



## Review

State-of-art of the legislation on odour emissions with a focus on the Italian studies<sup>☆</sup>Gaetano Settimo<sup>a</sup>, Pasquale Avino<sup>b,c,\*</sup><sup>a</sup> Department of Environment and Health, Italian National Institute of Health, Viale Regina Elena 299, IT-00161, Rome, Italy<sup>b</sup> Department of Agricultural, Environmental and Food Sciences (DiAAA), University of Molise, Via De Sanctis 1, Campobasso, IT-86100, Italy<sup>c</sup> Institute of Atmospheric Pollution Research (IIA), National Research Council (CNR), Rome Research Area-Montelibretti, IT-00015, Monterotondo Scalo, Italy

## ARTICLE INFO

Handling Editor: Eddy Y. Zeng

## Keywords:

Odour emissions

Olfactometry

e-nose

Sensors

GC-MS

VOCs

Waste

Legislation

European Union

## ABSTRACT

This review would like to point out the state-of-art of the European legislation for the odour pollution determination and management. Odour is generated by a mixture of more or less volatile and persistent compounds that surround us in daily life. European directives impose the use of corresponding technical standards for the application of the limits imposed. The different approaches (chemicals and/or olfactometries) and integrated evaluation methods for measuring and characterizing odour, even if in a very different way in the European territories, will be reviewed and commented. Specifically, the authors will describe and comment the main procedures for odour concentration determination (e.g., multigas sensors, electronic sensors for odour monitoring). It is important to note that the definition of odour does not take into account if an odour is “good” or “bad”, but only if it exists. The limit value must guarantee a total equivalent level of environmental protection and does not involve a greater polluting load in the environment. Further, a deep revision of the Italian situation in terms of legislation and studies, will complete the paper.

## 1. Introduction

The odour emissions (stack, diffusive, fugitive) from settlement sites of artisan communities and complex industrial centers with considerable potential represents one of the nuisance elements that the population perceives most. In particular, the term “stack emission” is related to direct or indirect release of substances, vibrations, heat or noise from individual or diffuse sources from the installation into air, water or land. On the other hand, “diffusive emissions” are emissions of pollutants into the environment through any kind of duct, pipe, stack, chimney, funnel, flue, etc., including emissions from open-top biofilters whereas fugitive emissions are emissions from “point” sources. Basically, the emissions are associated to factors of potential danger to the perception of odours even though they do not always constitute a toxicological problem. Sometimes, they are the cause of competition, social and political conflicts and the non-acceptance of the industries that spread them throughout the territory (Nimby Syndrome) (Rosenkranz and Cunningham, 2003). In general, it is a problem that is not easy to interpret and is constantly evolving: it requires knowledge on the sources of

pollution, on the methods of containment, and diffusion to start the interventions.

An odour is generated by a mixture of more or less volatile, more or less persistent compounds that surround us in our daily life. In particular, as defined by the European technical standard, odour is the sensation perceived by means of the olfactory organ in sniffing certain volatile substances (European Committee for Standardization, 2022). Further, it should also be considered the definition of emission by the IPPC Directive (Directive, 2010/75/EU). In this case, pollution means the direct or indirect introduction, as a result of human activity, of substances, vibrations, heat or noise into air, water or land which may be harmful to human health or the quality of the environment, resulting in a damage to material property, or impairing or interfering with amenities and other legitimate uses of the environment. Odours do not always constitute a toxicological problem both for the nature and for the concentrations. The olfactory sensations and annoyances cause real health problems, such as headaches, gastric disorders, sleep, stress, etc. Measurements and characterizations are now widespread, even if in a very different way across European territories, with different

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Received 5 September 2023; Received in revised form 4 February 2024; Accepted 6 February 2024

Available online 7 February 2024

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approaches (chemical and olfactometric), with integrated evaluation methods and criteria (Brancher et al., 2017).

For the analytical point of view, the analytical methods allow the qualitative and quantitative identification and recognition of the concentration ( $\text{mg m}^{-3}$  or  $\mu\text{g m}^{-3}$ ) of the organic and inorganic compounds present in the air. They prove to be very useful in evaluating the effectiveness of biofilter abatement systems, because they allow you to identify on which chemical compounds they have had an effect on and therefore intervene on the system accordingly. The sensorial methods which are based on the direct measurement of the odour ( $\text{ou}_E \text{ m}^{-3}$ ) through the noses of the people of a commission-panel who instead act as evaluators carried out according to methodological indications. The analytical techniques do not give information about the olfactory effect of the air under examination, because even when the olfactory perception threshold of the individual compounds identified is known, it is not possible to attribute the additive effect to the components that make up the air under examination. It must be clear that each of these two approaches applied separately are only able to cover some aspects of the odour problem. The olfactory effect does not depend only on the compounds present in the air, but also on how it is perceived by the human sense of smell (i.e., subjective component).

Before approaching the text, a consideration is necessary. In this review the authors focused their attention on the European legislation with a stress on the Italian issue. The reason on our attentiveness on the Italian situation depends on the fact that in recent years, Italy has been the country that coordinated several WG norms on European commissions that led to the definition of EN 13725:2022 (i.e., Stationary source emissions - Determination of odour concentration by dynamic olfactometry and odour emission rate) and UNI 11761:2023 (Emissions and air quality - Instrumental odour measure using IOMS, Instrumental Odour Monitoring Systems) documents. Actually, several European countries have only recently equipped themselves with legislative instruments of a prescriptive nature, adopting the two different approaches with the identification of limit values both in terms of  $\text{mg m}^{-3}$  or  $\mu\text{g m}^{-3}$  and in  $\text{ou}_E \text{ m}^{-3}$ . In recent years, only the references contained in the technical standards issued by the respective standardization bodies were used. This review is addressed to the outdoor odour emissions from different industrial (e.g., from refineries, landfills, food industries, tanning industries) and no-industrial (e.g., from livestock farms) plants.

