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Critical method needs in measuring greenhouse gas fluxes

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Abstract

Reaching climate goals depends on appropriate and accurate methods to quantify greenhouse gas (GHG) fluxes and to verify that efforts to mitigate GHG emissions are effective. We here highlight critical advantages, limitations, and needs regarding GHG flux measurement methods, identified from an analysis of >13 500 scientific publications regarding three long-lived GHGs, carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). While existing methods are well-suited for assessing atmospheric changes and local fluxes, they are expensive and have limited accessibility. Further, we are typically forced to choose between methods for very local GHG sources and sinks and their regulation (m^2-scaled measurements), or methods for aggregated net fluxes at >ha or km^2 scales measurements. The results highlight the key need of accessible and affordable GHG flux measurement methods for the many flux types not quantifiable from fossil fuel use, to better verify inventories and mitigation efforts for transparency and accountability under the Paris agreement. The situation also calls for novel methods, capable of quantifying large scale GHG flux patterns while simultaneously distinguishing local source and sink dynamics and reveal flux regulation, representing key knowledge for quantitative GHG flux modeling. Possible strategies to address the identified GHG flux measurement method needs are discussed. The analysis also generated indications of how GHG flux measurements have been distributed geographically and across flux types, which are reported.

1. Introduction

1.1. The international UN approved greenhouse gas (GHG) assessment system

In a world that strives to reduce GHG emissions and mitigate global warming, there is a need for reliable methods to assess GHG emissions and how they change over time. Extensive efforts in the United Nations Framework Convention on Climate Change (UNFCCC) and Intergovernmental Panel on Climate Change (IPCC) have led to procedures for national reporting of anthropogenic emissions of multiple GHGs [1] and the Paris agreement highlighted the need for transparency and capacity building to enable accurate assessments of GHG emissions [2]. The guidelines for reporting national inventories has been rigorously developed for consistency based on the best available science [1, 3].

The implementation of the internationally accepted GHG reporting system based on national inventories, is a remarkable achievement. This system was carefully designed at multiple implementation levels and is adapted for a situation of severe data limitation and lack of resources for measurements in many countries. Accordingly, inventories can be made in different ways depending on availability of data. The most basic Tier 1 reporting approach does not need own GHG flux measurements but is based on emission factors (EFs), i.e. factors yielding the estimated emission of each GHG by source type if multiplied with more commonly available proxy data, referred to as activity data. Activity data can be e.g. sales

or energy statistics, or area cover for relevant land use. The EFs are determined from the best available scientific information at the time of their development by carefully selected international working groups, after evaluation by a rigorous peer-reviewed process organized by the IPCC, and approval by the UNFCCC [1, 3].

The use of EFs are often considered adequate for some GHG fluxes, such as CO_2 emissions that are directly linked to fossil fuel use, but EFs are questioned for many other emissions for multiple reasons. EFs often lack mechanistic basis and their quality depends on the data used to develop them and thereby on the flux measurement methods generating this data. In practice, EFs are static (it can take decades to refine them) and only representative for the locations, time periods, and conditions of the studies used to derive them [1, 3]. Accordingly, the Tier 1 approach results in rather uncertain estimates for many GHG sources and sinks. Therefore, countries that have enough resources are encouraged to develop more detailed Tier 2 or Tier 3 GHG assessments for their inventories, based on more local and frequent proxy data or GHG flux estimates. Tier 3 assessments often depend on *in situ* GHG flux measurements aiming to consider relevant local conditions.

The different Tier levels means that the international GHG assessment system based on national inventories, upon which the accountability of the Paris agreement rests, has important mechanisms for gradual improvement by a development towards Tier 3. However, a main bottleneck for this development is the scarcity of relevant GHG flux measurements, preventing many countries from making Tier 3 level assessments, and inhibiting verification that inventories are accurately reflecting reality. This bottleneck is well-known and has been highlighted broadly [4].

In order to address these challenges, the Paris agreement emphasizes establishing accurate measurement, reporting and verification (MRV) systems for GHG mitigation efforts (e.g. [2] Addendum 1, Section III, Paragraph 98 and Section IV, Paragraph 105e with reference to [5] Addendum 1, Paragraph 61). Similar verification systems are adopted for various efforts to mitigate emissions, in some contexts also referred to as measuring, monitoring, reporting, and verifying systems. This does not mean that all GHG fluxes must be measured everywhere at all times, but successful and transparent MRV systems depend on comprehensive and cost-effective capacity to quantify all relevant types of GHG fluxes at whatever locations and times suitable, to support accurate, validated models, and verify that local mitigation efforts work.

1.2. Examples of key GHG knowledge gaps and the critical role of appropriate GHG flux measurement methods

The scarcity of GHG flux measurements has resulted in multiple critical knowledge gaps associated with both the national inventories and our general GHG flux understanding. The latter is increasingly important as feedbacks from anthropogenic climate and land-use change can have large effects on many GHG emissions in nature, which thereby becomes relevant to the Paris agreement [6, 7]. The below examples are provided with underlying positive recognition of the GHG reporting system via national inventories, and of the past knowledge development regarding GHG fluxes in general, as extraordinary achievements. The intention is to highlight the critical role of GHG measurement methods behind main biases or knowledge gaps.

