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Quantifying Local to Regional Emissions of Methane Using UAV-based Atmospheric Concentration Measurements

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Summary

Changes in Earth's climate has been clearly observed during the last century, and is undeniably related to anthropogenic activities. The accurate prediction of our future climate is strongly linked to our ability to accurately measure and represent the amount of greenhouse gases (GHGs) present in the atmosphere. Following carbon dioxide (CO_2), methane (CH_4) is the second most abundant GHG, and it contains a tremendous warming potential for our climate; about 28 times greater than that of CO_2 on a 100-year time frame. Measuring CH_4 is crucial for knowing how much is currently residing in the atmosphere. Measuring alone is, however, not enough, and we require knowledge of how much CH_4 is currently being emitted, as well as knowledge of how much CH_4 is going to be emitted to the atmosphere by individual and different sources. To perceive the full picture, we also need to know the amount of CH_4 being removed from the atmosphere, by which processes, and their individual mechanisms. The global methane cycle is a complex structure with many links, and understanding its inner workings is necessary for accurate future climate predictions.

In the last decades methane has been an important GHG to study due to its large climate warming potential. However, magnitudes of individual methane sources are still characterized by large uncertainties. Many sources, such as individual factories or production facilities, can be difficult to quantify using conventional methodologies. They may be difficult to distinguish, or may be hard to access. In recent years, the use of drones, or unmanned aerial vehicles (UAVs), have become a promising technology for use in atmospheric GHG measurements. The goal of this thesis is to develop a UAV-based system for atmospheric measurements of GHGs and employ this system to quantify local to regional emissions of methane. This thesis begins by presenting the development of the UAV-based active AirCore system. Using this instrumentation, a technique is presented in chapter 3 to quantify emitted CH_4 and CO_2 from a point source, specifically a coal mine ventilation shaft. This methodology is then used in chapter 4 to estimate the regional coal mining emissions from the Upper Silesia Coal Basin, Poland.

Chapter 2 presents the development and testing of the UAV-based active AirCore system. The active AirCore system was inspired by the stratospheric AirCore sampler (Karion et al., 2010), but was customized to work in conjunction with a UAV, utilizing a small 45 [μm] pinhole orifice and a micropump to pull air into the tube while sampling, as opposed to the conventional filling of an AirCore using differences in pressure. This allows for sampling of air along the trajectory of the UAV while preserving the spatial resolution of the sampled air. Once an air sample had been procured, a Picarro Cavity RingDown Spectrometer (CRDS) G2401 was used to analyze the sampled air for atmospheric constituents, such as CO_2, CH_4, and carbon monoxide (CO). After extensive laboratory testing, the system was tested at a coastal site in the northern part of the Netherlands, close to a Integrated Carbon Observation System (ICOS) atmospheric station. Five consecutive flights over a 5-hour period were performed, starting early morning before sunrise. The first three flights were vertical profiles ranging in height from 300 to 500 [m], performed close to the 60 [m] tall ICOS tower, allowing for comparison between continuous CO_2, CH_4, and CO measurements from the tower and the vertical profiles from the UAV-based active AirCore system.

It was demonstrated that the build-up of the boundary layer was clearly observed over the three morning flights. A fourth flight flew from the atmospheric station across the Wadden sea dike, over a large patch of wetlands, and picked up enhanced levels of both CO_2 and CH_4. The fifth and final flight hovered next to the atmospheric station's 60 [m] air inlet for comparisons between the active AirCore and the continuous measurements of CO_2, CH_4, and CO, and found an R^2 value of 0.97 for CO_2 and 0.95 for CH_4. The determination of the spatial resolution was a key result of this chapter, and was determined to be comprised of four factors; analyzer smearing effects, GPS uncertainties, diffusion, and Taylor dispersion, where the analyzer smear effect was the dominant factor. At typical vertical and horizontal flight speeds (1.5 to 2.5 [m/s]), and average storage times between 10 and 40 minutes, the spatial resolution was determined to be between 24.7 and 48.9 [m] for CH_4, and between 24.1 and 46.0 [m] for CO_2. This chapter demonstrated the usefulness of the UAV-based active AirCore system, and its ability to capture both vertical and horizontal trace-gas profiles.

The third chapter focuses on the development of a methodology using the UAV-based active AirCore system to quantify the emitted CH_4 from a point source, specifically from a coal mine ventilation shaft. Anthropogenic exploitation of fossil fuel is one of the larger contributors to emitted atmospheric methane, and was between 2003 and 2017 responsible for about 35 [%] of the annually emitted methane world-wide (approximately 128 [Tg CH_4 year^{-1}]). From this, about 33 [%] (41 [Tg CH_4 year^{-1}]) originates from coal production, both from when coal is cracked from the bedrock as well as when the coal is being processed (crushed and pulverized). The released methane coming from the underground mines is diluted and vented directly into the atmosphere through ventilation shafts located on the surface. The exact amount of methane being emitted to the atmosphere in this manner is poorly characterized, and therefore contributes to the overall uncertainty of the methane cycle. **Chapter 3** focuses on utilizing the UAV-based active AirCore system to quantify the emitted methane from a single ventilation shaft.

In the summer of 2017, as part of the Carbon and Methane Mission (CoMet) 0.5, fifteen UAV-based active AirCore flights were performed downwind of a single ventilation shaft in the Upper Silesia Coal Basin (USCB) over the span of two days. The flights were performed downwind the source, and the UAV-based active AirCore system was flown perpendicular to the wind direction, attempting to transect the emitted plume at separate height levels. This effectively created a 2D plane ('curtain') of measured CH_4 mole fractions. A criteria list was made to filter successful from unsuccessful flights, namely that flights require wind speeds above 2 [m/s] and to be perpendicular to the wind direction (within 15 [°]). Out of the fifteen flights, eight fulfilled the criteria for emission rate quantification. Two different methodologies were used to quantify the emitted methane and determine the source strength; a mass balance approach, and an inverse Gaussian approach. To test the methodologies, a simulated data experiment was made with the aim of mimicking the observed flight profiles, while exactly knowing the plume parameters and source strength. The two methodologies were applied to the simulated data, and achieved an accuracy of within 10 [%] of the true value for the mass balance approach, and within 3.6 [%] for the inverse Gaussian approach. The number of transects was investigated using the simulated data, showcasing the advantage of being able to transect the emitted plume at several height levels, as compared to using a single transect. The simulated data was also used to study the influence spacing between each flight transect would have. It was found that a spacing between the transects of less than 2.5 multiples of σ_z (vertical dispersion parameter of the plume), as well as the plume's center-height being 2.5 multiples of σ_z away from the fight's center transect, drastically increased the methodology's quantification capability. From the simulated data tests, the inverse Gaussian approach yielded a more accurate quantification of the true source strength than the mass balance approach.

Following the simulated data experiments, the two quantification methodologies were applied to the eight flights that passed the criteria list. The quantified emission rate during day 1 was estimated as 4.9 ± 1.4 [kt CH_4/year] and 4.3 ± 3.1 [kt CH_4/year] on day 2 using the inverse Gaussian approach, with the quantified emission rates ranging from 1.1 - 9 [kt CH_4/year]. For the mass balance approach, the quantified emission rate on day 1 was 3.7 ± 2.3 [kt CH_4/year], and 6.9 ± 5.0 [kt CH_4/year] on day 2. The range of quantified CH_4 emission rates was 0.5 - 14.5 [kt CH_4/year]. Due to strong correlation between CH_4 and CO_2 ($R^2 > 0.69$), the CO_2 emission rate could also be inferred. The CO_2 emission rate was estimated to be 2.9 ± 0.4 [kt CO_2/year] on day 1, and 2.9 ± 2.1 [kt CO_2/year] on day 2 for the inverse Gaussian approach. The mass balance obtained 2.0 ± 1.1 [kt CO_2/year] and 4.6 ± 3.4 [kt CO_2/year] for the same two days, respectively. A limitation for these quantification strategies is unstable wind conditions. Since the sampling in not instantaneous, and takes place over the span of 10 - 12 minutes, unstable winds can cause the plume to meander over the course of the flight. This may lead to the plume being picked up several times, or sometimes not at all. The inverse Gaussian approach also assumes that the plume has a perfect Gaussian shape. Since we use single-pass transects at different altitudes, sufficient averaging time at a certain altitude is lacking, and adds significant uncertainty to the estimation. However, the observations from multiple transects at different heights provide additional constraints on the location and the shape of the plume, especially concerning the location of the center of the plume. The methodology presented in chapter 3 is demonstrated to be a valuable tool in obtaining and verifying point source CH_4 emissions using the active AirCore system.