## 2. State-of-art of the European and national analytical procedures

Before starting with the analytical procedures, the authors would like to show some considerations on the advantages and disadvantages of the distinct odour nuisance characterization and quantification techniques. The techniques indicated in the European standards concern the application of a sensory olfactometric-technician evaluation, which uses a dilution tool (olfactometer), to evaluate an odour at different levels of concentration, in a controlled way to a group of evaluators. The dynamic olfactometry allows you to determine the concentration of odour of a sample of air relating to the sensation caused by the sample directly on a panel of appropriately selected people. The odour concentration is expressed in European units per cubic meter ( $\text{ou}_E \text{ m}^{-3}$ ), it is the number of dilutions with neutral air necessary to bring the sample concentration to the concentration of the detection threshold of panel odours. The odour concentration ( $\text{ou}_E \text{ m}^{-3}$ ) is statistically the same as the dilution factor of the perception threshold: that is, a concentration of  $50 \text{ ou}_E \text{ m}^{-3}$  means that the sample has been diluted fifty times to reach the panel threshold. Then there are methods that involve chemical analyzes with sorts, or the use of the Instrumental Odour Monitoring Systems (IOMSs) that continuously allow you to know the extent of the odour impact. IOMSs are tools designed to simulate human odour, which characterizes an odour mixture in its entirety. Therefore, it makes a chemical analysis of the analyzed mixture, providing in real time the

results of the olfactory imprint need specific training. Finally, the analytical method used to measure odour compounds is the gas chromatography coupled to mass spectrometry (GC-MS).

Before dealing with the regulatory aspect of a strictly legal nature, it is necessary to introduce technical standardization (produced by bodies such as ISO, CEN or UNI). This is often intended as a voluntary but non-mandatory technical reference.

The European Directives impose the use, for the application of the imposed limits, of the corresponding technical standards published by the CEN, becoming the reference.

The technical standards that currently refer to odours are.

- UNI EN 13752:2022 Stationary source emissions - Determination of odour concentration by dynamic olfactometry and odour emission rate (EN 13725:2022);
- VDI 3518-3:2019 Multigas sensors (Verein Deutscher Ingenieure.);
- IEEE P2520.1 - Standard for Baseline Performance for Odour Analysis Devices and Systems (IEEE, P2520.1);
- CEN TC264 WG41 - Electronic sensors for odour monitoring (CEN/TC264/WG41);
- UNI 11761:2019 - Emissions and air quality - Instrumental Odour Monitoring Systems (UNI 11761, 2019).

The first standard, EN 13725:2022, defines what an odour is: it is defined as what is detected by at least 50 % of the participants in an odour test. It should be noted that this definition does not take into account whether a smell is “good” or “bad” but only that it exists, and this can objectively present application problems because, instead, the law often asks to limit the “bad smells”. First, it should be defined the European Odour Unit ( $\text{ou}_E$ ) which is defined as the amount of odour(s) that, when evaporated into one cubic meter of neutral gas at standard conditions, elicits a physiological response from a panel (detection threshold) equivalent to that elicited by one European Reference Odour Mass (EROM), evaporated in  $1 \text{ m}^3$  of neutral gas at standard conditions. Absurdly, assuming that the (annoying) odour limit in a residential area could be  $5 \text{ ou}_E \text{ m}^{-3}$ , this would mean that if there is a garden near the house that releases  $10 \text{ ou}_E \text{ m}^{-3}$  of odour (from flowers and plants), it would violate the law. This type of information, therefore, does not take into account the ‘quality’ of the smell. The limit value must guarantee “an equivalent level of protection of the environment as a whole and not lead to higher pollutant loads in the environment”. The best approach to the problem is preventive, adopting structural and management solutions that limit the impact of emissions.

The second standard, VDI 3518-3:2019, and the third, IEEE, P2520.1, are related to the management of the so-called IOMS, commonly known as “electronic noses” (e-nose). The attention on these devices is increasing very much because, precisely due to the importance that odours have by now, the need to have automatic devices that can continuously monitor the state of the air is very strong, instead of having to depend on sampling and manual analyses, much more complicated, long and expensive. These standards propose that the electronic nose be managed in a very similar way to the instruments used for air quality, i.e., characterizing them for their ability to select a specific spectrum of chemical compounds. However, albeit with positive sides, this approach has the serious limitation of losing the connection with the innate human sensitivity in tests conducted by olfactometric method, which is however imposed by law. The fourth standard, CEN/TC264/WG 41, despite the title, is concentrated on the ‘training’ aspects of electronic noses and would like to bridge the limits of the previous two. In fact, once the IOMS device has been calibrated it is necessary to determine a correlation between the response provided and the ‘reference’ measurements provided by odour method; this process is, in fact, called training. The standard is currently in the drafting stage and its publication is not expected for 5 years.

The last standard, UNI 11761:2023, in the international framework tries to describe the steps necessary to qualify an IOMS system. It

prescribes that.

- the manufacturer submits the instrument to a series of laboratory checks aimed at characterizing the instrument, so that a potential customer can choose the device that best serves the purpose;
- the instrument, once installed in field, is characterized by comparison with measurements carried out by olfactometric method;
- a series of checks are imposed during the life of the instruments to guarantee that it functions regularly, i.e., that the correlation characteristics between the measurements provided and the odours detected are still valid.

The standard follows the lines of the EN 14181 standard (EN 14181:2014) as regards the management of SME systems (Systems for the Measurement of Emissions) as regards the quality management process of the measurements provided.

### 3. Legislative and methodological aspects

#### 3.1. The European situation

At European level, the EU has issued the directive on integrated pollution prevention and control (IPPC) which has been updated several times (96/61, 2008/1 and 2010/75), fully implemented in Italy with the Legislative Decree 59/2005 and with 46/2014 which defined the concept and application of the Best Available Techniques (BAT) (Legislative Decree 46/2014, 2005).

Several European countries have proceeded over time to issue acts and regulations relating to odour emissions. This has also contributed to the implementation of monitoring campaigns in order to keep under control the odour impact of refineries, landfills, food industries, livestock farms, tanning industries, purifiers, mechanical-biological treatments (MBTs), aerobic, anaerobic digestion, composting, etc.