1.2.1. GHG models depend on flux measurements

Various types of models have been developed for GHG flux estimation [1], but they all critically, directly or indirectly, depend on representative and reliable flux data. Models are only as accurate as the data they are built upon and validated against. Unfortunately, the spatiotemporal resolution of available flux data is often insufficient for robust model validation [4, 8]. In many cases, data may be entirely missing at national and local scales, leading to a heavy reliance on assumptions that data from other locations or time periods are valid. Hence, behind current GHG inventories and climate models, there are many layers of models and assumptions, considerable uncertainties, and a scarcity of local *in situ* measurements [4, 9, 10]. The data scarcity leads to fundamental uncertainty that cannot be resolved without adequate flux measurements.

1.2.2. EFs—good enough for some fluxes but not all

It should be noted that some key GHG fluxes, such as the large CO_2 fluxes from combustion of fossil fuels, may be assessed adequately via EFs from indirect data on energy consumption [1]. However, although these fluxes triggered climate change and have to remain a main target for flux mitigation, they are not the only GHG fluxes of importance. Anthropogenic fluxes other than CO_2 from fossil fuel combustion, together with the distributed landscape GHG fluxes influenced by expanding and intensified land use (often expressed as agriculture, forestry, or other land use; AFOLU), and climate feedbacks [6, 7], jointly account for the greatest uncertainty in global GHG budgets [11]. For comparison cumulative land-use change emissions have, although recognized to be difficult to quantify accurately, been estimated to contribute up to ca 30% of the total

cumulative anthropogenic CO_2 emissions 1850–2020 [11, 12].

1.2.3. Anthropogenic and natural fluxes need joint consideration

The national inventories only include flux types approved by the UNFCCC as anthropogenic and having enough associated flux data. Accordingly, the inventories estimated via EFs are subsets of the total fluxes. This does not match the need to quantify how all main GHG flux types change over time to assess effects on global temperature, mitigation success, or anthropogenic perturbations on fluxes classified as natural. Anthropogenic perturbations including climate feedbacks, on the balance between the very large gross C fluxes driven by organic matter respiration and photosynthesis, could greatly influence net landscape C emissions and have great consequences for the efforts to fulfil the Paris agreement. Consequently, both anthropogenic and natural GHG fluxes and their change over time need adequate quantification.

Further, specific anthropogenic emission sources are often constrained by inversions where *a priori* flux estimates from all known sources and sinks are used to match the atmospheric levels over time [13, 14]. In this context it is problematic that major GHG fluxes have not been recognized as globally important until recently, e.g. CO_2 and CH_4 fluxes from inland waters (e.g. reservoirs, lakes, ponds, rivers, streams, and ditches) [15, 16], CH_4 emissions through tree stems [17, 18], CH_4 uptake by tree canopies [19], and loss of GHG sinks when agricultural practices transform natural soils [20, 21]. Not knowing or accounting for major sources or sinks—regardless of anthropogenic or natural—can result in considerable bias [14]. Thereby, limitations of flux measurement methods can result in incomplete *a priori* flux estimates, uncertain relative importance of different sources and sinks, and ambiguities on what are the most effective ways to reach climate goals.

1.2.4. The AFOLU flux measurement challenges

Assessing fluxes distributed across landscapes require measurement methods that can identify and quantify local and episodic emissions in representative ways. Such fluxes can contribute large shares of the total emissions. For example, streams and rivers, estimated to only cover 0.5% of the continental area, are hotspot CO_2 emitters [22] found to release 1.8 Pg CO_2–C yr^{-1} to the atmosphere (comparable with the land sink or ocean uptake of carbon) [15]. Similarly, CH_4 fluxes from areas termed flooded land in the national inventory terminology, including human-made inland waters, can represent large climate sensitive hot-spot fluxes [23–25] that are anthropogenic but were previously unaccounted for. In terrestrial environments, moisture dynamics control the soil GHG exchange [26–28], which makes local land use management in agriculture or forestry important for GHG fluxes. The agricultural situation was expressed in a recent US Department of Agriculture report: 'The most accurate way of estimating emissions is through direct measurement, which often requires expensive equipment or techniques that are not feasible for a single landowner or manager. On the other hand, lookup tables and estimation equations alone often do not adequately represent local variability or local conditions.' [29]. The GHG mitigation plans in some countries to generate negative emissions by enhancing AFOLU sector CO_2 sinks, further stresses the need for effective measurement capacity to verify locally the extent to which negative emissions are achieved and sustained. Overall, proper flux measurement capacity is necessary to identify and quantify the AFOLU fluxes of importance across time and space.

1.2.5. Examples of measurement challenges regarding CH_4

There are multiple examples of biased CH_4 inventories. At the global level the GHG reporting can be validated by comparing the total bottom-up national inventories with atmospheric concentration changes. Such a comparison based on isotopic composition, recently revealed that bottom-up inventories underestimated the anthropogenic CH_4 emissions from fossil fuel extraction, distribution, and use, by as much as 60% [30]. A potentially related finding is that the oil and natural gas CH_4 emissions from various leakages associated with extraction and distribution, are not fully accounted for by EFs: Local measurements yielded fossil CH_4 emissions 1.5- to 2-fold greater than official inventory estimates in the US [31]. Effective measurements to target such leakages could effectively mitigate emissions once known [32, 33]. Similar findings of novel measurements allowing quick mitigation of identified leakages were recently reported from a sewage treatment plant and a biogas production facility [34]. A recent multi-sector synthesis concluded that quickly mitigating CH_4 emissions could reduce global warming rates in the order of 30% and 0.25 °C the coming decades [35]. This exemplifies the need of appropriate measurement capacity to enable effective emission mitigation.

