detection [98,99]. The GC–MS systems can accurately identify and quantify different types of substances even at very low concentrations. In addition, they have a high potential for differentiating individual substances. The different GC–MS solutions on the market differ in the performance of the gas and steam analysers [100,101]. The main assumption of portable gas chromatograph manufacturers is to create a small-sized device with detection capabilities comparable to laboratory chromatography. Microsensors are often used in this kind of device due to their low carrier gas consumption, small size, low power consumption for operation, and quite good sensitivity compared to standard sensors. Examples of microsensors include the solid-state thermal conductivity detector (TCD or SSD) [102–104], photoionisation detector (PID), [105,106], surface acoustic wave (SAW) detector [107–109], and the flame ionisation detector (FID) [99,110–112].

GC–MS is one of the most sought-after methods for identifying chemical substances. In the literature, this method is referred to as the gold standard [113–115]. Integration of these systems requires two independent technologies. The mixtures are separated into their individual components using GC and MS. The GC–MS combines fast analysis, high

resolution, excellent quantitative results, and moderate costs. MS systems require few samples to provide qualitative and quantitative data [116].

The most critical component of the gas chromatograph system is the separation column. Traditional conventional columns have a circular cross section, while microcolumns usually have a rectangular cross section [107]. Portable chromatographs usually use open columns. The low thermal weight of the open tubular column GC allows the analysis of the temperature program at relatively low power when the heating is limited to the column itself, as with resistance-heated columns. Thanks to this arrangement of the columns, a significant temperature increase is possible, even if the device is operated with a battery. The resulting fast analysis speeds increase the ability to perform GC analyses in the field [117].

Compared with the traditional GC system, the separation performance of the μ GC system decreases greatly, mainly because the channel length of the chromatographic column is reduced, and the control accuracies of temperature and carrier gas pressure are much less than those of the traditional GC system [118]. Chen et al. proved that for low-carbon mixtures, the integrated μ GC chip has a separation effect similar to that of heating with a column thermostat. With low power consumption and a small size, the integrated μ GC chip may be applied to a portable gas chromatograph for detecting the low-carbon mixtures [119].

Studies conducted by Wang et al. [99] on μ FID showed its much better sensitivity and higher signal-to-noise ratio due to its ability to operate at high polarisation voltage than conventional FID. In their study, the researchers also obtained a reduction in gas consumption of about 30% compared to the conventional counterpart. The authors note that features such as reduced operating costs and energy savings make this type of detector suitable for portable GCs when conducting field and in situ studies.

Zampetti et al. [120], in their work, presented the development and application of micro cartridge-based μ PID in chromatography. This system is characterised by its small size, light weight (about 1 kg), ease of handling, low production cost (about 1000 €), simple operation, low power consumption, and good accuracy and selectivity for BTX substances.

Miresmailli et al. [121] performed a quantitative analysis to compare the results obtained with a portable GC equipped with a SAW detector (zNoseTM, Electronic Sensor Technology, Newbury Park, CA, USA), an electronic nose, and a conventional GC. The researchers' results show correct identification of compounds by zNoseTM and consistent results with conventional GC, despite environmental conditions, which is extremely important for fieldwork. Due to its fast analysis time of about 3 min per complete cycle, it can be applied to monitoring changes in VOCs emitted by plants and other organisms [122]. The main disadvantage of this analysed portable GC is its relatively short column (at about 1 m), limiting the resolution of peaks when there are many of them in the sample.

A few years later, a study on the same device was performed by Li et al. [123]. During the analysis of different aromatics, they compared the results of VOCs with a carbon content of 4 to 16 molecules obtained using two instruments: a portable GC with SAW detector and a conventional GC. The results obtained from the two devices show a correlation of 95%, which, according to the authors, allows the conventional GC to be replaced by a portable GC in aromatics testing.

Another portable GC zNose[®] (model 4300) was presented in work by Meciariova et al. [124]. The researchers used the device to analyse the composition of indoor air in a model room. The device used allows the determination of VOCs with a carbon content of 4 to 24 molecules. Similarly to Miresmailli et al. [121], the authors point out a weakness of the device, in the form of a short column, which reduces the resolution of VOCs with similar retention times.

