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Establishing an Eionet Group of In Situ Data Experts

Report on how EIONET and EEA can contribute to the urban *in situ* requirements of a future Copernicus anthropogenic CO₂ observing system

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The purpose of the overall work of the Eionet Group of Copernicus In Situ Data Experts is to advise and assist the EEA regarding its cross-cutting coordination of the Copernicus In Situ Component. The composition of the Copernicus Eionet Group of In Situ Data Experts ensures that the following two thematic in situ data domains are embraced to the greatest extent possible: (a) observational data related to meteorology, oceanography, atmospheric chemistry and air quality, and climate; and (b) geospatial and environmental data themes as defined in Annexes I to III of the INSPIRE Directive (except themes 13, 14 and 15).

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EXECUTIVE SUMMARY

To protect the integrity of the Paris Agreement, the European Union is currently elaborating a measurement-based system to independently monitor fossil fuel CO₂ emissions of nation states, large cities and even industrial complexes. The foreseen *Copernicus CO₂ Monitoring and Verification Support (MVS) capacity* constitutes a unique and unprecedented global inverse modelling framework that will rely heavily on a space-based observation component. However, as outlined in the successive reports by the European Commission's CO₂ Monitoring Task Force, *in situ* observations of CO₂ and other co-emitted tracers will also form an essential system component.

In 2019, the CO₂ Monitoring Task Force published its *Green Report* describing the specific needs and high level requirements of the *in situ* component of the Copernicus CO₂ MVS capacity. While the report acknowledges the high measurement standards and solid foundation provided by existing *in situ* observation networks, the document concludes that these networks currently do not meet all the operational requirements for the Copernicus CO₂ MVS capacity. Sustainability issues and insufficient geographical coverage, in particular a lack *in situ* measurement data from urban areas, were highlighted as substantial limitations. The report furthermore *recommended to exploit all existing relevant in situ capabilities and extend them as appropriate.*

The European Environment Information and Observation Network (EIONET), which of course serves, *inter alia*, as a data repository for *in situ* air quality measurements reported by EU member states, represents a relevant *in situ* network due to the CO₂ MVS capacity need for urban measurement data on co-emitted species. Furthermore, looking ahead to a potential future scenario when big European cities start to implement greenhouse gas (GHG) measurements for local emissions monitoring, additional opportunities beyond air quality measurements may arise for EIONET. This report of the *EIONET Group of Copernicus in situ Data Experts* examines how EIONET in its current form could be *exploited*, and how it could be *extended* to help contribute to needed urban *in situ* observation data to the Copernicus CO₂ MVS capacity. This report furthermore examines *in situ* contribution opportunities for the European Environment Agency (EEA) given the EEA's responsibility for developing EIONET and for coordinating *in situ* activities under the Copernicus programme.

Measurements of urban air quality in the EU under the Air Quality Directive (AQD; 2008/50/EC, amended by Dir. 2015/1480/EU) and the 4th Daughter Directive (4DD; 2004/107/EC) are regularly uploaded on the central data repository of the EEA. Due to the fact that these measurements and the subsequent sharing of the data are embedded in EU legislation, it would seem that a potential stream of this data into the CO₂ MVS capacity could be sustained in the long-term. This report therefore recommends that the architects of the CO₂ MVS capacity further define what is needed in terms of *in situ* tracer measurements and evaluate the extent to which the legislated EU MS measurements reported to EIONET are sufficient. Such an evaluation should take place in the near-term considering the European Commission's plans to evaluate current air quality legislation. As stated in the EU's Green Deal, the Commission will propose to strengthen provisions on monitoring. Considering this prospect, it would therefore make sense to establish/increase dialogue between the CO₂ Monitoring Task Force and the air quality community. Such dialogue could help identify improvements in air pollution measurements that would benefit both local air quality monitoring and the CO₂ MVS capacity and formulate proposals for subsequent legislative amendments.

Regarding *in situ* urban GHG measurements, the initial road ahead for EIONET is more difficult to navigate. Nonetheless, this report argues that a paradigm shift with respect to urban emissions monitoring is gathering momentum. Given both the importance of urban climate change mitigation on the one hand and the uncertainties in city-scale emissions inventories on the other, interest has been growing within the research community regarding how *in situ* measurements could be integrated into urban GHG emissions monitoring systems. Moreover, it now appears that this interest has found its way into the policy arena.

This report provides a technical review of CO₂ and CH₄ emissions monitoring methods based on surface mixing ratio measurements, total column mixing ratio measurements and flux measurements. The review demonstrated that all these measurements would fulfil respective *in situ* requirements of the Copernicus CO₂ MVS capacity, contributing to the validation of space observations in and around cities and/or the system's city-scale emissions estimates. The review furthermore elaborated on the benefits to climate change mitigation monitoring in the respective cities and how these methods could be implemented to monitor local emissions.

If these urban GHG measurements do indeed make the transition from science to policy, this may constitute a welcome development for the Copernicus CO₂ MVS capacity. Not only would target areas be increasingly covered by such *in situ* measurements, the vested interest cities have in the data may ensure the sustainability of the measurement operations. A further opportunity may therefore arise for EIONET, especially if the cities themselves plan to directly implement such measurements in a similar way as they implement their air quality monitoring programmes.

This report nonetheless argues that as emissions monitoring methods based on *in situ* GHG measurements are still maturing, EEA should consider how it could help expand and sustain urban *in situ* GHG measurements within the research networks, in particular ICOS. Considering the organisational structure of the research and development phase of the Copernicus CO₂ MVS capacity so far, it is likely that ICOS will ultimately be responsible for coordinating the system's *in situ* component and thus the urban sub-component. There is however, scope for EEA to support ICOS on this front, given that EEA's mandate to coordinate *in situ* activities under the Copernicus programme includes an explicit role to *manage partnerships with data providers*. Specifically, EEA could consider facilitating dialogue between ICOS and municipal climate change mitigation planners in large European cities to support ICOS in implementing its strategy on urban observations. Such cooperation could enable identification and utilisation of synergies between the expansion strategy of ICOS needed to fulfil the *in situ* requirements of CO₂ MVS capacity and the plans/wishes of cities to improve local GHG emissions monitoring. A vested interest of cities in directly using the GHG measurement infrastructure for local emissions monitoring may for example facilitate mobilisation of additional funding needed to expand and sustain the urban *in situ* observations, as well as open opportunities for co-location within existing sensor networks that are operated within the local air quality monitoring programme. It would therefore be timely for EEA to consider how it could help facilitate this dialogue through EIONET national reference centres on e.g. air quality and climate change mitigation and/or through contacts within fora such as the European Covenant of Mayors.

1. INTRODUCTION

The 2018 Intergovernmental Panel on Climate Change (IPCC) special report stresses that only by achieving zero global net greenhouse gas (GHG) emissions by ca. 2050 can global warming at the end of the century be restricted to 1.5 °C above pre-industrial levels (IPCC, 2018). Limiting global warming to 2°C would require reaching net zero emissions by 2070. As such, regardless of the scenario, huge and rapid GHG emissions reductions are required to meet the Paris Agreement goal.

Given what is at stake (IPCC, 2014), and the short-time frame for action, the world cannot afford to lose sight of progress on mitigating climate change. As such, systems that monitor GHG emissions and removals form a fundamental element of the mitigation effort. As outlined in the Paris Agreement (UNFCCC, 2015) and the 2018 decision on the transparency framework (UNFCCC, 2018), reporting of national emissions inventories will continue to constitute the information basis for monitoring progress towards mitigation goals. However, in contrast to previous protocols under the United Nations Framework Convention on Climate Change (UNFCCC), there is scope under the Paris Agreement for space-based measurements of GHGs to provide independent verification of the national inventory estimates as well as additional data to be directly utilised within the global stocktake under article 14 (Aganaba-Jeanty and Huggins, 2019).

Recognising this potential opportunity and the need to protect the credibility of this and future climate agreements, a 2015 European Commission report concluded that it would be *relevant and timely* to develop a measurement-based system to monitor fossil fuel carbon dioxide (CO₂) emissions under the EU's Copernicus programme (Ciais et al., 2015). The foreseen *Copernicus CO₂ Monitoring and Verification Support (MVS) capacity* constitutes a unique and unprecedented global inverse modelling framework and would of course rely heavily on a space-based observation component. However, as outlined in the successive reports elaborating this vision (Ciais et al., 2015; Pinty et al., 2017, 2019), *in situ* observations of CO₂ and other tracers will also form an essential system component.

In 2019, the European Commission CO₂ Monitoring Task Force published the green report describing the specific needs and high level requirements of the *in situ* component of the Copernicus CO₂ MVS capacity (Pinty et al., 2019). While the report acknowledges the high measurement standards and solid foundation provided by existing *in situ* observation networks, the authors nonetheless concluded that these networks currently do not meet all the operational requirements for the Copernicus CO₂ MVS capacity. The report furthermore highlights the lack of *in situ* measurement data from urban areas and other emissions hot spots as a substantial limitation.

Although the European Environment Information and Observation Network (EIONET) is not explicitly mentioned in the green report, the report does refer to the need for measurement data of tracer gases from air quality networks. Furthermore, looking ahead to a future when big European cities may start to implement GHG measurements for local emissions monitoring, opportunities to contribute beyond urban air quality measurements could arise for both EIONET as a network, and the European Environment Agency (EEA) that is responsible for *inter alia* developing EIONET and coordinating *in situ* activities under the Copernicus programme. This study of the *EIONET Group of Copernicus in situ Data Experts* provides a preliminary examination of how EIONET and EEA could contribute the Copernicus CO₂ MVS

capacity in terms of *in situ* urban observations. After providing an overview of the Copernicus CO₂ MVS capacity and the needs and requirements for the *in situ* component (chapter 2), the main chapter of this report (chapter 3) describes how EIONET and EEA could contribute to the system in terms of *in situ* observation data from urban areas. Specifically, the chapter provides an account of what EIONET in its current form can provide, and how a contribution from EIONET and EEA could be enhanced if cities begin to adopt measurement-based systems for routine monitoring of GHG emissions. The report then draws to a close with conclusions and perspectives in chapter 4.

2. COPERNICUS CO₂ MONITORING AND VERIFICATION SUPPORT CAPACITY AND THE ROLE OF *IN SITU* MEASUREMENTS

In 2015, in anticipation of the 21st session of the Conference of the Parties (COP-21) to the UNFCCC, the European Commission set up and tasked an expert group *to assess the need and opportunity for an independent European space-borne observation capacity for CO₂ to monitor and to verify the compliance of parties to international climate agreements*. Later that year in October, the expert group published its final report, which concluded that such a capacity would be indeed relevant and timely, and should moreover be developed within the EU's earth observation programme, Copernicus (Ciais et al., 2015). The rationale behind these recommendations was the risk that the anticipated international climate agreement could be undermined if the monitoring of anthropogenic CO₂ emissions relied solely on the reported emissions inventories of the respective parties. The report outlined the varying quality in inventories between countries and emphasised the increasing uncertainty in global CO₂ emissions estimates due to growing contribution of emissions from non-Annex I countries, whose emissions inventories are generally more uncertain. The report therefore provided a review of how emissions estimates can be constrained using top-down inversions based on atmospheric observations of CO₂ and outlined the current and future capabilities of space-based greenhouse gas measurements. Based on this review, the report proposed a notional system design for a European capacity to independently monitor fossil fuel CO₂ emissions and set out a preliminary road map for its development.

In response to the report, the European Commission subsequently established a *CO₂ Monitoring Task Force* to build upon the vision set out in the 2015 report and to elaborate on the system elements required to set up and implement an operational *Copernicus CO₂ Monitoring and Verification Support (MVS)* capacity. Analysed against climate policy needs, the first report of the Task Force outlined what the CO₂ MVS capacity should be capable of and what system architecture and constituent elements would be required to enable these capabilities (Pinty et al., 2017). As described in the 2017 report, the CO₂ MVS capacity should be capable of assessing national emissions and 5 year changes in these emissions to assist the global stocktake process of the Paris Agreement. Significantly, the CO₂ MVS capacity should furthermore be capable of detecting and monitoring hot spot emissions (power/industrial plants, megacities) and assessing changes in these emissions against local reduction targets. To deliver these services, an overall system design was proposed, whereby prior information (emissions inventories, model parameters, and economic statistics) and atmospheric observations (meteorological variables, CO₂ concentrations and other auxiliary measurements) feed into an integration core component containing the atmospheric transport and chemistry models and biogeochemical models. It is in this component, where model-, observation- and prior data are assimilated (minimisation of predefined cost functions) and

inverse estimations of hot-spot and national fossil fuel emissions are generated. Finally, the report also points out the role of data access, archiving and distribution functions within the system.

Given that the proposed MVS system is unprecedented, it is clear that all system elements will require further elaboration and subsequent development if the system is to deliver the desired emissions estimates at the required accuracy and temporal and spatial resolutions. Looking ahead to prototype implementation, a number of research and development initiatives linked to the *CO₂ Monitoring Task Force* (the CO₂ Human Emissions (CHE)¹ and VERIFY² projects) have been making progress on various aspects on the foreseen system. Indeed, it was announced at the 2019 joint CHE-VERIFY General Assembly that pending a follow-up project the development is *on track for an early prototype with limited capabilities by 2021, a full prototype by 2023, and an operational system by 2026*³. Of course, due to the unprecedented global disruptions caused by the COVID-19 pandemic, caution should be taken when interpreting the future time frame for the CO₂ MVS capacity's subsequent further development and eventual operation. Nonetheless, in June 2020 an online Copernicus CO₂ workshop⁴ was held whereby representatives of the European Commission and the institutions involved in developing the CO₂ MVS capacity (European Centre for Medium-Range Weather Forecasts; ECMWF, European Space Agency, ESA; and European Organisation for the Exploitation of Meteorological Satellites, EUMETSAT) gave perspectives on the system's development. At this workshop, ECMWF announced that the CoCO₂ project it will lead to develop the prototype system is to go ahead between 2021 and 2023. Furthermore, with respect to the dedicated satellite mission, CO₂M, ESA announced its expectations that preliminary satellite designs be in place by mid-2021 and that two of the three satellites be ready for launch by mid-2025.

A key focus is of course the observation element of the CO₂ MVS capacity, given that it is against these atmospheric measurements that the emissions estimates will be constrained. As set out in the 2015 and 2017 reports, satellite CO₂ observations will constitute an essential stream of observation data into the CO₂ MVS capacity. Nonetheless, both reports highlighted the importance of *in situ* observations of CO₂ and other tracers.

Recently, the CO₂ Monitoring Task Force published a subsequent report on the CO₂ MVS capacity that focused specifically on the needs and high level requirements for *in situ* measurements (Pinty et al., 2019). High-quality and sustained streams of *in situ* measurement data will be essential for the Copernicus CO₂ MVS capacity, serving a number of purposes:

- Validation of the satellite observations as well as the temporal variability of the prior emissions;
- Direct utilization to inversely model fossil fuel emissions and natural fluxes or to calibrate processed-based parameters and/or the integration model; and
- Validation of the outputs i.e. emissions estimates from the CO₂ MVS capacity

A number of *in situ* networks currently collect and archive various, high-quality and relevant atmospheric measurements e.g. point concentrations-, isotopes-, and turbulent fluxes of CO₂ and other GHGs (ICOS); measurements of total atmospheric column CO₂, methane (CH₄), and

¹ <https://www.che-project.eu/>

² <http://verify.lsce.ipsl.fr/>

³ <https://www.che-project.eu/news/successful-joint-che-verify-general-assembly>

⁴ <https://www.copernicus.eu/en/events/events/online-monitoring-anthropogenic-co2-emissions-copernicus>

carbon monoxide (CO) concentrations (TCCON, and COCCON); balloon-based measurement of vertical CO₂, CH₄, and CO profiles (AirCore); and measurements of aerosol optical depth (AeroNet). Indeed the latest Task Force report acknowledges the high measurement standards and potential utility of these networks. However, the Task Force concluded that these networks currently do not meet all the operational requirements for the CO₂ MVS capacity. The analysis revealed critical limitations in terms of both the current geographical coverage and sustainability of the measurements. Given that some of these networks were originally set up to focus on GHG fluxes from the terrestrial biosphere, priority targets of the CO₂ MVS capacity i.e. areas in and round urban centres and large industrial facilities are critically under-sampled. Furthermore, as sites and stations within these research networks are supported by respective national research funding sources, long-term operation of the individual measurement facilities cannot be ensured.

The report therefore recommends that *in situ* measurement networks in Europe be extended to increase coverage of urban and industrial areas. Given the pressing need to validate the concentrations (in particular the upwind-downwind concentration gradients) observed from space, the need for *in situ* total column CO₂ and CH₄ measurements in and around large cities and industrial complexes is emphasised. In terms of separating the natural and fossil fuel emissions of CO₂ the report furthermore recommends the implementation of radiocarbon (¹⁴C) measurements and/or co-location of low cost CO₂ sensors with air quality stations measuring co-emitted species from fossil fuels such as CO and nitrogen oxides (NO_x).

The 2019 report does not explicitly mention EIONET; however, the report does state that it is *recommended to exploit all existing relevant in situ capabilities and extend them as appropriate*. The report furthermore recommends *taking stock of existing and future air quality networks and infrastructures and, where possible, collocating these with the newly planned stations*. Finally, the report makes multiple references to the need for coordinated governance of the *in situ* element to ensure sustained streams of high quality *in situ* measurement data into the CO₂ MVS capacity. It is therefore timely and relevant to begin examining the role that EIONET, as a central data repository for mandatory reporting of *in situ* atmospheric measurement data in Europe, could play in the CO₂ MVS capacity. It is furthermore appropriate to consider a coordinating role that could be played by EEA. In addition to being responsible for developing EIONET and coordinating the network's activities with representatives of the participating countries, EEA has been entrusted by European Commission with the coordination of the *in situ* component of the Copernicus programme⁵.

3. THE POTENTIAL ROLE OF URBAN EIONET *IN SITU* MEASUREMENT DATA IN THE COPERNICUS CO₂ MVS CAPACITY

The European Environment Information and Observation Network (EIONET)⁶ is a formal partnership network of 38 member and cooperating countries of the EEA. Responsibility for developing the network lies with the EEA, while activities of the network are coordinated by the EEA in collaboration with representatives, so called National Focal Points (NFPs), of the member- and cooperating countries. Unlike research networks, the exchange and sharing of environmental data is embedded in EU legislation (EU member states) or high-level agreements between the EU and respective non-EU EIONET member and cooperating

⁵ <https://insitu.copernicus.eu/about>

⁶ <https://www.eionet.europa.eu/>

countries. While various EIONET data could potentially feed into the CO₂ MVS capacity (e.g. the official national GHG emissions inventories as data streams into the prior information core element), the focus of this study here is the *in situ* atmospheric measurement data which EIONET could provide from urban areas.

As mentioned in the previous chapter, the most recent report of the CO₂ Task Force states that it is *recommended to exploit all existing relevant in situ capabilities and extend them as appropriate*. Therefore this chapter is structured to describe how EIONET in its current form could be *exploited* and how it could be *extended*. Chapter 3.1 gives an overview of the most obvious potential *in situ* data contribution to the CO₂ MVS System - the *in situ* urban air quality measurements which are reported to EIONET as per EU air quality legislation. Chapter 3.2 provides an examination of potential opportunities for EIONET in terms of *in situ* GHG (particularly CO₂ and CH₄) observation data from European cities. Finally, chapter 3.3 concludes with a preliminary roadmap identifying how a contribution of EIONET to the *in situ* component of the Copernicus CO₂ MVS system could be facilitated. This final subchapter describes not only the opportunities for facilitating potential urban *in situ* data streams from EIONET, but also how EEA and its national contacts within EIONET may contribute to the general expansion and sustainability of urban *in situ* infrastructure required for the CO₂ MVS capacity.