The need to consider the problem of odour emissions with extreme attention has led several European countries to a series of legislative initiatives and recommendations. The aspects relating to odours emissions are managed through a suitable location of the plants, the definition of guidelines for plants and activities, the imposition of limits on emissions. In all European countries, in the procedure for issuing the Integrated Environmental Authorization (IEA), the competent Authority must consider the references of the guidelines, ad hoc regulations and BAT Reference Documents, to fix the emission values in  $\text{uo}_E \text{ m}^{-3}$  and for some pollutants in  $\text{mg N}^{-1} \text{ m}^{-3}$ .

Below is an examination of the measures adopted by some European countries (Diaz et al., 2019).

Germany reports in its legislation, Technische Anleitung zur Reinhaltung der Luft-TA Luft und Bundes Immissionsschutz Gesetz (BlmSchG), the reference to the guidelines Feststellung und Beurteilung von Geruchsmissionen (Geruchsim-missions-Richtlinie-GIRL (1994–2008) which indicates in points 3. Evaluation criteria and 3.1 Input values the input values expressed in relative frequencies of odour hours (defined as the odour frequency is the odour load emitted over time, i.e. the percentage (%) of hours in a year in which the odour is perceived; it is measured in ou-hours) for the different areas classified as residential, commercial/industrial and agricultural (applies only to odours caused by livestock facilities), referring to specific standards (VDI 3880, EN 13725 and 16841

1). This guideline indicates the % of hours per year for 5 areas.

Denmark has developed the Guidelines for Air Emission Regulation - Limitation of air pollution from installations which contains reference values between  $5 \text{ ou}_E \text{ m}^{-3}$  and  $10 \text{ ou}_E \text{ m}^{-3}$  not to be exceeded (Danish Environmental Protection Agency).

In the case of farms, the Technical Report on New Odour Guidelines for Livestock have been published which present criteria applicable exclusively in the context of the impact assessment for the establishment, expansion or modification of farms.

- $5 \text{ ou}_E \text{ m}^{-3}$  for urban and recreational areas;
- $7 \text{ ou}_E \text{ m}^{-3}$  for agglomerations in rural areas (six or more residential buildings);
- $15 \text{ ou}_E \text{ m}^{-3}$  for expansion or modification.

The Netherlands, already in the early 70s was working on the regulation of odour emissions generated by the livestock sector (in particular pig farms). In this context, minimum distances were identified between residential areas and farms according to the number of animals (Brattoli et al., 2011).

It subsequently published the national guidelines Nederlandse Emissie Richtlijn (NeR) using the same methodology developed in Germany (TA-Luft) (InfoMil, 2012). The NeR was of particular interest because it contained emission factors for specific activities. It reported odour concentration ranges between 0.5 and  $25 \text{ ou}_E \text{ m}^{-3}$ . The NeRs were in turn modified in 1995 by the Minister van Volkshuisvesting, Ruimtelijke Ordening en Milieubeheer (VROM) and then completely withdrawn in 2016.

France has reported the general requirements to limit the annoyance of odours in the law sur l'Air et l'Utilisation Rationnelle de l'Énergie (LAURE) of 30 December 1996 and in the decree of 2 February 1998 Code Permanent Environnement et Nuisance. These prescriptions are supplemented by the indications present in the circular of 17 December 1998. The decree relating to the "prescriptions générales applicables aux installations soumises à déclaration sous la rubrique n. 2170" of 7 January 2002 established measures and prescriptions on odours for each odour emission source (Decree 2170/2002).

In the decree 22 april 2008 "relatif aux installations de compostage, complètement par la circulaire da 6 mars 2009" (Circulaire du, ), the role of odour emission measurements was highlighted and indications were reported for.

- minimum distances from homes (from 50 to 200 m);
- air quality objectives of  $5 \text{ uo}_E \text{ m}^{-3}$  not to be exceeded more than 175 h/year (2 % of the time) within 3 km of the plant;
- for new installations, an impact study including:
- list of the main odour emissions;
- study of atmospheric dispersion in order to ensure compliance with the air quality objectives;
- initial odour situation of the site;

for existing installations.

- a list and quantification of the main odour emissions;
- a study of atmospheric dispersion in order to ensure the achievement of the air quality objective;
- limit value of  $5 \text{ mg N}^{-1} \text{ m}^{-3}$  for  $\text{H}_2\text{S}$ ;
- limit value of  $50 \text{ mg N}^{-1} \text{ m}^{-3}$  per  $\text{NH}_3$ .

Briefly resuming the European situation, the authors would like to evidence that at the European level, the EU has issued the directive on the Integrated Pollution Prevention and Control (IPPC Directive) which has been repeatedly updated by 96/61, through 2008/1 and up to the current 2010/75, fully implemented in the several Member States. Particularly, Italy ruled the Legislative Decrees 59/18 and 46/2014 which defined the concept and application of the BAT; Greece emitted the joint ministerial decisions 36060/1155/E.103/2013 and 1450/B/14-6-2013 in the Law on Environmental Protection 1650/1986; France ruled a decree on 24 September 2013 related to "prescriptions générales applicables aux installations relevant du régime de l'enregistrement au titre de la rubrique no 2910-B de la nomenclatures des installations classées pour la protection de l'environnement"; the Netherlands emitted the general provisions of the legal environment of 6 November 2008 incorporating into the environmental law through two modification acts; Spain ruled the Law 16/2002 of 1 July on the integrated prevention and reduction of pollution, subsequently modified by

the Royal Decree Law no. 1 of 16 December 2016. This strategic approach is based both on the prevention, reduction and, as far as possible, the intervention on the causes of pollution for a real decrease in the polluting load. Among these parameters odour emissions play an important role. It should not be forgotten that the limits on emissions are mainly technological and are taken into account in the BAT.

In Europe, Germany is the only country that, more than the others, has a legislative tradition on odours and quantification in objective terms (Technische Anleitung zur Reinhaltung der Luft-TA Luft and Bundes Immissionsschutz Gesetz, BImSchG) which identified, among other things, a limit value for the odour concentration in emission equal to 300 ou m<sup>-3</sup>, value taken up in other legislative acts.

