

JRC TECHNICAL REPORT

Air pollutants of emerging concern

*Potential air pollutants and
how they could be monitored
under future air quality
directives*

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Contents

Abstract.....	1
Executive summary.....	3
1 Introduction.....	5
2 Black Carbon.....	7
2.1 Properties & sources.....	7
2.2 Reasons for concern.....	7
2.3 Measuring Black Carbon.....	7
2.4 Measurements availability, coverage and use.....	8
2.5 Towards more systematic monitoring.....	9
3 Ammonia.....	11
3.1 Properties & sources.....	11
3.2 Reasons for concern.....	11
3.3 Measuring Ammonia.....	11
3.4 Measurements availability, coverage and use.....	12
3.5 Towards more systematic monitoring.....	12
4 Ultra-fine particles.....	13
4.1 Properties & sources.....	13
4.2 Reasons for concern.....	13
4.3 Measuring Ultra Fine Particles.....	13
4.4 Measurements availability, coverage and use.....	13
4.5 Towards more systematic monitoring.....	14
5 Methane.....	15
5.1 Properties & sources.....	15
5.2 Reasons for concern.....	15
5.3 Measuring Methane.....	17
5.4 Measurements availability, coverage and use.....	17
5.5 Towards more systematic monitoring.....	19
6 Levoglucosan.....	20
6.1 Levoglucosan properties and sources.....	20
6.2 Reasons for concern.....	21
6.3 Measuring Levoglucosan.....	21
6.4 Measurements availability coverage and use.....	21
6.5 Towards more systematic monitoring.....	23
7 Conclusions.....	25
References.....	27
Measure units.....	32
List of figures.....	35

Abstract

The report presents information about four pollutants that are not currently regulated by the European Air Quality legislation and an atmospheric marker for biomass burning. They were selected for the prominent role they could play in the revision of the current Ambient Air Quality Directives. For each compound, their physico-chemical properties and major sources are described. Current knowledge about the impacts of these pollutants on health and the environment is then summarised, together with a description of existing measurement methodologies, and of current availability of supranational monitoring data and their main current uses. Finally, possible further exploitation of data and the advantages of more systematic monitoring for the zero pollution targets are discussed together with an indication of the resources needed for suitable monitoring devices and networks. We conclude that systematic monitoring of (equivalent) black carbon and ammonia would be beneficial for both human health and ecosystems because both species play prominent roles in air pollution and can currently be monitored using robust and cost-effective measurement techniques.

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Executive summary

The report presents relevant information about four air pollutants not currently regulated by EU European air quality legislation and an atmospheric marker for biomass burning. The additional air pollutants addressed are ultrafine particles (UFP), black carbon (BC), ammonia (NH₃) and methane (CH₄), and the marker for biomass burning: levoglucosan. The selection of these additional air pollutants is based on their health and environmental impacts together with the maturity and robustness of current measurement techniques. The chemical and physical properties of these air pollutants are presented, together with their most important sources. Current knowledge about the impacts of these pollutants on health and the environment is then summarised, together with a description of existing measurement methodologies and of current availability of supranational monitoring data and their main current uses. Finally, the benefits of more systematic monitoring for the zero pollution targets related to air pollution, and a possible further exploitation of the resulting monitoring data are discussed, together with an indication of the resources needed for suitable monitoring devices and networks.

Policy context

In 2019 the European Commission launched the European Green Deal (European Commission, 2019), followed in May 2021 by the Zero Pollution Action Plan (European Commission, 2021b). The Action Plan includes pollution reduction targets for 2030, including two related to air quality: to both reduce the number of premature deaths caused by air pollution by more than 55% and to reduce the share of ecosystem areas in the EU where biodiversity is at risk due to air pollution by 25%. In this context, the Commission is designing a monitoring and outlook framework for the zero pollution ambition, which could in the future take up additional monitoring data on pollutants of emerging concern (European Commission, 2021a). At the same time, the Commission is looking into expanding the monitoring requirements of the Ambient Air Quality Directives to include additional harmful air pollutants and relevant health information currently not covered (based on recommendations by the WHO, e.g. for black carbon or ultrafine particles) as part of the ongoing review of the Directives. This report aims to contribute to this process.

Main findings

The report compiles relevant information about a selection of air pollutants of emerging concern in Europe including reasons to measure their concentration in ambient air, such as their impact on health, their role in the formation of secondary pollution and their impacts on other environmental endpoints (ecosystems, soils, climate, water, etc.). The most important sources of those air pollutants are described, as well as the current state of measurement techniques and available data. Key information, including the main reasons of concern and the state of monitoring in Europe, is summarised in a synthetic table included in the conclusions.

Two of the air pollutants selected (NH₃ and CH₄) should be addressed to further reduce already regulated air pollutants (mostly particulate matter -PM and ozone -O₃) due to their roles as secondary pollution precursors. NH₃ is also known to influence soil and water eutrophication and acidification. The other two air pollutants (UFP and BC) may be related to specific impacts on human health. Looking at the current status of monitoring of these pollutants, we found that only a relatively small number of monitoring stations are reporting data to easily accessible international data repositories, complicating the use of existing data to underpin studies on the health effects of these pollutants, as well as efficient policy design.

Enhanced measurements of all four air pollutants and the marker would provide valuable information, but considering robustness and resources needs of the available measurement techniques, JRC proposes to consider monitoring two additional air pollutants under future EU legislation: black carbon and ammonia. The assessment of black carbon is highly relevant for emission inventories, exposure assessments and source apportionment of particles. The available multi-wavelength light absorption measurement technique has been used in research monitoring since many years by now and can be considered operational and robust. A better understanding of ammonia in management of air pollution from secondary particulate matter and its environmental impact needs to be supported by additional assessments. Diffusive sampling of ammonia and subsequent analysis presents a cost-effective way of assessing this air pollutant.

Related and future JRC work

This report has been prepared in the framework of the JRC institutional project “Pressures on Air”, a foresight activity, that aims to identify and provide advance warning of upcoming impacts on European air quality (Monforti-Ferrario et al., 2019) from emerging pollutants. A significant part of the information in the report,

directly originates from JRC work and its collaboration in international and research networks. The JRC is running the European Commission's Atmospheric Observatory, which monitors aerosol physical and chemical properties and gaseous air pollutants concentrations. The Observatory is part of UNECE CLRTAP, EMEP, WMO-GAW and ACTRIS networks and of the European ICOS infrastructure. Development, validation and quality assurance of air pollutant monitoring has its home in the JRC's European Reference Laboratory for Air Pollution (ERLAP).

More information can be found on

[Atmospheric Pollution Monitoring @ JRC](#)

[European Reference Laboratory for Air Pollution @ JRC](#)

[Greenhouse gases @ JRC](#)

1 Introduction

Air pollutants are generally defined as substances in gaseous, liquid, solid or mixed phases that can harm human health and/or ecosystems. They are either directly emitted into the atmosphere (primary pollutants) or result from reactions of different substances in the atmosphere (secondary pollutants).

In the European Union, a number of relevant air pollutants are currently regulated by a set of Directives and other legislative acts aimed at avoiding, preventing or reducing harmful effects on human health and the environment by keeping the pollution levels across the EU territory within the limits of air quality standards, currently set in the Ambient Air Quality Directives (AAQD) 2008/50/EC and 2004/107/EC. These standards set limit values for concentrations of pollutants in ambient air: particulate matter (PM), both PM₁₀, (i.e., particles smaller than 10 µm in aerodynamic diameter) and PM_{2.5} (particles smaller than 2.5 µm in aerodynamic diameter), sulphur dioxide (SO₂), nitrogen dioxide (NO₂), carbon monoxide (CO), benzene (C₆H₆), ozone (O₃), Polycyclic Aromatic Hydrocarbons (PAH, traced by BaP – benzo(a)pyrene) and a number of metals (Pb, As, Cd and Ni).

Moreover, some of the pollutants regulated by the AAQDs, together with some additional pollutants not considered in the AAQDs are subject to restrictions and limitations as far as their emissions in the atmosphere are concerned. Directive 2016/2284 on the reduction of national emissions of certain atmospheric pollutants (formerly “NEC Directive”) sets nation-wide reduction targets for EU Member States for SO₂, nitrogen oxides (NO_x), Volatile Organic Compounds (VOCs), ammonia (NH₃) and PM_{2.5}.

Finally, a set of sector-specific regulations imposes additional emission limits from transport and Non-Road Mobile Machinery (PM, NO_x, CO and hydrocarbons), maritime transport (sulphur content of fuels), industrial emissions and medium-size combustion plants (SO₂, NO_x, CO, PM) and other sectorial regulations such as paint products.

This report focuses on some pollutants currently not monitored under the AAQDs. The pollutants considered are Black Carbon (BC), i.e., the fraction of the atmospheric carbonaceous Particulate Matter absorbing visible light, ammonia (NH₃), Ultra Fine Particles (UFP), i.e., the fraction of PM with diameter below 0.1 µm in aerodynamic diameter and methane (CH₄). Besides these pollutants the report also considers levoglucosan, a compound that according to current understanding does not pose direct threats to humans or ecosystems *per se* but is instead a tracer of biomass burning, both spontaneous or related to agriculture waste burning and wood burning in electricity and heat production, a practice of increasing importance with well-known impacts on air quality and health. (Monforti-Ferrario, F. and Belis, C., 2018)

Although designated here as “of emerging concern”, all these compounds have been already subject of discussions both in the scientific and policy debate. The necessity and possibility of extending regulations to include or strengthen their control is being examined, with a view to limiting their possible specific health impacts or their role as precursors of secondary pollutants.

The World Health Organization has recently assessed the impact of both UFP and BC on human health (WHO, 2021). However, because of a continuing lack of quantitative evidence of the effect of these two pollutants on health impacts, WHO has formulated good practice statements rather than specific air quality target levels, inviting for more systematic measurements. Ammonia national emissions ceilings already exist (Directive 2016/2284), while methane has been recently the subject of a specific EU Methane Strategy (European Commission, 2020a) that proposes legislative and non-legislative actions in the energy, agriculture and waste sectors, both in the EU and globally. Finally, the use of biofuels and bioenergy in transport, in electricity and heat generation, and its overall sustainability including health impacts mediated by air pollution, has been highlighted as a trade-off between air quality and climate mitigation policies (see e.g., Camia et al., 2021), recent impact assessments conducted by the European Commission for the revisions of the Renewable Energy Directive¹ and of the Energy Taxation Directive², as well as the European Court of Auditors³).