3.1. Exploiting EIONET – EIONET as a provider of *in situ* measurements of air pollutants

EIONET represents a relevant network of *in situ* measurement data due to the need for urban measurement data on co-emitted species. EIONET of course serves as a data repository for *in situ* measurements of air quality and could therefore function as a source of *in situ* observational data on relevant co-emitted air pollutant tracers into the CO₂ MVS capacity. It is also important to point out that if utilised, EIONET would provide a sustainable stream of these *in situ* observations given that the measurement and reporting of this data are embedded in EU legislation, which in turn has been transposed into the national legislation of the EU member states (MS). Furthermore, the European Commission, the EEA, and the European Parliament engage external institutions to review the national implementation of European air quality legislation (e.g. Nagl et al., 2016, 2018, 2019).

Air quality assessment in EU MS follows the provisions of the Air Quality Directive (AQD; 2008/50/EC (European Union, 2008), amended by Dir. 2015/1480/EU (European Union, 2015)) and the 4th Daughter Directive (4DD; 2004/107/EC (European Union, 2005)). These directives cover the pollutants sulphur dioxide (SO₂), nitrogen dioxide (NO₂), nitrogen oxides (NO_x), PM₁₀⁷, PM_{2.5}⁸, Ozone (O₃), carbon monoxide (CO), benzene, and the heavy metals lead (Pb), cadmium (Cd), arsenic (As), and nickel (Ni) and benzo(a)pyrene (B(a)P) in PM₁₀. There are further specific measurement obligations for ozone precursors, PM_{2.5} compounds, additional polycyclic aromatic hydrocarbon (PAHs) species and deposition of heavy metals and PAHs.

⁷ 'PM₁₀' shall mean particulate matter (PM) which passes through a size-selective inlet as defined in the reference method for the sampling and measurement of PM₁₀, EN 12341, with a 50 % efficiency cut-off at 10 μm aerodynamic diameter.

⁸ 'PM_{2.5}' shall mean particulate matter (PM) which passes through a size-selective inlet as defined in the reference method for the sampling and measurement of PM_{2.5}, EN 14907, with a 50 % efficiency cut-off at 2,5 μm aerodynamic diameter.

The primary method for air quality assessment is measurement. Additional assessment methods are modelling and objective estimation, which may supplement monitoring, and which may replace monitoring in zones with low pollution levels (see below).

Requirements for minimum numbers of monitoring stations per zone and basic requirements for their spatial distribution are laid down in Annex V (all pollutants but ozone) and Annex IX (ozone) of the AQD and Annex III of the 4DD). The minimum number of monitoring sites depends on

- The population of the zone
- The pollution level in relation to the “assessment thresholds”.

The number of monitoring sites may be reduced and assessment amended by modelling or indicative measurements in zones where the concentration is between the lower and the upper assessment threshold, and replaced by modelling or objective estimation techniques in zones where the pollution level is below the lower assessment threshold.

Assessment thresholds are laid down in relation to limit values and to the uncertainty of the “low quality” assessment techniques indicative measurement, modelling, and objective estimation, and roughly correspond to about 60 % and 40 %, respectively, of the limit value.

For NO₂, benzene, CO, PM₁₀ and PM_{2.5}, the number of traffic related monitoring sites and the number of urban background monitoring sites shall not differ by more than a factor of 2. The ratio of number of PM₁₀ and PM_{2.5} monitoring sites shall be within 0.5 and 2.

Annex III (all pollutants but ozone) and Annex VIII (ozone) of the AQD and Annex III of the 4DD provide siting criteria for monitoring stations.

Measurement is targeted at two primary objectives:

- Monitoring the maximum concentration the population in a zone is exposed to, with respect to the averaging time of the respective limit or target values, and giving a minimum spatial extent (e.g. 250 m street length).
- Monitoring the exposition of the general population.

Due to the aforementioned population and exposure criteria, EU MS are obliged to increase their sampling effort in their cities. In many instances, more stations are operated in the larger cities than prescribed in the legislation (Table 1).

Table 1: Examples for NO₂ monitoring sites in European cities: Minimum required number according to AQD Annex V, actual number of monitoring sites operating in 2017 as documented in Nagl et al. 2019.

City	Annex V	Actual number in 2017
Berlin	7	17
Paris	10	37
Rome	7	15
Vienna	5	16
Warsaw	5	5

Beyond prescriptions on number of sampling sites, the legislation also obliges the EU MS to observe certain specific siting criteria and to employ reference measurement methods. Detailed local siting criteria provide e.g. provisions for free air flow and the distance from

obstacles, the maximum distance from kerb (10 m) for traffic-orientated stations, and the inlet height (1.5 – 8 m). Reference methods are laid down in Annex VI of the AQD and Annex V of the 4DD. The MS have to apply either the reference method or another method for which the equivalence to the reference method has been proven (criteria for equivalence testing are given).

Table 2: Reference methods according to AQD Annex VI.

Pollutant	Standard	Reference method
SO ₂	EN 14212:2005	Ultraviolet fluorescence
NO ₂ , NO _x	EN 14211:2005	Chemiluminescence
PM ₁₀	EN 12341:1999	Gravimetry
PM _{2,5}	EN 14907:2005	Gravimetry
CO	EN 14626:2005	Non-dispersive infrared spectroscopy
O ₃	EN 14625:2005	Ultraviolet photometry

For all pollutants other than PM₁₀ and PM_{2,5}, almost all measurement sites in the EU are operated by the reference method. In the case of PM₁₀ and PM_{2,5} the reference method is a manual method which produces daily average concentrations with several weeks delay, whereas equivalent monitors produce hourly (or half-hourly) concentrations in real-time (i.e. continuously). Therefore, in many cases equivalent continuous monitors are employed so that ambient concentrations of PM can be monitored in near-real-time by city planners and the public. In Austria almost all PM sites are operated with equivalent PM monitors, with ca. 25 % equipped additionally with the reference instruments.

Data quality objectives are given in Annex I of the AQD and Annex IV of the 4DD. The data quality objectives specify minimum time coverage (usually the calendar year), minimum data capture (90 % for most pollutants) and requirements for the measurement uncertainty. These depend on the pollutant and are stricter for fixed measurements than for indicative measurements, modelling and objective estimation.

The administrative organisation of air quality assessment in EU MSs roughly depends on the country size. In large MS⁹ (but also in some smaller ones like Austria), air quality monitoring is the task of regional/provincial/state authorities. In such cases, particular tasks e.g. regional background monitoring, or specific measurements like ozone precursors or PM compounds may still be performed by authorities at the national level. In contrast, smaller MS typically operate one national monitoring network. Finally, in some MS, municipal or industrial monitoring networks are included in the national air quality assessment system.

Despite potentially decentralised structures in terms of the measurement network, quality assurance and international reporting are the tasks of national authorities or legally assigned scientific/expert institutions. Reporting provisions for AQ data to the EEA are laid down in the Commission Implementing Decision 2011/850/EU (European Union, 2011). The reporting obligations are listed in Annex I of the Decision as datasets B to K and cover:

- Meta-information for AQ assessment (datasets B, C, D):
 - Zones (dataset B)
 - Assessment regimes (dataset C)

⁹ The major exception was UK, which now is no longer an EU MS.

- Meta-data for monitoring stations (dataset D), e.g.:
 - Geographical co-ordinates, elevation,
 - Measurement method and equipment type
 - Sampling time and interval
 - Analytical methods,
 - Modelling and objective estimation techniques
 - Classification in relation to predominant emissions
 - Area classification
 - In case of traffic stations: distance from kerb, traffic volume, emissions
 - In case of industrial stations: distance from plant, emissions
 - Details monitoring network operator.
- Near-real-time data (to be submitted hourly) (dataset E2): Covers all pollutants with continuous measurements, i.e. SO₂, NO₂, NO_x, Ozone, and CO; and PM₁₀, PM_{2,5}, and Benzene in case of continuous measurement.
- Validated data for all pollutants regulated in the AQD and the 4DD (to be submitted annually) (dataset E1):
 - Hourly time series for pollutants with hourly (i.e. continuous) measurements (SO₂, NO₂, NO_x, Ozone, and CO; and PM₁₀, PM_{2,5}, and Benzene in case of continuous measurement);
 - Daily time series for pollutants with daily sampling (PM₁₀, PM_{2,5}, PM compounds, VOCs/Benzene, and heavy metals);
 - Time series of variable temporal resolution for pollutants with sampling periods of weeks or months (Benzene, concentrations of heavy metals and PAHs), deposition of heavy metals and PAHs).
- Attainment information (dataset G) for all pollutants for which limit values and target values are laid down.
- Information on air quality management (datasets H to K)

Reporting of the above data according to Dec. 2011/850/EU is obligatory for EU MSs, while most EEA members and some cooperating countries also follow these reporting provisions (albeit with different coverage of pollutants and data-sets). The data have to be uploaded on EEA's central data repository¹⁰ using a consistent xml format specified by the EEA¹¹.

After receiving validated meta- and assessment data (deadline 30th September for the previous year's data for datasets B to E and G; 31st December for datasets H to K), EEA performs central data checks for completeness, outliers and plausibility, comparing values with previous years data. Feedback¹² and, if necessary, completion or correction are subsequently

¹⁰ [https://aqportal.discomap.eea.europa.eu/toolbox-for-e-reporting/repository/#All official deliveries to date for any country](https://aqportal.discomap.eea.europa.eu/toolbox-for-e-reporting/repository/#All%20official%20deliveries%20to%20date%20for%20any%20country)

¹¹ See EEA user guide: https://www.eionet.europa.eu/aqportal/doc/UserGuide2_AQD_XML_v3.4.1.pdf

¹² Feedback is summarised on <https://aqportal.discomap.eea.europa.eu/products/feedback-on-submissions/>.

requested from MS. However, it is important to point out that there is no regular check (neither by European Commission nor EEA) on the implementation of QA (quality assurance)/QC (quality control) procedures for the measurements or analyses.

The EEA uses the verified, reported air quality data to monitor air quality at the European scale, providing the European Commission with information on attainment/exceedance of limit or target values as well as producing freely accessible European Air Quality reports¹³ and country fact-sheets¹⁴. The EEA also provides public access to the reported measurement data¹⁵.

Considering the need of the CO₂ MVS capacity for urban *in situ* measurement data on co-emitted air pollutants, this report argues that EIONET should be seriously considered as a potential source of this data. As the air quality data viewers^{16,17} demonstrate, not only are the required *in situ* data being measured and shared within EIONET, but also a formal structure is in place which could potentially ensure that the stream of data is sustained. As such, the continued elaboration of the CO₂ MVS capacity should define what exactly is needed in terms of *in situ* tracer measurements and evaluate to what extent EIONET, in its current form (i.e. the current reference measurement methods, siting criteria etc.), meets these particular system requirements.

3.2. Extending EIONET – opportunities to coordinate *in situ* GHG measurements in European cities

Unlike the air quality measurements described previously, there is no European legislation currently obliging/formally encouraging the EIONET countries to measure and report GHG concentrations, isotopes or fluxes within their national territories. There is therefore no current mandate for EIONET to serve as repository for such data. Until now measurements of CO₂ and other GHGs have been conducted within the scientific sphere, with data exchanged and stored via international research networks such as AirCore¹⁸, the COllaborative Carbon Column Observing Network (COCCON, (Frey et al., 2019a)), FLUXNET¹⁹, the Global Atmosphere Watch Programme (GAW)²⁰, the Integrated Carbon Observation System (ICOS)²¹, the Network for the Detection of Atmospheric Composition Change (NDACC)²² and the Total Carbon Column Observing Network (TCCON)²³. However, recent developments indicate that *in situ* measurements of GHGs in cities may be about to enter the policy sphere at national and municipal levels. Under such a scenario, opportunities for EIONET with respect to urban GHG observations may arise.

¹³ https://www.eea.europa.eu/publications#c7=en&c11=5&c14=air-quality-reports&c12=&b_start=0&c5=air

¹⁴ <https://www.eea.europa.eu/themes/air/country-fact-sheets/2019-country-fact-sheets>

¹⁵ <https://www.eea.europa.eu/themes/air/explore-air-pollution-data>

¹⁶ <https://www.eea.europa.eu/data-and-maps/dashboards/air-quality-statistics>

¹⁷ <https://www.eea.europa.eu/data-and-maps/explore-interactive-maps/up-to-date-air-quality-data>

¹⁸ <https://www.esrl.noaa.gov/gmd/ccgg/aircore/>

¹⁹ <https://fluxnet.fluxdata.org/>

²⁰ <https://public.wmo.int/en/programmes/global-atmosphere-watch-programme>

²¹ <https://www.icos-cp.eu/>

²² <https://www.ndsc.ncep.noaa.gov/>

²³ <https://tccodata.org/>

3.2.1. The emerging need for measurement-based monitoring of city GHG emissions

As concentrated hot spots of human activity, cities have a huge impact on the climate system. Although urban areas constitute only ca. 3 % of the total land area (Liu et al., 2014), 55% of the global population currently resides in urban areas (United Nations, 2019). According to central estimates from the latest IPCC assessment report, urban areas are responsible for 76% of CO₂ emissions from global final energy consumption and 43 % of CO₂ emissions from global primary energy consumption (Seto et al., 2014). In terms of aggregated GHG emissions, Marcotullio et al. (2013) estimated that emissions of CO₂, nitrous oxide (N₂O), CH₄ and sulphur hexafluoride (SF₆) coming from within urban areas contribute 36.8 % of the global aggregate total expressed in CO₂-equivalent (CO₂e). When factoring in additional emissions from energy generated outside of urban areas according to final energy consumption, the urban contribution was estimated at 48.6 %.

Increasing recognition of the impact of cities on the climate system has seen a partial yet significant reconfiguration of the international policy response to climate change (Rosenzweig et al., 2010; Watts, 2017). Where the focus of climate change mitigation has traditionally been at the nation state level, recent years have seen cities become increasingly engaged in international climate policy with many cities signed up to collective transnational frameworks for reducing GHG emissions. At the time of writing, there are 96 megacities affiliated to the C40 network²⁴, over 1750 local and regional governments are members of the *ICLEI – Local Governments for Sustainability* network²⁵, and more than 10,000 cities are signed up to the *Global Covenant of Mayors for Climate and Energy*²⁶ (GCoM). According to the recent climate targets submitted by the respective municipalities, the GCoM signatories are aiming to deliver combined emissions reductions (savings against a business-as-usual scenario) of 2.3 Gt CO₂e in 2030 and 4.2 Gt CO₂e in 2050 (Global Covenant of Mayors for Climate & Energy, 2019). In Europe, a recent meta-analysis found that 586 of the 885 *core cities*²⁷ across the former EU-28 have prepared formal local climate change mitigation plans (Reckien et al., 2018). The study reports that 174 cities from Denmark, France, Slovakia and the UK have prepared these plans due to national legislation that obliges the respective municipalities to compile and report such local climate plans. The local mitigation plans of the remaining 412 cities were developed either autonomously and/or under the auspices of transnational urban climate networks such as those listed above. The review thus illustrates that there is both a top-down international and national desire, as well as a bottom-up municipal aspiration to realise climate change mitigation at the city-level.

Successful collective- and individual implementation of urban climate change mitigation will of course depend on systems that quantify and monitor how local activity impacts climate (Wright et al., 2011). Despite compelling arguments for a consumption-based, footprint approach (i.e. estimates which include all emissions associated with energy and material consumption including both upstream and downstream processes occurring outside city boundaries), there seems to be prevailing top-down and bottom-up preference for activity-based emissions inventories which estimate those emissions occurring within a city's borders (scope 1) as well

²⁴ <https://www.c40.org/>

²⁵ <https://www.globalcovenantofmayors.org/>

²⁶ <https://www.iclei.org/>

²⁷ Core cities refer to those cities listed in the Eurostat urban audit database:
<https://ec.europa.eu/eurostat/web/cities/data/database>

as a proportional share of emissions from electricity and heat energy which is consumed within the city yet generated outside of the city (scope 2). For example, while the GCoM advocate flexible monitoring approach *to suit differentiated local circumstances and needs* (Global Covenant of Mayors for Climate & Energy, 2018), the European Covenant of Mayors (originally an independent Covenant of Mayors initiative which helped establish the GCoM) explicit recommends the activity-based inventory approach which includes scope 2 emissions (Bertoldi, 2018). A recent global synthesis of self-reported city CO₂ emissions data found that 343 cities employ scope-1 GHG emissions inventories, with 187 cities providing both scope 1 and scope 2 estimates (Nangini et al., 2019). The preference for such inventory approaches is perhaps unsurprising given that for decades such systems have formed the information backbone of climate policy at both national and international levels. However, recent developments in both the scientific and political spheres suggest that a paradigm shift with respect to emissions monitoring is well underway. The hitherto reliance on inventories for quantifying and reporting national GHG emissions is being challenged, with scholars calling for formal inclusion of remote- and *in situ* atmospheric GHG observations to constrain national emissions estimates (Aganaba-Jeanty and Huggins, 2019; Leip et al., 2018). Indeed policy-makers at international and national levels are acting upon these recommendations. While the European Commission and its CO₂ Monitoring Task Force continue to elaborate the Copernicus CO₂ MVS capacity vision (Ciais et al., 2015), nation states are looking to develop/expand national capacities for integrating *in situ* GHG measurements into their respective emissions monitoring systems. The most recent *National Inventory Reports* of Australia (Australian Government, 2020), Switzerland (FOEN, 2020) and UK (Ricardo-AEA Ltd., 2020) document the current use of measurement-based inversions to validate national emissions estimates of fluorinated gases. Furthermore, research projects in Germany (Kaiser-Weiss et al., 2019), Switzerland (Müller et al., 2019), New Zealand (Mikaloff Fletcher et al., 2019) and UK (Palmer et al., 2018) are currently laying the experimental foundations for respective measurement-based systems to independently estimate national emissions of CO₂ and other GHGs. In addition to concrete developments in terms of national, regional and global emissions monitoring, evidence suggests that a tentative paradigm shift at the municipal level is also be underway.

Despite the potentially more restricted budgets available to municipalities compared to nation states, the need for measurement verification at the city level is arguably more acute. Emissions inventories at the city scale are considered to be significantly more uncertain than national inventories. The issues causing biases in national inventories i.e. poorly characterized emission factors, potentially unaccounted sources and sparse and unreliable activity data (Leip et al., 2018) are typically amplified at the municipal level. Despite the emergence of guidance (Bertoldi, 2018) and tools (e.g. the Global Protocol for Community-Scale Greenhouse Gas Emission Inventories (WRI, 2014)) seeking to standardise the compilation of city inventories, it seems that for most cities the required activity statistics are not generated at the local level (Wu et al., 2016). Therefore, in most cases official city emissions are estimated by either compiling inventories using local activity data which have been derived from a proxy-based disaggregation of national statistics or by applying proxy-based disaggregation of the respective national emissions inventories. Such a proxy-based disaggregation, be it a desegregation of national input statistics or national emissions, thus introduces an additional and significant node along which errors can propagate (Christen, 2014; Gately and Hutyra, 2017). Furthermore, in contrast to national emissions inventories reported under the UNFCCC or the EU's Monitoring Mechanism Regulation (MMR) 525/2013 (European Union, 2013), city

GHG inventories are not subject to independent and thorough reviews. Though some degree of quality control on the reported emissions inventories does take place within the European Covenant of Mayors Framework (Kona et al., 2018), the lack of a comprehensive and transparent transnational reporting framework means that there is no formal verification process in place (Bertoldi et al., 2018). As such, urban scale emissions data and inventory methodologies are not scrutinised and the respective cities are not prompted to make subsequent improvements.