The fourth chapter deals with quantifying the regional CH_4 emissions in the USCB by sampling air from several coal mine ventilation shafts using the UAV-based active AirCore system, and quantifying their individual source strengths. The USCB, located in the southern region of Poland, is home to many coal mines, and was in 2017 responsible for more than 447 [kT CH_4 year^{-1}]. The total annual European emissions in 2017 were, according to the European Pollutant Release and Transfer Register (E-PRTR), 1642 [kT CH_4 year^{-1}], making the USCB responsible for 27.3 [%] of Europe's annual emissions. With the large uncertainties that are associated with these methane emissions, the USCB is an important region to study and to quantify the emissions. Similar to the measurement campaign presented in chapter 3, this measurement campaign was also part of the Carbon Dioxide and Methane Mission (CoMet), designated CoMet 1.0. This was a joint campaign held between May and June, 2018, and collaborated a variety of research groups. Different measurement techniques included aircraft measurements, mobile van measurements, Fourier Transform Infrared (FTIR) spectroscopy measurements, and UAV measurements.

In **chapter 4**, the UAV-based active AirCore is used to quantify the emitted methane from several coal mine ventilation shafts in the USCB, using the technique presented in chapter 3. Isotopic data (^{13}C-CH_4 and ^{2}H-CH_4) from each flight is used to compare with isotopic signatures from the same, or close-by, ventilation shafts in the region obtained by other research groups, and is used to verify that the emitted CH_4 is originating from the ventilation shafts. These signatures range from -59.4 to -41.0 [‰] for ^{13}C-CH_4, and from -212 to -142 [‰] from ^{2}H-CH_4. Five separate shafts were sampled over 36 successful flights, with the number of successful flights per shaft ranging from 2 to 14. The quantified CH_4 emission rate from each ventilation shaft was highly variable, which was also reflected in high-frequency (1 hour averaged) inventory measurements from the companies themselves. The mean average quantified CH_4 emission rate was 6.5 ± 2.3 [kt CH_4/year], while the mean average CH_4 emission rate using the high-frequency inventory was 10.6 ± 2.9 [kt CH_4/year]. A key finding here was the statistical necessity for more than 2 flights when doing the emission rate quantification to be able to obtain the best possible estimate on the emission rate.

A comparison between the quantified emission rates and direct annual emissions from E-PRTR highlighted the difficulty in comparing snapshot measurements with mean annual emissions, showing a low correlation (R^2 of 0.06). A better correlation was found by comparing daily-averaged quantified emission rates and inventory emissions (from hourly data), with the R-square value being 0.23. Shaft-averaged emissions saw the best correlation, with an R-squared of 0.86 for the inverse Gaussian approach, and 0.76 for the mass balance approach. The linear fit curves were used to scale the quantified emission rate to the full regional emissions. Three upscaling approaches were used: scaling the E-PRTR annual inventory using the linear curve, multiplying the quantified shaft-averaged emission rate with active ventilation shafts in the area, and using the hourly emission inventory to derive a quantified emission rate using the linear curves. The first approach obtained emission rates of 332.6 [kt CH_4/year] and 268.2 [kt CH_4/year], for the inverse Gaussian and mass balance, respectively. The second approach obtained regional emission estimates of 324.5 ± 147.5 [kt CH_4/year] using the inverse Gaussian, and 318.6 ± 188.8 [kt CH_4/year] using the mass balance. The third approach resulted in regional emission estimates of 446.9 ± 133.2 [kt CH_4/year] using the inverse Guassian, and 346.9 ± 103.4 [kt CH_4/year] using the mass balance. Up-scaled regional CO_2 emissions were also quantified, and were found to be between 0.2 and 0.3 [Mt CO_2/year] for both approaches. This is roughly 1 [%] of the annual E-PRTR (2017) inventory of 35.3 [Mt CO_2/year]. Fiehn et al. (2020) quantified the regional CO_2 emissions to be 38.2 ± 22.7 [Mt CO_2/year] and 35.3 ± 11.7 [Mt CO_2/year]. We conclude that coal mine ventilation shafts are not a large regional CO_2 source, but that the inventory is missing a source of ∼ 1 [%]. The results found in this study illustrate the possibility of using the UAV-based active AirCore system to quantify regional emission estimates, which is particularly useful in locations that are difficult to access and where other methods prove difficult to use.

This thesis shows that accurate measurements of GHGs can be readily obtained without great loss of the spatial resolution using the UAV-based active AirCore system, and that this system can be used in combination with atmospheric dispersion modeling to obtain facility to regional level GHG emission estimates. **Chapter 5** discusses the important findings of this thesis in relation to other studies, and presents advancements that can be made using the active AirCore system in the future. The UAV-based active AirCore system provides a unique possibility to combine the versatility and flexibility of a UAV with atmospheric composition measurements, and can continue to be a valuable asset in quantifying and verifying methane emissions, and other GHGs, on both facility and regional levels.

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Samenvatting

Veranderingen in het klimaat op aarde zijn de afgelopen eeuw duidelijk waargenomen en zijn onmiskenbaar gerelateerd aan antropogene activiteiten. De nauwkeurige voorspelling van ons toekomstige klimaat is sterk verbonden met ons vermogen om de hoeveelheid broeikasgassen in de atmosfeer nauwkeurig te meten en weer te geven. Na CO_2 is CH_4 het meest voorkomende broeikasgas en bevat het een enorm opwarmingspotentieel voor ons klimaat; ongeveer 28 keer groter dan die van CO_2 over een tijdsbestek van 100 jaar. Het meten van CH_4 is natuurlijk een must om te weten hoeveel er momenteel in de atmosfeer aanwezig is. Meten alleen is echter niet genoeg, en we hebben kennis nodig van hoeveel CH_4 momenteel wordt uitgestoten, evenals kennis van hoeveel CH_4 door individuele en verschillende bronnen in de atmosfeer zal worden uitgestoten. Om het volledige plaatje te zien, moeten we ook de hoeveelheid CH_4 weten die uit de atmosfeer wordt verwijderd, door welke processen, en hun individuele mechanismen. De wereldwijde methaancyclus is een complexe structuur, en het beter begrijpen is noodzakelijk voor nauwkeurige toekomstige klimaatvoorspellingen.

In de afgelopen decennia is methaan een belangrijk broeikasgas geweest om te bestuderen vanwege het grote klimaatopwarmingspotentieel. De omvang van individuele methaanbronnen kent echter nog steeds grote onzekerheden. Veel bronnen, zoals individuele fabrieken of productiefaciliteiten, kunnen moeilijk te kwantificeren met behulp van conventionele methoden. Ze zijn moeilijk te onderscheiden zijn of moeilijk toegankelijk zijn. In de afgelopen jaren is het gebruik van drones of onbemande luchtvaartuigen een veelbelovende technologie geworden voor gebruik bij atmosferische broeikasgasmetingen. Het doel van dit proefschrift is om een op onbemande luchtvaar-tuigen (UAV) gebaseerd systeem te ontwikkelen voor atmosferische metingen van broeikasgassen, en het systeem te gebruiken om lokale tot regionale emissies van methaan te kwantificeren. Dit proefschrift begint met de ontwikkeling van het UAV-gebaseerde actieve AirCore-systeem. Met behulp van deze instrumentatie wordt in hoofdstuk 3 een techniek gepresenteerd om de uitgestoten CH_4 en CO_2 te kwantificeren vanuit een puntbron, met name een ventilatieschacht van een kolenmijn. Deze methodologie wordt vervolgens in hoofdstuk 4 gebruikt om de regionale kolenmijnemissies uit het Opper-Silezië kolenbekken, in Polen te schatten.