The European Green Deal, and the Zero Pollution Action Plan (ZPAP) announced legislative action on air pollution and confirmed the Commission’s commitment to reduce its health impacts. The ZPAP sets pollution reduction targets for 2030, including two related to air quality: to reduce the number of premature deaths caused by air pollution by more than 55% and to reduce the share of ecosystem areas in the EU where biodiversity is at risk due to air pollution by 25% (European Commission, 2021b). An integrated perspective across policies will be

¹ SWD(2021) 621 final

² SWD(2021) 641 final

³ See paragraph 59 of Court of Auditors (2018) Special Report No. 23 on ‘Air pollution: Our health still insufficiently Protected’, https://www.eca.europa.eu/Lists/ECADocuments/SR18_23/SR_AIR_QUALITY_EN.pdf

needed in order to maximise efficiency in pursuing the other zero pollution targets that also relate to water and soil. In this framework, a monitoring and outlook framework for the zero pollution ambition is being designed, considering also the possibility of including data on “emerging pollutants” relevant not only for air quality but also for soil and water. (European Commission, 2021a)

At the same time, the Commission is looking into expanding the monitoring requirements of the Ambient Air Quality Directives to include additional harmful air pollutants and relevant health information currently not covered (based on recommendations by the WHO, e.g. for black carbon or ultrafine particles) as a part of strengthening air quality monitoring, modelling and plans. (European Commission, 2020b). This report aims to contribute to this process, assembling key pieces of information about a selection of pollutants and markers.

Within this scope, this report presents a consistent set of information on the air pollutants and the marker listed above: each of them is presented, highlighting chemical and physical properties, sources, and reasons for the emerging concern, be it their direct impact on human health and ecosystems or their role in the formation of regulated secondary pollutants. Measurement methodologies are also described together with current availability of supranational monitoring data and the main current uses of the information provided by measurements, both from international networks (if existing) or specific measurement campaigns. Finally, possible further exploitation of data and advantages provided by more systematic monitoring for the zero pollution targets are discussed together with an estimate of the resources needed for suitable monitoring devices and networks.

The information provided could be used to frame further discussions and contribute to future prioritisation in the evolution of air quality monitoring networks. In our opinion enhanced measurements of all four air pollutants and the marker would provide valuable information for studies on the health effects of these pollutants, as well as efficient policy design. However, considering the available measurement techniques, their robustness and the resources needed for monitoring and data evaluation, we propose to concentrate on two additional air pollutants in the near future, namely black carbon and ammonia.

2 Black Carbon

2.1 Properties & sources

By definition, Black Carbon (BC) is the fraction of the atmospheric carbonaceous Particulate Matter (PM) that absorbs visible light. Black Carbon actually also absorbs near infrared (at least in the 750-1000 nm range) and near ultraviolet (at least in the 350-380 nm range) and mainly consists of graphitic Elemental Carbon (EC). In the atmosphere, BC mostly occurs in particles smaller than 300 nm of diameter⁴.

BC results from the incomplete combustion of carbon-containing fuels, from methane (with relatively low emission factors) to coal and biomass (with higher emission factor). According to the JRC's Emissions Database for Global Atmospheric Research (EDGAR) (Crippa, M. et al., 2021), EU countries emitted in 2015 178.6 kt of BC, 34.7% originating from residential sector, 32.7% from transport (including road resuspension) and 21.4% from construction and all types of industries excluding the power and heat production sector, responsible for just 1.9% of emissions. It is worth mentioning that Diesel engines emit much more BC than petrol- or gas- fired engines, but most of this Black Carbon is captured by Diesel-exhaust particle filters when present. Biomass burning (wild fires, domestic fireplaces, etc...) combustion also leads to the emission of Black Carbon (6% of EU emissions in 2015).

2.2 Reasons for concern

Despite the notable decrease in emissions observed in the latest decades (-37% in 2015 in comparison to 1990 in the EU) in line with the general reduction in primary PM emissions, BC has attracted specific concerns because epidemiological studies have shown a stronger association of health adverse effects with BC concentrations than with any other variable related to particulate air pollution (WHO, 2013). The US Environmental Protection Agency (EPA) also investigated BC health effects (US EPA, 2019) finding evidence of possible associations between BC and different impacts, with the strength of the association varying among impacts, but also stating that EC or BC is not better associated with respiratory, cardiovascular effects and mortality than PM mass concentration.

However, there is currently no consensus within the scientific community regarding the cause of this association. Some studies indicated that short-term exposure to pure Elemental Carbon does not lead to measurable clinical effects for concentrations up to 70 µg/m³ (Mills et al., 2011). It has been suggested that the association of Black Carbon with health effects could actually originate from the presence of highly toxic species adsorbed on Black Carbon cores (Casseo et al., 2013), or from the fact that Black Carbon is a tracer for combustion sources which also emit toxic substances. The hazards and risks of inhalation of poorly soluble particles of low toxicity is still debated (Borm and Driscoll, 2019)

However, WHO emphasised again in its most recent edition of Air Quality Guidelines (WHO, 2021) the concern over the potential impacts on health of black carbon and the links between black carbon particles and cardiovascular health effects and premature mortality, for both daily (short-term) and annual (long-term) exposures, suggesting that further systematic monitoring of black carbon could improve the evaluation of the health risks posed by primary combustion particles, complementing the information provided by PM_{2.5} mass concentrations.

2.3 Measuring Black Carbon

Black Carbon atmospheric concentrations cannot be measured as such. However various methodologies can be applied to measure metrics which are related to BC. The two most common ones are described below.

- Elemental Carbon (EC)

EC can be operationally measured in particulate matter deposited on filters using thermal methods based on EC non-volatility (EN 16909). The inter-laboratory comparisons organised by the JRC for 15 years showed that the reproducibility standard deviation for EC filter loadings (µg/cm²) is about ±15%.

A synthetic reference material for Elemental Carbon is currently being developed by the JRC.

- Particulate matter light absorption (or equivalent black carbon, eBC)

⁴ It is worth noticing that BC is different from elemental carbon (EC) that is already required by 2008/50/EC directive for which EN standards exist.

Light absorption coefficients can be obtained from on-line automatic optical measurements. Common techniques primarily consist of the measurement of light attenuation by particulate matter deposited on a filter tape. Measurements are generally performed at different light wavelengths. The aerosol light absorption coefficient can be determined within $\pm 10\%$ to $\pm 25\%$ (reproducibility standard deviation), depending on the measurement technique and the main issue regards the current lack of reference material for aerosol light absorption measurements.

Light absorption coefficients can potentially be converted into BC mass concentrations using a conversion factor (the mass absorption cross-section). However, several studies (e.g. Zanatta et al., 2016) showed that the mass absorption cross section is highly variable in time and space because light absorption from aerosols is not only related to BC concentration, but also to other factors such as the presence of brown carbon, and iron oxides, non-absorbing coatings, absorbing particle number size distribution. Nevertheless, equivalent Black Carbon (eBC) mass concentrations can be calculated from light absorption coefficients by using a constant arbitrary mass absorption cross-section as a conversion factor.

2.4 Measurements availability, coverage and use

Both the Elemental Carbon concentration and aerosol light absorption have been monitored at some atmospheric research observatories (including at JRC-Ispra) for 15 years. EC and BC data from AQ National networks have been reported to the European Environmental Agency (EEA) since 2010 and 2013, respectively.

- *Elemental Carbon (EC)*

The measurement of Elemental (and Organic) Carbon in $PM_{2.5}$ at rural background sites is requested by the Ambient Air Quality Directive 2008/50/EC. These variables are currently measured continuously or semi-continuously (e.g. 3 days per week) at 25 EMEP⁵ and ACTRIS⁶ sites and at several other sites across Europe. EC measurement data from EMEP and ACTRIS sites are openly accessible from EBAS (<https://ebas.nilu.no>). EC data archived by the EEA can also be accessed from the expert viewer dashboard⁷. Other EC data are potentially available from national agencies. A large amount of EC measurement data from on-spot campaigns and research projects is also accessible from the literature.

- *Light absorption or equivalent Black Carbon (eBC)*

The aerosol light absorption coefficient is one of the core variables within ACTRIS and an EMEP level 2 variable as well. It is currently continuously measured at more than 30 EMEP and ACTRIS sites across Europe and many more globally, particularly in the USA. Continuous time series of multi-wavelength aerosol light absorption obtained at ACTRIS and EMEP sites are openly accessible from EBAS database (<https://ebas.nilu.no>). Aerosol light absorption is also generally measured in field campaigns addressing specific air pollution questions. These data are more and more frequently made available by authors as they publish their papers.

Currently all BC data sets actually contain eBC data. Data from 30 to 60 stations, distributed in four Member States (Netherlands, Belgium, Finland and Sweden) and in the United Kingdom (UK) are available for years 2010-2020 from the EEA AirBase data bank. Measurements have also been performed at several other (rural to urban) sites across Europe for more than a decade, but a comprehensive inventory of these additional data sets is difficult in the absence of structured reporting to open international databases.

⁵ EMEP is the co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe. EMEP data are available in open access from EBAS (ebas.nilu.no), a database hosting observation data of atmospheric chemical composition and physical properties, operated by NILU, Norwegian Institute for Air Research. Depending on their completeness, EMEP measurements are associated with different "levels": level 1 measurements include parameters required to describe basic aspects of tropospheric chemistry and deposition rates and standard meteorological parameters. Level 2 measurements can include inter alia PM components, heavy metals and Persistent Organic Pollutants (POPs). Level 3 measurements are research-driven and can include e.g., vertical profiles of ozone and aerosols, dry deposition fluxes and others.