### 3.2. State-of-art of the legislation in Italy

In Italy, the legislation relating to odour emissions has been lacking for many years and the complex events that the entire sector has experienced have greatly slowed down the study and development of periodic monitoring and prescriptions from an environmental point of view. In fact, knowing the emission level allows, in addition to verifying compliance with the prescriptions, the correct functioning of the system.

In recent years, in Italian legislation through successive decrees, a series of new concepts relating to odour emissions have been included in Legislative Decree 152/06 both in terms of definitions and in terms of authorizations and controls. In this way, these additions have tried to fill that gap and give concrete and clear answers with the introduction of specific articles concerning the conveyed or diffused emissions having effects of an odour nature (articles 268 and 272-bis) (Legislative Decree 152/2006). These updates constitute elements of primary importance for operating more effective interventions for measures for the prevention and limitation of odour emissions in the issuance of the

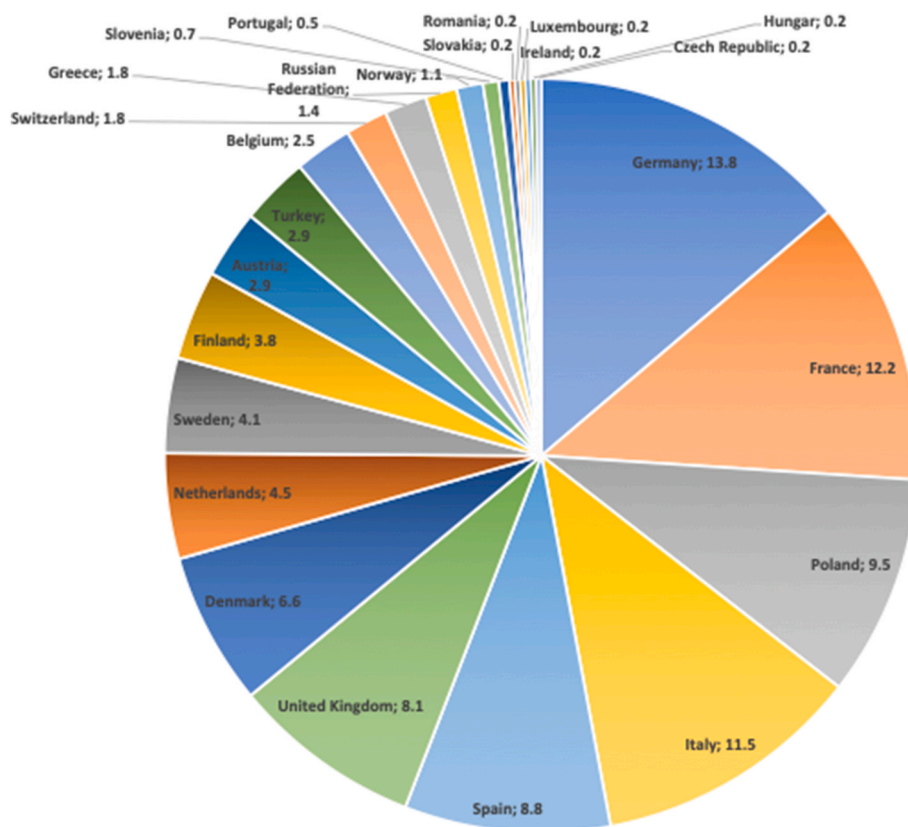
authorization for emissions into the atmosphere and can allow for better social acceptance of the plants and activities.

Many regions (Apulia, Veneto, Piedmont, Basilicata, Lombardy, Sicily and Abruzzo) had indicated a first series of technical indications with very diversified legally binding approaches and settings, even before the publication of Legislative Decree 152/06 as amended, on a limited but significant number of specific chemical parameters (e.g., detection of concentrations of NH<sub>3</sub> and H<sub>2</sub>S, etc.) with the relative methods to be used for measuring and controlling odour emissions. Among the numerous legislative indications, the ones that most influenced the sector were those of the Lombardy region and subsequently of Apulia.

With the Guidelines for existing plants for activities falling within the IPPC categories on waste management in Legislative Decree 372/99, "Guidelines containing the criteria for the identification and use of the best available techniques pursuant to art. 3 paragraph 2 of Legislative Decree 372/99, the value of 300 uoE m<sup>-3</sup> accompanied by the periodicity of the surveys appeared for the first time in national legislation. This limit has been taken up by some regions in their documents concerning the assessment of odour emissions.

### 4. State-of-art of the research in Italy

Although the Italian legislation is lacking in this topic, from a scientific point of view, Italy shows a high number of publications on the topic, precisely 11.5 % (i.e., 51 papers) of the total publications in relation to EU member states (Fig. 1): only Germany (13.8 %, 61 papers) and France (12.2 %, 54 papers) show a number of major publications. On the other hand, still on the Scopus database, it is reported that, the works published by EU member countries, # 442 (including UK, Turkey, Norway, Switzerland and the Russian Federation, which are # 66), are



**Fig. 1.** Percentage of country contributions (%) to the total amount of publications (# 442) on odour emissions in Europe from 2000 to 2022 (source: Scopus, search: 12 April 2023; keyword used "odour emission" with the limitation to the European countries). The search includes Norway, Russian Federation, Switzerland, Turkey, and the UK, which left UE on 31 January 2021.



in number greater than the works published by the United States (262 papers), China (157 papers) and Australia (66 papers): this data indicates the considerable interest of the EU in this topic which has important repercussions on the health of the population and on the ecosystem in general.

The Italian law delegates to each region the definition of the limits of odour emission through environmental authorizations and some regional regulations. On the other hand, it should be highlighted the absence of specific working groups as present in other air quality topics, first of all, for example, the National Study Group (GdS) on Indoor Air Pollution established in 2010 at the Italian Institute of Health (IIS). This study aims to summarize and present the main studies published by Italian authors on this important topic.