Hoofdstuk 2 presenteert de ontwikkeling en het testen van het UAV-gebaseerde actieve AirCore-systeem. Het actieve AirCore-systeem is geïnspireerd op de stratosferische AirCore (Karion et al., 2010), maar is aangepast om te werken in combinatie met een UAV. Het gebruikt een kleine gaatjesopening van 45 [μm] en een micropomp om actief een luchmonster lucht in de buis te trekken, in tegenstelling tot het conventioneel passief vullen van een AirCore met drukverschillen. Dit zorgt voor bemonstering van lucht langs het traject van de UAV met behoud van de ruimtelijke resolutie van de bemonsterde lucht. Nadat een luchtmonster was verkregen, werd een Picarro-caviteit-ringdown-spectrometer (CRDS) G2401 gebruikt om de bemonsterde lucht te analyseren op atmosferische bestanddelen, zoals CO_2, CH_4 en CO. Na uitgebreide laboratoriumtests werd het systeem getest op een kustlocatie in Nederland, vlakbij een ICOS atmosferisch meetstation. Er werden vijf opeenvolgende vluchten uitgevoerd over een periode van 5 uur, die vroeg in de ochtend voor zonsopgang begonnen. De drie eerste vluchten waren verticale vluchten variërend in hoogte van 300 tot 500 [m], uitgevoerd in de buurt van de ICOS-toren, waardoor een vergelijking mogelijk was tussen continue CO_2-, CH_4- en CO-metingen vanaf de toren en de profielen van CO_2, CH_4 en CO van de actieve AirCore. Er werd aangetoond dat de opbouw van de grenslaag duidelijk werd waargenomen over de drie ochtendvluchten. Een vierde vlucht vloog vanaf het atmosferische station over de Waddenzeedijk, over een groot stuk wetlands, en pikte duidelijk verhoogde niveaus van zowel CO_2 als CH_4 op. De vijfde en laatste vlucht zweefde naast een van de luchtinlaten voor continue metingen van CO_2, CH_4 en CO voor het atmosferische station, met een R^2-waarde van 0,97 voor CO_2 en 0,95 voor CH_4. De bepaling van de ruimtelijke resolutie was een belangrijk resultaat van dit hoofdstuk en er werd vastgesteld dat deze uit vier factoren bestond; uitstrijkeffecten van de analysator, GPS-onzekerheden, diffusie en Taylor-dispersie, waarbij het uitstrijkeffect van de analysator de dominante factor was. Bij typische verticale en horizontale vliegsnelheden (1,5 tot 2,5 [m/s]) en gemiddelde opslagtijden tussen 10 en 40 minuten, werd de ruimtelijke resolutie bepaald tussen 24,7 en 48,9 [m] voor CH_4, en tussen 24,1 en 46,0 [m] voor CO_2. Dit hoofdstuk demonstreerde het nut van het UAV gebaseerde actieve AirCore-systeem en zijn vermogen om zowel verticale als horizontale sporegasprofielen vast te leggen.

Het derde hoofdstuk richt zich op de ontwikkeling van een techniek die gebruik maakt van het UAV-gebaseerde actieve AirCore-systeem om de CH_4 emissies te kwantificeren van een puntbron, specifiek van een ventilatieschacht van een kolenmijn. Antropogene exploitatie van fossiele brandstoffen is een van de grootste oorzaken van de emissie van methaan in de atmosfeer en was tussen 2003 en 2017 verantwoordelijk voor ongeveer 35 [%] van de jaarlijks uitgestoten methaan wereldwijd (ongeveer 128 [Tg CH_4 jaar^{-1}]). Hiervan is ongeveer 33 [%] (41 [Tg CH_4 jaar^{-1}]) afkomstig uit steenkoolproductie, zowel vanaf het moment dat de steenkool uit het gesteente wordt gekraakt als tijdens de verwerking (geplet en verpulverd). Het vrijgekomen methaan dat uit de ondergrondse mijnen komt, wordt verdund en rechtstreeks in de atmosfeer afgevoerd via ventilatieschachten aan de oppervlakte. De exacte hoeveelheid methaan die op deze manier in de atmosfeer wordt uitgestoten, is slecht gekarakteriseerd en draagt daarom bij aan de algehele onzekerheid van de methaancyclus. **Hoofdstuk 3** richt zich op het gebruik van het UAV gebaseerde actieve AirCore-systeem om het uitgestoten methaan uit een enkele ventilatieschacht te kwantificeren. In de zomer van 2017 werden, als onderdeel van de Carbon and Methane missie (CoMet) 0.5, vijftien UAV gebaseerde actieve AirCore-vluchten uitgevoerd benedenwinds van een enkele ventilatieschacht in het Upper Silesia Coal Basin (USCB) gedurende twee dagen. De vluchten werden tegen de wind in uit-ge-voerd en het UAV-actieve AirCore-systeem werd loodrecht op de windrichting gevlogen, in een poging de emissie pluim op verschillende hoogtes te doorsnijden.

Dit creëerde effectief een 2D-gordijn (vlak) van gemeten CH_4-molfracties. Er is een lijst met criteria gemaakt om succesvolle vluchten van mislukte vluchten te onderscheiden, namelijk dat vluchten een windsnelheid van meer dan 2 [m/s] vereisen en loodrecht op de windrichting staan (binnen 15 [°]). Van de vijftien vluchten voldeden er acht aan de criteria voor kwantificering van de flux. Er zijn twee verschillende methoden gebruikt om de bronsterkte te bepalen; een massa-balansbenadering en een inverse Gauss-benadering. Om de methoden te testen, werd een pseudo-data-experiment gedaan met als doel de waargenomen vluchtprofielen na te bootsen, terwijl de pluimparameters en de bronsterkte precies bekend waren. De twee methodologieën werden toegepast op de pseudo-gegevens en bereikten een nauwkeurigheid van binnen 10 [%] van de werkelijke waarde voor de massabalansbenadering, en binnen 3,6 [%] voor de inverse Gauss-benadering. Het aantal kruisende vluchtsegmenten werd onderzocht met behulp van de pseudo-gegevens, wat het voordeel aantoonde van het kunnen snijden van de pluim op verschillende hoogte in plaats van het gebruik van een enkel transect. De inverse Gauss-benadering leverde een nauwkeuriger kwantificering van de werkelijke bronsterkte op dan die van de massabalansbenadering. Na de pseudo-gegevensexperimenten werden de twee methoden toegepast op de acht vluchten die aan de criterialijst voldeden. De bronsterkte gedurende dag 1 was 4,9 ± 1,4 [kt CH_4/jaar] en 4,3 ± 3,1 [kt CH_4/jaar] op dag 2 met behulp van de inverse Gauss-benadering, variërend van 1,1 - 9 [kt CH_4/jaar]. Voor de massabalansbenadering was de bronsterkte op dag 1 3,7 ± 2,3 [kt CH_4/jaar] en 6,9 ± 5,0 [kt CH_4/jaar] op dag 2. Het spreiden van CH_4-fluxen was 0,5 - 14,5 [kt CH_4/jaar]. Vanwege de grote correlatie tussen CH_4 en CO_2 ($R^2 > 0{,}69$), kon ook de CO_2-flux worden afgeleid. De CO_2-flux werd dus geschat op 2,9 ± 0,4 [kt CO_2/jaar] op dag 1 en 2,9 ± 2,1 [kt CO_2/jaar] op dag 2 voor de inverse Gauss-benadering. De massabalans resulteerde in respectievelijk 2,0 ± 1,1 [kt CO_2/jaar] en 4,6 ± 3,4 [kt CO_2/jaar] voor de twee dagen. Een beperking voor deze methoden zijn onstabiele windomstandigheden. Aangezien de bemonstering niet onmiddellijk plaatsvindt maar binnen 10 - 12 minuten, kunnen onstabiele winden ervoor zorgen dat de pluim tijdens de vlucht gaat slingeren. Dit kan ertoe leiden dat de pluim meerdere keren of soms helemaal niet wordt opgepakt. De inverse Gauss-benadering gaat er ook van uit dat de pluim een perfecte Gauss-vorm heeft. Omdat we single-pass transecten op verschillende hoogten gebruiken, ontbreekt het aan voldoende middelingstijd op een bepaalde hoogte, en dit vergroot de onzekerheid. aanzienlijk onzekerheid toe aan de schatting. De waarnemingen van meerdere transecten op verschillende hoogtes zorgen echter voor extra beperkingen op de locatie en de vorm van de pluim, vooral met betrekking tot de locatie van het centrum van de pluim. De methodologie die in hoofdstuk 3 wordt gepresenteerd, blijkt een waardevol hulpmiddel te zijn bij het verkrijgen en verifiëren van CH_4-emissies uit puntbronnen.