⁶ The Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) is the pan-European research infrastructure (RI) producing high-quality data on short-lived atmospheric constituents. Currently the ACTRIS community involves more than 100 research organisations from 22 European countries. www.actris.eu

⁷ <https://www.eea.europa.eu/data-and-maps/dashboards/air-quality-statistics-expert-viewer>

Figure 1 - European ACTRIS and EMEP stations measuring light absorption coefficient (Source: actris.nilu.no)



The main use of the aerosol light absorption data is currently in climate change research (long-term trend in aerosol direct radiative forcing). Equivalent Black Carbon data were also recently used to determine the COVID lockdown's effect on emissions across Europe (Evangelizou et al., 2021). Aerosol light absorption (or equivalent Black Carbon) data are one of the core variables requested by Copernicus Atmosphere Monitoring Service (CAMS) for data assimilation and model validation. Elemental Carbon data are mainly used in combination with aerosol light absorption data to derive the elemental carbon absorption cross section, a key variable to model the radiative impact of air pollution from emission inventories.

2.5 Towards more systematic monitoring

Possible expansion of BC/EC measurements will have to consider both, direct instrumentation costs and additional costs related to calibration and the resources needed to perform the analysis.

The instrument costs for measuring BC and EC are quite different:

For EC: The current (2021) cost of an off-line thermal Carbon Analyser is about 50 k€. Off-line analyses of Elemental (and organic) Carbon are quite time consuming. Therefore, the running cost of an off-line carbon analyser very much depends on the local labour cost. It can be estimated to range between 3 and 12 k€/yr/station.

For eBC: The current (2021) cost of an advanced multi-wavelength absorption photometer is about 30 k€. Such an instrument needs little maintenance and service. Running costs (< 1k€/yr) are mainly related to the use of the sampling filter tape.

The current (2021) cost of an on-line thermal carbon analyser is about 60 k€. In principle such an instrument needs little maintenance and service. Running costs (ca. 1k€/yr) are mainly related to the periodic change of the sampling filter and spare parts.

Absorption photometers have been operated for many years at numerous atmospheric research observatories and monitoring stations, such as ACTRIS and EMEP. The main obstacle to a wider implementation of equivalent Black Carbon measurements (using an absorption photometer) is the difficulty in performing calibrations, due to the lack of reference material for such measurements. International standards for performing these measurements are also currently lacking, but on-going metrological projects (e.g., EURAMET Black Carbon project) are expected to trigger the development of a standard method in the coming years.

Measurements of Elemental Carbon have also been performed at numerous research atmospheric observatories and monitoring stations for many years. The main limitation to more systematic monitoring of Elemental Carbon (including urban sites) is the time needed for filter sampling and analyses. Other disadvantages include the handmade style of purpose-built current analysers, the need of a proper analytical

laboratory (including gas cylinders, ultra-pure water, clean bench, etc...) and the current lack of reference material (close to be finalised). On-line (organic and) elemental carbon analysers also need gas cylinders. Their performances in inter-laboratory comparison are not always satisfactory.

- Potential contribution to Zero Pollution objectives.

As discussed, the evidence of specific impacts of BC on human health is growing, and deeper knowledge of its sources and levels in the urban atmosphere will help to better focus actions aiming to support clean air and zero pollution objectives. Clearly, with human health being the main concern, monitoring strategies will need focus on urban areas, to better identify by means of appropriate source apportionment methodologies, the most relevant sources to act on.

Sensitive measurements will be also needed: Black Carbon and Elemental Carbon are currently significant but minor constituents of Particulate Matter: for instance, in 2015, BC emissions accounted for 8% of PM₁₀ emissions in EU27 and it is also a tracer of incomplete combustion processes, which might also generate the co-emission other more harmful species.

Finally, it is worth noting that Black Carbon is a key element of the public perception of air pollution because it is the only atmospheric pollutant visible to humans, at least at the source point of super-emitters.

3 Ammonia

3.1 Properties & sources

Ammonia (NH_3) is a reduced nitrogen species. It is highly volatile soluble in water (60 times as much as SO_2 but 3.000 times less than nitric acid HNO_3) and sticky to surfaces (slightly more than SO_2 , but 5–10 times less than HNO_3). Ammonia is a weak Lewis basis. It reacts quickly with Lewis acids as e.g., organic acids, sulphuric acid (H_2SO_4) and nitric acid (HNO_3) in the atmosphere to give origin to ammonium nitrate (NH_4NO_3) and sulphates (NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$), which are important components of secondary particulate.

In Europe, the main source of ammonia by far is agriculture (93% of NH_3 emissions in 2015, according to EDGAR), principally livestock husbandry and manure spreading. The natural biosphere is also a source of ammonia. However, traffic related emissions of ammonia by both gasoline engines and Diesel engines equipped with DeNox catalytic (or selective catalytic reduction, SCR) systems are currently largely underestimated (Suarez-Bertoa and Astorga, 2016; Suarez-Bertoa et al., 2017 and references therein).

3.2 Reasons for concern

The health hazard of exposure to atmospheric NH_3 is relatively low at typical ambient (outdoor) concentrations (maximum a few tens of ppb, 24-hr average). For instance, the Occupational Safety and Health Administration fixes the limit value at workplaces to 50 ppm for 8 hours, while the EPA Chronic Inhalation Reference Concentration (RfC) for effects other than cancer is equal to 650 ppb⁸.

Instead, concerns about the health impacts of ammonia are due to its key role in the formation of secondary organic aerosol, especially of ammonium nitrate. In particular, ammonia plays a major role in the formation of secondary aerosol (NH_4NO_3) in the western part of Europe (Clappier et al., 2021). Consequently, reducing NH_3 emissions is an efficient way to abate $\text{PM}_{2.5}$ mass concentrations across large areas of the EU. (Gu et al., 2021) recently confirmed that a decrease of ammonia emissions on a global scale would be a more cost-effective strategy to control $\text{PM}_{2.5}$ concentrations than strategies focusing on nitrates. From a measurement perspective, European aerosol phenomenology papers indicate that ammonium nitrate accounts on average for 18% of PM_{10} , 13% of $\text{PM}_{2.5}$ (Putaud et al., 2010), and 17% of non-refractory PM_1 at regional and urban background sites (sampling artefact free data, (Bressi et al., 2021)).

It should be noted that ammonium nitrate contributions to PM generally increase in winter when PM levels also increase. Without ammonia in the gas phase, there would be no ammonium nitrate in the particulate phase (PM), and nitric acid would be removed from the atmosphere by dry or wet deposition.

Moreover, ammonia (and ammonium) deposition fluxes contribute to the eutrophication of water bodies, and to the acidification of soils (via nitrification). Indeed, (EEA, 2019) has reported how eutrophication is one of the reasons that achieving good environmental status of European marine waters remains unlikely, while (Maas and Grennfelt, 2016) have demonstrated how further reductions of ammonia emissions are necessary to keep critical loads⁹ for nitrogen eutrophication in Europe under control.

3.3 Measuring Ammonia

Ammonia can be measured by diffusive sampling, as described in the EN standard method for measuring gas phase ammonia (EN 17346:2020): diffusive sampling (denuder), extraction, and determination of ammonium (NH_4^+) in solution. Gas phase ammonia can also be measured from impregnated filters placed downstream of total particulate filters (filter packs) and subsequent extraction and analysis by e.g., ion chromatography. Other methods include on-line chemiluminescence (after thermal conversion to NO), cavity ring-down spectroscopy (IR) based monitors, and optical remote sensing techniques like Differential Optical Absorption Spectroscopy (DOAS) and Differential Absorption (LIDAR¹⁰) (Gasmi Cherifi, 2021)

The many technical issues related to off-line ammonia measurements mainly relate to risks of contamination for diffusive or filter pack sampling (analytical methods are robust). In the case of thermal conversion and chemiluminescence, the converter efficiency and selectivity could also be questionable.

⁸ iris.epa.gov/static/pdfs/0422_summary.pdf

⁹ According to EEA, 2020 a critical load represents the upper limit of one or more pollutants deposited on the Earth's surface that an ecosystem, such as a lake or a forest, can tolerate without its function or its structure being damaged.

¹⁰ Light Detection and Ranging

Cavity ring-down spectroscopy is *a priori* a very selective and accurate technique, but instruments and data output processing require experience and attention.

3.4 Measurements availability, coverage and use

Ammonia is not currently included in the standard set of variables measured in ACTRIS. However, it is an EMEP core variable (level 1 variable). It has been monitored at a number of EMEP sites from the 1990's and it is currently continuously measured at 43 EMEP sites in 13 countries across Europe by a mixture of denuders and filter packs. The spatial coverage spans from 37° to 69° N, and from 9° W to 44° E, plus 2 sites in the Arctic. Continuous time series of ammonia concentrations are openly accessible from the EMEP database (<https://ebas.nilu.no>). Time resolutions range from 1 hour to 1 month.

Ammonia measurements have also been performed for instance at a series of sites in northern Italy (e.g., Lombardy region) and data for ammonia are potentially accessible via regional environment protection agencies. However, being reported only partially to the EEA (less than 20 sites are currently reported), NH₃ data sets obtained by regional environment protection agencies across Europe might be difficult to dig out.

Ammonia data obtained during specific field campaigns might be available from scientific papers, but accessing a sufficiently large amount of data from the scientific literature (to ensure a certain representativeness) looks unaffordable.

So far ammonia concentration measurements are mainly used for assessing deposition fluxes on ecosystems, especially in protected areas. Ammonia concentration measurements have also been used to check emissions inventories and models (e.g. (Dore et al., 2020); (Sutton et al., 2003), Van Damme et al., 2021). Remote sensing instrumentation is attractive from emissions inventories perspective (Farren et al., 2020).

3.5 Towards more systematic monitoring

Ammonia off-line analyses are basically cheap in terms of investment (if analytical instruments for NH₄⁺ determination do not have to be especially purchased for NH₃ measurements) and consumables, but are time demanding. The overall cost is probably in the range 10 – 50 € per sample.

The cost of a thermal converter – chemiluminescence NH₃ monitor ranges between 12 and 15 k€, and that of a cavity ring-down NH₃ spectrometer is about 60 k€ with running costs close to 1k€/yr.