For the reasons described above, interest has been growing within the science community regarding how *in situ* measurements could be integrated into urban GHG emissions monitoring systems (Christen, 2014; Duren and Miller, 2012; Lin et al., 2018). Indeed, the following subchapter 3.2.2 will present numerous examples of the intense research that has been ongoing. Crucially however, it now appears that this interest has found its way into the policy arena. The US National Institute of Standards and Technology (NIST) has established three urban measurement tests beds²⁸ (Indianapolis, Los Angeles and the Northwest corridor) to develop and demonstrate the technology needed to monitor urban GHG emissions (Mueller et al., 2017). Furthermore, in Paris, a city which has been at the forefront of urban GHG measurement research (Bréon et al., 2015; Frey et al., 2019b; Staufer et al., 2016; Vogel et al., 2019; Wu et al., 2016), a pilot initiative has been launched to make such measurements useable for city's planners (Benkhelifa and Dubreil, 2019). The goal of these projects to translate science into usable solutions for city managers is underlined by the endorsements by the Integrated Global Greenhouse Gas Information System (IG3IS)²⁹. In 2018, IG3IS was formally launched by its host institution, the World Meteorological Organization (WMO). Among IG3IS's four stated objectives, is objective #3 which essentially aims to support subnational entities such as cities with implementing GHG measurement-based approaches to improve local emissions monitoring. Working towards this particular objective, IG3IS organised an online workshop in June 2020³⁰ to begin the process of developing an international standard for urban GHG monitoring and assessment. The workshop brought together international experts and scientists in the field of urban GHG inventory- and measurement methods, who are currently drafting a Best Practise Guide on available urban emissions monitoring methodologies and how they can be best implemented. While this Best Practice Guide will target the research community developing emerging methods, the ultimate aim is to stimulate the development of standards for operational implementation as the biannual updates of the best practices converge into widely accepted methodologies.

These developments thus indicate that cities, or at least capitals and large metropolises, are beginning to consider such measurement-based options. While there is an evident global interest in accurately quantifying emissions from cities, it could be argued that a local-level interest in improving urban emissions monitoring may stem from cities themselves. Due to underlying sociodemographic factors, public belief in climate change and support for mitigation action tends to be more widespread within urban populations (Howe et al., 2015; Mildemberger et al., 2016; Otto and Gugushvili, 2020). Therefore, if local election victories are secured on the back of climate change mitigation commitments (Martelli et al., 2018), cities may start to feel increasing pressure from their electorates to deliver effective and provable local action. Beyond verifying/improving aggregate city emissions estimates, *in situ* measurements can

²⁸ <https://www.nist.gov/topics/greenhouse-gas-measurements/urban-test-beds>

²⁹ <https://ig3is.wmo.int/en/outcomes/projects>

³⁰ <https://ig3is.wmo.int/en/events/towards-international-standard-urban-ghg-monitoring-and-assessment>

help to quantify local emissions at more useful temporal and spatial scales (e.g. the street and building scale). Such information of course can improve the process of identifying-, implementing- and monitoring mitigation strategies and the approach would be consistent with the Smart City revolution (Lin et al., 2018). Finally, by helping to provide verifiable emissions and emission reductions estimates, cities may consider measurement-based emissions monitoring an opportunity to facilitate participation within anticipated emissions trading schemes (Wu et al., 2016).

At this juncture, it wise to consider how a scenario, in which large European cities begin to implement *in situ* GHG measurements as part of their emissions monitoring systems, could influence the development of the Copernicus CO₂ MVS capacity. According to the *EIONET Group of Copernicus in situ Data Experts* survey, five of the 15 responding air quality national reference centres (NRCs) believe that large cities in their countries may start to establish measurement-based local GHG emissions monitoring within the next decade (ANNEX I). At first glance, the scenario may indeed present an opportunity to address some of the current limitations in the *in situ* component of the MVS system. Not only could the coverage of urban measurements increase but also the sustainability of the measurements could be enhanced due to the vested interest the cities have in the data. If cities begin implementing measurements of GHGs as they do measurements of air pollutants, it is pertinent to ask what this could mean for EIONET. To help address this question, the following subchapter provides a technical overview of in situ measurement-based options available for quantifying urban CO₂ and CH₄ emissions and discusses how they could be implemented within municipal monitoring systems.

3.2.2. Technical review of *in situ* measurement methods for estimating urban CO₂ and CH₄ emissions

The following subchapter reviews three types of *in situ* CO₂ and CH₄ measurement methods:

- Surface measurements of CO₂ and CH₄ (3.2.2.1)
- Ground-based measurements of total column CO₂ and CH₄ concentrations (3.2.2.2)
- Measurements of CO₂ and CH₄ fluxes (3.2.2.3)

For each of the measurement methods the review provides an overview of the measurement techniques and the methods for inferring local emissions, as well as a presentation of city case studies where the measurements have been tested. A synthesis subchapter (**Error! Reference source not found.**) then discusses how these specific measurements could contribute to the CO₂ MVS capacity and how they may be formally employed within municipal monitoring systems.

3.2.2.1. Surface measurements of CO₂ and CH₄

This chapter presents the different approaches most frequently used to measure dry-air mole fraction (mixing ratio) in ambient air and to derive fluxes of CO₂ and CH₄. Here different categories of instruments used in each approach are presented according to their accuracy and purchase costs.

3.2.2.1.1. Measurement techniques and instruments

High-Accuracy instruments (HA)

The World Meteorological Organization (WMO)/Global Atmosphere Watch (GAW) has recommended the use of the Non-Dispersive Infrared (NDIR) detection method to measure CO₂ with an uncertainty within the range of 0.01 to 0.1 ppm with 0.05 ppm as a typical value for most field systems (WMO, 2018a). For atmospheric CH₄, Gas-Chromatography (GC) with Electron Capture Detector (ECD) is the recommended technique for atmospheric CH₄ determination with an uncertainty less than 1 pbb (WMO, 2009). The high accuracy is achieved by frequent calibrations with standards traceable to WMO Central Calibration Laboratory (CCL³¹, NOAA). Within WMO/GAW, the conventional reference scale refers in particular to the calibration scale used within the GAW network. In the case of CO₂, CH₄, N₂O and CO, this scale is implemented as a family of gas cylinders maintained at the CCL. The Swiss Federal Laboratories for Materials Science and Technology (Empa) in Dübendorf, Switzerland, that hosts WMO/GAW World Calibration Centre for Surface Ozone, Carbon Monoxide, Methane and Carbon Dioxide (WCC-Empa) is now recommending the use of the Cavity ring-down spectroscopy (CRDS) technique to measure CO₂ and CH₄, which has the enormous advantage of being able to simultaneously measure CO₂ and CH₄. The CRDS technique shows a better performance than NDIR technique (Zellweger et al., 2016) and it requires a much lower frequency of calibrations given a very high linearity and stability of the instrument compared to NDIR (for CO₂) and GC-ECD (for CH₄) techniques (Gomez-Pelaez et al., 2019). A further advantage of the CRDS technique is that, in addition to measuring CO₂ and CH₄, it simultaneously measures CO with great precision, allowing determination of the CO₂/CO ratio to help discern the type of CO₂ source.

The Integrated Carbon Observation System (ICOS³²) only uses CRDS instruments (ICOS-ATC, 2016; Yver Kwok et al., 2015) meeting the WMO quality assurance and calibration protocols.

European GAW stations and ICOS atmosphere stations (Figure 1) provide CO₂ and CH₄ mole fraction data that can be used as reference for seasonal and inter-annual CO₂ and CH₄ variations in Europe, providing unique high-quality reference background values.

³¹ <https://www.esrl.noaa.gov/gmd/ccl>

³² <http://www.icos-ri.eu>

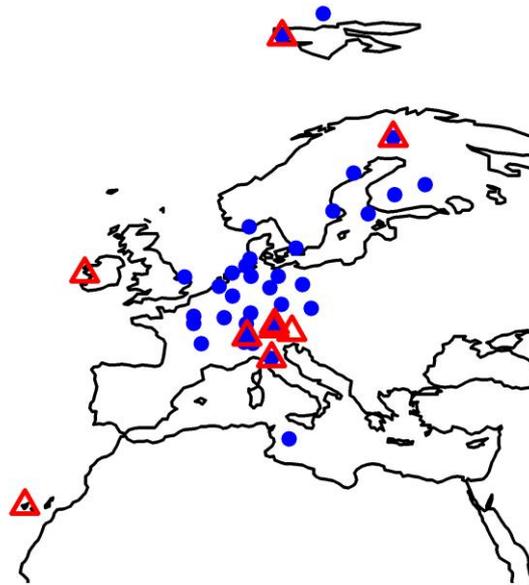


Figure 1: Location of the European GAW stations of global importance (red triangles) and the ICOS atmosphere stations (blue dots).

Other high accuracy techniques to measure CO₂ and CH₄, such as Off-axis CRDS (Crosson, 2008) or Off-Axis Integrated Cavity Output Spectroscopy (OA-ICOS) (Baer et al., 2012), and Tunable Infrared Laser Direct Absorption Spectroscopy (TILDAS), implemented with long path Astigmatic Multi-pass Absorption Cells (AMAC) (McManus et al., 2011), are mainly used for research purposes. These instruments achieve precisions lower than 0.03 ppm for CO₂. The high precision and accuracy CO₂ and CH₄ instruments, used in research activities and in regional or global background station networks, work with the same WMO references, currently X2007 for CO₂, and X2004A for CH₄, calibrated by the WMO CCL. The cost of these instruments is above 80 kEuro, except of those using the NDIR technique that are in the range of 25-40 kEuro. The high cost of the HA instruments is more than offset by the saving of expensive calibrations (the equipment is very stable and shows a great linear response), and the lower cost for operators since they do not need to be highly trained technicians (the instruments are very easy to manage and maintain, once installed).

CO₂ Low-Cost Sensors (LCSs)

Battery powered miniaturised versions of optical absorption methods (non-dispersive infrared -NDIR- sensors) are the common low-cost option (between 3 and 0.5 kEuro) (Kunz et al., 2018; WMO, 2018b) to set up relatively dense networks for determining the spatial and temporal distributions of CO₂ mole fractions in urban areas. These instruments, normally referred to as Low-Cost Sensors (LCSs), use the same technique of some reference instruments. Therefore, the evaluation of their performance can be straightforward since it is based on simple principles such as path length and CO₂ absorption properties (WMO, 2018b). The accuracy of LCSs ranges from 10 to 50 ppm, significantly lower than that of high-precision NDIR analyser or CDRS instruments.

The main advantage of CO₂ LCSs is of being of smaller size, much lower weight, having reduced power consumption, and lower cost, normally one or two orders of magnitude lower than a comparable high-accuracy instrument. However, the main drawback is that the response of CO₂ LCSs is highly dependent on temperature (T), pressure (P) and relative

humidity (RH) when the sampled air is not dried. Furthermore, the responses of each individual sensor may have a different dependency with P, T, and RH. For this reason, the sensor units must be individually assessed with test measurements in environmental chambers, or in previous intensive outdoors campaigns using changing actual T, P and RH data. Later, a verification of the applicability of the determined data correction parameters through comparison of the resulting sensor signal with reference instruments during field tests is required (WMO, 2018b).

Most of the efforts on LCSs for CO₂ have focused on the characterization of performance by comparison with reference instruments used under field conditions, using multiple linear regression models. Spinelle et al. (2017) evaluated the performance of two types of CO₂ LCSs from field tests. Different statistical and machine learning approaches were used for correcting for ambient T and RH effects. They obtained best results using Artificial Neural Networks (ANN), where sensor uncertainty was in the 18-25 ppm range for ambient concentrations between 370 ppm and 490 ppm CO₂. They also reported that the CO₂ uncertainty increased to unacceptable values >100 ppm when a less sophisticated linear regression model was applied. Zimmerman et al. (2018) analysed also the performance of different data models for the correction of a LCS signal operating under field conditions, finding that machine learning techniques, and specifically Random Forest models, outperformed multiple linear regression models. In this study the absolute mean error of the CO₂ LCS measurements was 10 ppm during a 16-week testing period.

In addition, it should be taken into account that the evaluation of the sensor performance is made in a specific site where both the training and test data are extracted, and that the calibration models usually also include CO as an interference factor. The CO₂/CO ratios may be different from one place to another depending on the nearby sources of CO₂ at each site. Therefore, although the tests finally show us an excellent agreement between the LCS and the reference instrument in the place where the LCS has been characterized, it cannot be guaranteed that the modelling carried out is equally valid for another site where the ratio CO₂/CO can be different (WMO, 2018b). Therefore, when sensors are then deployed away from reference instruments, strategies for continuous quality assurance and quality control of the sensors must be implemented, as reported by Kunz et al. (2018).

CO₂ Lower-Cost Medium Precision instruments (LCMP)

There is an intermediate group of low-cost sensors that are more precise and more expensive than LCSs, but in any case the costs are of an order of magnitude lower than the first-class analysers for research (e.g. CRDS). These so-called low-cost medium-precision (LCMP) instruments are associated with an approximate price of 5 kEuro. Hummelgård et al. (2015) introduced and tested some LCMP instruments. Although accuracy < 2 ppm can likely be achieved by LCMP sensors after applying calibrations, it can be improved by using machine learning techniques such as ANN.

According to Arzoumanian et al. (2019), the LCMP sensors are also sensitive to atmospheric P, T, and RH conditions, and the sensors respond linearly to CO₂ when measuring calibration tanks with CO₂ dry air mole fraction between 330 and 1000 ppm. However, the regression slope differs between individual sensors and changes with time.

These authors performed an empirical correction by measuring in parallel with the LCMP sensor and a HA instrument (CRDS) for a period of 6 months. The empirical “calibration” method consists of using a multivariable regression approach, based on predictors of air T, P

and RH, showing that the residual errors are within ± 1 ppm. This method explains well the observed drifts of the LCMP sensor on timescales of up to 1–2 months when trained against 1–2 weeks of HA instrument data series.

Provided that LCMP sensors can be regularly calibrated against one reference HA instrument, these sensors could be used to measure CO₂ mole fraction in dry air required for top-down CO₂ flux estimates. Several authors, such as Turner et al. (2016) and Wu et al. (2016), estimated that a ca. 5 % precision of CO₂ fluxes derived from relatively dense networks (i.e. every 2 km) of LCMP sensors can be achieved provided that CO₂ observations errors have a random error of less than 1 ppm on hourly measurements and that these errors are uncorrelated in time and space between stations. Broquet et al. (2018) reported that a typical spatial resolution of 2 km using LCMP CO₂ analysers (± 1 ppm or less) would be sufficient to provide an error reduction of up to 50% on the emissions of a large city. Therefore, it can be assumed that the target performance for the LCMP sensors should be an uncertainty of 1 ppm in hourly CO₂ values.

The total cost of networks using LCS or LCPM sensors can be notably increased, depending on the number of units used, on the infrastructure and logistics costs, including communication/data transfer, on time consumed for prior characterization of each and every sensor, and on the maintenance of periodic calibrations against reference instruments (e.g. CRDS) to correct possible instrument ageing and drifts.

Regarding mobile applications, special attention must be paid to possible performance reduction due to poorly controlled flow conditions and to the sensor response time.

CH₄ Low-Cost sensors (LCSs)

Regarding low-cost sensors for CH₄, there has been less research compared with the work on CO₂ LCS sensors and only a few publications exist (Collier-Oxandale et al., 2018; Eugster and Kling, 2012; Suto and Inoue, 2010). According to these studies the CH₄ LCS sensor is based on a metal oxide semiconductor as the gas sensor, a material that is also sensitive to CO and certain hydrocarbons.

Collier-Oxandale et al. (2018) found that the CH₄ LCS sensor tested had an accuracy of ± 1.7 ppm in a laboratory setting for minute-averaged data. Eugster and Kling (2012) tested a new LCS under real-world conditions over Toolik Lake, Alaska, to determine its suitability for preliminary studies. They tested two low-cost solid-state gas sensors being compared against a high-quality OA-ICOS instrument. The LCSs revealed a high sensitivity for T and RH similar to that of the specifications given by the manufacturer. The CH₄ concentration calibration checks were always within the uncertainty of the calibration tank available at Toolik Field Station (1.894 ± 95 ppm). Suto and Inoue (2010) tested a new portable instrument for atmospheric CH₄ measurement that was developed based on a tin dioxide natural gas leak detector, by comparing with a classical gas chromatograph with a flame ionization detector (FID). They reported an excellent linear relationship between the low cost sensor and the GC-FID, with a standard deviation of 4 ppb (1s). According to these authors the results of the comparison between the low-cost sensor and the GC-FID system at the environmental temperature in the laboratory matched within ± 10 ppb. It should be borne in mind that these tests have been carried out in the laboratory with calibration gases, and not in ambient air under real conditions.

As with CO₂ LCSs, the CH₄ LCSs need the corrections by T and RH. However, the fact that they are also affected by other hydrocarbons and their impact cannot yet be quantified, is a

major drawback. On the other hand, although in principle it is possible to apply corrections to eliminate the interference of water vapour, these would be unrealistic because of complicated adsorption/desorption processes on the wall material at different temperatures.

All these preliminary results indicate that the CH₄ LCSs are in a very incipient stage of development, with very high uncertainties, and cannot be used operationally as is the case with CO₂ LCSs.

Stable isotope measurements of atmospheric CO₂ and CH₄

Stable isotope measurements of atmospheric CO₂ and CH₄ require complex and high-cost instrumentation used for research activities. The stable carbon isotopes can be obtained with on-site analysers or by using air samples that are stored in flasks for a week and then analysed in central laboratories.

Commercial CRDS analysers provide continuous measurements of $\delta^{13}\text{C}$ in CO₂ at < 0.1‰ precision (Markovic et al., 2018), and the ratio of stable isotopes ^{13}C to ^{12}C in CH₄ with a precision of < 0.1‰ in 1 h (Rella et al., 2015). In addition, commercial TILDAS instruments (operating at mid-IR spectral region) provide real-time measurements in ambient air of $\delta^{13}\text{C}$ in CH₄ with a precision < 1‰ in 1 s, and isotopic ratio of ^{13}C in CO₂ with a precision < 0.1‰ in 1 s, and better than 0.03‰ in 1 minute (Nelson et al., 2008).

In Europe, the ICOS-ERIC infrastructure provides analysis of stable isotopes in CO₂ from air samples collected at all ICOS class-1 stations at weekly interval using automated samplers into glass flasks and transferred for analysis to: 1) the Flask and Calibration Laboratory³³ in Jena (hosted by the Max-Planck-Institute for Biogeochemistry), where stable isotopes of CO₂ $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ are quantified, and 2) the Central Radiocarbon Laboratory³⁴ in Heidelberg (operated by the Institute of Environmental Physics of the University of Heidelberg) that quantifies the radiocarbon content of CO₂ ($\Delta^{14}\text{C}$ in CO₂) and develops methods to derive the fossil fuel contribution to atmospheric CO₂.

3.2.2.1.2. Inferring emissions

Fluxes and emissions cannot be directly measured rather they are inferred from the measurements of CO₂ and CH₄ dry-air mole fractions.

A first simple approach to obtain CO₂ and CH₄ fluxes would be to measure the gradients of dry-air mole fractions of CO₂ and CH₄ between stations located upwind and downwind outside the ends of a city, and a third one located in the central part of the city (known as “mass-balance” approach). In addition, an adequate resolution meteorological model is required that provides the air flow through the city. However, pointed out by Rigby et al. (2008), the experimental design of one-point measurements of dry-air mole fraction of CO₂ does not allow for an absolute quantification of urban-wide emissions since the boundary layer height may change substantially over time and between rural and urban locations, which usually explains most of the observed variability. For example, Christen (2014) found that CO₂ mole fractions in rural stations are higher than measurements in cities, especially on summer nights, due to a more stably stratified and shallower rural boundary layer that accumulates biogenic CO₂ from

³³ <https://www.icos-cal.eu/fcl>

³⁴ <https://www.icos-cal.eu/crl>

soil and above-ground respiration. Only in winter was an urban enrichment of up to 10 ppm identified.