Het **vierde hoofdstuk** richt zich op *regionale* CH_4-emissies in de USCB door luchtmonsters te nemen uit *verschillende* kolenmijnventilatieschachten met behulp van het UAV-gebaseerde actieve AirCore-systeem, en het kwantificeren van hun individuele bronsterkten. Het USCB, gelegen in de zuidelijke regio van Polen, is de thuisbasis van vele kolenmijnen en was in 2017 verantwoordelijk voor meer dan 447 [kT CH_4 jaar^{-1}]. De totale jaarlijkse Europese emissies in 2017 waren, volgens het European Pollutant Release and Transfer Register (E-PRTR), 1642 [kT CH_4 jaar^{-1}], waarmee het USCB verantwoordelijk is voor 27,3 [%] van de Europese jaarlijkse uitstoot. Met de grote onzekerheden die met deze methaanemissies gepaard gaan, is het USCB een belangrijke regio om de emissies te bestuderen en te kwantificeren. Net als in hoofdstuk 3, maakte deze meetcampagne ook deel uit van de Carbon Dioxide and Methane missie (CoMet), aangeduid als CoMet 1.0. Dit was een gezamenlijke campagne die werd gehouden tussen mei en juni 2018, en waarin werd samengewerkt met verschillende onderzoeksgroepen. Verschillende meettechnieken omvatten vliegtuigmetingen, metingen van mobiele bestelwagens, Fourier Transform Infrared (FTIR) spectroscopiemetingen en UAV-metingen. In **hoofdstuk 4** wordt de UAV-actieve AirCore gebruikt om het uitgestoten methaan te kwantificeren van verschillende ventilatieschachten van kolenmijnen in de USCB, met behulp van de techniek die in hoofdstuk 3 wordt gepresenteerd.

Isotopen ratio gegevens (^{13}C-CH_4 en ^{2}H-CH_4) van elke vlucht worden gebruikt om te vergelijken met isotopensignaturen van dezelfde of dichtbij gelegen ventilatieschachten in de regio, verkregen door andere onderzoeksgroepen. Dit wordt gebruikt om te verifiëren dat de uitgestoten CH_4 afkomstig is van de ventilatieschachten. Deze signaturing variëren van -59,4 tot -41,0 [‰] voor ^{13}C-CH_4, en van -212 tot -142 [‰] van ^{2}H-CH_4. Er werden vijf afzonderlijke schachten bemonsterd, waarbij het aantal succesvolle vluchten varieerde van 2 tot 14, afhankelijk van de ventilatieschacht. De gekwantificeerde CH_4-flux van elke ventilatieschacht was zeer variabel, wat ook tot uiting kwam in hoogfrequente (1 uur) emissie inventarisaties van de bedrijven zelf. De gemiddelde gekwantificeerde CH_4-flux was 6,5 ± 2,3 [kt CH_4/jaar], terwijl de gemiddelde CH_4-flux met behulp van de hoogfrequente inventarisaties 10,6 ± 2,9 [kt CH_4/jaar] was. Een belangrijke bevinding hierbij was de statistische noodzaak voor meer dan 2 vluchten bij de kwantificering van de flux, om de best mogelijke schatting te kunnen maken van de geëmitteerde flux, in vergelijking met de hoogfrequente inventarisatie.

Een vergelijking tussen de gekwantificeerde fluxen en de directe jaarlijkse emissies van E-PRTR benadrukte de moeilijkheid om snapshot-metingen te vergelijken met de gemiddelde jaarlijkse emissies, en toonde een niet significante correlatie (R^2 van 0,06). Een betere correlatie werd gevonden door de daggemiddelde gekwantificeerde gekwantificeerde fluxen en inventarisaties (uit uurlijkse gegevens) te vergelijken en er werd een R-kwadraat van 0,23 gevonden. Ventilatieschacht gemiddelde emissies gaven de beste correlatie, met een R-kwadraat van 0,86 voor de inverse Gauss-benadering en 0,76 voor de massabalansbenadering. De lineaire fitcurves werden vervolgens gebruikt om de gekwantificeerde flux op te schalen naar regionale emissies. Er zijn drie opschalingsbenaderingen gebruikt: het schalen van de E-PRTR jaarinventarisatie met behulp van de lineaire curve, het vermenigvuldigen van de gekwantificeerde schachtgemiddelde flux met actieve ventilatieschachten in het gebied, en het gebruik van de uurlijkse emissie-inventarisatie om een gekwantificeerde flux af te leiden met behulp van de lineaire curven . De eerste benadering leverde fluxen op van 332,6 [kt CH_4/jaar] en 268,2 [kt CH_4/jaar], voor respectievelijk de inverse Gauss- en massabalans. De tweede benadering verkreeg regionale fluxen van 324,5 ± 147,5 [kt CH_4/jaar] met behulp van de inverse Gauss, en 318,6 ± 188,8 [kt CH_4/jaar] met behulp van de massabalans. De derde benadering resulteerde in regionale fluxen van 446,9 ± 133,2 [kt CH_4/jaar] met behulp van de inverse Guassian, en 346,9 ± 103,4 [kt CH_4/jaar] met behulp van de massabalans. Opgeschaalde regionale CO_2-emissies werden ook gekwantificeerd en bleken voor beide benaderingen tussen 0,2 en 0,3 [Mt CO_2/jaar] te zijn. Dit is ongeveer 1 [%] van de jaarlijkse E-PRTR (2017)-inventaris van 35,3 [Mt CO_2/jaar]. Fiehn et al. (2020) kwantificeerde de regionale CO_2-uitstoot op 38,2 ± 22,7 [Mt CO_2/jaar] en 35,3 ± 11,7 [Mt CO_2/jaar]. We concluderen dat de ventilatieschachten van kolenmijnen geen grote regionale CO_2-bron zijn, maar dat in de inventaris een bron van ~ 1 [%] ontbreekt. De resultaten illustreren de mogelijkheid om het UAV-actieve AirCore-systeem te gebruiken om regionale emissieschattingen te kwantificeren, wat vooral handig is op moeilijk bereikbare locaties en daar waar andere methoden moeilijk te gebruiken blijken te zijn.

Dit proefschrift laat zien dat nauwkeurige metingen van broeikasgassen gemakkelijk kunnen worden verkregen zonder groot verlies van de ruimtelijke resolutie met behulp van het UAV-gebaseerde actieve AirCore-systeem. En dat dit systeem kan worden gebruikt in combinatie met atmosferische dispersiemodellering om emissie schattingen op regionaal niveau te verkrijgen. schattingen van de uitstoot van broeikasgassen. **Hoofdstuk 5** bespreekt de belangrijke bevindingen van dit proefschrift in relatie tot andere studies, en presenteert mogelijk nieuwe innovaties aan, en toepassingen van, het actieve AirCore-systeem. Het op UAV gebaseerde actieve AirCore-systeem biedt een unieke mogelijkheid om de veelzijdigheid en flexibiliteit van een UAV te combineren met metingen van de atmosferische samenstelling, en kan een waardevolle aanwinst blijven bij het kwantificeren en verifiëren van methaanemissies en andere broeikasgassen, zowel op lokaal als op regionaal niveau.

B

Bibliography

Abdel-Rahman, A. A. (2008). On the Atmospheric Dispersion and Gaussian Plume Model. In Mastorakis, N. E., Poulos, M., Mladenov, V., Bojkovic, Z., Simian, D., Kartalopoulos, S., Varonides, A., and Udriste, C., editors, 2nd International Conference on Waste Management, Water Pollution, Air Pollution, Indoor Climate (WWAI'08), pages 41–50. WSWAS. 14

Allen, G., Hollingsworth, P., Kabbabe, K., Pitt, J. R., Mead, M. I., Illingworth, S., Roberts, G., Bourn, M., Shallcross, D. E., and Percival, C. J. (2019). The development and trial of an unmanned aerial system for the measurement of methane flux from landfill and greenhouse gas emission hotspots. Waste Management, 87:883 – 892. 9, 10, 40, 45, 58, 79, 110, 111, 113, 118

Allibert, G., Abeywardena, D., Bangura, M., and Mahony, R. (2014). Estimating body-fixed frame velocity and attitude from inertial measurements for a quadrotor vehicle. In 2014 IEEE Conference on Control Applications (CCA), pages 978–983. 119

Andersen, T., De Vries, M., Vinkovic, K., Kers, B., Peters, W., Necki, J., Swolkien, J., Roiger, A., and Chen, H. (2021). Quantifying methane emissions from coal mining ventilation shafts using a small Unmanned Aerial Vehicle (UAV)-based active AirCore system. Atmospheric Environment X, 0:1. In review. 80, 81, 83, 93, 94, 100