The examples of the UK and to a certain extent the Lombardy Region in Italy, show that deploying and managing a relatively dense NH₃ monitoring network is feasible. Nevertheless, possible issues could include high costs and difficulties to maintain high time coverage when using expensive techniques such as cavity ring-down spectroscopy, while data quality can be an issue with other less expensive techniques.

- Potential contribution to Zero Pollution objectives

A more developed monitoring framework for ammonia would help improve understanding of its role as a precursor of PM, especially in the urban areas where the human health impact of air pollution tends to be highest.

NH₃ emissions are regulated by Directive 2016/2284. Although, 23 Member States (MS) remained below their national ceilings for ammonia, the projected compliance against the 2020-2029 emissions reduction commitments (ERCs) from Directive 2016/2284 looks less promising, with 13 MS currently projected to exceed their ERCs (raising to 22 MS currently projected to exceed their ERC for 2030¹¹). In many countries NH₃ emissions have decreased only slightly, or in some cases increased, since 2005 (EEA, 2021, European Commission, 2021e). A further development of the monitoring network in especially ecologically sensitive areas, including water bodies, would support a more detailed understanding of reasons underlying the still too large deposition fluxes currently observed and could contribute to actions towards achieving the ZP target to reduce by 25% ecosystems where biodiversity is threatened by air pollution.

¹¹ Even with additional pollution control measures implemented, 15 MS are projected to miss their ERC for ammonia in 2030.

4 Ultra-fine particles

4.1 Properties & sources

Ultra-fine particles (UFP) are characterised by a diameter <100 nm. As a result of source and sink strengths, particles with diameters ranging 50-100 nm are usually the most abundant in the air, except at traffic sites where primary particles (10 – 25 nm) often dominate, and during nucleation events (gas to particle conversion) where new particles (a few nm) form and grow.

The chemical composition of primary and new particles depends on the emission sources and precursors, respectively. Particles in the range 50-100 nm are mostly made of a refractory core (initially emitted as a primary particle) surrounded by a shell composed of secondary organic or inorganic particulate matter.

Primary particles are mainly produced by incomplete combustion processes. Other sources include friction (e.g., break disks vs pads, trams' wheels vs rails). The oxidation of gas phase pollutants to condensable vapours leads to the formation of clusters which can grow up (by condensation and coagulation) to stable particles. Except during nucleation events, most atmospheric particles result from primary particle emission and subsequent condensation of condensables¹² on their surface.

4.2 Reasons for concern

Particles in the approximate range 10 -100 nm mainly deposit in lungs' alveoli, while particles larger than 300 nm or smaller than 10 nm deposit mainly in the nose, larynx and pharynx. When deposited in the alveoli, 10 -100 nm particles are not easily detected by macrophages and therefore can accumulate.

The WHO has recently assessed the growing amount of epidemiological evidence linking exposure to UFP with health, both short-term (visits in emergency departments, hospital admissions, respiratory symptoms, pulmonary inflammation, etc.) and long-term effects on mortality and morbidity, and concluded that, although growing, "the body of epidemiological evidence was not yet sufficient to formulate AQ guidelines" for UFP particle number concentrations, but concerns were considered sufficient to formulate appropriate "good practice" advice (WHO, 2021).

4.3 Measuring Ultra Fine Particles

Since UFP largely dominate particle number size distributions (PNSD), their number concentration can be estimated by measuring the total particle number concentration. Total particle number can be measured by condensation particle counters (CPC) in which particles are exposed to highly saturated vapours (of butanol or water) and grow large enough to become detectable by a (visible) laser beam. Currently, CEN specifications (CEN TS16976) do not recommend the use of CPC working with water as condensation fluid.

CPC are delicate instruments and in need of expert operators and data evaluators. The efficiency and performance of CPCs strongly depends on the temperature difference between the saturation and the condensation chambers. The detection efficiency of all CPCs on the market drops below about 20 nm and the detection efficiency curve depends on the brand, model, and varies from unit to unit. Even if the detection efficiency curve is known, the total particle number concentration cannot be precisely determined if the number size distribution is unknown.

CPCs cannot be in-house calibrated. They must be calibrated at specialised calibration centres by comparison with electrometers. Moreover, the use of butanol as a working fluid can lead to potentially harmful workers' exposure if CPCs exhausts are not properly treated.

In research, total particle number concentrations are measured to cross check PNSD measurements only.

4.4 Measurements availability, coverage and use

Particle number size distributions (PNSD) have been measured at an increasing number of ACTRIS atmospheric observatories since the end of the 1990's (4 sites) until now (>30 sites). Currently, data are available from 7°W

¹² It is worth reminding that significant differences exist in the way countries report this kind of emissions. Some countries include "condensable" organics in their emissions, while others exclude such organics. This difference is important since inclusion of organics can sometimes increase reported PM_{2.5} emissions even by a factor of 3 or sometimes more.

to 26°E, and from 68°N to 35°N. Particle number concentrations measurements under air quality monitoring networks started somewhat later (end of the 2000's).

Particle number size distributions measured at ACTRIS atmospheric observatories are openly available from ebas.nilu.no (about 30 sites), together with data obtained by the GUAN network. PNSD are also commonly measured in research field campaigns addressing specific particulate pollution sources and processes. However, getting access to a sufficiently large amount of data from the scientific literature looks unaffordable.

Total particle number concentrations might be monitored by national or regional Environment Protection Agencies in various countries across Europe. Not being reported to the EEA, such datasets could only be accessed after making contact with relevant persons in each country or region.

PNSD are mainly used in research activities dealing with the sources and processes governing particle number concentrations (e.g., impact of COVID-19 lockdowns) and have been used also to assess the (very) few models dealing with particle numbers. The extent of the use of particle number (concentrations or size distributions) in studies dealing with the health impact of particulate pollution is still to be investigated.

4.5 Towards more systematic monitoring

The cost of an instrument for determining the PNSD of ultrafine particles (and beyond) is about 100 k€. The instruments also need to be run by qualified staff for checking flows, check particle sizing and other specialised duties. Condensation particle counters themselves cost about 30 k€, running costs are limited to the butanol fluid (approx. 1 k€/yr), while maintenance is to be performed by qualified operators.

The examples of Germany and Spain show that deploying and managing a PNSD monitoring network is feasible. However, beyond costs, issues to be tackled include the use of radioactive sources in the bipolar chargers (PNSD), the use of butanol as working fluid and the uncertainty introduced by the efficiency curve dropping below 20 nm (UFP concentration), the lack of reference materials for calibration and the need for particle loss corrections.

- Potential contribution to Zero Pollution objectives.

Particle number size distributions are necessary to determine the dose of particulate matter deposited along respiratory tracts. Therefore, PNSD (or the number size distribution of e.g., particles containing a non-soluble core) could be a key factor in determining the health impact of particulate air pollution and its enhanced monitoring, especially in urban areas, will support a better tailoring of policies aimed at reaching the overall ZP target of decreasing the impact of air pollution on human health. Conversely, UFP number concentration is not necessarily a good indicator of pollution, as biogenic emissions of e.g., isoprene and terpenes are known to lead to huge numbers of new particles during nucleation events.

5 Methane

5.1 Properties & sources

Methane is a hydrocarbon with chemical formula CH_4 and is the lightest alkane. At standard temperature and pressure, it is an invisible and odourless gas. Methane reacts easily with oxygen in combustion, and it is flammable in air at concentrations between about 5% and 17%.

Methane is the most abundant organic chemical in the Earth's atmosphere and is present in the troposphere at concentrations usually well below the range of flammability, while its accumulation in restricted areas is a well-known cause of explosions and represents a major industrial and domestic hazard. According to the National Oceanic and Atmospheric Administration (NOAA) Global Monitoring Laboratory the global average atmospheric concentration of methane in June 2021 reached a seasonally adjusted value of 1.889 ppb, more than 150% higher than preindustrial era values. Since 2007 the atmospheric methane level has risen sharply. It began growing by about 8 ppb/y in 2007, after a period of stability at the start of the millennium. The growth rate accelerated in 2014 (13 ppb/y) and it reached the value of 15 ppb/y in 2020 (Ed Dlugokencky, NOAA/GML¹³). The trend of globally averaged methane concentrations does not seem to be influenced by the COVID-19 pandemic, considering the relatively long lifetime of methane in the troposphere (9.1 years, see (IPCC, 2021)).

Sources of methane can be grouped into three categories: biogenic, thermogenic and pyrogenic. Biogenic sources are represented by microbial methanogenesis operating in anaerobic environments such as natural wetlands and rice paddies, oxygen-poor freshwater reservoirs (such as dams), digestive systems of ruminants and termites, and organic waste deposits (such as manure, sewage and landfills). Thermogenic CH_4 is a fossil fuel. It is vented from the subsurface into the atmosphere through natural features (such as terrestrial seeps, marine seeps and mud volcanoes), and through the extraction of coal, oil and natural gas. Pyrogenic CH_4 is produced by the incomplete combustion of biomass and soil carbon during wildfires, and of biofuels and fossil fuels.

In the EU, CH_4 originates mainly from the agricultural sector (enteric fermentation and manure management, representing 45 % of CH_4 emissions in 2018), waste treatment and disposal (representing 30 % of CH_4 emissions in 2018) and production of coal and gas (representing 18 % of CH_4 emissions in 2018). In 2018, the top five emitting countries, responsible for 58% of EU CH_4 emissions were France (14%), Germany (13%), Poland (13%), Spain (9%) and Italy (9%).