1.3. Scope

As illustrated by the above examples: While some GHG emissions can be estimated from EFs or other modeling without local measurements, capacity and means to accurately quantify and localize many types

of GHG fluxes, are important to understand changes in atmospheric levels and make effective policy and mitigation priorities. Overall, many of our main GHG-related challenges and knowledge gaps depend on the scarcity of GHG flux measurements, which we hypothesize is linked to which GHG flux methods are available and their characteristics. This study therefore investigates how well the available methods for GHG flux measurement match societal needs and commitments to the Paris agreement focusing on the question:

What measurement methods are available for quantifying GHG fluxes, what are their benefits and limitations, and how accessible are they for broad societal use?

This general question can be translated to case-specific questions. For example: Given the goals of negative emissions in agriculture and forestry—are there cost-effective and easily used methods for land-owners or authorities to determine if fields or forest plots subjected to practices to increase the C-uptake actually generate expected negative emissions? Are there cost-effective and easily used methods for facility-owners or authorities to evaluate if mitigation strategies in wastewater treatment, biogas, natural gas, oil refinery, or other facilities, actually lead to reduced emissions from the facility or infrastructure in focus?

2. Methods

2.1. GHG flux measurement methods

Information about different GHG flux measurement methods were gathered from a systematic analysis of titles and abstracts of GHG flux-oriented papers, followed by in-depth reading of selected papers (described below). The intention was to generate an overview of current methods, including a description of measurement principles and information about spatiotemporal resolutions of measurements, potential applications, advantages, limitations, and accessibility outside academia.

The application of many of the GHG flux measurement methods requires consideration of local conditions on a case-by-case basis. It is not possible in the scope of this study to analyze case-specific applications of all the methods. We therefore focus our analysis on the fundamental case-independent principles and characteristics of the GHG flux measurement methods.

To make the systematic literature analysis of GHG flux measurement methods, we searched the Web of Science database by topic using the following search string:

TS = ((((greenhouse gas* AND flux*) OR (greenhouse gas* AND emission*)) AND measure*) OR (((greenhouse gas* AND flux*) OR (greenhouse gas* AND emission*)) AND map*))

The string was selected after testing multiple word combinations and making preliminary assessments of titles to ensure that the search was sufficiently broad to include a variety of methods measuring GHGs in different environments. The literature search which defined the frame of this review was performed on 26 July 2020 and yielded 11 992 papers published between 1990 and July 2020. Sub-searches on known common measurement approaches e.g. eddy covariance, remote sensing, flux chambers, etc gave 200—>700 hits each, confirming that they were well represented.

The 11 992 hits were divided into groups of 500 papers (the last group containing 492). Six researchers divided the papers and performed a title scan. The papers were classified according to the likelihood that the paper would include detailed method description(s) (by the priority codes: 0 = not likely, 1 = unclear, 2 = probably as part of methods description, 3 = high—methods in focus, and 4 = very high—method development/comparisons in clear focus). For the 3481 papers with priority codes 2–4, the abstracts were read. A focus on GHG flux measurements was confirmed in 2041 papers which were assessed further to retrieve information on flux types, method class, environment, gas, spatial scale, mobility, platform, biome and continent (table S1).

During the title and abstract review process, multiple meetings took place to cross-calibrate interpretations and usage of specific words, terms, or content, and discuss the assignment of the priority codes. Methods to quantify GHG fluxes are often based on combinations of measurements of GHG concentrations and ancillary variables, such as time, temperature, pressure, and air motion. While a review of specific techniques to measure ancillary variables is beyond the scope of this paper, we focused on overall methods to quantify GHG fluxes (including the above-mentioned measurements of all relevant variables). Accordingly, methods solely used for determining GHG concentrations, or GHG accounting based on EFs and activity data were disregarded. Studies based entirely on secondary data from other studies or modelling, without indicating any actual original measurement efforts, were also omitted. Each title and abstract did not contain information in all categories; hence the number of studies

for each category is different. For example, information on studied GHGs, environments and continents of study, was clearly indicated in the titles and/or abstracts of 1941, 1996 and 1308 papers, respectively.

The papers that were classified with priority 3 or 4 (i.e. with methods in focus, i.e. 577 papers), were studied in greater detail to to review principles and features of the flux measurement methods. The draft of this overview was sent by email to an independent reference group of 21 persons known for their method expertise and to others hearing about the study and expressing interest. This group was also encouraged to spread the draft to others with potential interest to provide feedback (box S1 in supplementary material show the letter). The method overview was updated and adjusted according to the responses. Given the large number of studies considered, and this external verification with representatives of multiple organizations within and outside academia, it seems likely that the main types of currently used GHG flux measurement approaches have been identified.