In practice, the simplest approach used is the one known as a "top-down" estimate. This approach consists of determining the CO₂ and CH₄ emission estimates from urban areas from the dry-air CO₂ and CH₄ mole fraction measurements with a network of *in situ* instruments combined with high-resolution transport modelling. The most commonly used principle is to measure atmospheric CO₂ and CH₄ dry-air mole fraction gradients at stations between the upwind and downwind vicinity of an emitting area and infer the emissions that are consistent with those observed gradients and their uncertainties, using an atmospheric transport model (Arzoumanian et al., 2019). Logically, there are different sampling strategies, and the measurement accuracies differ among these approaches.

The present *in situ* surface observation networks do not sample the atmosphere with a sufficient spatial resolution to resolve the complexity of CO₂ and CH₄ sources and sinks existing in a city. For this reason the estimation of natural GHGs fluxes from atmospheric measurements constitutes a highly underdetermined mathematical inverse problem (Bergamaschi et al., 2018). Therefore, a more complex approach for determining CO₂ and CH₄ emissions in cities is based on inverse techniques (Bayesian statistical inference), which require the combination of three elements: 1) an a priori estimate of the CO₂ and CH₄ fluxes provided by the available emission inventories; 2) measurements of atmospheric CO₂ and CH₄ mole fractions from a network of stations within and around cities; and 3) atmospheric transport models to translate information on surface fluxes into atmospheric CO₂ and CH₄ concentration gradients. In this way, the new computed CO₂ and CH₄ fluxes (considered as "true emissions") improve and correct a prior estimate of the CO₂ and CH₄ emission budgets, to better fit observed concentration gradients. The ultimate goal of the inversion method is to reduce the uncertainty on CO₂ and CH₄ fluxes in the city, by assessing the consistency of the three sources of information, and diagnosing the uncertainty in the estimate of the new inverted ("posterior") emissions (Bergamaschi et al., 2018; Wu et al., 2016).

Discrimination of types of CO₂ sources

When we refer to the term "CO₂ fluxes" both direct anthropogenic CO₂ emissions and natural CO₂ fluxes are included (Bergamaschi et al., 2018). So, one important research focus on urban and industrial "hot-spots" of CO₂ emissions with high-resolution observations is using tracers that help us separate the signal of fossil fuel CO₂ sources (Bergamaschi et al., 2018). This is especially important in the current urbanization trend in which power plants and other industries are being "incorporated" into large metropolitan urban areas. Because these smaller sources are not detected from space, and because total CO₂ also in hot-spot areas contain significant contributions from ecosystem fluxes, the successive CO₂ reports (Ciais et al., 2015; Pinty et al., 2017, 2019) propose to complement satellites by continental scale sampling of ¹³C and ¹⁴C in CO₂ and other fossil fuel CO₂ tracers such as co-emitted NO_x and CO (Ammoura et al., 2014; Levin et al., 2003; Levin and Karstens, 2007; Lopez et al., 2013; Pataki, 2003; Vogel et al., 2013), whose ratios to CO₂ depend on the sectors of activity.

Although CO₂ fossil fuel fluxes are typically dominant at the annual scale, ecosystem CO₂ fluxes can be significantly larger at any given point in time. For this reason Vogel et al. (2013) propose to use radiocarbon, or proxy gases like ¹⁴C-calibrated CO to separate their signals, and hence for inversions to constrain fossil fuel CO₂ emissions. However, in a city the number of possible CO₂ sources is high. Stable isotopic composition of CO₂ can be used to discern

between sources such as natural gas and gasoline combustion, as well as biogenic respiration (Pataki et al., 2005). For example $d^{13}\text{C}$, the normalized ratio between the heavier $^{13}\text{CO}_2$ and the lighter $^{12}\text{CO}_2$, is substantially lower in CO_2 emitted when burning natural gas (primarily for heating) than in CO_2 from gasoline combustion and plant respiration (Tans, 1981). Therefore, Djuricin et al. (2010) proposed a multi-isotopologue approach determining simultaneously $d^{13}\text{C}$, $d^{18}\text{C}$ and $d^{14}\text{C}$ in CO_2 and further use CO as a tracer to attribute multiple sources of CO_2 in urban air. However, this type of complementary measurements have the problem that the isotopic signatures of fossil fuels may change over time, as their values depend on the geographic origin of the imported fuels (Bush et al., 2007).

Discrimination of types of CH_4 sources

In the case of CH_4 , the main potential sources are basically leaks in storage systems and pipelines of natural air, which are not expected to be significant, and emissions from farmlands, landfills and wetlands that are normally located outside the cities. The stable isotopes in CH_4 , such as $^{13}\text{C}/^{12}\text{C}$ and D/H ratios (δD), provide additional constraints on different sectors, since different CH_4 sources have characteristic isotopic ratios (Bergamaschi et al., 2018). For example, natural gas shows δD values between -180‰ and -200‰ , while CH_4 emitted by wetlands and landfills has associated δD values $\sim -300\text{‰}$.

3.2.2.1.3. City case studies

A reasonable question is: how many CO_2 and CH_4 mole fraction measuring stations do we need in a city to improve CO_2 and CH_4 emissions knowledge? Actually, there is no clear answer since the ideal monitoring strategy depends on the particular goals and characteristics of the city, as well as the type of application and objectives that we pursue. For some applications, high-cost, low-density instrumentation is necessary, and in others, low-cost, high-density platforms are more effective (Shusterman et al., 2018). Below we summarize the most relevant experiments/networks carried out to estimate CO_2 fluxes using surface measurements in urban areas.

The Rotterdam experiment (Super et al., 2017)

In this experiment, the authors explored the use of a simple method to constrain urban emissions using only two measurement sites, one upwind and one downwind of the city of Rotterdam (Netherlands). They used HA CRDS instruments at both locations to measure atmospheric mole fractions of CO_2 , CH_4 and CO.

To get a flux estimate from continuous surface observations, the mass balance approach using a one-box model was applied (see section 3.2.2.2.2) considering constant the height of the box (h). This method assumes that the emissions are well-mixed throughout the boundary layer by the time it reaches the downwind site and that h is taken as a constant during transport. The authors used a monthly value of h based on measurements at the nearby Cabauw testbed site from ceilometer backscatter profiles, and assumed this value to be maximal and relatively constant during the afternoon, when well-mixed afternoon conditions are valid.

The authors showed that the observed gradient of CO and CO_2 ratio, $\Delta\text{CO}/\Delta\text{CO}_2$, agreed well with the CO/ CO_2 emission ratio in the emission database for metropolitan Rotterdam, in a large observed range, proving that continuous observations of CO and CO_2 at two sites could provide interesting information about the different CO_2 source sectors and can be used for an initial estimate of the CO_2 fluxes. Furthermore, the results indicate that the mass balance

approach provided good results for the cases in which the main flow is aligned with the transect of the upwind and downwind stations.

The Paris experiment (Bréon et al., 2015; Staufer et al., 2016; Wu et al., 2016)

As described by Bréon et al. (2015), a first attempt to estimate the CO₂ emissions from Paris was done using five stations equipped with HA CRDS instruments that provided continuous CO₂ mole fraction measurements: one analyser was located in the heart of Paris, at the summit of the Eiffel Tower, 300 m above the surface, two of the instruments were located to the north and north-east of the Paris area in a mixed urban – rural environment, and complemented by two ICOS-France stations installed in rural background conditions. This experiment was carried out in the framework of the CO₂-Megaparis and ICOS-France projects between August 2010 and July 2011.

Anthropogenic and biogenic CO₂ fluxes from a high-resolution local emission inventory (AIRPARIF) and an ecosystem model, respectively, were compared with an inversion model. The comparison of the measurements against the atmospheric transport simulation driven by the a priori CO₂ surface fluxes showed significant differences upwind of the Paris urban area indicating that distant sources (mainly from urbanised and industrialised areas north-east of Paris) were contributing to the CO₂ mole fraction variability. On the other hand, the direct model – measurement comparison showed that the CO₂ variability at the top of the Eiffel Tower is large and poorly represented by the model for most wind speeds and directions. That meant that the model was unable to reproduce the CO₂ variability at the centre of Paris. For that reason, and because it was necessary to determine the CO₂ budgets for the whole city, the strategy was changed to consider the assimilation of upwind – downwind CO₂ mole fraction hourly gradients between the sites at the north-eastern and south-western edges of the urban area, only. In this case the inversion significantly improved the agreement between measured and modelled CO₂ mole fraction gradients. Realistic emissions of CO₂ were retrieved for two 30-day periods, indicating a significant overestimate by the AIRPARIF inventory.

However, the drastic data selection used in this study limited the ability to continuously monitor Paris fossil fuel CO₂ emissions. For example, the inversion results for specific months such as September or November 2010 were poorly constrained by too few CO₂ measurements (Staufer et al., 2016). The fast mixing by the atmosphere and the complex structure of urban CO₂ emissions of Paris require high-resolution atmospheric transport models and a denser network of continuous CO₂ mole fraction measurements to select gradients induced by emission plumes that can be captured at the scale of the model (Bréon et al., 2015; Wu et al., 2016). A new approach, with an expanded observation network, could improve the knowledge of the CO₂ emissions inside the city.

In fact, in the Paris experiment, just using two HA CRDS CO₂/CH₄ stations, the CO₂ emissions from the Paris megacity could be retrieved with an accuracy of $\pm 20\%$ on monthly budgets (Staufer et al., 2016). However, a denser network of stations, as such designed in the study by Wu et al. (2016) for the retrieval of CO₂ emissions per sector for this megacity, using 10 LCMP sensors of 1 ppm accuracy on hourly time steps, has shown the error of the annual emission budget could be reduced down to a 10% uncertainty. Since the distribution of CO₂ emissions is highly heterogeneous in space and variable in time in urban areas, Wu et al. (2016) also found in the Paris experiment that even more stations were needed to obtain a detailed separation of emissions into different source-sectors, such as road transport and residential CO₂ emissions. With 70 LCMP stations, the uncertainties in the inverted emissions

were reduced significantly over those obtained using 10 stations: by 32% for commercial and residential buildings, by 33% for road transport, by 18% for the production of energy by power plants, and by 31% for total emissions.

The BEACO₂N experiment (Shusterman et al., 2016, 2018; Turner et al., 2016)

The BEACO₂N (the Berkeley Atmospheric CO₂ Observation Network) is an ongoing greenhouse gas and air quality monitoring campaign operating across the city of Oakland, California in the San Francisco Bay Area since late 2012. The current network is comprised of ~50 “nodes” stationed on top of schools and museums at approximate 2 km intervals equipped with CO₂ sensors showing intermediate performances between those of LCS and LCMP sensors. The CO₂ LCSs dataset is supplemented by a HA CRDS instrument.

A major concern for a long-term field deployment with this type of sensor is systematic uncertainty resulting from a combination of gradual temporal drift, given the limited access to validation and calibration infrastructure, and constant biases from the “true” value perhaps incurred abruptly upon installation.

The CO₂ measurement at a given site was the sum of the real regional and local influences at the said site as well as the systematic uncertainties. To derive corrections they removed the CO₂ urban background signal from the data record by subtracting the weekly minimum CO₂ concentrations recorded at a reference site within the network domain. Preliminary results successfully captured hourly, daily, and seasonal CO₂ signals of urban environments on spatial scales that cannot be accurately represented by atmospheric transport models alone. They also demonstrated that high-resolution surface networks showed significant qualitative and quantitative differences among the diurnal cycles at individual nodes on spatial scales that cannot yet be accurately captured by atmospheric transport models, confirming the necessity of a high-density approach when attempting to represent the variability of a complex urban environment.

Potential CO₂ sources can be identified and quantified simply by comparing signals from adjacent nodes. A higher CO₂ level measured at the downwind node relative to its upwind neighbour indicates the presence of a CO₂ emitter in between the two. It may even be possible to make a first estimate of CO₂ emissions in each grid taking into account wind fields. Turner et al. (2016) found that using high-resolution surface networks with LCMP sensors, improved the results provided by smaller networks using HA analysers in estimating urban CO₂ emissions and mapping the sources.

The Salt Lake City experiment (Lin et al., 2018; Mitchell et al., 2018a)

This research effort was focused in the Salt Lake City, metropolitan region, Utah, which is the locus for one of the longest-running urban CO₂ networks in the world (Utah Urban Carbon Dioxide Network (UUCON)). The experiment started in 2001 by researchers at the University of Utah (Mitchell et al., 2018a; Pataki, 2003). They used CO₂ observations from HA NDIR and OA-ICOS instruments calibrated with reference gases traceable to the WMO calibration scales. Details of the instrumentation, data, calibration, and QA/QC methods can be found in Pataki et al. (2003; 2006) and Mitchell et al. (2018a). This network was enhanced with novel mobile observations from van-based, on-road measurements, platforms on light-rail public transit trains that commenced in December 2014 and are ongoing (Mitchell et al., 2018b), and with modelling that includes atmospheric simulations and high-resolution emission inventories.

The mobile instrument deployed on electricity-powered light-rail trains repeats observations along dedicated rail lines, providing coherent spatiotemporal patterns across the city. For example, CO₂ mole fractions have shown to decrease from the urban core toward the southwestern part of the valley. Furthermore, this observational mobile platform allows real-time measurements with low operating costs while avoiding self-contamination from vehicle exhaust.

As basic results, these authors found that the average diurnal cycle of CO₂ is modulated by both atmospheric mixing and socioeconomic activity, while the CO₂ seasonal cycle is driven by meteorological, biological, and anthropogenic factors. In summer, a deeper summertime planetary boundary layer dilutes emissions reducing the measured CO₂ mole fraction. Moreover, there is a removal of CO₂ during this season because of the vegetation growing season. In winter the vertical mixing is suppressed resulting in higher CO₂ mole fraction with especially pronounced events during prolonged meteorological stagnation episodes. Winter emissions also increase as a result of natural gas combustion for heating.

Other results show that rapid increasing daytime emission rates during the summer occurred in areas with initially low population density that underwent conversion of rural land to suburban developments, while emissions were stable in the urban core despite population increases, consistent with prior modelling efforts examining changes in on-road emissions over space and time. These authors consider that detecting and understanding patterns in suburban areas is important as they tend to have a higher household carbon footprint than urban centres, and this should be taken into account for monitoring network design.

The Vancouver experiment (Lee et al., 2017)

For the Vancouver experiment (Canada) a method for directly measuring CO₂ emissions using a mobile sensor network in cities at a fine spatial resolution was developed and tested. First, a compact, mobile system was built using a LCMP NDIR CO₂ analyser combined with open-source hardware to control, georeference, and log measurements of CO₂ mole fraction on vehicles. Second, two measurement campaigns, one in summer and one in winter, were carried out. Five mobile sensors were deployed within a 1x12.7 km transect across the city. The sensors were operated for 3.5 h on predefined routes to map CO₂ mixing ratios at street level, which were then averaged to 100x100 m² grid cells. In both campaigns, CO₂ mole fractions were highest in the grid cells of the downtown core and along arterial roads and lowest in parks and well vegetated residential areas. Third, an aerodynamic resistance approach to calculate emissions was used to derive CO₂ emissions from the gridded CO₂ mole fraction measurements in conjunction with CO₂ mole fractions and fluxes collected from a 28 m eddy covariance tower located within the study area. Estimation of CO₂ emissions for each grid cell is based on the aerodynamic resistance approach, which postulates that the molar flux of CO₂ for a given area and time is equal to the difference of the molar concentration at the height above the roughness sublayer (tower CO₂ measurements) and screen level at 2 m height (mobile sensors CO₂) divided by the aerodynamic resistance of CO₂. In this experiment, an independent emissions inventory was developed for the study area using building energy simulations from a previous study and routinely available traffic counts. The comparison on a grid-by-grid basis showed linearity between CO₂ mixing ratios and the emissions inventory. Also, 87% (summer) and 94% (winter) of measured grid cells showed a difference within ± 1 order of magnitude, and 49% (summer) and 69% (winter) show an error of less than a factor 2.

Although associated with considerable errors at the individual grid cell level, the study demonstrates a promising method for using a network of mobile sensors and an aerodynamic resistance approach to rapidly map greenhouse gases at high spatial resolution across cities.

The Madrid experiment (Frey et al., 2019b; García et al., 2019)

Another interesting example of instrumental synergies to derive urban CO₂ and CH₄ enhancements is the Madrid experiment (Frey et al., 2019b; García et al., 2019), which combined surface CO₂ and CH₄ HA CRDS measurements (fixed and mobile) with column-average mole fractions from a city-scale column network. This experiment is explained in detailed in section 3.2.2.2.3.

The Carbosense experiment (Empa³⁵)

A quite different approach is that used by Carbosense project (Switzerland) in which 300 low-cost CO₂ sensors distributed over Switzerland provide an unprecedented density of continuous CO₂ observations. For the city of Zurich, Empa aims to link the sensors data to an atmospheric dispersion model based on the CO₂ emission inventory of the city (Müller et al., 2019). These emission sources include various modes of traffic, industry and heating in residential buildings. By combining these simulations with the sensor data, Empa will be able to display the city's current CO₂ emissions almost in real time. As far as we know no results have been published so far.

In order to summarize the above case studies, the main characteristics of these experiments are presented in Table 3.

Table 3: Overview of the different city experiments inferring CO₂ emission fluxes from surface CO₂ observations.

Experiment	Instruments	Methods	Main goals
Rotterdam	HA CRDS	Mass balance	Estimation of CO ₂ fluxes
Paris	HA CRDS and LCMPs	Inversion modelling	Estimation of CO ₂ fluxes and inventory verification
BEACO ₂ N	LCSs, LCMPs and HA CDRS	Comparison between neighbouring stations and atmospheric models	Characterization of CO ₂ emissions
Salt Lake City	HA NDIR and OA-ICOS (mobile platforms)	Comparison between neighbouring stations and atmospheric models	Characterization of CO ₂ emissions and their temporal evolution
Vancouver	HA NDIR	Comparison of mobile transects and synergy with Eddy-covariance measurements	Estimation of CO ₂ fluxes and inventory verification
Madrid	HA CRDS (fixed and mobile platform)	Mass balance	Characterization of CO ₂ and CH ₄ emissions
Carbosense	LCSs	Inversion modelling	Characterization of CO ₂ emissions in near real time

From these experiments we conclude that a combination of a few permanent stations equipped with HA instruments, supplemented by denser networks of LCMP sensors deployed on a permanent basis or during field campaigns in certain periods of the year (for example, 1-2 months in summer and winter), atmospheric transport models, and emissions inventory, could

³⁵ <https://www.empa.ch/web/s604/carbosense4d>

provide high temporal and spatial resolution information on CO₂ emissions in big cities on a continuous basis.

3.2.2.2. Ground-based total column measurements of CO₂ and CH₄

Urban CO₂ and CH₄ emissions can be inferred from measurements of the mixing ratios of the respective gases integrated over the total atmospheric column. Implemented within inverse modelling frameworks or simple mass-balance approaches, measurements of temporal and spatial variations in total column-averaged dry-air mole fractions can be used to derive local net surface-atmosphere fluxes of CO₂ and CH₄ as well as estimations of relative sink and source-sector contributions. This chapter presents the main measurement techniques and instruments for measuring total column-averaged CO₂ and CH₄ mole fractions from the ground and the methods with which these measurements can be implemented to derive emissions estimates. Finally, some examples of case studies where such measurements were successfully used to estimate city CO₂ and CH₄ emissions are described.