The authors wish to underline that this review focuses on papers that are considered most significant and innovative in terms of both the analytical methodology for the determination of odours and the results obtained. Furthermore, before approaching, the authors would also like to remember that volatile organic compounds (VOCs) emitted from industrial sources and characterized by unpleasant odour include mercaptanes, sulphides and disulphides, amines, carboxylic acids, aldehydes, ketones, aliphatic and aromatic hydrocarbons, etc. (Czerny et al., 2011; Yuwono and Lammers, 2004). The odour classification is made comparing a signal with a pattern acquired previously in a training phase, particularly important and delicate, consisting in the analysis of different gas samples of known olfactory quality diluted at different odour concentration ( $C_{od}$ ) values (Kaeppeler and Mueller, 2013).

With this in mind, the first paper reported in Scopus regards the odour emissions from landfills by Davoli et al., in 2003 (Davoli et al., 2003). The authors developed an analytical method based on solid phase microextraction (SPME) preconcentration using a three-phase fiber (divinylbenzene, DVB/carboxen/polydimethylsiloxane, PDMS, 50–30  $\mu$ m) followed by gas chromatography-mass spectrometry analysis, for analyzing about 100 VOCs. Blank tests and samples for characterizing the odour emissions in a 210,000  $m^2$  municipal solid waste (MSW) landfill-site, were collected by means of Nalophan bags, 9 L (copolymers of terephthalic ester), in accordance with the CEN TC264/WG62 guidelines. Using this approach, the authors managed to analyze a wide range of compounds below 0.1 ppb (except dimethyl disulfide, 50 ppb) from the highly polar volatile fatty acids to non-polar hydrocarbons. Finally, the results were interpreted by multivariate statistics, in particular the principal components analysis (PCA) and the cluster analysis (CA) based on Euclidian distance, for evaluating similarities and dissimilarities between emissions and ambient air samples: for instance, the authors established that limonene was a tracer of fresh wastes, whereas *p*-cymene was characteristic of leachate and biogas. Even if the authors themselves reported that, at that time, the odour emission characterization was an open topic for different reasons (e.g., the complexity of the relationship among the different compounds, the lack of knowledge of some occurrence in atmosphere, the instrumental limitation), the results reached in this paper were interesting: the good analytical procedure (below 1 ppb) allowed the efficiency assessment of the scrubber installed in that landfill for removing malodorous compounds (below 24 %, not very high according to the authors' considerations).

In 2004 Canovai et al. (2004) published an intriguing paper describing the investigation of odour emission from two municipal solid waste (MSW) biological treatment plants located in Albano, close Rome, and in Milan (Canovai et al., 2004). In particular, the authors focused their attention on the analysis of operational parameters as temperature, pH, humidity, loss of pressure of the biofilter affecting the biofiltration efficiency, for both chemical parameters and odours compound concentration, measured by means of odour panel evaluation technique, and the efficiency of the biofiltration system for several compounds present in air emissions, analyzing organic substances by means of gas chromatography/mass spectrometry. Further, the authors tested an innovative bioscrubber pretreatment of the air flow coming from the

aerobic reactor for reducing the odour emissions from the biofilter. Actually, they started from the consideration that the odour emissions are a crucial point in the MSW biological treatment plant and an uncorrected management of these emissions could be a serious risk of conflict with the resident population. The use of biofiltering processes based on a patented material composed of bivalve mollusk shells, allowed to the authors to reduce the odour emissions in the range 50–98 %.

Pinzari et al. (2004) developed an electronic nose for detecting fungi actively growing on paper samples (Pinzari et al., 2004). Actually, even if their main scope was to build a device for verifying the mould presence in libraries, archives, museums, they investigated if different types of papers could influence the odour emission signals by mould. For this aim, they performed the measurements by an e-NOSE 4000 (Neotronics) evaluating the resistance change of a group of conducting polymer (basically, polypyrrole) sensors when VOCs are produced. The results reported in the paper are satisfactory: paper samples affected by moulds and those unaffected can be identified by means of measuring their odour fingerprint. For confirming the results, the authors applied the cluster analysis (CA) and the principal component analysis (PCA) for evidencing the different groups formed during the stress-tests.

The paper publication on odour emission were growing up in that period as well as the understanding that this was a really important issue. In fact, also Zarra et al. (2009a) dealing problems related to the sanitary environmental engineering plants, considered it as a main issue in relationship to the protests by the exposed population (Zarra et al., 2009a). They were the first to reveal that no regulations were present in Italy. They also stated that the traditional analytical approach for investigating the odour mixtures (e.g., use of chemical markers or chromatographic techniques) was too expensive and time consuming whereas it was necessary a continuous assessment in terms of air quality monitoring. They, focusing their attention on the olfactory annoyance and not on the olfactory perception, developed an original tool based on the portable GC-MS Hapsite equipment (Inficon, NY, USA), an innovative EPA-certified analytical instrument, for identifying and characterizing volatile compounds causing odour annoyance. The GC-MS Hapsite was used directly on-site. From a qualitative point of view, the authors managed to analyze almost 38 compounds belonging to different classes of compounds such as ketones (acetone, 2-butanone, 2-eptanone 3-methyl-2-pentanone acetophenone 4-methyl-3-penten-2-one), hydrocarbons (undecane, methylchloride, dodecane, *p*-cymene, decane, 1-ethyl-2-methylbenzene, 1,3,5-trimethylbenzene, *o*-xylene, *p*-xylene, 1,2,4-trimethylbenzene, cyclohexane, styrene, ethylbenzene, toluene), terpenes (limonene, 3-carene,  $\beta$ -pinene,  $\alpha$ -pinene), alcohols (ethanol, 2-ethyl-1-ethanol), volatile fat acids (ethyl acetate, isopropyl acetate, butyl acetate, isobutyl acetate, formic acid), others (octamethyl-cyclo-tetrasiloxane, 2-methyl-1,3-butadiene, 2-propenoic acid, 2-methyl-methyl-ester, chloroform, tetrahydrofuran). On the other hand, the authors achieved semi-quantitative analyses using a direct ratio with internal standards commercialized by Inficon. For composting plant, the authors stated that 2-butanone and limonene could be considered key compounds linked to specific production processes. Few years later, the same research group with the addition of Giuliani published another paper regarding the use of a multisensory array system for detecting the Odour Emission Capacity (OEC) in a wastewater treatment plant (WWTP) (Giuliani et al., 2013). The device is made of 2 specific gas sensors ( $NH_3$  and  $H_2S$ ), 12 metal oxides non-specific gas sensors (selected on the basis of the odour substances to be investigated) and 2 internal conditions control sensors (humidity and temperature), placed in a fluid dynamics chamber (Zarra et al., 2009b; Viccione et al., 2012; Zarra et al., 2021). The authors compared the results obtained by means of this multisensory array with those measured by dynamic olfactometry: they evaluated a very low difference, lower than 5 % whereas, simultaneously, stated that such device significantly reduced the costs and the analysis time. This same device was used and the results discussed in a paper on the odour measures in a WWTP: a research group of