To incorporate the possible method development published during the review process another supplementary Web of Science literature search according to above was made for the period of 27 July 2020 to 13 April 2022, generating 2661 additional papers. Based on information in the titles and abstracts, these papers were examined only for possible new GHG flux measurement methods not identified in the above analyses, and the new information obtained was incorporated in the method descriptions and analyses.

3. Results and discussion

3.1. What GHG flux measurement methods have been used?

Overall, the large number of papers fitting the search string indicates extensive work on performing and improving GHG flux measurements. The identified GHG flux measurement methods are presented in table 1 and box S2 (supplementary material), along with descriptions of the method principles, advantages, limitations, accessibility, and example references. These descriptions also aim to supplement previous reviews which provides more technical details on groups of methods [8, 36, 37]. This study also focuses more on feasibility for broad societal use than past reviews.

The GHG flux measurement methods can be broadly categorized as enclosure-based or open. In enclosure-based methods, the control volume (the air volume to which the flux is determined) is constrained physically by an enclosure, and the footprint area (the area from which the flux enters the control volume) is thereby also well-defined. The change of GHG concentrations over time in the enclosure quantifies the flux. The underlying principles are straight-forward and optimizations for different applications, and to reduce measurement complexity, can often be made. As an example, small inexpensive GHG sensors are in some cases sufficient in enclosure methods [38, 39]. This removes the need of expensive analytical instrumentation and could make GHG flux quantification affordable in most parts of the world [40]. Two main limitations of enclosure-based approaches are (a) that enclosures prevent open exchange of gas and material with the surroundings, which may influence the GHG flux, and (b) that enclosures, for practical reasons, often have small footprints which make extrapolations to larger areas uncertain. Method development for enclosure methods often aims at overcoming or minimizing these limitations.

Open methods are based on measurements in open air and are thereby considered non-invasive. This, combined with the much larger footprint areas than enclosure-based methods, and possibilities of automation, has contributed to their increasing popularity. Eddy covariance, tracer studies, various types of inverse modelling, and open mass balances, are examples. Open methods require information about GHG concentrations, air movements, and sometimes other ancillary variables, to estimate flux and footprint location and size. The limitations of open methods include: (a) higher demands on measurement accuracy and precision and consequently more expensive and power demanding equipment, (b) greater overall complexity, (c) greater know-how needs to evaluate data, and often, (d) dependency on assumptions and simplifications regarding how the air moves in the boundary layer near the measured surface. A large and variable footprint area, continuously changing location and size with wind speed and direction, can make it challenging to distinguish fluxes from different sources in the footprint and separate the variability in time from variability in space. There are many types of open methods—some estimate flux from single point measurements, while others from multi-point measurements or column densities of gases along lines. In addition, there are different ways to determine air motion and resulting mass transport, which are critical for GHG flux quantification.

Enclosure-based and open methods can contribute to large-scale GHG emission assessments in different ways. Measurements targeting specific sources at local scales are typically extrapolated by source area and summed to generate bottom-up assessments for different source categories [37]. At larger scales, open measurements of overall atmospheric GHG content, combined with air transport models and estimates of atmospheric GHG residence times, generate top-down assessments.

Table 1. Brief characteristics of the identified method types. More details are provided in box S2(a)–(g) in the supplementary material.