3.2.2.2.1. Measurement techniques and instruments

Fourier Transform Infra-Red (FTIR) spectrometry is currently the sole operational remote sensing technique providing total column amounts of key atmospheric trace gases such as GHGs or ozone-related compounds (i.e. fluorine and chlorine species). In FTIR spectrometry, the source radiation (typically Sun for atmospheric ground-based measurements) is modulated by an interferometer and all optical frequencies are recorded simultaneously in the measured interferogram (Griffiths and de Haseth, 2007). Then, a mathematical Fourier transform is used to retrieve the atmospheric absorption spectrum from the interferogram. By analysing the pressure broadening effect on the measured solar infrared absorption spectra through inversion schemes, the FTIR technique can provide atmospheric concentrations of many different trace gases simultaneously and with high precision (De Mazière et al., 2018; Wunch et al., 2011).

Nowadays, the globally-distributed high-resolution FTIR spectrometers mainly operate in the framework of two international networks for atmospheric composition monitoring: NDACC, Network for the Detection of Atmospheric Composition Change³⁶ (De Mazière et al., 2018); and TCCON, Total Carbon Column Observing Network³⁷ (Wunch et al., 2011) (Figure 2). Whilst NDACC mainly aims to establish long-term databases for detecting changes and trends in atmospheric composition and to understand their impacts on the Earth's atmosphere, TCCON is more focused on GHG research, improving our understanding of the carbon cycle and source-sink attribution. Nonetheless, both networks share the same primary purpose of providing reference validation datasets for space-based observations (e.g. SCIAMACHY, MIPAS, ACE, TANSO, AIRS, IASI, TROPOMI, OCO-2, OCO-3) (Buchwitz et al., 2015; De Mazière et al., 2018; Sha et al., 2019; Wunch et al., 2011 and references therein). Recently, these high-resolution FTIR observations have been extended by COCCON, COllaborative Carbon Column Observing Network³⁸ (Frey et al., 2019a), which is a research infrastructure of portable, compact, and low-resolution ground-based FTIR spectrometers set up as a supplement of TCCON (Figure 2).

³⁶ <https://www2.acom.ucar.edu/irwg>

³⁷ <https://tcccon-wiki.caltech.edu/>

³⁸ <https://www.imk-asf.kit.edu/english/COCCON.php>

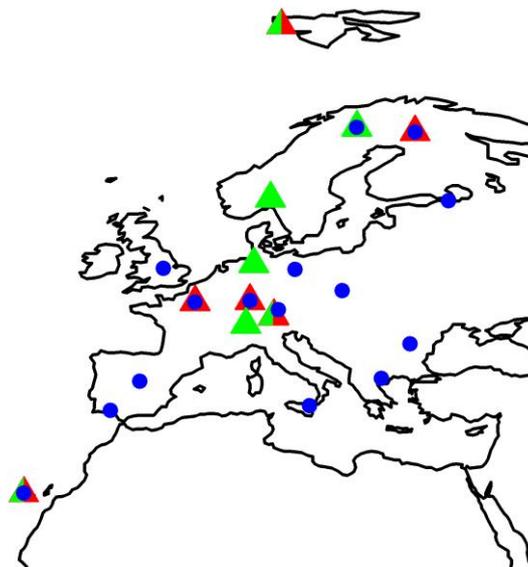


Figure 2: Location of the European fixed NDACC stations (green triangles), TCCON stations (red triangles), combined NDACC/TCCON stations (green/red triangles) and COCCON stations (operational and field campaigns in blue dots).

For the interest of the Copernicus CO₂ MVS capacity, TCCON operationally provides total column-averaged dry-air mole fractions of CO₂, CH₄, and CO (denoted as XCO₂, XCH₄, and XCO, respectively), standardized and calibrated onto the WMO GHG *in situ* scale. The measurement precision for the TCCON XCO₂ and XCH₄ products varies from site to site, but it is generally less than 0.25% (Wunch et al., 2011). COCCON is also able to provide standardized and WMO-calibrated XCO₂, XCH₄ and XCO data with very high accuracy and precision meeting the TCCON requirements. The COCCON products have shown to be slightly biased high with respect to coincident TCCON observations, providing scale factors ($1 \hat{f}$) of only 1.0098 (0.0015) and 1.0072 (0.0024) for XCO₂ and XCH₄, respectively (Frey et al., 2019a). Finally, NDACC routinely measures total column amounts and low-resolution vertical profiles of CH₄ and CO as well as NO₂ at most of NDACC sites. Although CO₂ and the stable isotopologues of CO₂ and CH₄ are not required NDACC species, they can be potentially retrieved from middle infrared FTIR spectra (Bader et al., 2017; Barthlott et al., 2015; Buzan et al., 2016; Reuter et al., 2012).

As Pinty et al. (2019) pointed out, these ground-based total column measurements are essential components to identify, characterize, and mitigate the impact of instrumental, geophysical, or methodological biases on the products delivered by the space segment of the Copernicus CO₂ MVS capacity (i.e. the Sentinels CO₂ monitoring constellation). Particularly, as the report on the CO₂ MVS capacity furthermore emphasises, the ground-based XCO₂ and XCH₄ from TCCON and COCCON FTIR instruments provide the needed transfer standard between the space-based estimates and the WMO standards. Although only the TCCON and COCCON FTIR observations are explicitly mentioned in this report, it is worth to highlight here the potential of the ground-based FTIR measurements, acquired in the framework of NDACC, to contribute to the GHG research and, thus, to the CO₂ MVS capacity.

In terms of instrumental operation and installation, the only logistic requirement is that the direct solar path is free of obstacles that could shadow the incoming solar beam into the ground-based FTIR spectrometers. Most TCCON and NDACC stations are fully automated

inside fixed scientific containers or laboratories, while COCCON instruments typically operate outdoor and require an operator, which limits the availability of observations. However, recent developments (i.e. automated enclosure) lead to a fully automated measurement system, providing the foundation for a permanent GHG monitoring network (Heinle and Chen, 2018). Furthermore, although research studies have probed the added value of placing COCCON devices on mobile platforms (e.g. ships, vehicles) (Luther et al., 2019 and references therein), the recommended configuration is that instruments be installed on fixed locations for operational purposes (i.e. field campaigns or long-term performance). Regarding maintenance, the FTIR spectrometers are complex instruments that require skill and experience, especially those operating with TCCON and NDACC networks. Optic re-alignments, among other tasks, are periodically needed to ensure high performance as well as regular measurements against gas standards must be done to characterize the instrumental stability. Particularly, TCCON also recommends a calibration with in-situ instruments on-board research aircrafts or balloon-based vertical profiles (i.e. AirCore), where available, or, otherwise, against travelling and traceable COCCON FTIR instruments (Hedelius et al., 2017). Finally, given the documented robustness of the low-resolution FTIR spectrometers used within COCCON, this infrastructure suggests these instruments be only sent to COCCON central facilities for a biennial instrumental inspection (Frey et al., 2019a).

Purchase prices of high-resolution FTIR spectrometers, such as those compatible either with NDACC or TCCON (research instruments), typically start at around 300 kEuro, while complete high-resolution FTIRs, spanning from near to middle infrared spectral region and operating within both networks, can reach 600 kEuro. The low-resolution FTIR instruments, such as those being used within COCCON, are commercially available from 80 to 100 kEuro. Other compact, portable and medium-to-low resolution systems, such as other versions of FTIR instruments, laser heterodyne and grating spectrometers, or fibre Fabry-Perot interferometers, are currently under investigation and could be available for GHGs monitoring in a near future (El Kattar et al., 2019; Sha et al., 2019; and references therein).

3.2.2.2.2. Inferring emissions

Total column observations are directly linked to total emissions and are sensitive to surface-atmosphere fluxes. McKain et al. (2012) suggested that column-integrated measurements can offer a promising route for improved detection of CO₂ emissions from major source regions, such as urban environments, completing or partially obviating the need for extensive surface measurements near these areas. Compared to other measurement techniques, urban enhancements in columns are relatively insensitive to vertical redistribution of tracer mass (e.g., due to growth of the planetary boundary layer) (Chen et al., 2016) and also less influenced by the redistribution of surface emissions by small-scale processes (Lauvaux and Davis, 2014). Such measurements may therefore allow for more precise trend detection of emissions from urban regions and for accurate verification of emission inventories of CO₂ and other GHGs (McKain et al., 2012).

Similarly to surface measurements, column-integrated observations can be employed in combination with inverse modelling frameworks to derive local net surface-atmosphere fluxes of CO₂ and CH₄ as well as estimations of relative sink and source-sector contributions (Hedelius et al., 2018; Olsen and Randerson, 2004; Viatte et al., 2017). However, column observations are more compatible with the scale of atmospheric models and hence have been shown to be a powerful source of observational information to constrain local surface fluxes at

regional scales and to mitigate uncertainties in atmospheric inversion systems (Lauvaux and Davis, 2014; Lindenmaier et al., 2014; McKain et al., 2012). The use of ground-based column measurements appeared, for example, to decrease the biases associated with carbon cycle processes (Olsen and Randerson, 2004) and the impact of vertical transport errors on the flux retrievals (Gerbig et al., 2008; Lauvaux and Davis, 2014) or to constrain significantly the boundary concentrations, a critical limitation in current regional inverse flux estimates (Lauvaux and Davis, 2014).

One potential drawback, however, is that, depending on location, column observations are sensitive to surface emissions over a very wide range of spatial scales, spanning nearby emissions and all those upwind in the urban, continental, and hemispheric domains (Chen et al., 2016). To mitigate this limitation, small-scale networks of ground-based FTIR instruments measuring gradients of column amounts are often used (typically compact low-resolution spectrometers since the large high-resolution instruments are not easily relocated) (Chen et al., 2016; García et al., 2019; Hase et al., 2015a; Luther et al., 2019; Toja-Silva et al., 2017; Viatte et al., 2017; Vogel et al., 2019). The horizontal gradients within a region are influenced by the local net emission strength across the domain and atmospheric mixing (Vogel et al., 2019), being relatively insensitive to surface fluxes upwind of the domain and, thus, providing favourable inputs for regional flux inversions (Chen et al., 2016).

This type of deployment strategy also allows the total emissions of CO₂ and CH₄ to be derived using the top-down mass-balance approach rather than inverse modelling systems. This simple column method is able to resolve CO₂ and CH₄ enhancements (as small as 0.5 ‰) from small-scale emitting sources (Hase et al., 2015a). This is possible because the detection of enhancements is based on the differences between simultaneous and precise column-averaged observations across local and regional areas (typically at upwind and downwind stations of the target emitting sources). In its simplest form, this approach basically depends on the upwind-downwind concentration gradients and wind fields, but it requires making assumptions on the homogeneity of the sources and conservative transport in the domain (e.g. homogenous wind shear in the planetary boundary layer) (Chen et al., 2016; Viatte et al., 2017). The instrumental deployment to be implemented depends on the size of area of interest, the wind regime and sources' characteristics. For compact sources, such as urban areas, the optimal setup consists of 4 or 5 instruments circling the source, which allows possible changes of wind regime to be taken into account. Note that the triangulation or identification of sources requires changes in wind direction when using a static network of sensors.

Recent studies point out that the mass-balance column approach could be improved by collecting ancillary data, such as wind vertical profile measurements co-located to FTIR instruments, to help define the boundary conditions and refine the estimated emissions fluxes (Luther et al., 2019; Viatte et al., 2017). Likewise, combining these differential column observations with high-resolution chemistry and transport models enhances the capability of attributing observed variability to both atmospheric dynamics and local emission patterns as well as identifying sink and source-sector contributions (Toja-Silva et al., 2017; Viatte et al., 2017; Vogel et al., 2019; Zhao et al., 2019). Traditionally, the mass-balance approach has been used with column-integrated observations, but city-scale emissions fluxes have also been derived using this method from surface measurements (3.2.2.1.2) and aircraft data (Mays et al., 2009; Peischl et al., 2013; Super et al., 2017; Zimnoch et al., 2010).

Analogously to surface measurement campaigns, other approaches rely on the use of additional co-emitted tracers, such as CO, NO₂, NH₃, Ethane (C₂H₆), or stable isotopologues

of CO₂ and CH₄, which can play a pivotal role in helping to assess regional CO₂ and CH₄ emissions and to separate the atmospheric signals related to the different flux processes (i.e. biogenic and anthropogenic flux components) (Kille et al., 2019; Lindenmaier et al., 2014). This is also the basic principle to infer CH₄ emission fluxes, and other GHGs, from column-averaged observations using the tracer–tracer anomaly correlation technique (Wong et al., 2016, 2015, Wunch et al., 2009, 2011, 2016, 2019). This simple method uses the regression slopes between observed trace gas anomalies (e.g. CH₄/CO₂ or CH₄/CO) in the atmosphere to calculate the target gas emissions (e.g. CH₄) based on the more accurately known emissions of the correlate (e.g. CO₂ or CO) (Wong et al., 2016). This permits therefore the derivation of the relative emissions of the two trace gases without the use of transport models and does not require the sources to be co-located (Wong et al., 2016). However, as potential drawbacks, normally the needed known emissions are taken from emission inventories, which determines the accuracy of the estimated emission fluxes, and the spatial distribution of the target gas emissions are assumed to be similar to those for the tracers (Wunch et al., 2019). The latter is typically true for CO in the case of Europe, where the CO signals are dominated by urban/industrial combustion processes (CO contributions from biomass burning are considerably small) or for NO₂.

The following subchapter summarizes examples of case studies where column-integrated measurements were successfully used to estimate city CO₂ and CH₄ emissions.

3.2.2.2.3. City case studies

Many studies have probed the capability of ground-based FTIR column-averaged observations to monitor atmospheric CO₂ and CH₄ concentrations, to detect and quantify their emissions from regional-urban scales to point sources as well as to inform and verify emission inventories. Particularly, citywide GHG column measurements have been carried out during field campaigns or long-term operation, e.g., in North and Central America: Los Angeles Basin (Hedelius et al., 2018; Wong et al., 2016, 2015, Wunch et al., 2009, 2016), Boston (Chen et al., 2016); Indianapolis (Franklin et al., 2017), Toronto (University of Toronto), Mexico City (Baylón, 2017); in South America: Manaus (Dubey et al., 2014); in Europe: Bialystock (Deutscher et al., 2015), Berlin (Hase et al., 2015a; Zhao et al., 2019); Bremen (Notholt et al., 2014), Bucharest (Dandocsi et al., 2019), Karlsruhe (Hase et al., 2015b), Leicester (Humpage et al., 2019), Madrid (Frey et al., 2019b; García et al., 2019), Munich (Heinle and Chen, 2018; Toja-Silva et al., 2017), Orléans (Warneke et al., 2014), Paris (Té et al., 2014; Vogel et al., 2019), St. Petersburg (Gavrilov et al., 2014), Thessaloniki (Topaloglou et al., 2020), Yekaterinburg (Rokotyan et al., 2015); in South East Asia: Darwin (Griffith et al., 2014a), Wollongong (Griffith et al., 2014b), Hefei (Liu et al., 2018), Saga (Kawakami et al., 2014), Tsukuba (Morino et al., 2016), Tokyo (Frey et al., 2017); and Africa: Jinja (Uganda, University of Leicester). These examples span from megacities such as Los Angeles Basin, Mexico City, Paris or Tokyo, to small urban areas with more than 100.000 inhabitants. However, they also illustrate the irregular spatial distribution of the ground-based column observations, since the important metropolitan areas of South America, Africa and Central Asia are currently not covered.

The South Coast Air Basin experiment (Hedelius et al., 2018; Wong et al., 2016, 2015, Wunch et al., 2009, 2016)

One of the most comprehensively analysed areas has been the South Coast Air Basin (SoCAB), which is one of the most urbanized regions in North America, centred on the Los Angeles megacity and home to more than 40% of the population in California (USA). Wunch et al. (2009) measured for the first time diurnal changes of XCH_4 , XCO_2 , and XCO from TCCON data in the SoCAB, and used CO_2 emission inventories and the tracer-tracer anomaly method, to estimate emissions of CH_4 (and CO). Their top-down CH_4 flux estimates confirm that urban emissions are a significant source of CH_4 and in fact may be substantially higher than those currently reported by bottom-up CH_4 emission inventories. Posterior works, using similar strategies, confirm the underestimation of the state government's bottom-up CH_4 emission inventory (California Air Resources Board, CARB) by 18–61% in this area (Wong et al., 2016, 2015). Hedelius et al. (2018) further improved the SoCAB emission analysis by implementing the column-averaged observations into an inverse modelling framework. They documented XCO_2 enhancements observed over the SoCAB compared to an external background of the order of 2–3 ppm. Likewise, their estimates of total annual CO_2 fluxes are on the low end of previous values (evaluated from aircraft, space-based, inverse models or bottom-up inventories) and about 28%–47% less than inventory records used in tracer–tracer flux estimate works (Wong et al., 2015; Wunch et al., 2009). The agreement for net CH_4 fluxes is generalized in literature. Other studies in this area were focused on quantifying the loss of natural gas by using column measurements of CH_4 and C_2H_6 (Wunch et al., 2016).

However, most of these studies did not have simultaneous upwind and downwind column data, which allow for concentration gradients to be determined and thus the emission fluxes using the mass-balance approach. The simplest form of this method has been used by Chen et al. (2016) to measure XCH_4 gradients across the largest dairy-farming area in the SoCAB (about 50 km² area in Chino city) from a small-scale COCCON FTIR network. They found that the observed emissions from the XCH_4 gradient of ~2 ppb are comparable to the emissions estimated at Chino during the CalNex campaign (Peischl et al., 2013), which were determined using the bottom-up method and an aircraft-based mass balance approach. However, this version of the differential approach, using only upwind and downwind measurements, reduces the flux estimates to only steady-wind conditions (constant wind speed and direction). In Viatte et al. (2017), the analysis of Chino was extended combining the FTIR column observations and an advanced atmospheric modelling system, which allow for describing the spatial distributions of CH_4 emissions and deriving optimized XCH_4 emission fluxes.

The Berlin experiment (Hase et al., 2015a; Zhao et al., 2019)

In Europe, several field campaigns specifically designed to identify urban CO_2 and CH_4 sources and quantify urban enhancements have been carried out during the last years. In 2014 in Berlin, Hase et al. (2015a) demonstrated that an array of ground-based COCCON FTIR spectrometers allows the reliable detection of XCO_2 and XCH_4 enhancements due to local emissions in the range of 1‰ (~0.5 ppm and ~2 ppb for XCO_2 and XCH_4 , respectively). In Zhao et al. (2019) these column-averaged measurements were compared to high-resolution chemistry transport simulations, attributing the observed variability to different emission processes. These authors documented that the observed enhancement of XCH_4 is highly dependent on human activities, whilst the XCO_2 enhancement in the vicinity of Berlin is dominated by anthropogenic behaviour rather than biogenic activities.

The Paris experiment (Frey et al., 2019b; Vogel et al., 2019)

A similar deployment strategy was used in 2015 to characterize the Paris metropolitan area (Frey et al., 2019b), which is the most densely populated region in France. Based on a modelling framework and the observed column-averaged concentrations, Vogel et al. (2019) showed that the strong decrease in XCO₂ during daytime can be linked to net ecosystem exchange, while a significant enhancement compared to the background is caused by XCO₂ from fossil-fuel emissions within the Paris metropolitan area, but this is often compensated by net ecosystem exchange. The data also confirm previous results by models that XCO₂ gradients caused by a megacity do not exceed 2 ppm, which supports the requirements for the space-borne elements of the Copernicus CO₂ monitoring constellation of less than 1 ppm precision on individual soundings and absolute biases lower than 0.5 ppm (Ciais et al., 2015; Pinty et al., 2019).