Italian and Japanese scientists reported the development of the innovative prototype of e-nose patented by University of Salerno (Naddeo et al., 2016).

Finally, in 2016 Zarra et al. (2016) published a paper regarding the characterization of odour emissions from two large liquid waste treatment plants (LWTPs) (Zarra et al., 2016). They proposed an Odour Concentration Index (OCI) as a tool for identifying the main activities for regulating the main odour sources. For this scope the authors used a dynamic olfactometry according to the EN 13725:2003 (Van Harreveld, 2014; Polvara et al., 2023) and determined the odour concentration as  $\text{OU m}^{-3}$ . The air samples were collected by means Nalophan bags and analyzed within 14 h from the sampling.

Between 2010 and 2022, different papers from Capelli's group were published, basically starting from the development of an electronic nose (e-nose) for characterizing the environmental odours (Dentoni et al., 2012a), even in consideration of the different wind speeds, simulating different conditions occurring on field (Dentoni et al., 2012b). In 2012, the research group investigated the odour emission from a solid surface: they simulated the diffusion of an odour at known concentration dissolved into a liquid phase located below the layer of solid material (Capelli et al., 2012a). On the other hand, the same group published a study on a correct model for assessing the odour emissions from liquid passive area sources, namely from WWTPs (Lucernoni et al., 2017). For this aim the authors used a wind tunnel (body  $25 \times 50$  cm base section, 8 cm high) connected to the inlet and outlet of a chamber where the compounds were volatilized. After, the compounds were analyzed by means of a gas chromatography-thermal conductivity detector (GC-TCD). Simultaneously, comparison tests were carried out sampling gaseous samples with Nalophan bags and analyzing by Dynamic Olfactometry (EN 13725:2022). Even if the system gave interesting results with VOCs of easy detection such as acetone and butanone, the authors selves stated that it would have been better to evaluate this methodology with specific experiments involving generic odour emission monitoring. In a paper came out in 2021, Capelli's group dealt the issue related to the drift, i.e., the progressive deviation over time of sensor responses (Bax et al., 2021), a crucial point in the odour monitoring. They reviewed previous campaigns, identifying and removing the drift connected to sensor aging or external factors, and performed other campaigns for testing the correction efficiency. The e-nose used, namely EOS507F, commercialized by Sacmi s. c. and developed in collaboration with the Olfactometric Laboratory of the Politecnico di Milano, was equipped with 6 Metal Oxide Semiconductor (MOS) gas sensors, characterized by high sensitivity, and by systems for humidity regulation and reference air and by an automatic calibration system. The OPLS algorithm proposed was able to counterweigh the drift effects both on calibrant dataset and on real environmental samples whereas the authors suggested that the system periodical recalibration were necessary in any case.

A very interesting Horizon project titled "Distributed Network for Odour Sensing Empowerment and Sustainability (D-NOSES)" was devoted to this issue: two papers regarded the Italian contribution to it (Polvara et al., 2021; Lotesoriere et al., 2021). In the first paper, Capelli et al., (2016a) used a sensorial analysis, the dynamic olfactometry, for evaluating the human exposure to odour samples at increasing concentrations. Looking at the results, the authors stated the exposure risk for olfactometric assessors in relation to odour landfill emissions. In the second paper, the authors performed intensive experimental campaigns where monitoring of odours was carried out along with an impact odour assessment by dispersion modelling (Cusano et al., 2010). The authors evidenced how about 60 % of the odour observations well-matched with the municipal WWTP location but also that the malodorous compound release (i.e., aldehydes) was not due to WWTP but from other sources present in the investigated area.

Finally, it should be underlined two reviews published by this group. One, highly cited (162 times), regards a discussion on the techniques to be assumed for measuring odours in field (Capelli et al., 2013). The

authors stated that chemical measurements are advantageous (easier and more reliable than the dynamic olfactometry) but they cannot be directly correlated to the odour perception in the environment (Dincer et al., 2006; Dincer et al., 2007). Further, the authors also underlined that the chemical analysis, despite being a reliable and consolidated procedure, manages to give accurate responses only when the emission is characterized by a single compound (Capelli et al., 2012), thus the odour is directly proportional to its concentration. In the second paper, more recent, the authors deal the issue related to the effect of the micrometeorology for estimating the odour emissions (Lotesoriere et al., 2022).

As just reported above, nasty odour emission is often cause of citizens' protests about local government. So, it becomes necessary to give as precise and accurate results as possible. Cangialosi et al. (2018) dealt such issue in a paper where reported measures of odour emissions from a sanitary landfill in different operative conditions and compared the results with  $\text{H}_2\text{S}$  measures *in situ* and dynamic olfactometry used as benchmark (Cangialosi et al., 2018). The authors tested a commercial e-nose equipped with 32 sensor (MSEM32® by Sensigent, Cal.) and also used automatic air samplers (OdorPrep® by Labservice Analytica). The results partially satisfied the authors: first odour and  $\text{H}_2\text{S}$  measures did not show any correlation and different interferences were recorded (e.g., operation regime, weather conditions, presence of other malodorous activities in the area).