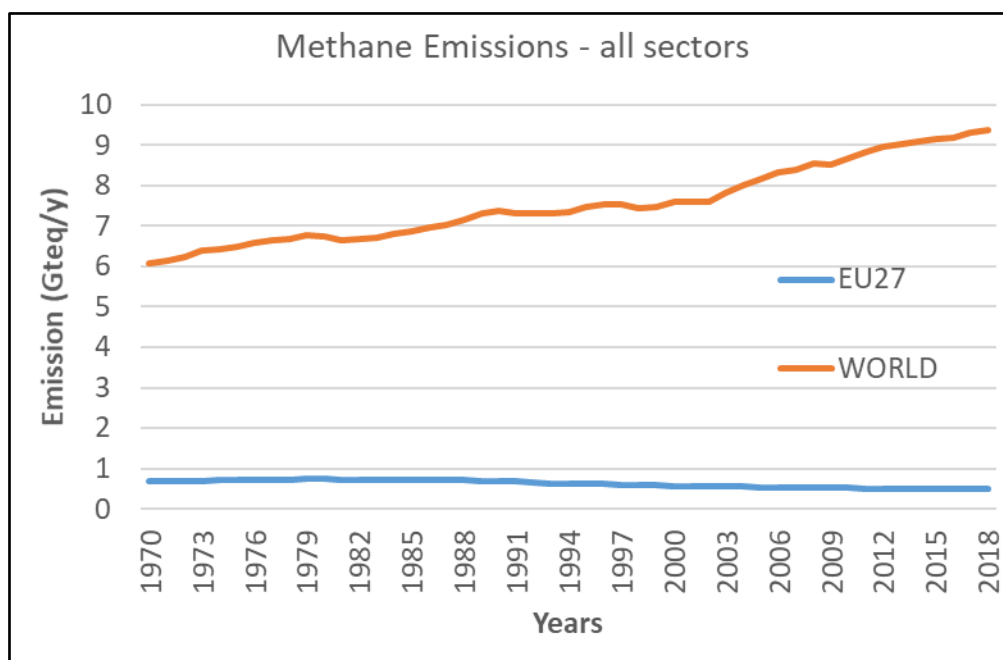
5.2 Reasons for concern

Methane is a powerful greenhouse gas (GHG) with a Global Warming Potential (GWP) equal to 82.5 on a 20-year time scale and 29.8 on a 100-year time scale (IPCC, 2021). It also plays a major role in the formation of tropospheric ozone (O_3) and stratospheric water vapour, which indirectly add approximately 0.2 Wm^{-2} to its climate forcing. Similarly to Volatile Organic Compounds (VOCs), methane acts as a precursor to ozone formation providing a methyl peroxy-radical triggering the oxidation of NO to NO_2 , a compound that can be photolyzed resulting in the production of a free atom of oxygen. The free oxygen atom (O) can then react with molecular oxygen (O_2) to produce ozone.

Being a powerful GHG, several CH_4 emission inventories have been developed for instance in the frame of the reporting obligations of the Paris agreement and in the climatic research field. According to the latest EDGAR estimates (see Figure 2), CH_4 global anthropic emissions were in 2018 54% higher than in 1970 and 27% higher than in 1990. In contrast, EU27 emissions were in 2018 about 30% lower than both in 1970 and 1990 and, as EU share of global emissions, decreased from 11% in 1970 to 9% in 1990 and to 5% in 2018.

¹³ gml.noaa.gov/ccgg/trends_ch4/

Figure 2 - Global and EU27 methane emissions evolution between 1970 and 2018 (Source: EDGAR)



Besides the impact of methane on climate, the main reasons of concern for methane are related to its role in the formation of tropospheric ozone. As also declared by the European Commission in appendix to Directive 2016/2284 “there is a strong air quality case for keeping the development of methane emissions in the Member States under review in order to reduce ozone concentrations in the EU and to promote methane reductions internationally”. Ozone is in turn a very powerful GHG, and a pollutant which is harmful to human health at current atmospheric concentrations. (Sicard et al., 2021) have analysed the trends in European O_3 levels driving the conclusion that “the control strategies of O_3 precursor emissions were effective in rural areas” while an average increase of premature deaths related to ozone of 0.55 deaths per 10^6 inhabitants was estimated for the time period 2000–2017, triggering a renewed attention to O_3 mechanism of formation and precursors emissions control in cities. Moreover, ozone is known to have a relevant impact on crops productivity and forest health and AOT40¹⁴, the main indicator used for quantifying these effects, has shown a slow but clear decreasing trend in southern Europe and continental Europe and has remained steadily below the EU target value in northern Europe between 2000 and 2014 (Proietti et al., 2021).

In order to better map methane emissions in cities and their potential role in the urban ozone cycle, several recent observations have focused on urban areas and/or industrial sites. For instance, one study (Maazallahi et al., 2020) has focused on the cities Hamburg (Germany) and Utrecht (Netherlands) and detected local methane emissions equal to 440 ± 70 t/y from all sources in Hamburg and 150 ± 50 t/y in Utrecht, with an emission share ranging between 50% and 90% to be caused by the distribution of natural gas across the city. A similar situation has been found in the US east coast, where the five major cities areas (Washington, New York, Boston, Philadelphia and Baltimore) emit 0.85 Tg CH_4 /year, of which again a large share (0.75 Tg CH_4 /year) is attributed to natural gas (Plant et al., 2019).

In both cited cases, and in others reported in literature an important mismatch has been found between estimated urban methane emissions and the equivalent emissions from available local emission inventories and consequently concerns have been risen on the actual coverage of CH_4 measurement networks. These discrepancies with emission inventories are a well-known cause of the difficulties that chemical transport models typically have in estimating ozone formation.

On the global scale, ozone production triggered by non-methane VOCs normally dominates the methane-induced production because of shorter reaction times, especially in polluted areas. (Heyes and Schopp, 1995).

¹⁴ AOT40 is defined as the sum of the differences between hourly ozone concentration and the 40 ppb threshold for each hour when the concentration exceeds it.

However, the theoretical maximum yield of O_3 from methane oxidation is 5 O_3 molecules per each CH_4 molecule oxidised and it would occur when NO_x levels are sufficiently high that the peroxy radicals react exclusively with NO and all formaldehyde formed photolyses by radical path.

Ozone induced through CH_4 oxidation has been shown to play an important role in O_3 background concentration production. For instance, (Fiore et al., 2008) has shown how a global emission decrease of 10% in methane emissions would lead to a decrease in ozone background by 0.4 to 0.7 ppb. Moreover, a study from the JRC (Van Dingenen, R. et al., 2018) has estimated CH_4 -induced O_3 concentrations range 5 to 8 ppb (annual country average) across Europe with a theoretical maximum mitigation potential (corresponding to an ideal total abatement of anthropogenic CH_4 emissions) leading to a decrease of ozone related crops damage by 26%, and of ozone caused mortality by 20% at the global level (in Europe decreases to crop damages and to mortality would reach 40% and 34%, respectively).

It is worth noting that the design of containment measures for methane and other ozone precursors needs to remain focused on both urban and global perspectives. Indeed, the relatively long time life of ozone in the troposphere results in its transport towards more remote areas and even across continents (Yan et al., 2018).

5.3 Measuring Methane

Until recently, CH_4 in-situ measurements have been performed by gas chromatography equipped with flame ionisation detectors (GC/FID). In the last few years, spectroscopic techniques such as direct absorption spectroscopy, cavity ring-down spectroscopy (CRDS), cavity enhanced off-axis integrated cavity output spectroscopy (OAICOS) and Fourier transform infrared (FTIR) spectroscopy have become commercially available for monitoring of atmospheric CH_4 . Currently, many traditional GC/FID systems for methane monitoring are being replaced by modern spectroscopic instruments based on CRDS technology. This new technique requires less frequent calibration and offers better repeatability (approx. 0.25 ppb), a larger dynamic range and faster response times without the need to use N_2 carrier gas and H_2 fuel gas that are needed to operate a GC/FID (Zellweger et al., 2016). Moreover, instrumentation based on the CRDS technique is much more robust and reliable.

Recently, relatively low-cost technologies, including e.g., infrared cameras have been also developed for the purpose of identifying methane leaks from pipelines and industrial facilities, although their set-up does not necessarily allow a precise estimate of concentrations.

In-situ CH_4 observations are usually taken on top of tall towers, in mountainous terrain or in remote environments. Measurements have different sampling frequency and can be grouped into discrete air samples (flasks) collected about once a week and continuous (5-10 seconds/hourly) in situ measurements. The continuous measurements contain more information about sources and sinks than the weekly flask data. However, denser spatial coverage is typically achieved through analysis of flask air samples, such as those collected in the global NOAA/ESRL cooperative air sampling network.

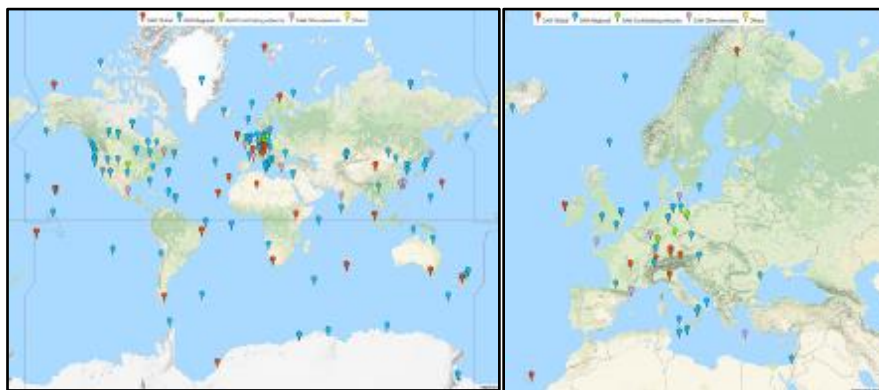
Long-term in-situ observations of methane concentration are used by atmospheric models to provide estimates of total CH_4 emissions, i.e. including both anthropogenic and natural sources, at global, regional and national scales (Bergamaschi et al., 2018). Direct measurements of CH_4 in the atmosphere began in 1978 and reached global coverage after 1983 through networks operated by Integrated Carbon Observation System (ICOS), National Oceanic and Atmospheric Administration (NOAA), Advanced Global Atmospheric Gases Experiment (AGAGE), Commonwealth Scientific and Industrial Research Organization (CSIRO) and University of California Irvine (UCI). To ensure that measurements of CH_4 from different networks meet the Global Atmospheric Watch (GAW) compatibility target of ± 2 ppb and can be included in model studies without causing bias, a common standard scale maintained at NOAA is used. Moreover, quality control strategies, such as 'round-robin' comparisons of air in cylinders, same air comparisons, and comparison of simultaneous measurements using different methods, are implemented to ensure the highest accuracy and precision of measurements.