The Madrid experiment (Frey et al., 2019b; García et al., 2019)

In 2018 in Madrid (highest population density city in Spain), the column setup was further improved including mobile surface CO₂ and CH₄ records (Frey et al., 2019b; García et al., 2019). A small-scale network of ground-based COCCON FTIR spectrometers were operated on the outer perimeter of Madrid metropolitan area along the prevailing wind axis measuring gradients of column-averaged CO₂ and CH₄ amounts, while two mobile surveys took place each day using a flight-ready HA CRDS analyser to measure air CO₂ and CH₄ mole fractions also along the perimeter of the Madrid City. This deployment was completed with a fixed HA instrument for surface continuous measurements and ancillary data (meteorological surface and vertical records and aerosol load observations). Similarly to Paris and Berlin, fossil fuel emissions were found to be mainly responsible for CO₂ emissions with urban enhancements of ~1 ppm, while CH₄ enhancements were strongly linked to waste treatment. Due to the proximity of a waste disposal site to the Madrid metropolitan area, very large XCH₄ enhancements up to 140 ppb were observed during the campaign, which are consistently detected also by the mobile surface records. These significant CH₄ plumes were confirmed in a second field campaign in 2019, where column-averaged observations were specifically taken across the potential CH₄ sources identified in 2018. The results show how using a combination of surface and column-averaged techniques highly enhances the capacity to better characterize diffuse and punctual CO₂ and CH₄ city sources.

Point source experiments

Column-integrated observations, alone or complemented by other measurements, allow for determining point source emissions at finer scales, such as volcanoes, coal mining, cattle and dairy feedlots, natural gas leaks, fracking oil wells, coal-fired power plants as well as sporadic events, such as pollution plumes, transient peaks, or instabilities across the planetary boundary layer (Butz et al., 2017; Chen et al., 2016; Kille et al., 2019; Lindenmaier et al., 2014; Luther et al., 2019; Mellqvist et al., 2010; Toja-Silva et al., 2017; Viatte et al., 2017).

For example, Mellqvist et al. (2010) studied plumes from industrial complexes, and Lindenmaier et al. (2014) examined plumes from two coal-fired power plants and discriminated them, combining column and surface observations. Toja-Silva et al. (2017) used column gradient measurements and a computational fluid dynamics model to quantify emissions from a natural-gas-based power plant in Munich. Luther et al. (2019) deployed a mobile COCCON FTIR spectrometer to quantify CH₄ emissions from hard coal mines.

3.2.2.3. Measurements of CO₂ and CH₄ fluxes

The previous sections describe how surface and total column mixing ratios of CO₂ and CH₄ can be measured *in situ* and how these measurements can be used to infer urban emissions of the respective gases. This chapter describes the methods for measuring and processing net CO₂ and CH₄ fluxes in the urban boundary layer and how these *in situ* data can be analysed to infer emissions.

3.2.2.3.1. Measurement techniques and instruments

Turbulent exchange is the process by which atmospheric scalars (heat, gases, and aerosols) are vertically mixed through the boundary layer. This mixing is driven by eddy circulations caused by wind shear and buoyancy forces generated by surface drag and surface heating, respectively (Kaimal and Finnigan, 1994). In micrometeorological research, turbulent exchange is typically sampled in the so-called surface layer, the layer constituting the lower 10 % of the boundary layer (Foken, 2017). In urban environments, measurement equipment are generally installed on towers that reach above the roughness layer (above 2 times the average height of roughness elements i.e. buildings) into the lower surface layer (Feigenwinter et al., 2012; Grimmond et al., 2002).

Each measurement station of course constitutes a point measurement i.e. a measurement of net turbulent exchange at the measurement location. However, under well-mixed and ideal measurement conditions (measurement in the surface layer above a flat, expansive and horizontally homogeneous surface), the vertical and horizontal variation in turbulent exchange should not be substantial (Kaimal and Finnigan, 1994). Furthermore, under well-mixed and ideal measurement conditions this point measurement of turbulent exchange is assumed to be in pseudo-equilibrium with surface-atmosphere exchanges integrated over a footprint extending upwind. Therefore, in the case of turbulent CO₂ and CH₄ fluxes it is assumed that measured net vertical exchanges are representative of the upwind emissions and removals integrated over the footprint (Baldocchi, 2003; Foken et al., 2012b).

Measuring turbulent exchange requires integrated systems consisting of sensors which measure the respective scalars, and instruments which measure wind speeds. One option is to employ gradient systems, which by sampling scalars at two or more height locations can derive vertical scalar gradients. The respective net turbulent exchange is then estimated by multiplying the vertical scalar gradients by turbulent exchange coefficients derived from parallel the wind measurements (Foken, 2017). While variations of these systems are still implemented today, it is fair to say that most stations measuring CO₂ and CH₄ fluxes over the last two decades have been employing eddy covariance systems (Baldocchi, 2020). In contrast to the gradient method, eddy covariance provides a direct quantification of turbulent CO₂ and CH₄ exchange by measuring high-frequency (> 10 Hz) gas concentrations and wind speeds and calculating the 30 or 60 minute covariances between the respective scalar (e.g. CO₂, CH₄) and the vertical wind speed.

To measure the CO₂ and CH₄ concentrations, eddy covariance systems typically (e.g. within the FLUXNET and ICOS networks) rely on non-dispersive infra-red spectroscopy or cavity enhanced absorption (e.g. cavity ring-down spectroscopy or off-axis integrated cavity output spectroscopy). In contrast to surface measurement sensors described in 3.2.2.1.1, eddy covariance gas analysers have to be able to provide highly precise measurements at frequencies of 10 Hz or more. These gas analysers are then complemented by 3D ultrasonic

anemometers measuring horizontal and vertical wind speeds, and ultrasonic temperature (Munger et al., 2012; Rebmann et al., 2018).

Purchase prices of complete eddy covariance systems such as those operating within FLUXNET and ICOS (research instruments) typically start at around 40kEuro. Systems measuring CO₂ fluxes range from about 40kEuro to 80kEuro, while systems capable of measuring both CO₂ and CH₄ fluxes range from ca. 100 to 200 kEuro. In terms of maintenance, ICOS recommends that the gas analysers be checked and recalibrated against gas standards at least every 3 months and the whole system be sent for factory inspections/recalibrations every two years (Rebmann et al., 2018).

3.2.2.3.2. Inferring emissions

In contrast to surface- and total column mixing ratio measurements, net emissions can be directly inferred from the measured turbulent fluxes. As mentioned previously, under well-mixed and ideal measurement conditions, it is assumed that the measured turbulent CO₂ and CH₄ fluxes are in pseudo-equilibrium with the upwind emissions and removals integrated over the footprint (Baldocchi, 2003; Foken et al., 2012b). These *well-mixed* conditions (i.e. the periods when the state of the boundary layer permits maximum equilibrium between the measured turbulent fluxes and the upwind net emissions at the surface) are identified by applying standardised quality control procedures (Foken et al., 2012a; Mauder et al., 2013). Although a substantial proportion of data are filtered, typically between ca. 20 and 50 % of the half-hourly/hourly measured fluxes can be retained for subsequent analysis and calculating monthly and annual budgets. Aggregating to monthly and annual net emission estimates can be done by calculating average values from the available fluxes or by applying gap-filling routines and summing over the complete dataset (Falge et al., 2001; Papale, 2012).

Both the half-hourly/hourly fluxes and aggregated net emissions are representative of net surface emissions with the flux footprints; however, flux footprints never overlap perfectly with the respective city borders. In fact, most urban flux stations provide net emissions estimates at a neighbourhood scale than a city scale (Christen, 2014; Feigenwinter et al., 2012). Therefore to derive representative city-scale estimates, multiple stations are typically required to sample different parts of the city (Soegaard and Møller-Jensen, 2003). Nonetheless, given that the spatial extent of flux footprints increases with increasing measurement height, employing tall-tower systems (> 100 m) can help reduce the number of stations required for city-scale estimates (Helfter et al., 2016; Matthews and Schume, 2019; Song et al., 2013; Ueyama and Ando, 2016).

To derive source-sector estimates modelling is required. In theory, an inverse modelling approach can also be applied to eddy covariance where instead of trying to constrain the prior emissions estimates to reproduce the observed CO₂ and CH₄ mixing ratios and/or gradients only, the model is optimised to reproduce the observed turbulent fluxes. This is however seldom done, with most studies rather employing flux partitioning routines to divide the measured net fluxes between different component sources and sinks (Menzer and McFadden, 2017). For instance, to estimate annual emissions from space heating and traffic, simple linear functions of measured air temperature and continuous traffic counts can be fitted to the data and subsequently solved over the years' time series (Kleingeld et al., 2018).

The following subchapter presents a number of case studies to demonstrate the various ways in which urban flux measurements can be applied to estimate city CO₂ and CH₄ emissions.

3.2.2.3.3. City case studies

This review of English-language, scientific literature identified 37 cities/towns where eddy covariance has been applied to measure local net fluxes of CO₂. For 3 of these cities, eddy covariance measurements of CH₄ have also been applied (Table 4).

Table 4: List of cities for which studies conducting urban eddy covariance measurements of CO₂ fluxes have been published. For each measurement system that has been operated, the measurement height and reference are given. Therefore the same city can be listed more than once if multiple systems have been employed. If measurements of CH₄ have also been conducted, the respective cities are marked with a (*).

City/Town	Measurement Height (m)	Reference
Chicago, USA	27	(Grimmond et al., 2002)
Edinburgh, UK	69.5	(Nemitz et al., 2002)
Copenhagen, Denmark	40	(Soegaard and Møller-Jensen, 2003)
Basel, Switzerland	39	(Schmutz et al., 2016)
Basel, Switzerland	19	(Lietzke and Vogt, 2013)
Marseille, France	43.9	(Grimmond et al., 2004)
Marseille, France	34.6	(Grimmond et al., 2004)
Tokyo, Japan	29	(Moriwaki and Kanda, 2004)
Tokyo, Japan	52	(Hirano et al., 2015)
Mexico city, Mexico	37	(Velasco et al., 2005)
Mexico city, Mexico	42	(Velasco et al., 2009)
Melbourne, Australia	38	(Coutts et al., 2007)
Melbourne, Australia	40	(Coutts et al., 2007)
Münster, Germany	65	(Schmidt et al., 2008)
Helsinki, Finland	31	(Vesala et al., 2008)
Helsinki, Finland	60	(Järvi et al., 2018)
Vancouver, Canada	28.8	(Christen et al., 2011)
Baltimore, USA	37.2	(Crawford et al., 2011)
Łódź, Poland*	37	(Pawlak et al., 2011)
Florence, Italy*	33	(Gioli et al., 2012)
Beijing, China	47	(Liu et al., 2012)
Beijing, China	140	(Song et al., 2013)
Beijing, China	280	(Song et al., 2013)
London, UK*	190	(Helfter et al., 2011)
London, UK	46.4	(Björkegren and Grimmond, 2018)
Shanghai, China	80	(Ao et al., 2016)
Indianapolis, USA	30	(Davis et al., 2017)
Telok Kurau, Singapore	20.7	(Velasco et al., 2013)
Swindon, UK	12.5	(Ward et al., 2013)
Arnhem, the Netherlands	23	(Kleingeld et al., 2018)
Sakai, Japan	111	(Ueyama and Ando, 2016)
Sakai, Japan	30	(Ueyama and Ando, 2016)
Sakai, Japan	16.2	(Ueyama and Ando, 2016)
Porto, Portugal	32	(Salgueiro et al., 2020)
Aveiro, Portugal	12	(Salgueiro et al., 2020)
Seoul, South Korea	30	(Hong et al., 2019)
Cheongju, South Korea	19	(Hong et al., 2019)
Heraklion, Greece	27	(Stagakis et al., 2019)
Vienna, Austria	144	(Matthews and Schume, 2019)
Innsbruck, Austria	38.6	(Karl et al., 2017)
Toulouse, France	48.1	(Goret et al., 2019)
Essen, Germany	21	(Kordowski and Kuttler, 2010)
Minneapolis–Saint Paul, USA	40	(Peters and McFadden, 2012)
Montreal, Canada	25	(Bergeron and Strachan, 2011)
Montreal, Canada	25	(Bergeron and Strachan, 2011)
Leece, Italy	14	(Conte et al., 2018)
Auckland, New Zealand	11	(Weissert et al., 2016)

Phoenix, USA	15	(Pérez-Ruiz et al., 2020)
Murray-Salt Lake Valley, USA	36	(Ramamurthy and Pardyjak, 2011)
Phoenix, USA	22.1	(Song et al., 2017)

* For London and Florence, the CH₄ measurements are described in the references given in the table. For Łódź, the CH₄ measurements are described separately in another paper (Pawlak and Fortuniak, 2016).



Figure 3: Location of the European urban eddy flux stations identified in this review.

In terms of CO₂, most of the studies cited in Table 4 reported that the measured net CO₂ emissions varied according to dynamics in local heating demand and traffic intensity. For those cities outside of the tropics and subtropics such as Basel (Schmutz et al., 2016), Beijing (Liu et al., 2012), Florence (Gioli et al., 2012), Łódź (Pawlak et al., 2011), London (Helfter et al., 2016), Montreal (Bergeron and Strachan, 2011), Osaka (Ueyama and Ando, 2016), Toulouse (Goret et al., 2019) and Vancouver (Christen et al., 2011), CO₂ fluxes were shown to increase with decreasing temperatures (< 15° C) reflecting increased local combustion for space heating. Furthermore, the above examples plus studies from Helsinki (Järvi et al., 2012), Heraklion (Stagakis et al., 2019), Mexico city (Velasco et al., 2005), and Tokyo (Hirano et al., 2015) report strong positive correlations between measured CO₂ fluxes and local traffic counts sampled by automatic traffic recording stations, with weekday diurnal trends typically reflecting the morning and late-afternoon rush-hour periods. Studies investigating CH₄ in Florence (Gioli et al., 2012), London (Helfter et al., 2016), and Łódź (Pawlak and Fortuniak, 2016) have also shown fluxes to vary with local heating demand; however, the above studies indicate the measured CH₄ fluxes are not just the result of CH₄ released from the (incomplete) combustion process but also (and to a potentially larger extent) because of increased leakage of natural gas from pipe supply networks during the high consumption periods.

By tracking the temporal variations of the fluxes, one can not only infer but also quantify contributions from respective sources and sinks. In Arnhem, linear models of measured air temperature and continuous traffic counts were fitted to the flux data and subsequently solved to provide annual estimates of CO₂ emissions from space heating and road transport, respectively (Kleingeld et al., 2018). A similar approach was applied for a study in Florence, although emissions from combustion in buildings for space-, warm water- and cooking heat were modelled using linear functions fitted to gas consumption data (Gioli et al., 2012). More elaborate models can be fitted to the data to estimate emissions at a finer source-sector

resolution including additional source and sink contributions. A study of a suburban area of Minneapolis demonstrated an algorithm that estimated contributions from combustion for space heating and warm water/cooking separately, as well as additional biogenic contributions from photosynthesis and respiration fluxes (Menzer et al., 2013). Such methods of course rely on auxiliary data (traffic counts, natural gas consumption, and weather data) recorded over the time series. Nonetheless, such source-sector splits can also be derived by analysing the spatial variations of the measured fluxes. Due to changing wind directions and turbulence regimes, flux footprints vary for each measured 30/60 min flux. In Heraklion (Stagakis et al., 2019) and Vancouver (Crawford and Christen, 2015), analysis of flux footprints were analysed to determine how measured CO₂ fluxes varied with varying spatial contributions from roads, buildings and vegetation allowing a quantitative attribution of measured net fluxes to these spatial elements.

At this point it is necessary to reiterate that the measured net fluxes as well as the aggregated budgets and source sector estimates are representative of the respective flux footprints. As such, even with carefully selected placement of the measurement systems, it is impossible to measure fluxes which include all sources and sink contributions from within a city's borders and exclude all influence from outside the city. Furthermore, due to logistical restrictions most urban eddy covariance systems are set up on top of telecommunication masts or on masts on top of buildings at heights of ca. 10 – 50 m above the surface. Given the resultant footprint areas of ca. several ha to several km², these systems typically provide flux estimates at the neighbourhood to district spatial scale (Christen, 2014; Feigenwinter et al., 2012). To scale up to the city-scale thus requires multiple stations sampling the fluxes from different neighbourhoods/districts. In Copenhagen, this was accomplished by complementing a permanent measurement station in the city centre system with a mobile system that was moved around to sample different parts of the city outside the centre (Soegaard and Møller-Jensen, 2003). By modelling how the fluxes were influenced by the different land cover classes in the sampled footprints, the authors of this study were able to upscale to an average city CO₂ flux estimate for the whole metropolitan area of Copenhagen. An option with which the sampling effort (no. of stations) can be reduced is to employ a tall-tower approach. Given that the spatial extent of flux footprints increases with increasing measurement height, systems which measure fluxes at heights of > 100 m above the surface allow the sampling of footprints which are > 10 km² in area. In London (Helfter et al., 2011, 2016) and Vienna (Matthews and Schume, 2019), eddy covariance systems have been installed on the top platforms of large radio towers, operating at measurement heights of 190 and 144 m, respectively. In Sakai a system has been sampling CO₂ fluxes at 111 m above the surface from a mast built on top of a city skyscraper (Ueyama and Ando, 2016). Finally, in Beijing a 325 m meteorological tower has been utilised to measure CO₂ fluxes at heights of 140 and 280 m above the surface (Song et al., 2013).

For municipal policy makers and city planners, urban eddy flux measurements could provide a number of potential uses. Eddy covariance can provide independent measurement data with which such inventories can be validated and improved. In London (Helfter et al., 2016) and Copenhagen (Soegaard and Møller-Jensen, 2003), the reported annual aggregate CO₂ fluxes were in close agreement with the local inventory estimates thus providing confidence in the respective aggregate inventory estimates. Beyond verification, discrepancies between the measurements and inventories are also important to identify. In London (Helfter et al., 2016) and Florence (Gioli et al., 2012), the measured CH₄ were >2 and >7 times larger than the respective inventory estimates. These studies pointed towards missing/underestimated sources in the CH₄ inventory such as biogenic fluxes from waste water and fugitive emissions

due to leakage of natural gas from the pipe supply networks. Finally, given that eddy covariance provide 30-60 min flux estimates over the course of the year and for varying footprints, these data are ideal for testing and refining temporally resolved and gridded local emission inventories as demonstrated in Florence (Gioli et al., 2015).

Finally, in addition to improvement of local emissions inventories, arguments can be made for the direct use of these data in evaluating climate change mitigation strategies. Given that inventories depend on a wide range of statistical activity data, inventories do take time to compile (time to compile and synthesise the final input statistics and then time to compile the inventories). Given that eddy covariance measurements can be processed quickly by standardised calculation and quality control procedures, these measurements could fulfil a *nowcast* function, providing early indicators of e.g. year to year changes in emissions. Reflecting this potential nowcast function, a presentation at the online CHE-VERIFY Joint General Assembly in July 2020 demonstrated how COVID-19 lockdowns have impacted urban CO₂ fluxes measured in 11 different cities using the eddy covariance technique^{39,40}. Furthermore, analysing how the 30/60 min fluxes vary with air temperature and local traffic counts at different points in time could allow sectoral evaluation of mitigation policy seeking to reduce emissions through space-heating and traffic efficiency measures (Figure 4). By fitting such linear curves to the measured data, one can essentially calculate the emissions savings that have been achieved.