Principles (P) and applications (A)	Concerns (C) and benefits (B)
Static and flow-through flux chambers (box S2a)	
P: An enclosure is created over the target area and the change in amount of GHG with time in the enclosure represents the flux and is measured manually or by sensors. Static chambers (FC) are closed during the measurement period and new measurements are started by venting the chamber to regain initial conditions. Flow-through chambers (FTFC) have continuous through-flow of background gas and this gas flow rate plus GHG concentrations in and out from the chamber are measured. **A:** Useful for fluxes across surfaces that can be temporarily covered by an enclosure with acceptably flux bias. Typical applications are fluxes across soil-air, water-air, sediment-water, and vegetation-air interfaces. Parts of facilities can be used as flux chambers in themselves if GHG concentrations and residence times can be quantified (e.g. ring tanks on farms, or pipes, tanks, and rooms).	**C:** Requires that the area of interest can be enclosed. Careful design needed to minimize enclosure effects on flux measurements, and for some applications enclosure effects cannot easily be addressed. Small area coverage per chamber leading to uncertainty in flux estimation for large areas. FTFCs are sensitive to uncertainty in gas flow rates, pressure gradients, and gas concentration measurements, making precision, accuracy, and sensitivity lower than for FCs. The FTFC temporal response is delayed, and dynamics are smoothed out. **B:** Conceptually simple; all variables measured. Well-defined footprint. Easy replication to assess variability in space or time is straight-forward. FCs have high sensitivity, high mobility, and are comparatively simple to use and inexpensive in terms of equipment. Low education threshold. FTFCs can be operated continuously in some applications.
Flux at outlets of well-defined point sources (box S2b)	
P: Point source enclosure (EPS) are based on temporary covering of the emission source with e.g. a gas sampling bag. The filling rate gives the gas flow, which together with the concentration in the bag yields the flux. Open approaches at the point source (OPS) are based on measurements of both gas concentrations and the gas flow of the outlet air without enclosure. **A:** E.g. oil and gas industry, biogas production, gas pipelines, well confined combustion processes, ventilation outlets (mines, indoor air) and livestock emissions.	**C:** Efficient for known point sources only. Sensitive to precision and accuracy of GHG concentrations and air transport. **B:** Conceptually simple and straight-forward approaches for known point sources.
Ex situ fluxes—incubations (box S2c)	
P: Incubation approaches (EI), by which material of interest (e.g. sludge, manure, soil, sediment, water) are confined in incubation vessels under controlled and well-defined conditions. The gas concentration in the vessel is followed over time to determine flux. **A:** Suitable for studies on potential gas production or uptake rates under controlled incubation conditions and for studying how such processes are regulated.	**C:** Results reflect incubation conditions only—not *in-situ* conditions. Time-consuming and dependent on suitable facilities. There is often a need for many replicates in the incubations to handle uncertainties. **B:** Enable possibility to determine influence of various factors on the potential gas flux. This can reveal cause-effect relationships in flux regulation.
Micrometeorological approaches by point measurements in ambient air (box S2d)	
P: Fluxes calculated from gas concentration and transport in open air. Eddy covariance (EC) is based on the correlation between high frequency, high precision measurements of GHG concentrations and 3D movements of air (wind eddies). Relaxed eddy accumulation (REA), collect up-going and down-going air separately allowing use of a slower gas analyzer than in the EC approach. Flux estimation from gradients (GRAD), use transport rate coefficient and the difference in concentrations between two sampling points e.g. at different heights above ground. **A:** EC is commonly used for field scale net GHG exchange over time. REA or GRAD are alternatives when fast GHG analyzers are not available.	**C:** Footprint size and location moves continuously depending on wind speed and direction and temporal and spatial variability cannot always be separated. Difficulties to link fluxes to heterogeneous within-footprint features. EC and REA do not work at low wind conditions. Lateral fluxes can lead to bias. High-accuracy measurements to resolve small fluctuations in gas concentrations or air transport over short times can be a challenge and demands costly equipment. Data filtering, corrections, and gap-filing can be challenging. High complexity leads to high know-how needs for the QA/QC process reducing transparency. In GRAD approaches footprint sizes and locations differ among measurement points and dependency on modelled eddy diffusivities add uncertainty. **B:** No disturbance of the emitting surfaces. Often suitable for automated long-term measurements. Integrate net gas exchange over large areas. EC has become a standard technique for field scale greenhouse gas net exchange studies. For REA and GRAD, slower gas analyzers can be used.

(Continued.)

Table 1. (Continued.)

Principles (P) and applications (A)	Concerns (C) and benefits (B)
Open approaches based on column density, tracers or inverse modelling (box S2e)	
P: Perimeter facility line measurements (PFLM) are based on open path spectrometers (e.g. infrared, tunable diode laser, frequency comb laser spectrometers) with reflectors to measure GHGs along lines upwind and downwind of facilities to make flux estimates from mass balances. In tracer flux measurements (TFM), release of a separate tracer gas at a known rate at the source of the target gas allows flux estimation from the target to tracer gas concentration ratio downwind. Inverse modeling (IM) approaches track gas movement trajectories backwards from a sensor to the source area using e.g. Gaussian models or backward Lagrangian stochastic (bLS) dispersion modeling. **A:** PFLM seem primarily used for potentially high emission sources (agriculture, waste, oil and gas sectors). TFM are used in e.g. agriculture, waste, or energy sectors, while IM has been used both for well-known, small-scale emission sources, or as a part of large-scale assessments.	**C:** PFLM accuracy depend on knowledge of air movements and comparability between up-wind and down-wind measurements. TFM can be accurate if the target gas is emitted from a well-known point source, but situations with heterogeneous emissions or additional unknown sources of the target gas are challenging. There is also a dependence on appropriate weather and wind conditions and it is challenging to maintain long-term measurements. IM performance depends on sensors, area characterization, and turbulent transport model performance. **B:** No disturbance of the emitting surfaces. For PFLM a high degree of automation possible. TFM is a conceptually straight-forward and rather accurate for known point sources. IM is flexible and useful for many types of gases and source areas. Both point and diffuse sources can be captured.
Open approaches based on mass balances (box S2f)	
P: Mass balance approaches at local scales (MBLS) yield the net areal flux by the difference between export and imports of gas from/to a control volume. MBLS can be combined with PFLM. In atmospheric mass balances (AMB)—a large scale type of inverse modeling—the change over time of GHG concentrations across the whole atmospheric column is combined with atmospheric sink rate to estimate net fluxes of regions. Stable isotope information can be used to constrain different source types. The boundary layer budgeting (BLB) estimates flux from gas concentration changes during periods of boundary layer stratification periods. **A:** General approach used for local to large-scale emissions. AMB is used for global monitoring of atmospheric GHG content, and determination of total large-scale fluxes.	**C:** Depend on accurate concentration and air flow measurements, preferably at multiple points in 3D. In open control volumes (e.g. in open air) sufficient measurement locations to properly map gas concentrations and bulk medium flow to and from the control volume is critical. Measurements at multiple points in three dimensions around the control volume at adequate resolution is needed for best accuracy. Complex air mixing can make gas source attribution challenging. **B:** At small scales the bulk medium can be measured and no assumptions on boundary layer conditions are needed (MBLS). AMB is suitable to assess atmospheric GHG content. BLB can under favorable conditions represent a large-scale analogue to FC measurements.
Optical approaches with potential to map GHG concentrations and flux (box S2g)	
P: Passive optical approaches (POA) relying on emitted background radiation. Different infrared spectroscopic scanning techniques with relatively high spatial and spectral resolutions allows mapping of gas column densities, which together with wind patterns allow estimation of GHG fluxes. In other cases, separate independent wind velocity measurements are needed to derive fluxes. Background distances have to be measured. In active optical approaches (AOA), the radiation needed to detect gases is actively emitted from the instrument, and gas column densities are determined from the backscatter. **A:** POA and AOA are novel and under evaluation but are potentially widely useful. Hitherto, these types of instruments have often been tested for oil and gas industry emissions.	**C:** POA rely on background radiation i.e. requires temperature contrasts and does not work during some weather conditions. The high current costs of the hardware with high enough spectral resolution limits widespread use. Low accuracy if the spectral resolution is too low (separate gases cannot be distinguished). The performance details of AOA have in some cases not been released for instruments owned and operated by private companies illustrating a lack of transparency. Several AOA instruments are under development. Therefore cost, performance and future availability for various applications are at present unclear. **B:** Does not disturb the target area. Some hyperspectral POA generate high-resolution data for both gas column densities, distances and wind speed simultaneously allowing detailed flux quantification at high spatial resolution and resolving spatial variability in the scene. Capacity to detect new unknown sources and calculate their fluxes in many types of environments. AOA have the additional benefit of not depending on background temperature contrasts.