GC–MS systems are particularly in demand for military applications where the weight and speed of marking are critical [125].

3.3. Electronic Noses (E-Noses)

Electronic noses are instruments designed to mimic the human sense of smell and detect and characterise complex or straightforward odours. One way e-noses can work is to characterise gaseous mixtures by providing their odour pattern without identifying individual odour-forming compounds, like the human nose [126–128]. E-noses are equipped with sensor arrays where each element responds to several different chemicals or classes of chemicals. Selective operation of each component is not required, since the main goal of the sensor system is to have as much chemical diversity as possible, allowing the device to respond to as many chemicals as possible [95]. These devices are characterised by high sensitivity, and their main disadvantage is the restriction of concentration measurements to “learned” ones [129,130].

Portable electronic noses, often used in outdoor environments, require real-time calculations to obtain data analysis results. Common pattern recognition methods that rely on high-performance computers tend to fail in mobile devices, resulting in the need to develop online computing algorithms that can run on embedded e-nose portable systems [131].

The research conducted by scientists is focused on implementation of the e-nose with the possibility of high-performance recognition algorithms. However, as algorithms become more and more complex, making quick calculations becomes more and more difficult. An alternative could be to transfer data to servers for real-time computing, but it can make other problems and difficulties related to internet network coverage [132,133].

3.4. Gas Detectors with GC Function

An innovative solution for the in situ determination of VOCs is a gas detector equipped with a PID sensor (10.6 eV) and a chromatographic column. Dräger introduced such a solution in 2019. The PID sensor allows the detection of VOCs at ppm and ppb levels, being widely used in GC systems due to high sensitivity and a large dynamic range [134]. A xenon lamp within PID sensor generates 9.6 eV photon energy, deuterium lamps 10.2 eV, and krypton lamps 10.6 eV, while argon lamps produce 11.7 eV energy [135–137]. The device can operate in two modes. The first is detection, which allows to initially and broadly survey an area to pinpoint measurement points. It allows continuous and direct measurement of TVOCs. The second mode is the analysis mode, which allows making selective measurements for specific compounds selected from a list of approximately 40 compounds, including BTEX. Results are obtained after about 30 s, depending on the retention time of the test compound. The device shows readiness for the following determination after 60 s. The PID sensor is equipped with a hydrophobic membrane to protect it in industrial environments. Calibration of the device is carried out using isobutylene. The device allows setting the sensitivity to a substance using reaction factors. In fact, before each use of the device, a function test with the regulating (calibration) gas is required, which takes about 60 s and is valid for 24 h. A disadvantage of the device, as with portable GC, is that compounds with similar retention times cannot be determined simultaneously [138]. A PID sensor was also used for miniaturised GC to real-time detections of BTEX [139] and fast GC to total OH reactivity measurements [140]. Pang et al. [141] tested PID sensors as parallel secondary detectors in a GC–Q–TOF–MS system to evaluate response factors and response times relative to MS, and quantify this for seventeen species of VOCs and six toxic reagents used in insecticides and chemical weapons. Those researchers proved that simple PID sensors offer potential as GC detectors for toxic chemicals, tested here on organic sulphide and organic phosphonates.

Unfortunately, there is a lack of work in the scientific literature that compares the results obtained with the device with those obtained with conventional GC.

4. Summary and Future Research Work

The process of urbanisation contributes to increased VOC emissions. Many of these compounds have been shown to have very adverse effects on living organisms. Therefore, VOC monitoring is essential. There are many devices available on the market for measuring

VOCs, which differ in their technical parameters and the way they are used. Portable devices allow us to measure VOCs in situ, in order to obtain results virtually instantly. In the context of future work, it is interesting to compare results obtained from different devices in different facilities, because the operation of the devices and the reliability of the obtained results may depend on the characteristics of the plant and area. In situ measurement solutions are increasingly in demand due to their speed of analysis and ease of use.

5. Conclusions

This paper presents different methods for determining VOCs, which can be applied to studies in urban areas. Field studies using in situ methods allow more samples to be taken than laboratory methods and provide immediate information on the concentration levels of the test substances. This is critical to monitoring toxic substances in the air and ensuring the well-being of people.

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