5.4 Measurements availability, coverage and use

Because of its lifetime, much longer than the typical time scale of interhemispheric transport (1 year according to (Geller et al., 1997)), methane is relatively well-mixed in the troposphere and homogeneous background concentrations are observed across the globe. Nevertheless, methane concentrations can show both seasonal and geographical variations due to the presence of local emission hotspots and the occurrence of seasonal removal processes.

Most of the current surface stations reporting CH₄ concentrations to the World Data Centre for Greenhouse Gases (WDCGG) are located at marine sites and in North America, East Asia and Western Europe. The most important regions where the largest natural fluxes are located, including the Southern Ocean, tropical South America, tropical Africa, Siberia and the Arctic have very few observation sites. This severely limits the ability of global modelling to get reliable estimates of CH₄ fluxes over these regions. Most of measurements are coordinated through the Global Atmosphere Watch programme of the World Meteorological Organization and collected by the WDCGG. Stations reporting (or having reported in the past) to WDCGG are shown in Figure 3.

Figure 3 - Network of measuring stations reporting to World Data Centre for Greenhouse Gases (Source: WDCGG)



Background stations reporting to WDCGG provide a time coverage variable from hourly to weekly depending on the sampling methodology. A few stations provide continuous observations, sometimes dating back to 1980s, while most of the stations have started their observations in the 1990s or early 2000s. On the contrary observations from campaigns and other research projects do not usually provide systemic coverage.

Data from WDCGG are easily accessible through the website, and similarly the few data reported by German stations to EEA are available through the Agency data repository, while data from single measurement campaigns are usually available from literature publications through contacts with authors.

The European ICOS research infrastructure is a prominent example of coordinated and harmonised monitoring network for GHGs, including CH₄. The high quality and compatibility of CH₄ data are achieved through rigorous standardisation of instrumentation, sampling, calibration and quality assurance procedures as well as centralised data evaluation, that for CH₄ concentration data is performed by a dedicated Atmospheric Thematic Centre¹⁵. The ICOS Central Analytical Laboratories (CAL)¹⁶ provide calibration gases for the entire network to avoid calibration biases between the European stations. Moreover, regular inter-comparison with other networks worldwide is performed by the CAL in order to guarantee compatibility also at the global scale.

The ICOS atmosphere network includes 38 surface stations distributed over 13 European countries. Data from these stations are publicly available at the ICOS Data Portal¹⁷

Besides these global background networks, methane is also measured in regional studies often related to production basins and, more and more often, in urban areas. For instance, the MEMO² research network¹⁸ focuses on local and regional methane emissions throughout Europe and thanks to the use of mobile detectors is pursuing finer scale emission mapping of cities and industrial areas. Such a detailed work has allowed to detect and sometimes identify a somewhat unexpected number of relevant methane leaks e.g., throughout Paris, where fugitive emissions from the natural gas distribution system have been identified as the major urban source of methane, larger than sewage and furnaces combined. Mobile measurements have been also used to assess emissions from industrial areas e.g., in 130 gas processing sites in the USA (Mitchell et al., 2015).

Nevertheless, methane is not yet routinely measured in the European in urban or regional air quality network networks with the exception of Germany, where a dozen of stations have reported to EEA methane since 2013.

¹⁵ ICOS ATC, <https://icos-atc.lscce.ipsl.fr/>

¹⁶ ICOS CAL, <https://www.icos-ri.eu/icos-central-facilities/icos-cal>

¹⁷ <https://www.icos-cp.eu/data-products>

¹⁸ <https://h2020-memo2.eu/>

It is nevertheless worth reminding that as far as methane emissions related to coal and gas industry are concerned, the European Commission has proposed a number of emission reporting obligations for energy sector operators (European Commission, 2021d).

5.5 Towards more systematic monitoring

On-line methane measurements are nowadays usually performed using the Cavity Ring-Down Spectroscopy (CRDS) techniques. The instruments based on this technique are currently expensive (≥ 100 k€) but running costs are low (< 1 k€ /yr). Off-line methane measurements are commonly performed by GC-FID. GCs are not that expensive (ca. 50 k€) but maintenance costs are larger than for CRDS based instruments. Methane measurements need high accuracy and frequent round robin experiments are organised by e.g., the Central Calibration Laboratory (CCL) of the WMO. In order to assure measurements comparability and reliability, WMO recommends maximum discrepancies of ± 2 ppb at 1 sigma when background concentrations are investigated and a less stringent extended compatibility goal of ± 5 ppb for studies in which the highest precision and accuracy is not required. The cost of calibration scales range 1 – 3 k€/yr. Practical issues will be possibly related to calibration issues or the high costs of the systemic samples collection, especially if fine time resolution will be required.

- Potential contribution to Zero Pollution objectives.

Ozone is a major pollutant for which standards exist in the current EU legislation. WHO has also fixed more stringent interim targets and guidelines levels for ozone to further protect human health. In this context, extending the methane systemic monitoring infrastructure could allow completing the pool of ozone precursors observed directly, contributing to the pursuit of the ZP target on reducing the human health impact of air pollution. Moreover, better knowledge of the impact of ozone on ecosystems would be important in addressing the ZP target of protecting biodiversity from air pollution.

The monitoring infrastructure should be able to pinpoint emissions more accurately and quickly, and to find smaller point sources that can significantly increase CH_4 concentrations (at least on local scale). Indeed, although other precursors are also known to have a relevant role in ozone formation and scientific investigation have clearly shown the contribution of both long-range transport and stratospheric intrusions, there is consensus that methane induced ozone formation could not be neglected. From this perspective, more detailed information on methane distribution across the atmosphere is likely to shed additional light on ozone formation process, although it will be probably necessary to choose carefully the actual locations, possibly driven by the needs of the modelling tools aimed at investigating urban ozone phenomenology.

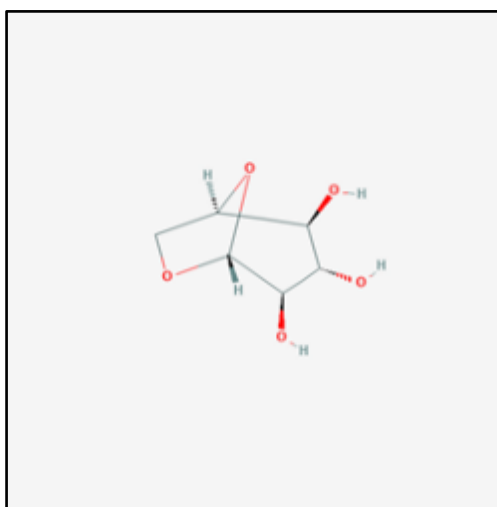
6 Levoglucosan

6.1 Levoglucosan properties and sources

Levoglucosan (1,6 anhydro- β -D-glucopyranose) is a product of cellulose pyrolysis at temperatures higher than 300 °C. It is typically the single most abundant species in wood burning aerosols. Other wood combustion products comprise its isomers like mannosan, galactosan and methoxylated phenols (Kuo et al, 2011)¹⁹

Levoglucosan has a molecular weight of 162.14 g/mol, a melting and a boiling point of 183 °C and 384 °C, respectively. It is soluble in water (62 g/l), has a density of 1.69 g/cm³ and a vapour pressure of 24.1 μ Pa. Its chemical structure (C₆H₁₀O₅) is represented in Figure 4.

Figure 4 - Levoglucosan chemical structure (Source: textbooks)



Most levoglucosan is mostly accumulated in particles with aerodynamic diameters under 2 μ m (Giannoni et al., 2012). Levoglucosan shows a significant hygroscopicity and the water adsorption attributed to its presence in wood burning aerosols is calculated to be up to 30 % of the organic mass at 90 % relative humidity (Mochida and Kawamura, 2004).

Initial studies of levoglucosan reactivity have shown its ability to resist both photochemical oxidation and acid catalysed hydrolysis in atmospheric aerosols and its relatively high stability of around 10 days without significant degradation (Fraser and Lakshmanan, 2000). However, its use in long-range transport studies has been questioned because of its reactivity and composition changes in aged emissions (Bhattacharai et al., 2019).

Biomass burning has become an important source of pollution and toxicity in the ambient air particulate matter (Karanasiou et al., 2021). Biomass burning has also an impact in climate change by emitting greenhouse gases and altering the scattering and absorbing of solar radiation (Bhattacharai et al., 2019). Numerous studies have demonstrated that the reduction of traffic in cities is not enough to decrease extensively carbonaceous concentrations and, consequently, other important sources as biomass burning need to be targeted to effectively reduce air pollution (Titos et al., 2017). Therefore, the identification and control of biomass burning is becoming of high importance for air policy objectives.

¹⁹ The wide range of reaction products of the wood combustion is the result of its complex structure. In wood, cellulose is the main component of the cell wall accounting for 40-45 % of the wood. Cellulose is a polymer of the glucose, linked with a type of acetal ether linkage (O-C-O-C). Each glucose unit still has three free hydroxyl groups able to react with other groups or with water to form hydrogen bonds. The β configuration of the cellulose leads to highly ordered crystalline structure, although amorphous regions are also present. Other main components of the wood cell wall are the hemicellulose (20-30 % of the dry weight wood) and Lignin (15-36 %). Hemicellulose is an amorphous structure made up of glucose, mannose, galactose, xylose, arabinose, o-methyl-glucuronic acid and galacturonic acid. Lignin is a complex phenolic polymer of alcohol units as p-hydroxyphenyl, guaiacyl, and syringyl lignins.

Biomass burning (BB) is associated with the principal source of levoglucosan emission (Simoneit et al., 1999). However, the percentage of levoglucosan changes with the composition of the biomass burnt and the combustion temperature. Still, levoglucosan and its isomeric ratios are often used to identify the combustion type, which can be locally explained in term of sort and proximity to the emission source. For instance, levoglucosan/mannosan ratio ranges from 5 in softwood to 30 in hardwood smoke, while the combustion of brown coals containing fossilised cellulose (i.e. xylitic lignite) shows very high ratios between 30 to 90. (Kuo, Louchouart, and Herbert, 2011)(Schmidl et al., 2008).