Enclosure-based and open methods were further classified and described in table 1 and box S2 (supplementary material) as follows:

Flux measurement methods	
• Static and flow-through flux chambers (enclosure-based)	Box S2a
• Methods for outlets of well-defined point sources (including open and enclosure-based methods)	Box S2b
• *Ex situ* fluxes—incubations (enclosure-based)	Box S2c
• Micrometeorological methods by point measurements in ambient air (open)	Box S2d
• Open methods based on column density, tracers or inverse modelling	Box S2e
• Open methods based on mass balances	Box S2f
• Optical methods with potential to map GHG concentrations and fluxes	Box S2g

Each measurement method has advantages and limitations, and the suitability depends on the flux type and environment in focus (table 1; box S2). Methods for point source fluxes can be reliably tested by controlled emissions. However, it remains challenging to assess how enclosure-based and open methods compare, despite numerous attempts [37, 41–45]. A fundamental challenge in cross-comparisons is the difference in the spatial scales of the methods (e.g. comparing footprint areas of m^2 versus km^2), making it difficult to evaluate the reasons behind potential GHG flux deviations among methods. Accordingly, the results are mixed. Some comparisons yielded relatively close correspondence (within 20%), while in others enclosure-based and open methods differed substantially (up to 88%) [37]. Importantly, reference methods that can reliably assess fluxes at multiple spatiotemporal scales simultaneously to cross-validate enclosure-based and open approaches remains to be developed and established.

The literature analysis showed that CO_2, CH_4 and nitrous oxide (N_2O) were measured in 51%, 62% and 58% of the papers, respectively (many studies measured multiple gases). Flux chambers were predominantly used (61% of the studies), while micrometeorological measurements, dominated by the eddy covariance approach, were used in 20% of the papers. Incubation assessments and remote sensing-based methods were used in 8% and 5% of the papers, respectively.

Agricultural and soil environments (excluding rice fields) and wetlands were the most studied environments, followed by other types of open land, forests, aquatic environments, and livestock-, rice-, manure-, and waste-related emissions (figure S1). The three main GHGs (CO_2, CH_4, and N_2O) have all been given relatively similar attention in all environments. However, most studies were performed in natural or AFOLU environments, and substantially fewer studies made *in situ* measurements of waste-, urban- and combustion-related fluxes. This indicates that emissions from many large GHG sources are modelled from very limited actual measurements. The lower number of measurements for CO_2 emissions from fossil fuel combustion is logical given low expected uncertainties for these emissions compared to other fluxes [46]. However, there are cases where waste-related, urban, and industrial- fluxes can be significantly biased, as recently exemplified for CH_4 emissions from a waste treatment plant [34]. The distribution of flux measurements among sectors and gases in our data stem from literature found with a search string focusing on methods and not representative coverage of all environments. Nevertheless, the results generate questions about our actual knowledge on fluxes and their development in the environments for which data appear scarce, which warrants further investigations.

Of the 1308 studies that clearly indicated the geographic locations in the titles and/or abstracts, 82% were performed in Asia, North America or Europe, illustrating skewed representation of certain regions (figure S2). Only 68 (5%) and 35 (2,5%) studies were performed in South America and Africa, respectively. This geographic imbalance is especially pertinent as it has been suggested that tropical environments are of fundamental importance for GHG emissions and climate feedbacks [47–49].