Andersen, T., Scheeren, B., Peters, W., and Chen, H. (2018). A UAV-based active AirCore system for measurements of greenhouse gases. Atmospheric Measurement Techniques, 11(5):2683–2699. 10, 45, 46, 47, 49, 64, 79, 110, 111

Andrews, A. E., Kofler, J. D., Trudeau, M. E., Williams, J. C., Neff, D. H., Masarie,

K. A., Chao, D. Y., Kitzis, D. R., Novelli, P. C., Zhao, C. L., Dlugokencky, E. J., Lang, P. M., Crotwell, M. J., Fischer, M. L., Parker, M. J., Lee, J. T., Baumann, D. D., Desai, A. R., Stanier, C. O., De Wekker, S. F. J., Wolfe, D. E., Munger, J. W., and Tans, P. P. (2014). CO_2, CO, and CH_4 measurements from tall towers in the NOAA Earth System Research Laboratory's Global Greenhouse Gas Reference Network: instrumentation, uncertainty analysis, and recommendations for future high-accuracy greenhouse gas monitoring efforts. Atmos. Meas. Tech., 7(2):647–687. 8, 79

Arndt, C., Leytem, A., Hristov, A., Zavala-Araiza, D., Cativiela, J., Conley, S., Daube, C., Faloona, I., and Herndon, S. (2018). Short-term methane emissions from 2 dairy farms in California estimated by different measurement techniques and US Environmental Protection Agency inventory methodology: A case study. Journal of Dairy Science, 101(12):11461 – 11479. 9

Atherton, E., Risk, D., Fougère, C., Lavoie, M., Marshall, A., Werring, J., Williams, J. P., and Minions, C. (2017). Mobile measurement of methane emissions from natural gas developments in northeastern British Columbia, Canada. Atmospheric Chemistry and Physics, 17(20):12405–12420. 9

Bandyopadhyay, A. (2009). Prediction of ground level concentration of sulfur dioxide using ISCST3 model in Mangalore industrial region of India. Clean Technologies and Environmental Policy, 11(2):173–188. 12

Bergamaschi, P., Houweling, S., Segers, A., Krol, M., Frankenberg, C., Scheepmaker, R. A., Dlugokencky, E., Wofsy, S. C., Kort, E. A., Sweeney, C., Schuck, T., Brenninkmeijer, C., Chen, H., Beck, V., and Gerbig, C. (2013). Atmospheric CH_4 in the first decade of the 21st century: Inverse modeling analysis using SCIAMACHY satellite retrievals and NOAA surface measurements. Journal of Geophysical Research: Atmospheres, 118(13):7350–7369. 7

Berman, E. S. F., Fladeland, M., Liem, J., Kolyer, R., and Gupta, M. (2012). Greenhouse gas analyzer for measurements of carbon dioxide, methane, and water vapor aboard an unmanned aerial vehicle. Sensors and Actuators B: Chemical, 169:128–135. 10, 19, 45, 110, 111

Borchardt, J., Gerilowski, K., Krautwurst, S., Bovensmann, H., Thorpe, A. K., Thompson, D. R., Frankenberg, C., Miller, C. E., Duren, R. M., and Burrows, J. P. (2021). Detection and quantification of CH_4 plumes using the WFM-DOAS retrieval on AVIRIS-NG hyperspectral data. Atmospheric Measurement Techniques, 14(2):1267–1291. 120

Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noël, S., Rozanov, V. V., Chance, K. V., and Goede, A. P. H. (1999). SCIAMACHY: Mission Objectives and Measurement Modes. Journal of the Atmospheric Sciences, 56(2):127–150. 7

Brady, J. M., Stokes, M. D., Bonnardel, J., and Bertram, T. H. (2016). Characterization of a Quadrotor Unmanned Aircraft System for Aerosol-Particle-Concentration Measurements. Environmental Science & Technology, 50(3):1376–1383. 10

Briggs, G. (1972). Discussion: Chimney plumes in neutral and stable surroundings. Atmospheric Environment, 6:507–510. 13

Briggs, G. A. (1969). Plume Rise: A Critical Survey. Technical report, United States. 13

Briggs, G. A. (1973). Diffusion estimation for small emissions. Preliminary report. Technical report, United States. 15

Brosy, C., Krampf, K., Zeeman, M., Wolf, B., Junkermann, W., Schäfer, K., Emeis, S., and Kunstmann, H. (2017). Simultaneous multicopter-based air sampling and sensing of meteorological variables. Atmospheric Measurement Techniques, 10(8):2773–2784. 10, 45, 79

Brownlow, R., Lowry, D., Thomas, R., Fisher, R., France, J., Cain, M., Richardson, T., Greatwood, C., Freer, J., Pyle, J., MacKenzie, A., and Nisbet, E. (2016). Methane mole fraction and δ^{13}C above and below the trade wind inversion at Ascension Island in air sampled by aerial robotics. Geophysical Research Letters, 43(22):11893–11902. 10, 19, 79, 111

Cambaliza, M. O. L., Bogner, J. E., Green, R. B., Shepson, P. B., Harvey, T. A., Spokas, K. A., Stirm, B. H., and Corcoran, M. (2017). Field measurements and modeling to resolve m^2 to km^2 CH_4 emissions for a complex urban source: An Indiana landfill study. Elementa: Science of the Anthropocene, 5(36):1–12. 118

Cambaliza, M. O. L., Shepson, P. B., Bogner, J., Caulton, D. R., Stirm, B., Sweeney, C., Montzka, S. A., Gurney, K. R., Spokas, K., Salmon, O. E., Lavoie, T. N., Hendricks, A., Mays, K., Turnbull, J., Miller, B. R., Lauvaux, T., Davis, K., Karion, A., Moser, B., Miller, C., Obermeyer, C., Whetstone, J., Prasad, K., Miles, N., and Richardson, S. (2015). Quantification and source apportionment of the methane emission flux from the city of Indianapolis. Elementa: Science of the Anthropocene, 3. 000037. 58

Caulton, D. R., Li, Q., Bou-Zeid, E., Fitts, J. P., Golston, L. M., Pan, D., Lu, J., Lane, H. M., Buchholz, B., Guo, X., McSpiritt, J., Wendt, L., and Zondlo, M. A. (2018). Quantifying uncertainties from mobile-laboratory-derived emissions of well pads using inverse Gaussian methods. Atmos. Chem. Phys., 18(20):15145–15168. 9, 62, 114

Chang, C.-C., Wang, J.-L., Chang, C.-Y., Liang, M.-C., and Lin, M.-R. (2016). Development of a multicopter-carried whole air sampling apparatus and its applications in environmental studies. Chemosphere, 144:484 – 492. 10, 45, 79

Chen, H., Karion, A., Rella, C. W., Winderlich, J., Gerbig, C., Filges, A., Newberger, T., Sweeney, C., and Tans, P. P. (2013). Accurate measurements of carbon monoxide in humid air using the cavity ring-down spectroscopy (CRDS) technique. Atmospheric Measurement Techniques, 6(4):1031–1040. 23

Chen, H., Kivi, R., Heikkinen, P., Kers, B., d. Vries, M., Hatakka, J., Laurila, T., Sweeney, C., and Tans, P. (2020). High-latitude balloon observations of CO_2/CH_4/CO using AirCore: evaluation of Sodankylä TCCON retrievals,. in preparation, unknown:unknown. 19, 20

Chen, H., Winderlich, J., Gerbig, C., Hoefer, A., Rella, C. W., Crosson, E. R., Pelt, A. D. V., Steinbach, J., Kolle, O., Beck, V., Daube, B. C., Gottlieb, E. W., Chow, V. Y., Santoni, G. W., and Wofsy, S. C. (2010). High-accuracy continuous airborne measurements of greenhouse gases (CO_2 and CH_4) using the cavity ring-down spectroscopy (CRDS) technique. Atmospheric Measurement Techniques, 3(2):375–386. 23

Collier-Oxandale, A., Casey, J. G., Piedrahita, R., Ortega, J., Halliday, H., Johnston, J., and Hannigan, M. P. (2018). Assessing a low-cost methane sensor quantification system for use in complex rural and urban environments. Atmospheric Measurement Techniques, 11(6):3569–3594. 119

Collier-Oxandale, A. M., Thorson, J., Halliday, H., Milford, J., and Hannigan, M. (2019). Understanding the ability of low-cost MO_x sensors to quantify ambient VOCs. Atmospheric Measurement Techniques, 12(3):1441–1460. 119