Beside the interest of the isomers' identification, the influence of combustion conditions on these ratios may limit its use to a qualitative approach when this concept is generally applied or used for long-range transport identification.

6.2 Reasons for concern

Levoglucosan by itself does not pose environmental or health risks. Nevertheless, due to its high abundance in biomass combustion and its relative stability in air, it is an important tracer of biomass burning. As an important source of particulate matter and other pollutants, BB is associated with all-cause and cardiovascular mortality and respiratory morbidity, being a high risk of respiratory morbidity on smoky days. In general, there is a poor estimation of BB exposure and a lack of adjustment of important confounders (weather conditions, air pollutants, etc). This supports the inclusion of levoglucosan as a marker for the monitoring of BB emissions (Karanasiou et al., 2021)

Furthermore, biomass burning aerosols are also affecting the climate on local to global scales by affecting scattering and absorption of incoming sunlight, cloud condensation nuclei (CCN), cloud's reflectivity and lifetime, transport of pollutant and water vapour to the stratosphere and changes in precipitation patterns (Schkolnik and Rudich, 2006). Biomass burning in Central Europe is estimated to be the source of 10-20 % of the PM. For instance, 2 $\mu\text{g}/\text{m}^3$ of PM₁₀ are due to biomass burning in winter, and 1 $\mu\text{g}/\text{m}^3$ is the average annual mean contribution in western Germany (Pratap et al., 2019).

6.3 Measuring Levoglucosan

Common measurement methodologies for levoglucosan include Ion Chromatography/High Performance Anion Exchange Chromatography coupled to Pulsed Amperometric Detector (HPAEC-PAD) or mass spectrometry, gas chromatography coupled to mass spectrometry, which usually requires the use of a derivatisation. The analysis of levoglucosan is frequently combined with other species of interest and the final selection of the method depends on the interest of other combined analytes (Schkolnik and Rudich, 2006).

General analytical uncertainties for levoglucosan range between 35 to 50 % for ambient air filters. Uncertainty around 52 % for Standard Reference Material, 90 % of the uncertainty is due to the analytical procedure (Louchouart et al., 2009).

The diversity of methods, uncertainty of the reference material and the multiple factors that can affect quantification (desorption efficiency, separation of isomers, calibration) support the need of an urgent standardisation of the analytical methods (Pomata et al., 2014, Schkolnik and Rudich, 2006, Louchouart et al., 2009). Currently, a working group of the CEN Technical Committee Air Quality is working on the standardisation of levoglucosan measurement in PM. Ion-chromatography and Gas Chromatography equipped with mass spectrometer (GC-MS) are being considered for this purpose.

6.4 Measurements availability coverage and use

Levoglucosan and related isomers are commonly used tracer of biomass burning organic aerosol (BBOA) in source apportionment, as well as methoxyphenols, by-products of the thermal degradation of lignin (Weber et al., 2019, Bonvalot et al., 2016, Maenhaut et al., 2016). Nevertheless, as already mentioned, there is a series of issues related to the analysis and marker representability (emission factor and atmospheric stability) that could question the extensive use of this marker to cover all purpose in the identification and quantification of biomass burning.

Since levoglucosan formation is a function of combustion temperature the use of available source emission rates for this compound cannot be extensively used (Pratap et al., 2019). For instance, the wood levoglucosan contribution is related to the combustion efficiency of woodstoves that can vary from 40-50 % to 15 % depending on combustion efficiency (Bertrand et al., 2017). In fresh emissions, the emission factors (EFs) of these compounds can vary with the type of fuel (hardwood, softwood, or herbaceous types. See e.g., (Schmidl

et al., 2008) the type of fire (open fire, fire places, woodstove, see Fine et al., 2004), or even the sampling setup varying from experimental stack, dilution tunnel, or ambient air (Nussbaumer, 2010).

Furthermore, levoglucosan can decrease by up to 40 % during aging, changing its contribution from 30%, fresh emission, to 10 %, aged emission, to the total organic aerosol mass (Bertrand et al., 2018).

All these limitations put under question the general use of this primary marker for apportionment of aged biomass burning emissions. Nevertheless, levoglucosan is still a unique marker for BB, its use has been demonstrated in multiple source apportionment studies and its analysis provides more benefits than uncertainties to the determination of the BB contribution to PM (Weber et al., 2019).

During the last decade, several campaigns and programs at European, National and Regional levels have used levoglucosan for the identification of the biomass burning contribution to particulate matter. For instance, the intensive measurement period campaigns carried out at the EMEP stations taking place in winter 2017/18 aimed to distinguish among fossil fuel and biomass combustion sources (Fagerli et al., 2020). EMEP has also carried out long time series of levoglucosan data at the Birkenes Observatory (southern Norway) since 2008.

In France, in the CARA program²⁰ (Favez et al., 2021), running since 2008 by Laboratoire Central de Surveillance de la Qualité de l'Air and the regional monitoring networks for the characterisation of PM, has levoglucosan analysed for BB identification (Weber et al., 2019). Similarly, levoglucosan measurement has also been implemented in several federal states in Germany in their regional networks. That is the case for example of Baden-Wuerttemberg, where levoglucosan was measured in 2020 in approximately 60 % of the samples and increasing to daily frequency during winter period.

Moreover, measurements campaigns in Northern countries in Europe, e.g., Finland, are also considering levoglucosan for source apportionment studies and BB identification from regional long-range transport to urban backgrounds (Saarnio et al., 2013).

Specific cases studies are frequently found in bibliography, showing spatial and seasonal variation of levoglucosan concentration in PM in different European regions and cities like Oslo, Rotterdam, Amsterdam, Groningen, Amersfoort, Munich, Augsburg, Barcelona, Girona or Sabadell (Jedynska et al., 2015), Grenoble (Tomaz et al., 2017), Milan (Hakimzadeh et al., 2020), Granada (Titos et al., 2017), Seiffen (Poulain et al., 2011), Alicante, (Galindo et al., 2021);(Elsasser et al., 2012) Tuscany, (Giannoni et al., 2012) Lombardy, (van Drooge and Ballesta, 2009) and Upper Silesia (Marynowski et al., 2020).

Levoglucosan has been also used in describing episodes of wildfires, for instance during the heat wave of 2018 in north-west Europe. Its comparison with Elemental Carbon and inclusion in Positive Matrix Factors (PMF) to apportion Elemental Carbon can allow distinguishing between fossil fuels and biomass combustion sources. These studies comprise intensive measurement period campaigns at EMEP stations during the winter season (i.e. 2017/18) (Fagerli et al., 2020). Cases studies based on macro tracer approach (using levoglucosan) have also been used to evaluate wood burning contribution to PM₁₀ (Galindo et al., 2021).

The use of levoglucosan, as a marker of wood combustion has been generalised in source apportionment studies and source attribution of PM pollution (Simoneit et al., 1999). Some monitoring network programs have included levoglucosan in their data collection for the assessment of local wood combustion pollution issues (Favez et al., 2021) and its monitoring has been also used to identify transboundary biomass burning from forest or agricultural fires (Saarnio et al., 2013). Levoglucosan in urban areas has been also measured and attributed to residential wood burning (Jedynska et al., 2015).

There is an important effort at global level to identify atmospheric mass concentration of levoglucosan. The highest levels of levoglucosan have been reported in East China, Russian Siberia, Rondonia (Brazil), Moscow and western USA mainly due to agriculture burning and forest fires. Being the lowest at the polar regions (Bhattacharai et al., 2019).

Data of levoglucosan measurements has been collected by regional networks or reference laboratories, describing trends of concentrations, spatial variations or local monitoring campaigns during specific periods of interest (Yttri et al., 2020). That is the case of the EMEP station where trends in some fix stations and collected data from intensive campaigns are available for levoglucosan. Reports can be downloaded from their corresponding publication websites. However, there is not a centralised collection of levoglucosan data at national or international level in Europe besides EMEP, like it is the case of other pollutants (Air-Base data).

²⁰ CARA, "Caractérisation chimique des particules" <https://www.lcsqa.org/fr/le-dispositif-cara> (in French)

The collection of systemic or occasional levoglucosan data depends on the purpose of these measurements. Pilot studies or temporary measurement campaigns will lead to occasional data that describe a certain temporal or spatial pollution episode. Trend data of levoglucosan are generally linked to fixed stations aiming to watch temporal variations, long-range or transboundary transport of pollution related to BB, providing a systemic collection of data.

A valid example of levoglucosan data collection is the CARA program of the French operational air quality network (Favez et al., 2021), what foresees three types of levoglucosan datasets:

- Long term monitoring at an urban background site in Grenoble that considers speciation of the daily PM₁₀ filters every three days.
- Periodical sampling campaigns: PMF studies at yearly basis carried out in more than 15 cities in France (Weber et al., 2019).
- Specific campaigns during pollution episodes, i.e., for model validation (Nordic-Structure-Bois, 2015).

The spatial variability of levoglucosan differs from those of other pollutants of anthropic origin like NO_x, PM_{2.5} or EC, as it has been found a high spatial correlation with levels of PAHs in samples, and a low correlation with traffic markers like hopanes (Jedynska et al., 2015). The spatial and temporal variability of levoglucosan data is consequently related to the variability of BB contribution to PM.

Temporal variations of levoglucosan content in PM can be related to seasonal variations (winter/summer) of the overall impact of the residential heating combined with periods of higher atmospheric stability (Hakimzadeh et al., 2020). These variations can be observed at daily or even hourly basis, identifying particular events like local agricultural burning activities or hours of intensive BB emissions (van Drooge and Ballesta, 2009).

Levoglucosan as a unique marker for wood combustion, its analysis has become essential in all source apportionment studies that consider biomass burning and high levels of levoglucosan in urban areas have been attributed to residential wood burning. This is characteristic in American and Europe cities (Fine, Cass, and Simoneit, 2004), (Jedynska et al., 2015) while the major source of levoglucosan in rural areas are attributed to burning of agricultural residues, household firewood combustion and long-range transport (Bhattarai et al., 2019). In forested areas, levoglucosan is an indicator of forest fires (Callegaro et al., 2018) while in marine and remote areas (Arctic, Antarctica, Himalaya) it is linked to long range transport (Kehrwald et al., 2012). Levoglucosan has been analysed also in ice, where it can remain stable and provide an historical record of BB events (Kehrwald et al., 2012). Specific campaigns during large pollution episodes, in which levoglucosan was measured, have also been used for source apportionment model evaluation (Kalogridis et al., 2018) (Yttri et al., 2020).