A general pattern is that few of the GHG flux measurement methods are easily accessible independently from academic research (box S2). The exceptions are some methods for point source emissions, in e.g. industry or in the waste handling sector, that are used by consultants providing GHG flux assessment services, and latterly also commercial remote sensing-based flux estimation for the largest point sources. Most of the methods are far too costly for local independent use, with portable gas analyzer equipment costing >10 000 euros. In addition, many methods depend on access to advanced laboratories for reference measurements and calibration. Available methods are also often research-oriented in terms of know-how, given that method details need to be adapted to and tested in each local setting and few methods can be used in similar ways for all types of environments. This makes most methods costly and inaccessible for local societies and individual actors wanting to monitor their fluxes or validate their mitigation efforts. Some approaches generating GHG concentration or column density data, e.g. by satellites, seems to become increasingly available to the public domain [50]. So far, remote sensing detects

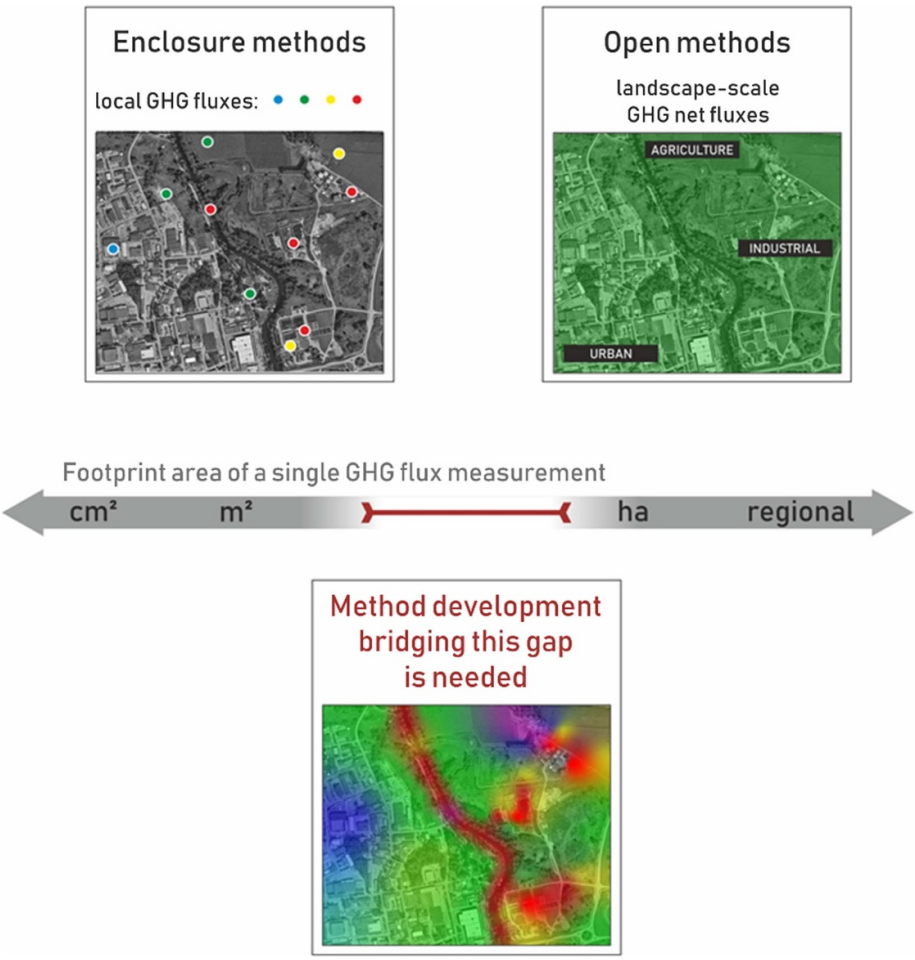

Figure 1. Illustration of the method gap limiting our understanding of source/sink attribution and regulation of GHG fluxes. Enclosure-based methods yield local flux measurements at m^2 scales that are challenging to extrapolate to large scales (upper left; different colors denote different flux levels). Open methods yield net flux over larger areas (upper right; the mean net flux for the whole area detected as denoted by the 'mean color'). Needed GHG flux measurement methods bridging this gap and detect fluxes at multiple scales simultaneously are visualized in the lower panel, where differences between local fluxes and their localization would be resolved over the entire landscape. Please note that methods to quantify GHG fluxes (i.e. not just GHG concentrations) are in focus here.

primarily the large emission sources and the generation of quality assessed gas flux rates (e.g. beyond the column density data) requires specialist knowledge. Commercial services therefore often target high emissions related to the energy sector and it remains challenging to verify mitigation efforts in most local settings. Hence, a key conclusion from this study is that development of more affordable, user-friendly, yet accurate GHG flux measurement methods, broadly accessible to societies, and being suitable for monitoring at multiple scales and for multiple flux types, are urgently needed to guide and verify mitigation efforts.

Moreover, this study highlights a critical method gap that concerns the lack of methods capable to detect fluxes at multiple scales simultaneously (figure 1). In order to link local GHG sources and sinks and their regulation to comprehensive landscape fluxes, which is essential for developing accurate dynamic landscape GHG flux models, measurement methods are required that provide simultaneous information across multiple spatial scales, and particularly from m^2 to ha or km^2 scales because m^2 scale understanding is often needed to understand flux regulation, while km^2 scale knowledge is needed for large-scale flux assessments. This scale shift largely corresponds to the identified gap between enclosure-based and open methods.