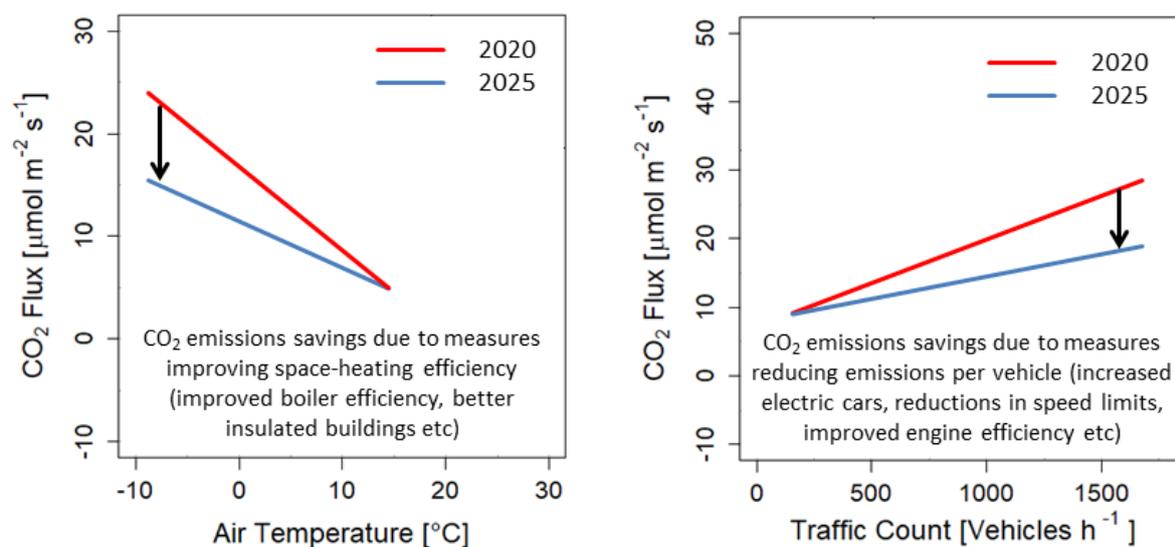


Figure 4: Conceptual graphs demonstrating how flux measurements can be used to directly evaluate climate change mitigation measures aimed at reducing CO₂ emissions from space heating (left) and road traffic (right).

3.2.2.4. Contribution to the CO₂ MVS capacity and recommendations on practical implementation to monitor local emissions

Urban *in situ* measurements of surface and total column CO₂ and CH₄ as well as measurements of urban CO₂ and CH₄ fluxes would fulfil respective *in situ* requirements of the Copernicus CO₂ MVS capacity. A pressing requirement is the *in situ* data stream that will validate the satellite observations of GHG concentrations and concentration gradients in and

³⁹ <https://www.che-project.eu/events/che-verify-joint-general-assembly-2020>

⁴⁰ https://verify.lsce.ipsl.fr/images/COVID_EC_Papale.pdf

around cities. In this case, the ground-based measurements of surface mixing ratios and, in particular total column mixing ratios, would offer the required independent *in situ* validation data. Ultimately, *in situ* validation data to verify the satellite-derived emissions estimates will also be required. In this respect, all of the measurement-based methods described above would offer suitable independent observation data to validate city-scale outputs of the Copernicus CO₂ MVS capacity. Separate inverse modelling of CO₂ emissions based on *in situ* measurements of surface and total column CO₂ would of course provide complementary estimates to compare with city-scale net CO₂ fluxes and anthropogenic CO₂ emissions (totals and source sector splits) derived from the Copernicus CO₂ MVS capacity. The same can also be said for the flux measurements. In this case, the eddy covariance measurements can be compared directly against the net CO₂ fluxes derived from the Copernicus CO₂ MVS capacity, while partitioned fluxes may offer useful independent source-sector CO₂ estimates.

In addition to utilisation within the Copernicus CO₂ MVS capacity, the above measurements could also benefit climate change mitigation monitoring in the respective cities. *In situ* measurements can verify/improve city emissions estimates as well as help to quantify local emissions at more useful temporal and spatial scales (Lin et al., 2018). It has thus been argued in this report that European cities, or at least capitals and large metropolises, may begin to consider implementing such measurement-based monitoring options. If so, local planners will be required to consider the design of their respective city measurement networks. As illustrated in the above review, the different types of available measurements can be implemented in various ways. While different methods have their respective strengths and weaknesses, it would be unwise to advocate one set of methods over the others. Indeed, any potential implementation will surely be dictated by local circumstances e.g. available budget, existing air quality/meteorological measurement infrastructure, technical know-how in the relevant municipal departments and/or partner universities/institutions, spatial distribution of sources and sinks and orography. Nonetheless, a few overall recommendations can be made.

With respect to integration within the existing air quality networks, it is clear that surface measurements would be most easily assimilated. In contrast to the total column and flux measurement devices, these instruments can be employed together with air quality systems to sample at the street level. Such integration would bring a number of benefits. Not only does co-location with measurements of co-emitted air pollutants improve the inverse estimate of anthropogenic CO₂ emissions by using the air pollutants as tracers (Levin and Karstens, 2007; Lopez et al., 2013; Vogel et al., 2013) but it also allows for utilisation of existing operational infrastructure (enclosures, power, UPS systems, data communication and technical maintenance). Nonetheless, it is important to point out that the existing air quality networks have been designed with both local human health and EU air quality legislation in mind. Therefore, it is advised to evaluate whether the existing network locations would be sufficient to deliver CO₂ and CH₄ emissions estimates at the desired temporal and spatial resolution and within an acceptable level of uncertainty.

In considering the sufficiency of surface CO₂ and CH₄ measurements at existing air quality station locations, it is strongly recommended to consider not just additional surface measurement sampling but all other possible extension options. The authors of this report advocate, where possible, to implement appropriate combinations of three measurement methods. While spatiotemporal patterns of surface CO₂ and CH₄ mixing ratios provide a solid basis to infer high-resolution local emissions, errors in the emissions estimates can occur due

to errors elsewhere in atmospheric transport models i.e. the model reproduces the surface observations; however errors in vertical and horizontal transport lead to errors in the inferred emissions. As such additional *in situ* measurements of column-averaged CO₂ and CH₄ mixing ratios and turbulent fluxes can provide additional useful constraints (upwind-downwind column-integration gradients, vertical turbulent transport) in the inversion. Indeed, assimilating multiple observations has proved to decrease the impact that vertical transport errors have on surface fluxes estimates and the boundary concentrations (Lauvaux and Davis, 2014; McKain et al., 2012). Previously, it would have been difficult to advocate such an approach utilising measurements of total column mixing ratios due to the high start-up-, maintenance- and operational costs of the high-resolution FTIR instruments. However, the recent emergence of portable, stable and more affordable low-resolution FTIR instruments may facilitate widespread application. This would be advantageous seeing as these systems, which sample gradients in the total column mixing ratios, are less likely to *miss* emissions from local large point sources. Given the tall stacks of such emissions, methods utilising surface measurements and/or flux measurements struggle to resolve emissions from these sources.

Finally, the review has demonstrated that research campaigns have been conducted in many of Europe's larger cities. Therefore, in addition to considering utilising cities' air quality network, local planners may also consider utilising measurement infrastructure and existing local expertise already in place from the cited research projects. Indeed, given the complexity of the methods and the ongoing work to make the methods operational, such partnerships with research would likely be beneficial in both the implementation and eventual operating phase. As mentioned previously, through initiatives of WMO IG3IS, as well as ICOS⁴¹, the international research community is working to develop best practise guidance and standardisation for applying urban GHG measurements to monitor local fluxes. As such, local research-policy partnerships in this area could help keep respective municipal climate planners informed of relevant developments and help steer the research and development process from the perspective of end-user needs.

3.3. Facilitating the contribution of EIONET to the *in situ* component of the Copernicus CO₂ MVS capacity

Considering the recently elaborated *in situ* requirements of the future Copernicus CO₂ MVS capacity and the changing landscape of urban monitoring of air quality and GHG emissions, an opportunity arises for EIONET to function as data stream into the *in situ* component. Realising this opportunity will of course depend on how various scenarios within, *inter alia*, European air quality- and urban climate change mitigation policy play out. Nonetheless, it is possible to already point out ways in which EIONET's potential contribution to the *in situ* component of the Copernicus CO₂ MVS capacity can be further elaborated.

Although EIONET is not explicitly mentioned, the recent CO₂ Monitoring Task Force report identifies and describes the need for *in situ* measurement data of tracer gases such as CO and NO_x from air quality networks (Pinty et al., 2019). This of course presents an obvious contribution that EIONET could make given that measurements of urban air quality in the EU under the Air Quality Directive (AQD; 2008/50/EC, amended by Dir. 2015/1480/EU) and the 4th Daughter Directive (4DD; 2004/107/EC) are regularly uploaded on EEA's central data repository. Due to the fact that these measurements and the subsequent sharing of the data

⁴¹ <http://www.icos-etc.eu/icos/working-groups/work-group?wgroup=19>

are embedded in legislation, it would seem that a potential stream of this data into the CO₂ MVS capacity could be sustained in the long-term. What is perhaps unclear is whether these measurements fulfil all the required criteria of the CO₂ MVS capacity e.g. accuracy, sampling density, temporal resolution. Answering this question is of course impossible because these precise criteria for the CO₂ MVS capacity still need to be elaborated. Therefore, one would propose that the architects of the CO₂ MVS capacity further define what is needed in terms of *in situ* tracer measurements and evaluate the extent to which the legislated EU MS measurements reported to EIONET are sufficient. Such an evaluation should take place in the near-term considering the European Commission's plans to evaluate current air quality legislation. As stated in the EU's Green Deal, the Commission will propose to strengthen provisions on monitoring (European Commission, 2019). Considering this prospect, it would therefore make sense to establish/increase dialogue between the CO₂ Monitoring Task Force, the Copernicus programme and the air quality community to explore potential improvements in air pollution measurements that would benefit both local air quality monitoring and the CO₂ MVS capacity formulate proposal for subsequent legislative amendments.

Regarding *in situ* urban GHG measurements, the initial road ahead for EIONET is more difficult to navigate. As this report indicates, these measurements are still in the process of making the transition from science to policy. Nonetheless, the fact that we are in the early stages may allow the EEA to be proactive in shaping EIONET's role with respect to *in situ* urban GHG measurements and the CO₂ MVS capacity.

A first step would be to conduct a thorough assessment at the municipal level to determine to what extent the local governments of Europe's large cities are considering these measurement-based options. Just as the air quality national reference centres (NRCs) have been contacted for their views by the *EIONET Group of Copernicus in situ Data Experts* (ANNEX I), the EEA together with counterparts from within Copernicus and the CO₂ Monitoring Task Force may consider reaching out to municipal representatives through EIONET and/or fora such as the European Covenant of Mayors. Such outreach could provide a clearer picture on which cities are considering to adopt *in situ* GHG measurements and when they will likely be implemented. Dialogue with this community could also reveal in what form the measurements could be implemented. If for instance cities will seek ownership of the process, directly implementing measurements in a similar way as they implement air quality monitoring, EIONET could play more of a substantial role. On the other hand, if local government departments will rather support or encourage universities/research institutes in implementing these measurements in their cities it would make sense that the research networks (ICOS, TCCON, COCCON etc.) coordinate the infrastructural implementation, the sharing of data and the subsequent stream into the CO₂ MVS capacity. Given that the emissions monitoring methods based on *in situ* GHG measurements are still maturing, it is likely that the latter represents the most likely scenario. Nonetheless, EEA could explore opportunities within and beyond its EIONET network to facilitate this process, particularly how EEA could cooperate with the ICOS network.

Considering the organisational structure of the research and development phase of the Copernicus CO₂ MVS capacity so far, it is likely that ICOS will ultimately be responsible for coordinating the system's *in situ* component and thus the urban sub-component. Building upon its prominent role in the CHE and VERIFY projects, ICOS will lead the work package on ground based measurements within the upcoming 2021-2023 CoCO₂ project to develop the

first system prototype⁴². At the June 2020 online Copernicus CO₂ workshop mentioned in chapter 2, a talk was given by ICOS announcing, *inter alia*, recent/coming addition of participating member countries, negotiations to integrate European TCCON stations into ICOS and a strategy to develop and integrate city observations. Given that EEA has been entrusted with the coordination of the *in situ* component of the Copernicus programme, there is scope for the EEA to support ICOS in implementing its strategy on urban observations. Indeed EEA's *in situ* mandate includes an explicit role to *manage partnerships with data providers*. The aforementioned outreach to- and dialogue with municipal climate change mitigation planners in large European cities could for instance be utilised to help ICOS implement the expansion and sustained operation of urban *in situ* GHG measurements and data streams. Such cooperation could benefit all sides, enabling identification and utilisation of synergies between the expansion strategy ICOS needed to fulfil the *in situ* requirements of CO₂ MVS capacity and the plans/wishes of cities to improve local GHG emissions monitoring. A vested interest of cities in directly using the GHG measurement infrastructure for local emissions monitoring may for example facilitate mobilisation of additional funding needed to expand and sustain the urban *in situ* observations, as well as open opportunities for co-location with existing sensors that operated within the local air quality monitoring programme. It would therefore be timely for EEA to consider how it could help facilitate this dialogue through EIONET national reference centres on e.g. air quality and climate change mitigation and/or through contacts outside of EIONET.

4. CONCLUSIONS AND PERSPECTIVES

The current development of the Copernicus CO₂ MVS capacity represents an opportunity for EIONET and EEA. Though still under construction, the European Commission's CO₂ Monitoring Task Force has been clear in stating that *in situ* measurements will form a fundamental component of the system. The CO₂ Monitoring Task Force also explicitly recommends that all existing relevant *in situ* capabilities be exploited and extended as appropriate.

By containing the measurement data reported by EU MS under the Air Quality Directive (AQD; 2008/50/EC, amended by Dir. 2015/1480/EU) and the 4th Daughter Directive (4DD; 2004/107/EC), EIONET is clearly a relevant *in situ* capability. In addition to urban *in situ* measurements of CO₂ and other GHGs, the CO₂ MVS capacity is set to rely heavily on urban *in situ* measurements of co-emitted air pollutants to help constrain city-scale anthropogenic CO₂ emissions estimates. Given the spatial sampling standards set out in the above legislation with respect to population density, EIONET provides a centrally validated dataset of *in situ* observations of tracer pollutants (e.g. NO_x, CO) from European cities that is updated annually. Furthermore, facilities are already in place to accept, store and visualise non-validated data on an hourly basis⁴³. The EIONET air quality data could thus be utilised by the CO₂ MVS capacity; however, further assessment is required to judge whether these potentially useful data are fit for purpose. It is therefore recommended that the CO₂ Monitoring Task Force, Copernicus and relevant actors in the European air quality community begin discussing this issue considering the plans in the EU's Green Deal to evaluate current air quality legislation. This may present an opportunity to explore potential improvements in air pollution measurements that would

⁴² <https://www.icos-cp.eu/event/964>

⁴³ <https://www.eea.europa.eu/data-and-maps/explore-interactive-maps/up-to-date-air-quality-data>

benefit both local air quality monitoring and the CO₂ MVS capacity and to propose subsequent draft amendments to the legislation.

Beyond air quality measurements, this report also explores the possibility that EIONET could, in the future, provide additional *in situ* measurements of urban GHGs. In the most recent CO₂ Monitoring Task Force publication, it was concluded that the research networks measuring GHGs *in situ* currently do not meet all the operational requirements for CO₂ MVS capacity. In particular, the Task Force highlighted a critical lack of urban measurements. While the Task Force and those within ICOS, TCCON, COCCON, etc., are no doubt exploring the extension of these networks to increase coverage within some of Europe's larger cities, an opportunity may arise for EIONET. Reflecting the current paradigm shift with respect to emissions monitoring, a scenario may develop whereby cities begin implementing *in situ* GHG measurements to improve monitoring of their local emissions.

This report demonstrates that substantial progress has been made in research into urban CO₂ and CH₄ emissions monitoring based on *in situ* measurements of surface mixing ratios and isotopes, total column mixing ratios and turbulent fluxes. Based on this progress, it is argued here that these methods are maturing for practical application and that the same rationale that has driven this research (i.e. the uncertainty in current city-scale inventories) may soon drive an uptake of these methods by local municipalities to improve their emissions monitoring systems. For the Copernicus CO₂ MVS capacity this may constitute a welcome development; however, for the measurements to meet the system requirements the implementation and eventual sharing of data would need to be coordinated. A further opportunity may therefore arise for EIONET, especially if cities are proactive in directly implementing such measurements in a similar way as they implement air quality monitoring. Of course these measurements are still in the process of making the transition from science to policy at the urban level. Therefore, the report suggests that it may be wise for

EEA to consider how the interests of municipal climate planners and the *in situ* research networks can be leveraged against one another. Specifically, EEA could consider facilitating dialogue between municipal climate change mitigation planners in large European cities and ICOS, given the likelihood that ICOS will ultimately be responsible for coordinating the *in situ* component of the Copernicus CO₂ MVS capacity.

The CO₂ MVS capacity will rely on coordinated governance of the *in situ* element to ensure sustained streams of high quality *in situ* measurement data into the system. Given that the CO₂ Monitoring Task Force recommendation that all existing relevant *in situ* capabilities be exploited and extended as appropriate, it is likely that EEA and its EIONET will have an important role to play in the *in situ* component of the CO₂ MVS capacity.

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ANNEX I: NRC QUESTIONNAIRE

To help evaluate the role EIONET could play within the CO₂ MVS capacity, a questionnaire was sent to the EIONET National Reference Centres (NRCs) for *Air Quality* and *Mitigation of Air Pollution and Climate Change*. The purpose of the questionnaire was to get an NRC perspective on the opportunities and challenges with respect to the adoption of in situ GHG measurement methods as part of local emissions monitoring.

The questionnaire, which consisted of seven multiple-choice questions, was sent out in January 2020 and the NRCs were given until 30th April 2020 to return the survey with their answers. In total 15 NRCs responded, with the results synthesised in the following figures.