Crosson, E. R. (2008). A cavity ring-down analyzer for measuring atmospheric levels of methane, carbon dioxide, and water vapor. Applied Physics B, 92(3):403–408. 22, 108

Cusworth, D. H., Duren, R. M., Thorpe, A. K., Pandey, S., Maasakkers, J. D., Aben, I., Jervis, D., Varon, D. J., Jacob, D. J., Randles, C. A., Gautam, R., Omara, M., Schade, G. W., Dennison, P. E., Frankenberg, C., Gordon, D., Lopinto, E., and Miller, C. E. (2021). Multisatellite Imaging of a Gas Well Blowout Enables Quantification of Total Methane Emissions. Geophysical Research Letters, 48(2):e2020GL090864. 120

Daube, C., Conley, S., Faloona, I. C., Arndt, C., Yacovitch, T. I., Roscioli, J. R., and Herndon, S. C. (2019). Using the tracer flux ratio method with flight measurements to estimate dairy farm CH_4 emissions in central California. Atmos. Meas. Tech., 12(4):2085–2095. 9

Dessler, A. (2015). Introduction to modern climate change. Cambridge University Press, 1st edition edition. 2

Dinger, A. S., Stebel, K., Cassiani, M., Ardeshiri, H., Bernardo, C., Kylling, A., Park, S.-y., Pisso, I., Schmidbauer, N., Wasseng, J., and Stohl, A. (2018). Observation of turbulent dispersion of artificially released SO_2 puffs with UV cameras. Atmospheric Measurement Techniques, 11:6169–6188. 63, 118, 119

Dlugokencky, E. (2020). Trends in Atmospheric Methane. NOAA/GML. www.esrl.noaa.gov/gmd/ccgg/trends_ch4/, Accessed: 2020-06-22. 2, 3, 79

Dlugokencky, E., Pieter, T., and NOAA/GML (2020). Trends in Atmospheric Carbon Dioxide. www.esrl.noaa.gov/gmd/ccgg/trends/, Accessed: 2020-06-22. 2, 3

Dlugokencky, E. J., Nisbet, E. G., Fisher, R., and Lowry, D. (2011). Global atmospheric methane: budget, changes and dangers. Philosophical Transactions: Mathematical, Physical and Engineering Sciences, 369(1943):2058–2072. 19

E-PRTR (2017). European Pollutant Release and Transfer Register (E-PRTR) - greenhouse gas overview 2017. https://prtr.eea.europa.eu/#/areaoverview. Accessed: 2020-09-30. 6, 45, 80, 91, 92, 98, 112, 116, 125, 131

Ehhalt, D., Prather, M., Dentener, F., Derwent, R., Dlugokencky, E., Holland, E., Isaksen, I., Katima, J., Kirchhoff, V., Matson, P., Midgley, P., and Wang, M. (2001). Atmospheric Chemistry and Greenhouse Gases. In: Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the

Intergovernmental Panel on Climate Change. Technical report, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. 3

Engel, A., Bönisch, H., Ullrich, M., Sitals, R., Membrive, O., Danis, F., and Crevoisier, C. (2017). Mean age of stratospheric air derived from AirCore observations. Atmospheric Chemistry and Physics, 17(11):6825–6838. 19, 20

EPA (2019). United States Environmental Protection Agency, Ventilation Air Methane (VAM) Utilization Technologies. https://www.epa.gov/sites/production/files/2019-11/documents/vam_technologies.pdf, Accessed: 2021-06-12. 117

Etheridge, D., Steele, L., Langenfelds, R., Francey, R., Barnola, J., and Morgan, V. (1998a). Historical CO_2 record from the Law Dome DE08, DE08-2, and DSS ice cores. In Trends: A Compendium of Data on Global Change. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., U.S.A. 2, 3

Etheridge, D. M., Steele, L. P., Francey, R. J., and Langenfelds, R. L. (1998b). Atmospheric methane between 1000 A.D. and present: Evidence of anthropogenic emissions and climatic variability. J. Geophys. Res., 103(D13):15–979. 2

Etminan, M., Myhre, G., Highwood, E. J., and Shine, K. P. (2016). Radiative forcing of carbon dioxide, methane, and nitrous oxide: A significant revision of the methane radiative forcing. Geophysical Research Letters, 43(24):12,614–12,623. 3, 44, 79

Fiehn, A., Kostinek, J., Eckl, M., Klausner, T., Gałkowski, M., Chen, J., Gerbig, C., Röckmann, T., Maazallahi, H., Schmidt, M., Korbeń, P., N cki, J., Jagoda, P., Wildmann, N., Mallaun, C., Bun, R., Nickl, A.-L., Jöckel, P., Fix, A., and Roiger, A. (2020). Estimating CH_4, CO_2, and CO emissions from coal mining and industrial activities in the Upper Silesian Coal Basin using an aircraft-based mass balance approach. Atmospheric Chemistry and Physics Discussions, 2020:1–33. 8, 46, 58, 80, 97, 98, 99, 100, 116, 125, 131

Fisher, R. E., France, J. L., Lowry, D., Lanoisellé, M., Brownlow, R., Pyle, J. A., Cain, M., Warwick, N., Skiba, U. M., Drewer, J., Dinsmore, K. J., Leeson, S. R., Bauguitte, S. J. B., Wellpott, A., O'Shea, S. J., Allen, G., Gallagher, M. W., Pitt, J., Percival, C. J., Bower, K., George, C., Hayman, G. D., Aalto, T., Lohila, A., Aurela, M., Laurila, T., Crill, P. M., McCalley, C. K., and Nisbet, E. G. (2017). Measurement of the ^{13}C isotopic signature of methane emissions from northern European wetlands. Global Biogeochemical Cycles, 31(3):605–623. 5

Fix, A., Amediek, A., Bovensmann, H., Ehret, G., Gerbig, C., Gerilowski, K., Pfeilsticker, K., Roiger, A., and Zöger, M. (2018). CoMet: an airborne mission to simultaneously measure CO_2 and CH_4 using lidar, passive remote sensing, and in-situ techniques. EPJ Web Conf., 176:1–4. 46, 79

France, J. L., Bateson, P., Dominutti, P., Allen, G., Andrews, S., Bauguitte, S., Coleman, M., Lachlan-Cope, T., Fisher, R. E., Huang, L., Jones, A. E., Lee, J., Lowry, D., Pitt, J., Purvis, R., Pyle, J., Shaw, J., Warwick, N., Weiss, A., Wilde, S., Witherstone, J., and Young, S. (2021). Facility level measurement of offshore oil and gas installations from a medium-sized airborne platform: method development for quantification and source identification of methane emissions. Atmospheric Measurement Techniques, 14(1):71–88. 58

Frankenberg, C., Meirink, J. F., van Weele, M., Platt, U., and Wagner, T. (2005). Assessing Methane Emissions from Global Space-Borne Observations. Science, 308(5724):1010–1014. 7

Frankenberg, C., Thorpe, A. K., Thompson, D. R., Hulley, G., Kort, E. A., Vance, N., Borchardt, J., Krings, T., Gerilowski, K., Sweeney, C., Conley, S., Bue, B. D., Aubrey, A. D., Hook, S., and Green, R. O. (2016). Airborne methane remote measurements reveal heavy-tail flux distribution in Four Corners region. Proceedings of the National Academy of Sciences, 113(35):9734–9739. 120

Fritz, B., Shaw, B. W., and Parnell, C. (2005). Influence of meteorological time frame and variation on horizontal dispersion coefficients in Gaussian dispersion modeling. Transactions of the ASABE, 48:1185–1196. 62, 113

Galperin, B., Sukoriansky, S., and Anderson, P. S. (2007). On the critical Richardson number in stably stratified turbulence. Atmospheric Science Letters, 8(3):65–69. 14

Gasbarra, D., Toscano, P., Famulari, D., Finardi, S., Di Tommasi, P., Zaldei, A., Carlucci, P., Magliulo, E., and Gioli, B. (2019). Locating and quantifying multiple landfills methane emissions using aircraft data. Environmental Pollution, 254:112987. 8

Global Carbon Project (2016). Global Methane Budget. 3, 4

Golston, L. M., Tao, L., Brosy, C., Schäfer, K., Wolf, B., McSpiritt, J., Buchholz, B., Caulton, D. R., Pan, D., Zondlo, M. A., Yoel, D., Kunstmann, H., and McGregor, M. (2017). Lightweight mid-infrared methane sensor for unmanned aerial systems. Applied Physics B, 123(6):170. 10, 45, 110, 111