The gap in suitable methods bridging key scales, and the high cost and complexity of available measurement methods, is directly linked to the specific questions expressed in section 1.3 including: Are there cost-effective and easily used methods for landowners, facility-owners, or authorities to determine GHG fluxes from local areas or facilities in response to changes over time? With the exception of the very large point sources being quantifiable by remote sensing, the answer in the surveyed literature is 'not yet'.

3.2. Ongoing developments

Multiple efforts towards improved measurement approaches are ongoing. There has been considerable progress in measuring atmospheric GHGs by satellite remote sensing at increased spatiotemporal detail [4, 50–54] and more is to be established via initiatives such as the MethaneSAT and the MERLIN LiDAR missions. Ground-based measurements at higher spatial resolution are repeatedly requested and attempted [4, 52, 55]. Initiatives to develop sensor networks for monitoring air pollution and some GHGs in cities are pioneering low-cost automated sensing systems successfully for e.g. CO_2 [56, 57]. The suitability of low-cost CH_4 sensors for such systems are under evaluation [e.g. 58, 59]. The rapid general development of Internet of things, automation, and artificial intelligence, is generating synergies and provides communication infrastructure for GHG sensor networks. The combination of vertical perimeter line measurements and mass balance approaches (box S2) are facilitated by recent mobile techniques to map GHG content in upwind and downwind air columns [60, 61]. This could become a very suitable approach to rapidly assess fluxes from many types of target areas based on straightforward and consistent principles and calculations, although some challenges regarding consideration of wind speed profiles and dispersion, and their interaction with GHG concentration profiles, remains. Emerging technologies to visualize GHGs *in situ* at high resolution [62, 63] are promising. Such techniques have the potential to bridge the highlighted method gap (figure 1). Further, efforts to coordinate and take advantage of the diversity of methods are also making key progress. One example is the Integrated Global Greenhouse Gas Information System (https://ig3is.wmo.int/en).

Accordingly, there is substantial progress and promising ongoing developments. Yet, direct flux measurements are still scarce compared to needs and reductions in costs and complexity are still required for broad local and independent GHG assessments, model validation, and mitigation verification. The highlighted method gap (figure 1) and the limited ability of GHG data to support a robust MRV process, require additional capacity development as also suggested elsewhere [64]. Major challenges remain that require exploration, development, and evaluation of many ideas in parallel. The remarkable success in quickly developing needed method capacity and transferring such capacity from research to broad societal use, associated with the Covid pandemic is inspiring and indicates that once awareness and societal priority are established, rapid progress of complex tasks are possible. If the promising recent developments and continued method development are supported and coordinated with societal needs, there are good chances for rapid progress enabling more frequent and accurate *in situ* GHG flux measurements.

3.3. Ways forward

With climate change already associated with more than 150 000 deaths yearly and the number expected to be 250 000 between 2030 and 2050 [65], development of methods better supporting effective and accurate flux monitoring, model validation, and mitigation verification, is key to manage one of the grand challenges facing humanity. This is well recognized and the IPCC and UNFCCC assessment frameworks are carefully considering transparency, completeness, consistency, and comparability [e.g. 66], and use of appropriate methodologies [67]. The established GHG reporting systems have been carefully and successfully organized to establish internationally standardized, consistent and comparable national GHG inventories and include mechanisms to stimulate and incorporate method improvements. There is awareness that we need better capacity to monitor *in situ* GHG fluxes in cost-effective ways at appropriate spatiotemporal resolution to connect the inventories to local *in situ* observations. However, methodological issues and how they hamper transparent progress towards the climate goals seem not yet widely debated outside expert communities.

Similar fundamental method challenges are common e.g. regarding water, food, healthcare, trade, and defense, and with examples from some of these sectors we discuss in the supplementary material a number of key elements to enforce and facilitate method development to address grand challenges. Six elements are discussed, including societal awareness and engagement, leadership, organizational structures and coordination, resource allocation, knowledge and communication, and capacity building (figure S3), highlighting examples of how they have been successfully managed towards improved method development [68–70].

4. Conclusions

This analysis of existing methods for measuring GHG fluxes highlights that development of methods being more suited for independent use beyond academia are needed. Based on the need of quick progress, and impressions from effective method development related with other societal challenges, the GHG flux measurement method development should not depend solely on the uncertainty and short time-perspectives associated with normal academic research funding. Quick progress requires a broader societal engagement calling for increased awareness of method-related bottlenecks, designated leadership, coordination, and resource allocation. In medicine, effective diagnostic methods are a prerequisite for

effective treatments. Similarly, society now needs improved capacity to diagnose local GHG flux regulation to effectively mitigate these fluxes or enhance key GHG sinks.

Data availability statement

The list of references surveyed for this study are available at the following URL/DOI: https://doi.org/10.48360/vsgy-d095.

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Author contributions

Conceptualization: D B Methodology, validation, and investigation: All. Formal analysis: J W and D B with contributions from all. Visualization: T O, J W, D B, with contributions from all. Supervision and project administration: J W and D B. Funding acquisition: D B. Original draft written by D B and J W with contributions from all. All were active in reviewing and editing the manuscript.

Conflict of interests

The authors declare no competing interests.

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