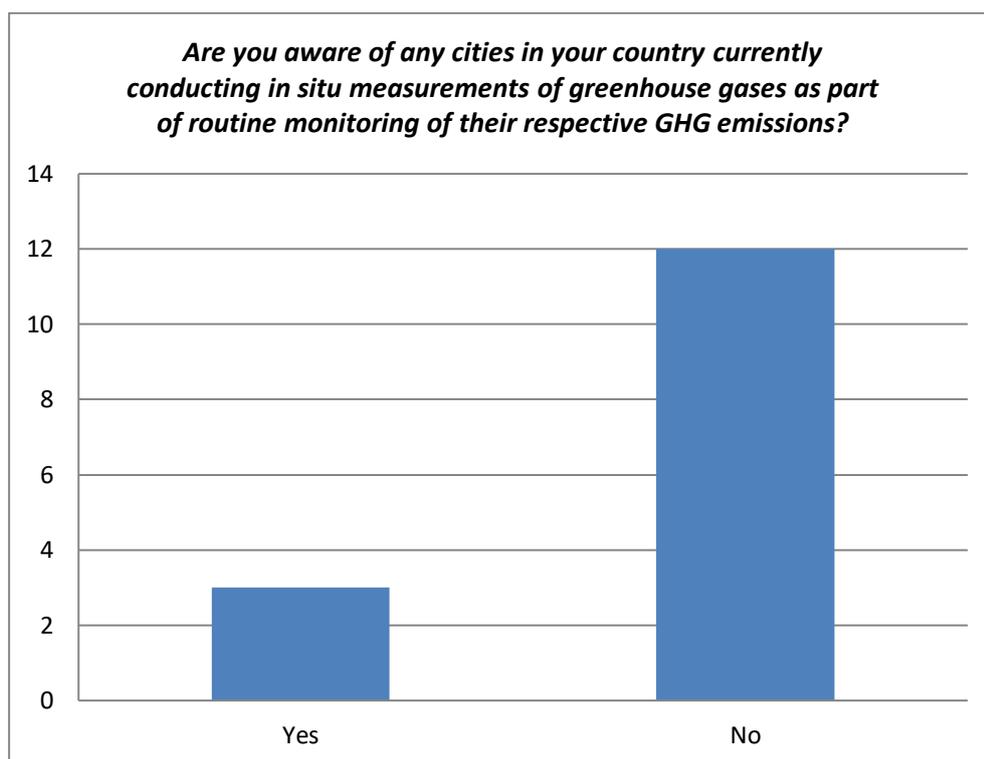


Figure A 1: NRC responses to question 1 - *Are you aware of any cities in your country currently conducting in situ measurements of greenhouse gases as part of routine monitoring of their respective GHG emissions?*

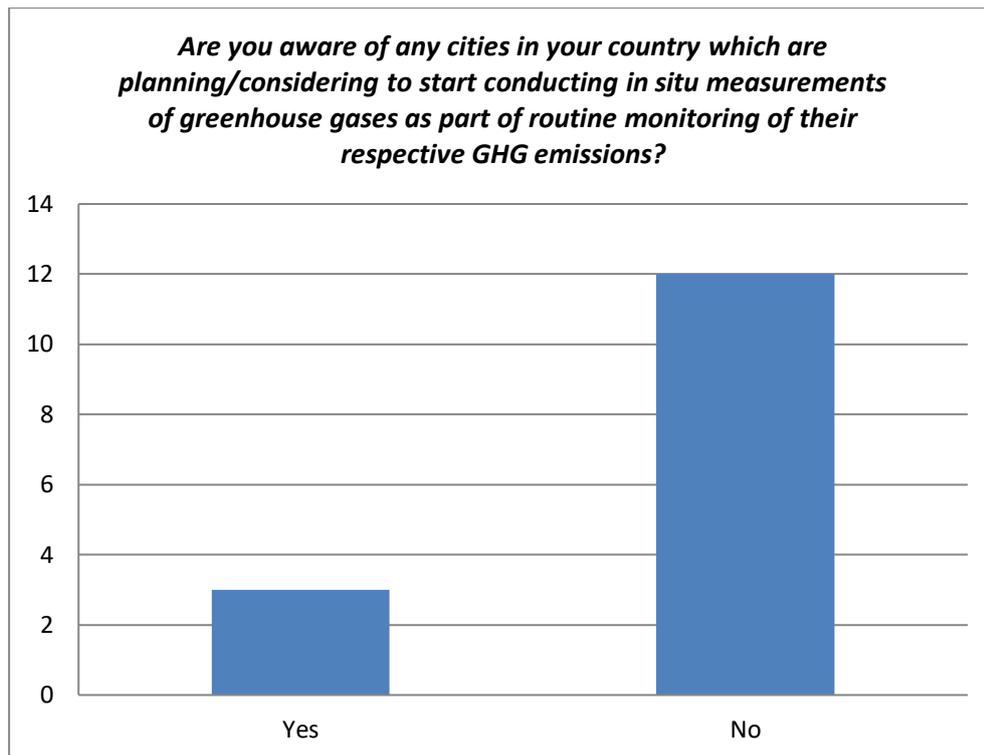


Figure A 2: NRC responses to question 2 - *Are you aware of any cities in your country which are planning/considering to start conducting in situ measurements of greenhouse gases as part of routine monitoring of their respective GHG emissions?*

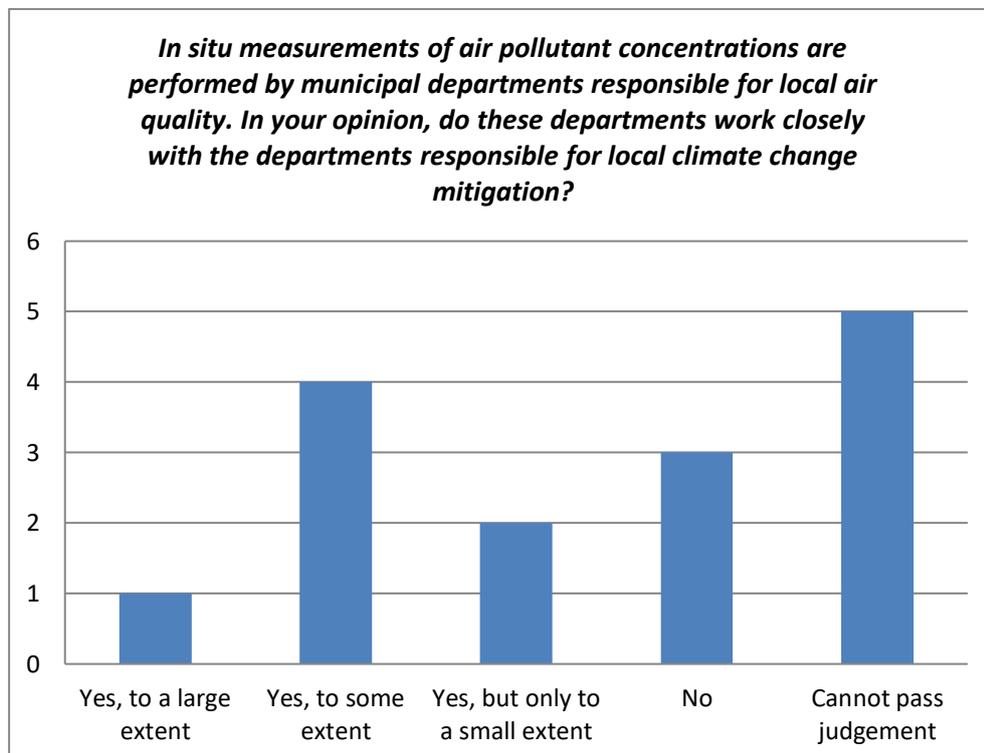


Figure A 3: NRC responses to question 3 - *In situ measurements of air pollutant concentrations are performed by municipal departments responsible for local air quality. In your opinion, do these departments work closely with the departments responsible for local climate change mitigation?*

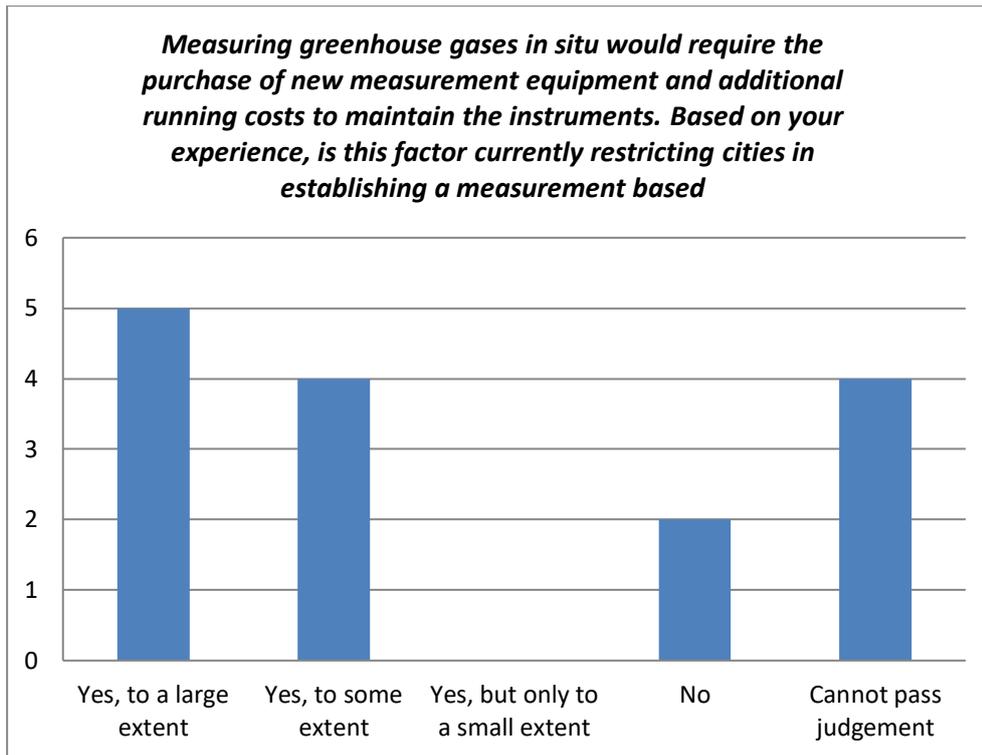


Figure A 4: NRC responses to question 4 - *Measuring greenhouse gases in situ would require the purchase of new measurement equipment and additional running costs to maintain the instruments. Based on your experience, is this factor currently restricting cities in establishing a measurement based system to monitor local greenhouse gas emissions?*

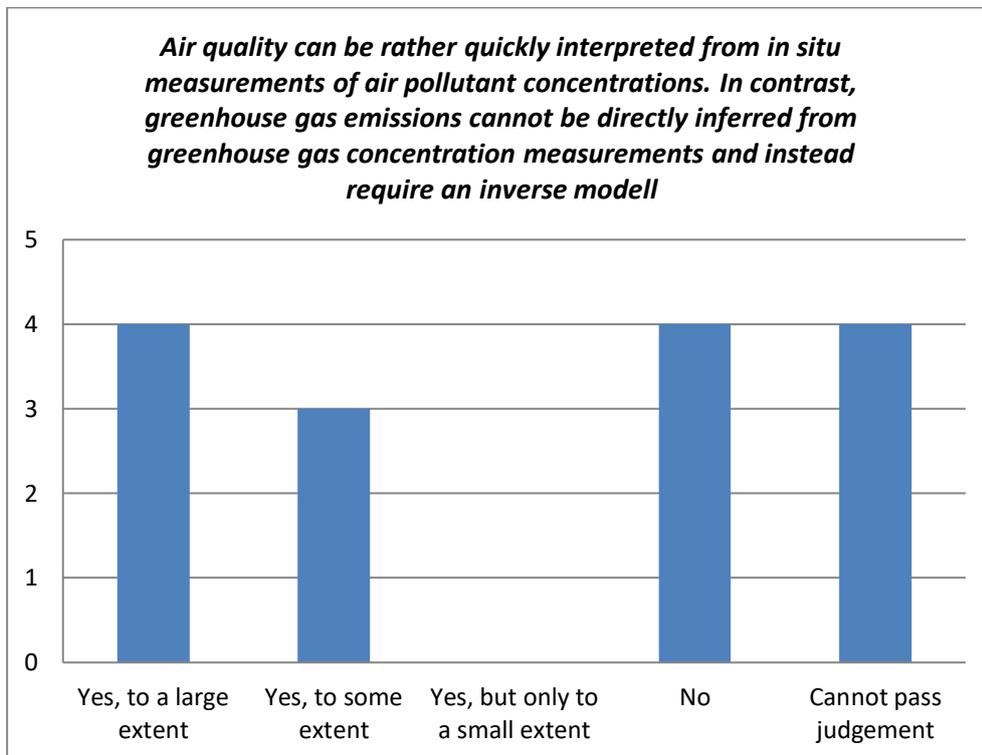


Figure A 5: NRC responses to question 5 - *Air quality can be rather quickly interpreted from in situ measurements of air pollutant concentrations. In contrast, greenhouse gas emissions cannot be directly inferred from greenhouse gas concentration measurements and instead require an inverse modelling framework to estimate local greenhouse gas emissions. Based on your experience, is this factor currently restricting cities in establishing a measurement based system to monitor local greenhouse gas emissions?*

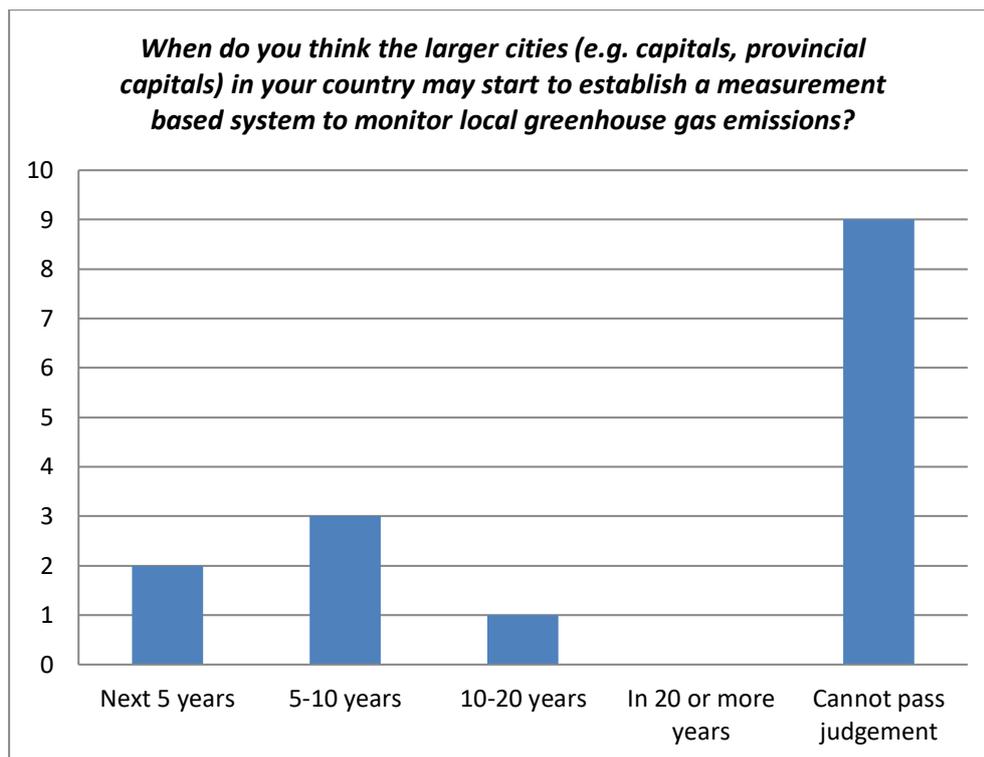


Figure A 6: NRC responses to question 6 - *When do you think the larger cities (e.g. capitals, provincial capitals) in your country may start to establish a measurement based system to monitor local greenhouse gas emissions?*

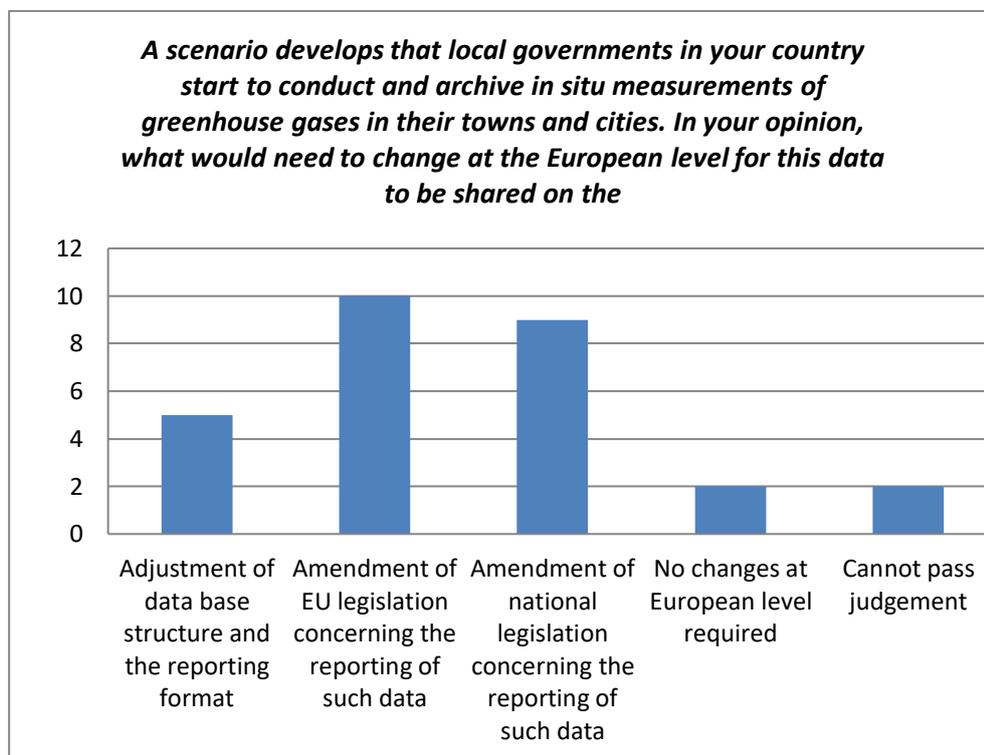


Figure A 7: NRC responses to question 7 - *A scenario develops that local governments in your country start to conduct and archive in situ measurements of greenhouse gases in their towns and cities. In your opinion, what would need to change at the European level for this data to be shared on the EIONET Portal?*

ANNEX II: LIST OF ABBREVIATIONS

ACE Atmospheric Chemistry Experiment

AirCore A sampling device that is usually released from the lower troposphere by a balloon

AIRPARIF Paris high-resolution local emission inventory

AIRS Atmospheric Infrared Sounder

AMAC Astigmatic Multi-pass Absorption Cell

ANN Artificial Neural Networks

AQ Air Quality

AQD Air Quality Directive

As Arsenic

B(a)P Benzo(a)pyrene

BEACO₂N Berkeley Atmospheric CO₂ Observation Network

CCL Central Calibration Laboratory

Cd Cadmium

CH₄ Methane

CO Carbon monoxide

COCCON COllaborative Carbon Column Observing Network

COP-21 21st session of the Conference of the Parties to the UNFCCC

CO₂ Carbon dioxide

CO_{2e} Equivalent carbon dioxide unit with which the radiative forcing of a given mixture and amount of greenhouse gas emissions is expressed an equivalent amount of CO₂ emissions with the same global warming potential.

C₂H₆ Ethane

CRDS Cavity Ring-Down Spectroscopy

ECD Electron Capture Detector

EC European Commission

EEA European Environmental Agency

EIONET European Environment Information and Observation Network

Empa Swiss Federal Laboratories for Materials Science and Technology

ERIC European Research Infrastructure Consortium

EU European Union

FID Flame Ionization Detector

FLUXNET Global network of linked regional networks of flux measurement sites

FTIR Fourier Transform Infra-Red

GAW Global Atmosphere Watch programme

GC Gas-Chromatography

GCoM Global Covenant of Mayors for Climate and Energy

GHG Greenhouse gas

G3IS Integrated Global Greenhouse Gas Information System

HA High-Accuracy

IASI Infrared Atmospheric Sounding Interferometer

ICLEI Local Governments for Sustainability network

ICOS Integrated Carbon Observation System

IPCC Intergovernmental Panel on Climate Change

LCS Low-Cost Sensor

LCMP Lower-Cost Medium Precision instrument

MIPAS Michelson Interferometer for Passive Atmospheric Sounding

MMR Monitoring Mechanism Regulation of European Union

MS Member states

MVS Monitoring and Verification Support

NDACC Network for the Detection of Atmospheric Composition Change

NDIR Non-Dispersive Infrared

NH₃ Ammonia

NIST National Institute of Standards and Technology of United States

NOAA National Oceanic and Atmospheric Administration of United States

NO_x Nitrogen oxides

NO₂ Nitrogen dioxide

N₂O Nitrous oxide

OA-ICOS Off-Axis Integrated Cavity Output Spectroscopy

OCO Orbiting Carbon Observatory

O₃ Ozone

P Atmospheric Pressure

Pb Lead

PAHs Polycyclic aromatic hydrocarbons

PM Particulate matter

PM_{2.5} Particulate matter which passes through a size-selective inlet with a 50 % efficiency cut-off at 2.5 µm aerodynamic diameter

PM₁₀ Particulate matter which passes through a size-selective inlet with a 50 % efficiency cut-off at 10 µm aerodynamic diameter

ppb Parts per billion

ppm Part per million

QA/QC Quality Assurance/Quality Control

RH Atmospheric Relative Humidity

SCIAMACHY SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY

SoCAB South Coast Air Basin

SF₆ Sulphur hexafluoride

SO₂ Sulphur dioxide

T Atmospheric Temperature

TILDAS Tunable Infrared Laser Direct Absorption Spectroscopy

TANSO Thermal And Near infrared Sensor for carbon Observation

TCCON Total Carbon Column Observing Network

TROPOMI TROPospheric Monitoring Instrument

UNFCCC United Nations Framework Convention on Climate Change

UUCON Utah Urban Carbon Dioxide Network

WCC-Empa WMO/GAW World Calibration Centre for Surface Ozone, Carbon Monoxide, Methane and Carbon Dioxide

WMO World Meteorological Organization

4DD 4th Daughter Directive