Grare, L., Lenain, L., and Melville, W. K. (01 Nov. 2016). The Influence of Wind Direction on Campbell Scientific CSAT3 and Gill R3-50 Sonic Anemometer Measurements. Journal of Atmospheric and Oceanic Technology, 33(11):2477–2497. 81

Greatwood, C., Richardson, T. S., Freer, J., Thomas, R. M., MacKenzie, A. R., Brownlow, R., Lowry, D., Fisher, R. E., and Nisbet, E. G. (2017). Atmospheric Sampling on Ascension Island Using Multirotor UAVs. Sensors (Basel, Switzerland), 17(6):1189. 10, 45, 79, 111

Green, A. E. S., Singhal, R. P., and Venkateswar, R. (1980). Analytic Extensions of the Gaussian Plume Model. Journal of the Air Pollution Control Association, 30(7):773–776. 14, 15

Hannun, R. A., Wolfe, G. M., Kawa, S. R., Hanisco, T. F., Newman, P. A., Alfieri, J. G., Barrick, J., Clark, K. L., DiGangi, J. P., Diskin, G. S., King, J., Kustas, W. P., Mitra, B., Noormets, A., Nowak, J. B., Thornhill, K. L., and Vargas, R. (2020). Spatial heterogeneity in CO_2, CH_4, and energy fluxes: insights from airborne eddy covariance measurements over the Mid-Atlantic region. Environmental Research Letters, 15(3):035008. 8, 79

Hartery, S., Commane, R., Lindaas, J., Sweeney, C., Henderson, J., Mountain, M., Steiner, N., McDonald, K., Dinardo, S. J., Miller, C. E., Wofsy, S. C., and Chang, R. Y.-W. (2018). Estimating regional-scale methane flux and budgets using CARVE aircraft measurements over Alaska. Atmospheric Chemistry and Physics, 18(1):185–202. 8

Hartmann, D. L., Klein Tank, A. M. G., Rusticucci, M., Alexander, L. V., Brönnimann, S., Charabi, Y., Dentener, F. J., Dlugokencky, E. J., Easterling, D. R., Kaplan, A., Soden, B. J., Thorne, P. W., Wild, M., and Zhai, P. M. (2013). Observations: Atmosphere and Surface, pages 161–218. Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1 edition. 19

Higaki, S., Oya, Y., and Makide, Y. (2006). Emission of Methane from Stainless Steel Surface Investigated by Using Tritium as a Radioactive Tracer. Chemistry Letters, 35(3):292–293. 48

Hoinaski, L., Franco, D., and de Melo Lisboa, H. (2016). Comparison of plume lateral dispersion coefficients schemes: Effect of averaging time. Atmospheric Pollution Research, 7(1):134–141. 12

Holmes, N. S. and Morawska, L. (2006). A review of dispersion modelling and its application to the dispersion of particles: An overview of different dispersion models available. Atmospheric Environment, 40(30):5902–5928. 12

Hu, H., Landgraf, J., Detmers, R., Borsdorff, T., Aan de Brugh, J., Aben, I., Butz, A., and Hasekamp, O. (2018). Toward Global Mapping of Methane With TROPOMI: First Results and Intersatellite Comparison to GOSAT. Geophysical Research Letters, 45(8):3682–3689. 7

ICOS (2020). Integrated Carbon Observation System, ICOS in a nutshell. 7, 109

IEA (2020a). International Energy Agency, Coal 2020. Accessed: 2021-05-20. 116, 117

IEA (2020b). International Energy Agency, Coal-fired power is not on track. Accessed: 2021-05-20. 116

IPCC (2013). Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Technical report, Cambridge University Press. 3

IPCC (2014a). Climate Change 2014: Impacts, Adaptation, and Vulnerability, Part A: Global and Sectoral Aspects, Contribution of Working Group II to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Technical report, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. 18, 44

IPCC (2014b). Climate Change 2014: Impacts, Adaptation, and Vulnerability, Part B: Regional Aspects, Contribution of Working Group II to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Technical report, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. 18

Jacob, D. J. (1999). Introduction to Atmospheric Chemistry. Princeton University Press. 5

Karion, A., Sweeney, C., Kort, E. A., Shepson, P. B., Brewer, A., Cambaliza, M., Conley, S. A., Davis, K., Deng, A., Hardesty, M., Herndon, S. C., Lauvaux, T., Lavoie, T., Lyon, D., Newberger, T., Pétron, G., Rella, C., Smith, M., Wolter, S., Yacovitch, T. I., and Tans, P. (2015). Aircraft-based estimate of total methane emissions from the barnett shale region. Environmental Science & Technology, 49(13):8124–8131. 58

Karion, A., Sweeney, C., Pétron, G., Frost, G., Hardesty, R. M., Kofler, J., Miller, B. R., Newberger, T., Wolter, S., Banta, R., Brewer, A., Dlugokencky, E., Lang, P., Montzka, S. A., Schnell, R., Tans, P., Trainer, M., Zamora, R., and Conley, S. (2013). Methane emissions estimate from airborne measurements over a western United States natural gas field. Geophysical Research Letters, 40(16):4393–4397. 8, 54, 79

Karion, A., Sweeney, C., Tans, P., and Newberger, T. (2010). AirCore: An Innovative Atmospheric Sampling System. Journal of Atmospheric and Oceanic Technology, 27(11):1839–1853. 19, 20, 38, 108, 121, 128

Khan, A., Schaefer, D., Tao, L., Miller, D. J., Sun, K., Zondlo, M. A., Harrison, W. A., Roscoe, B., and Lary, D. J. (2012). Low Power Greenhouse Gas Sensors for Unmanned Aerial Vehicles. Remote Sensing, 4(12):1355–1368. 10, 19, 45

Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J. G., Dlugokencky, E. J., Bergamaschi, P., Bergmann, D., Blake, D. R., Bruhwiler, L., Cameron-smith, P., Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E. L., Houweling, S., Josse, B., Fraser, P. J., Krummel, P. B., françois Lamarque, J., Langenfelds, R. L., Quéré, C. L., Naik, V., O'doherty, S., Palmer, P. I., Pison, I., Plummer, D., Poulter, B., Prinn, R. G., Rigby, M., Ringeval, B., Santini, M., Schmidt, M., Shindell, D. T., Simpson, I. J., Spahni, R., Steele, P. L., Strode, S. A., Sudo, K., Szopa, S., Van Der Werf, G. R., Voulgarakis, A., Weele, M. V., Weiss, R. F., Williams, J. E., and Zeng, G. (2013). Three decades of global methane sources and sinks. Nature Geoscience, 6(10):813. 4, 5, 19, 44, 45, 79, 107

Kostinek, J., Roiger, A., Eckl, M., Fiehn, A., Luther, A., Wildmann, N., Klausner, T., Fix, A., Knote, C., Stohl, A., and Butz, A. (2021). Estimating upper silesian coal mine methane emissions from airborne in situ observations and dispersion modeling. Atmospheric Chemistry and Physics, 21(11):8791–8807. 46

Krautwurst, S., Gerilowski, K., Borchardt, J., Wildmann, N., Galkowski, M., Swolkien, J., Marshall, J., Fiehn, A., Roiger, A., Ruhtz, T., Gerbig, C., Necki, J., Burrows, J. P., Fix, A., and Bovensmann, H. (2021). Quantification of CH_4 coal mining emissions in Upper Silesia by passive airborne remote sensing observations with the MAMAP instrument during CoMet. Atmospheric Chemistry and Physics Discussions, 2021:1–39. 46

Krautwurst, S., Gerilowski, K., Jonsson, H. H., Thompson, D. R., Kolyer, R. W., Iraci, L. T., Thorpe, A. K., Horstjann, M., Eastwood, M., Leifer, I., Vigil, S. A., Krings, T., Borchardt, J., Buchwitz, M., Fladeland, M. M., Burrows, J. P., and Bovensmann, H. (2017). Methane emissions from a Californian landfill, determined from airborne remote sensing and in situ measurements. Atmospheric Measurement Techniques, 10(9):3429–3452. 8, 54, 79

Krings, T., Gerilowski, K., Buchwitz, M., Hartmann, J., Sachs, T., Erzinger, J., Burrows, J. P., and Bovensmann, H. (2013). Quantification of methane emission rates from coal mine ventilation shafts using airborne remote sensing data. Atmospheric Measurement Techniques, 6(1):151–166. 54, 112