Autelitano and Giuliani (2018) reported the use of the AOS Cyranose® 320 (Cyranose Science) for determining the asphalt emissions at high temperature (90–200 °C) (Autelitano and Giuliani, 2018a). The C320 is made of 32 individual thin-film carbon black polymer composite chemiresistors: when the composite film is exposed to an analyte, the polymer matrix swells disrupting the conductive carbon black pathways and consequently increasing the sensors' electrical resistance. The results reported by the authors were particularly significant: mainly, a difference in the odour emissions can be evaluated when asphalt is heated at different temperatures. The authors underline that the main issue is still the development of standardized procedures and methodologies for understanding the different odour generated during the different asphalt production steps: this effort could give an important help for "building" a trained multisensory array useful in different industrial fields. The same authors published another interesting paper regarding the influence of chemical additives and wax modifiers on the odour emissions: also, in this paper they used the AOS Cyranose® 320 (Autelitano and Giuliani, 2018b). First, the device allowed to distinguish the different contribution: the authors achieved a different fingerprint for each additive. On the other hand, once the additive was added to the asphalt, no relevant effect was recorded, meaning that the bituminous binder odour was hidden or masked. The authors achieved the same behavior even when they tested odour suppressant agents.

A particular application was developed by Paris et al. (2020): they investigated the VOCs emitted by an orange with and without peel (Paris et al., 2020; Paris et al., 2022). The significant issue regards the use of author-produced activated carbon fiber (ACF) adsorbent tubes for sampling such compounds and subsequent GC-MS analysis. The results show that the orange aromatic profile is made of 42 VOCs whereas the profile of the peeled orange is rich of 101 VOCs.

A particular mention regards papers published by De Gennaro's research group. In more than two decades of activity this group issued more than twenty papers on this topic, more of them oriented to the determination of odour emissions from industrial plants. The first paper to be mentioned regards the measurements of odour compounds from a waste treatment plants (Bruno et al., 2007). The authors described an analytical protocol based on the sampling with thermal desorbable radial diffusive samplers Radiello® containing Tenax cartridges followed by GC-MS analysis. By this approach they managed to investigate some compounds representative of different species responsible of odour impact, namely 2-butanone,  $\alpha$ -pinene, tetrachloroethylene, dimethyldisulfide,  $\beta$ -pinene, limonene, phenol and benzoic acid, achieving

good limits of detection (LODs) ranging between 0.04 and 0.86  $\mu\text{g m}^{-3}$ , relative standard deviations (RSDs) below 7 % and recoveries above 97 % for all compounds. They applied this approach to a sampling campaign for monitoring the emission from a waste treatment plant near Taranto (South Italy): the results showing the odour emissions in the plant (odour concentration between 88 and 47,429  $\mu\text{g m}^{-3}$ ) and at 500 m from the disposal site (odour concentration between 2 and 210  $\mu\text{g m}^{-3}$ ) evidenced no landfill influence of the odour levels.

Even if they are dated, it should be mentioned three interesting reviews on the odour determination by olfactometry (Brattoli et al., 2011) or by chemical characterization (Brattoli et al., 2016; Giungato et al., 2018). In the first manuscript the authors described the advantages and disadvantages of using instrumental sensory methods and chemical sensors. They state that at that time dynamic olfactometry was the standardized method for odour concentration measurement but also it was affected by some limitations. E-noses have lower analysis costs, give rapid results and allow in-field continuous monitoring in the vicinity of sources and receptors. On the other hand, dynamic olfactometry gives punctual odour concentration values, it does not manage to evaluate an olfactory disturbance case due to the fact that it does not allow continuous and field measurements. On the contrary, in the other review the authors reviewed the gas chromatography-olfactometry (GC-O) technique for odour determination. In this case, the approach is different: the gas chromatography procedure allows to analyze the complex mixture and to identify the volatile components responsible for odour nuisance. The review deals with the different olfactometric detection method highlighting that they can be divided in three main categories: frequency detection methods (Pollien et al., 1997; Le Fur et al., 2003), dilution to threshold methods (Hallier et al., 2004; Delahunty et al., 2006), and direct intensity (Fan and Qian, 2006; Gürbüz et al., 2006) methods. Further, the authors reviewed the state-of-art of the applications in different field such as food, fragrance, medical, environmental and material issues. The third review gives an overview on analytical and sensorial techniques: the authors used the term “synergistic approach” for the identification of odours for food quality scopes, for managing malodour emissions in industrial plants and other similar situation where odours are important. In this synergistic approach the integration between analytical and sensorial methodologies are fundamental for obtaining more information as possible from the sampling.

In 2014, the group published a paper regarding the determination and assessment of olfactory annoyance (Brattoli et al., 2014). For this aim, the authors showed a methodology based on an integrating automatic remote system for detecting the olfactory perception of human receptors. The measurements were carried out in Taranto area, a location suffering of really severe environmental pollution due to a large industrial zone (i.e., in Taranto there are: an important harbor, a steel production plant which is the biggest metallurgic center in Europe, a petroleum refining, a cement plant, a military arsenal). The system, called Odortel® and developed with Labservise srl, consisted of the direct preoccupation of a cluster of population which communicated the odour perception using the telephone keypad and different colors for identifying the intensity levels, namely 1. faint odour (green color), 2. persistent odour (yellow color), 3. very strong odour (red color). Each person transferred his perception by digiting the number according to odour perceived. For testing the Odortel® application the authors used the air quality data from the monitoring stations, particularly  $\text{H}_2\text{S}$  and sulfur compounds data. The comparison between the alarm of an event occurred during the field campaign and the odour perception captured by population, confirmed the potentiality of this system. On the other hand, low-cost solid-state gas microsensors were used for landfill odour control and air-pollution monitoring (Penza et al., 2015). In particular, the authors monitored methane ( $\text{CH}_4$ ) and Non-Methane Hydro-Carbons (NMHC) by means of metal oxide (MOX) commercial gas sensors (4 MOX commercial devices: TGS 822, SP-AQ2, TGS 2600, TGS 2106 furnished by Figaro Engineering Inc., Japan). Such measurements were