The ratio between different levoglucosan isomers (mannosan, galactosan) can provide information on the type of source of biomass burning (hardwood, softwood, (Alves et al., 2017), (Louchouart et al., 2009), (Maenhaut et al., 2016). Levoglucosan/galactosan ratio was used to identify leaf burning emissions (Schmidl et al., 2008).

6.5 Towards more systematic monitoring

The measurement of levoglucosan is linked to PM source characterisation. Therefore, its systematic measurement will be easier in those networks in which speciation of PM is routinely performed.

Analytical costs depend on the analytical approach chosen for quantification. Ion chromatography seems the cheapest choice. Nevertheless, other techniques based on High Performance Liquid Chromatography and Mass Spectrometry (HPLC/MS) or GC/MS allow the simultaneous determination of other species that could be of interest for the PM characterisation. Levoglucosan analysis as part of PM speciation can be centralised to reference laboratories that provide the analytical support to national or regional networks, reducing subsequently the overall cost of the analytical implementation.

- Potential contribution to Zero Pollution objectives.

The use of levoglucosan as a tracer of BB represents an important tool for emission regulation at local and regional level, by identifying the contribution of BB to PM composition and its influence on other regulated pollutants associated with particulate matter as PAHs. However, due to the fact that levoglucosan is a marker that provides important but additional information, the systematic determination of levoglucosan makes sense when these data can be combined with additional measurements of PM composition and other correlated data that can provide a complete picture for source apportionment in the area of interest.

Traces of Levoglucosan in particulate matter are related to emissions coming from wood burning. This is of relevance in areas where the wood is typically used for domestic heating (Central and North Europe). In addition, it can be used as indicator of wildfire, agriculture residue burning and forest fire.

The implementation of systematic levoglucosan measurements in selected stations and the organisation of periodical measurement campaigns during appropriated periods of pollution episodes could help inform the control of BB emissions and may supply knowledge and tools for the development and implementation of an effective regulation of BB emissions in Europe, ultimately contributing to the achievement of ZP target for decreasing air pollution impact on human health.

On the same line, its measurement offers a potential to assess health effects of wood smoke separated from the traffic-related air pollution as well as the understanding of the biomass burning impact on climate change.

7 Conclusions

The following table summarises the features of the four pollutants and the biomass burning marker discussed in the report to allow for comparing key aspects of the “emerging” pollutants discussed here at a glance.

	Black Carbon	Ammonia	UFP	Methane	Levoglucosan
Nature	PM component	Gas	PM component	Gas	PM component
Reasons for concern	Health impact	Secondary PM formation Eutrophication Acidification	Health impact	Ozone precursor GHG	Biomass Burning tracer (source apportionment of particulate matter)
Reporting measuring stations in the EU	ACTRIS+EMEP (25) EEA (30-60)	EMEP (43) EEA (<20)	ACTRIS (30)	WDCGG ICOS (38)	EMEP intensive campaigns
Measurement methodology	Light absorption Thermal analysis	Diffusive sampling Filter packs	Condensation particle counters	Gas chromatography + flame ionisation detectors Spectroscopic techniques	Various Chromatography based techniques
Recent key documents	WHO (good practice)	Directive 2016/2284	WHO (good practice)	Methane strategy GHG mitigation policies	
Related zero pollution targets	Reducing PM health impact	Reducing (secondary) PM health impact Reduce threats to ecosystems, water bodies and soils (eutrophication and acidification)	Reducing PM health impact	Reduce air pollution health impact through ozone control Reduce threats to ecosystems (ozone)	Streamline energy and air quality policies
Directions for monitoring expansion	Densely populated areas	Source monitoring Sensitive ecosystems	Densely populated areas	Sources monitoring	Mirroring PM measurements

Table 1: Main features of the four pollutants and the biomass burning marker discussed (Source: JRC)

Regarding particulate matter, even in a scenario where concentrations of PM₁₀ and PM_{2.5}, already covered in the current legislation will continue to decrease, more attention should be focused on the specific threats posed by PM fractions and components suspected to be more directly linked with impacts to human health, namely UFP and BC. In accordance with the concept of WHO good practices, measures specifically tailored for these components would maximise the efficacy of further efforts in reducing the health consequences of particulate matter pollution impacts. Clearly, additional monitoring should be focused on monitoring the situation where most of the population lives and is exposed to air pollution.

Ammonia and methane pose direct threats *per se*, namely water eutrophication and climate warming, respectively. Nevertheless, they also play a relevant role in triggering the formation of secondary pollutants, i.e., ammonium nitrates and sulphates (also relevant components of PM) and ozone. The ambition of reaching very low levels of these pollutants naturally claims for a stricter control of their precursors, included the cited ones and for methane in particular, the Commission has called for keeping methane emissions in the EU Member States under review in an Annex to Directive 2016/2284. For both ammonia and methane enhanced measurement networks should provide more refined information of sources, with a particular attention to sources possibly presently underestimated, such as the urban ones. Newly collected knowledge will be relevant for improving the performance of instruments able to track back the origin of pollutants, such as inverse modelling.

Finally, the recent initiatives in the “fit for 55” package (European Commission, 2021c) have formalised the EU ambition to proceed rapidly in the pathway towards a decarbonised society without impacting on other environmental issues, including air quality. In this context, biomass burning, an energy source expected to play an important role in most energy scenarios, should be monitored for its contribution to air pollution, and levoglucosan measurements provide useful data in this sense.

Considering feasibility, simplicity and cost of the related measurement techniques, two possible priorities emerge. The JRC proposes to focus in the near future on the monitoring of black carbon by multi wavelength absorption technique, and of ammonia by diffusive sampling and subsequent analysis.

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Measure units

µm	micrometre
kt	kilotonnes
nm	nanometer
ppb	parts per billion
ppm	parts per million

Chemical elements and compounds

As	Arsenic
BaP	Benzo(a)pyrene
C ₆ H ₆	Benzene
Cd	Cadmium
CH ₄	Methane
CO	Carbon Monoxide
NH ₃	Ammonia
Ni	Nickel
NO ₂	Nitrogen dioxide
NO _x	Nitrogen Oxides
O ₃	Ozone
PAH	Polycyclic Aromatic Hydrocarbons
Pb	Lead
SO ₂	Sulphur Dioxide
VOCs	Volatile Organic Compounds

List of abbreviations and definitions

AAQD	Ambient Air Quality Directives
ACTRIS	Aerosol, Clouds and Trace Gases Research Infrastructure
AGAGE	Advanced Global Atmospheric Gases Experiment
AOT40	Accumulated exposure Over Threshold of 40 ppb
AQ	Air Quality
AQMN	Air Quality Monitoring Network
AR6	Sixth Assessment Report (IPCC)
ATC	ICOS Atmospheric Thematic Centre
BB	Biomass Burning
BBOA	Biomass Burning Organic Aerosol
BC	Black Carbon
CAL	ICOS Central Analytical Laboratories
CAMS	Copernicus Atmosphere Monitoring Service
CCL	Central Calibration Laboratory

CCN	Cloud Condensation Nuclei
CEN	European Committee for Standardization (Comité Européen de Normalisation)
CLRTAP	Convention on Long-range Transboundary Air Pollution
CPC	Condensation Particle Counters
CRDS	Cavity Ring-Down Spectroscopy
CSIRO	Commonwealth Scientific and Industrial Research Organization
DOAS	Differential Optical Absorption Spectroscopy
eBC	equivalent Black Carbon
EDGAR	Emissions Database for Global Atmospheric Research
EEA	European Environmental Agency
EMEP	European Monitoring and Evaluation Programme
EN	Europäische Norm (European Norm)
EPA	Environmental protection Agency
ERLAP	European Reference Laboratory for Air Pollution
EU	European Union (27 Members)
EURAMET	European Association of National Metrology Institutes
FTIR	Fourier Transform Infrared
GAW	Global Atmosphere Watch Programme
GC/FID	Gas Chromatography equipped with Flame Ionisation Detectors
GCOS	Global Climate Observing System
GHG	Greenhouse Gas
GUAN	GCOS Upper-Air Network
GWP	Global Warming Potential
HPAEC-PAD	High Performance Anion Exchange Chromatography coupled to Pulsed Amperometric Detector
ICOS	Integrated Carbon Observation System
IPCC	Intergovernmental Panel on Climate Change
JRC	Joint Research Centre
LIDAR	Laser Imaging, Detection, and Ranging
MEMO ²	MEthane goes MObile – MEasurements and Modelling
NOAA	National Oceanic and Atmospheric Administration
OAICOS	Off-Axis Integrated Cavity Output Spectroscopy
PM	Particulate Matter
PM ₁₀	Particles smaller than 10 µm in aerodynamic diameter
PM _{2.5}	Particles smaller than 2.5 µm in aerodynamic diameter
PMF	Positive Matrix Factors
PNSD	Particle Number Size Distributions
RfC	Reference Concentration
SCR	Selective Catalytic Reduction
SWD	Staff Working Document

UCI	University of California Irvine
UFP	Ultra Fine Particles
UNECE	United Nations Economic Commission for Europe
US	United States
WDCGG	World Data Centre for Greenhouse Gases
WHO	World Health Organization
WMO	World Meteorological Organization
ZP	Zero Pollution

List of figures

Figure 1 - European ACTRIS and EMEP stations measuring light absorption coefficient (Source: actris.nilu.no)	9
Figure 2 - Global and EU27 methane emissions evolution between 1970 and 2018 (Source: EDGAR)	16
Figure 3 - Network of measuring stations reporting to World Data Centre for Greenhouse Gases (Source: WDCGG)	18
Figure 4 - Levoglucosan chemical structure (Source: textbooks)	20

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