compared with those obtained by a portable gas sensor system, called NASUS, developed and assembled at ENEA laboratories. The authors stated that, even if the results were satisfactory and that the gas sensor arrays could be useful for measurements on-site, long-term studies in real scenarios were still necessary for a good evaluation of MOXs as technology for odour control and air quality monitoring. These further investigations were due essentially to the stability, cross-sensitivity and repeatability in the response whereas the low power supply autonomy could be dealt more easily. Further, the group published two papers on intensive study campaigns close to the same waste management plant investigated in previous studies and to one of the biggest crude oil and gas onshore European reservoirs, using different approaches (Giungato et al., 2016; Licen et al., 2020). Particularly, in the first paper they used electronic noses (ten metal oxide semiconductors, MOSs, and 32 polymer/black carbon, nano composite array, sensors), gas chromatography mass spectrometry/olfactometry (GC-O/MS; 45 compounds identified) and dynamic olfactometry whereas in the second paper they carried out the measurements by means of a two stages odour control map approach, applied to patterns of sensor signals of a commercial electronic nose implementing ten MOS sensors. In this latter paper the equipment was completed by a PID for measuring TVOCs expressed as isobutylene, and a meteorological station.

A particular interesting application was reported by the same group and regarded the perfume industry: namely, they showed the chemical characterization of essential oils and perfumes (Amenduni et al., 2016). The authors analyzed the VOC composition of commercial perfume and a natural-derived one, both branded by a floral scent. They mainly detected ocimene,  $\alpha$ -ionone and  $\alpha$ -isomethylionone in commercial perfume whereas  $\beta$ -hydroxyethylbenzene in natural one. At the same time, the authors arose an important alarm on the necessity of a stricter control (i.e., quality control) on the sale of perfumes: in fact, among the different VOCs they also identified the presence of toluene and benzyl alcohol which are harmful for the human health.

Finally, it deserves to be cited a paper by Verlicchi et al. (2019) which deals the odour emission issue in another way. The paper displays the main findings of a study carried out in Ecuador, particularly in the zootechnical sector which represents the most important economic activity in that country. The authors estimate the financial impact of such activity on the environment and simultaneously propose a treatment for the zootechnical farm wastewater. In the social and economic budget of the environmental problem the authors also considered the odour emission as an important factor to be solved (Verlicchi et al., 2019).

## 5. Conclusions

The problem of odorous emissions is becoming increasingly important, as are other forms of pollution: there is a need to develop adequate regulatory and legislative tools to support policies and control measures in order to guarantee adequate environmental quality.

The development of organic legislation on the subject required the acquisition of knowledge regarding prevention measures, operational methods, mitigation and abatement techniques, measurement systems and techniques to quantify the problem and the achievable objectives, which allow us to avoid or limit the occurrence of the phenomenon, as well as the factors that determine the spread of odours. Until a few years ago, the reference framework on the technical provisions for limiting odour emissions (criteria, parameters and methods of evaluating odours for the issuing of authorisations) were local interventions, despite the general agreement on the need for a framework law on odours with the identification of unique limit values at national and European level. In recent years there has been progress in the national legislation of various European countries to reduce the impact of odour pollution on the territories. These legislations present reference values for undertaking mitigation actions. Among other things, these legislations aim to develop methodologies for the evaluation of emission sources as well as to improve management practices of abatement processes and the



application of BAT. In this contest, Italy has played an important role both from the legislative point of view (because its regulations have been the foundation for the European standard norms) and from the scientific point of view. The Italian case studies have contributed to define the European standard method for olfactometry measurements.

According to the European standard guidelines regarding the analytical procedures three techniques for odour nuisance characterization and quantification were set up: analytical: chemical analyses; sensorial: dynamic olfactometry; senso-instrumental: electronic nose. The analytical approach based on chemical analyses by gas chromatography coupled with mass spectrometry (GC–MS) allows to determine qualitatively the occurrence of odour substances of known odour threshold level in a complex mixture, but without correlation with the overall effective odour due to masking and/or synergic effects in the mixture (Stuetz et al., 1999; Toropov et al., 2016). Similarly, in-situ H<sub>2</sub>S measurement by a gold-film analyser, considered a valuable odour assessment tool, seldom provides relationship among other odour chemicals present and their cumulative overall odour threshold (Bull and Fromant, 2014a; Fasolino et al., 2016). The sensorial approach (dynamic olfactometry, EN 13725, 2022), based on the statistical evaluation of the sensorial reaction of a panel of qualified examiners to the progressive dilution of an odour mixture, is probably the most affordable techniques, yielding the odour concentration (Cod in ou<sub>E</sub> m<sup>-3</sup> units) response to be compared with the limits in force; however, it cannot provide in-situ/real-time or online measurements. The senso-instrumental approach (electronic nose) (UNI 11761) is the only technique that allows continuous monitoring of odours *in situ* (Giuliani et al., 2012; Galang et al., 2022). The e-nose instrument depends on a suitable array of non-specific gas chemical sensors combined with a chemometric processing tool. On the other hand, at moment the Artificial Intelligence (AI) does not seem to be useful for evaluating the odour impact as the chemometric approach. In fact, the main issue related to the AI is “to educate” the system to understand and differentiate the odour sources instead the chemometric approach is able to do this. So, at the moment this approach is not so interesting but, collecting a lot of data and having several case-studies, it could be possible in a close future to make this option available and useful also in this field. Maybe, the main future benefit of using AI could regard the information to the population in case of accidents.

In recent years there has been progress in the national legislation of various European countries to reduce the impact of odour pollution on the territories. These legislations present reference values for undertaking mitigation actions. Among other things, these legislations aim to develop methodologies for the evaluation of emission sources as well as to improve management practices of abatement processes and the application of BAT. In this contest, Italy has played an important role both from the legislative point of view (because its regulations have been the foundation for the European standard norms) and from the scientific point of view (the Italian case studies have contributed to define the European standard method for olfactometry measurements).

#### CRediT authorship contribution statement

**Gaetano Settimo:** Writing – review & editing, Visualization, Validation, Software, Resources. **Pasquale Avino:** Writing – original draft, Supervision, Methodology, Data curation, Conceptualization.

#### Declaration of competing interest

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

#### Data availability

No data was used for the research described in the article.

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