Krings, T., Gerilowski, K., Buchwitz, M., Reuter, M., Tretner, A., Erzinger, J., Heinze, D., Pflüger, U., Burrows, J. P., and Bovensmann, H. (2011). MAMAP - A new spectrometer system for column-averaged methane and carbon dioxide observations from aircraft: Retrieval algorithm and first inversions for point source emission rates. Atmospheric Measurement Techniques, 4(9):1735–1758. 54

Krings, T., Neininger, B., Gerilowski, K., Krautwurst, S., Buchwitz, M., Burrows, J. P., Lindemann, C., Ruhtz, T., Schüttemeyer, D., and Bovensmann, H. (2018). Airborne remote sensing and in situ measurements of atmospheric CO_2 to quantify point source emissions. Atmospheric Measurement Techniques, 11(2):721–739. 54

Kumar, P., Broquet, G., Yver-Kwok, C., Laurent, O., Gichuki, S., Caldow, C., Cropley, F., Lauvaux, T., Ramonet, M., Berthe, G., Martin, F., Duclaux, O., Juery, C., Bouchet, C., and Ciais, P. (2020). Mobile atmospheric measurements and local-scale inverse estimation of the location and rates of brief CH_4 and CO_2 releases from point sources. Atmospheric Measurement Techniques Discussions, 2020:1–27. 9

Kump, L. R., Kasting, J. E., and Crane, R. G. (2009). The Earth System. Prentice Hall, 3rd edition edition. 2

Kunz, M., Lavric, J. V., Gasche, R., Gerbig, C., Grant, R. H., Koch, F.-T., Schumacher, M., Wolf, B., and Zeeman, M. (2020). Surface flux estimates derived from UAS-based mole fraction measurements by means of a nocturnal boundary layer budget approach. Atmospheric Measurement Techniques, 13(4):1671–1692. 9, 10, 79

Kunz, M., Lavric, J. V., Gerbig, C., Tans, P., Neff, D., Hummelgård, C., Martin, H., Rödjegård, H., Wrenger, B., and Heimann, M. (2018). COCAP: a carbon dioxide analyser for small unmanned aircraft systems. Atmospheric Measurement Techniques, 11(3):1833–1849. 10, 19, 45

Kuze, A., Suto, H., Nakajima, M., and Hamazaki, T. (2009). Thermal and near infrared sensor for carbon observation Fourier-transform spectrometer on the Greenhouse Gases Observing Satellite for greenhouse gases monitoring. Appl. Opt., 48(35):6716–6733. 7

Lagzi, I., Meszaros, R., Gelybo, G., and Leelossy, A. (2013). Atmospheric Chemistry. Eotvos Lorand University. 12, 52

Leelőssy, Á., Molnár, F., Izsák, F., Havasi, Á., Lagzi, I., and Mészáros, R. (2014). Dispersion modeling of air pollutants in the atmosphere: A review. Central European Journal of Geosciences, 6(3):257–278. 12

Loulergue, L., Schilt, A., Spahni, R., Masson-Delmotte, V., Blunier, T., Lemieux, B., Barnola, J.-M., Raynaud, D., Stocker, T. F., and Chappellaz, J. (2008). Orbital and millennial-scale features of atmospheric CH_4 over the past 800,000 years. Nature, 453(7193):383–386. 3

Lowry, D., Brownlow, R., Fisher, R., Nisbet, E., Lanoisellé, M., France, J., Thomas, R., Mackenzie, R., Richardson, T., Greatwood, C., Freer, J., Cain, M., Warwick, N., and Pyle, J. (2015). Methane at Ascension Island, southern tropical Atlantic Ocean: continuous ground measurement and vertical profiling above the Trade Wind Inversion. EGU General Assembly Conference Abstracts, 17, p. 7100. 10, 19, 79, 111

Luther, A., Kleinschek, R., Scheidweiler, L., Defratyka, S., Stanisavljevic, M., Forstmaier, A., Dandocsi, A., Wolff, S., Dubravica, D., Wildmann, N., Kostinek, J., Jöckel, P., Nickl, A. L., Klausner, T., Hase, F., Frey, M., Chen, J., Dietrich, F., Necki, J., Swolkińń, J., Fix, A., Roiger, A., and Butz, A. (2019). Quantifying CH_4 emissions from hard coal mines using mobile sun-viewing Fourier transform spectrometry. Atmos. Meas. Tech., 12(10):5217–5230. 46, 112, 115

Lyman, S. N., Tran, T., Mansfield, M. L., and Ravikumar, A. P. (2019). Aerial and ground-based optical gas imaging survey of Uinta Basin oil and gas wells. Elementa: Science of the Anthropocene, 7(43):1–13. 119

Maazallahi, H., Fernandez, J. M., Menoud, M., Zavala-Araiza, D., Weller, Z. D., Schwietzke, S., von Fischer, J. C., Denier van der Gon, H., and Röckmann, T. (2020). Methane mapping, emission quantification and attribution in two European cities; Utrecht, NL and Hamburg, DE. Atmospheric Chemistry and Physics Discussions, 2020:1–30. 9

Marshall, J. and Plump, A. (2008). Atmosphere, Ocean and Climate Dynamics - An introductory text. Academic Press, 1st edition edition. 2

Martinez, B., Miller, T. W., and Yalin, A. P. (2020). Cavity Ring-Down Methane Sensor for Small Unmanned Aerial Systems. Sensors, 20(2):1–11. 10, 11, 79, 110, 111

Massman, W. J. (1998). A review of the molecular diffusivities of H_2O, CO_2, CH_4, CO, O_3, SO_2, NH_3, N_2O, NO, and NO_2 in air, O_2 and N_2 near STP. Atmospheric Environment, 32(6):1111–1127. ID: 271798. 39

Membrive, O., Crevoisier, C., Sweeney, C., Danis, F., Hertzog, A., Engel, A., Bönisch, H., and Picon, L. (2017). AirCore-HR: A high resolution column sampling to enhance the vertical description of CH_4 and CO_2. Atmospheric Measurement Techniques Discussions, 10(6):2163–2181. 19, 20

Menoud, M., Röckmann, T., Fernandez, J., Bakkaloglu, S., Lowry, D., Korben, P., Schmidt, M., Stanisavljevic, M., Necki, J., Defratyka, S., and Kwok, C. Y. (2020). mamenoud/MEMO2_isotopes: v8.1 complete (Version v8.1.0) [Data set], Zenodo. https://doi.org/10.5281/zenodo.4062356. 86

Miller, S. M., Wofsy, S. C., Michalak, A. M., Kort, E. A., Andrews, A. E., Biraud, S. C., Dlugokencky, E. J., Eluszkiewicz, J., Fischer, M. L., Janssens-Maenhout, G., Miller, B. R., Miller, J. B., Montzka, S. A., Nehrkorn, T., and Sweeney, C. (2013). Anthropogenic emissions of methane in the United States. Proceedings of the National Academy of Sciences, 110(50):20018. 8

Monin, A. and Obukhov, A. (1954). Basic Laws of Turbulent Mixing in the Surface Layer of the Atmosphere. Contrib. Geophys. Inst. Acad. Sci. USSR,, 24:163–187. 14

Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., and Zhang, H. (2013). Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Technical report, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA. 4

Mønster, J., Kjeldsen, P., and Scheutz, C. (2019). Methodologies for measuring fugitive methane emissions from landfills – A review. Waste Management, 87:835–859. 118

Nara, H., Tanimoto, H., Tohjima, Y., Mukai, H., Nojiri, Y., and Machida, T. (2014). Emissions of methane from offshore oil and gas platforms in Southeast Asia. Scientific Reports, 4:6503–6509. 37

NASA (2020). NASA's Goddard Institute for Space Studies (GISS), Global Temperature. https://climate.nasa.gov/vital-signs/global-temperature/, Accessed: 2020-06-22. 2

Nathan, B. J., Golston, L. M., O'Brien, A. S., Ross, K., Harrison, W. A., Tao, L., Lary, D. J., Johnson, D. R., Covington, A. N., Clark, N. N., and Zondlo, M. A. (2015). Near-Field Characterization of Methane Emission Variability from a Compressor Station Using a Model Aircraft. Environmental Science & Technology, 49(13):7896–7903. 9, 10, 45, 79, 110, 111, 113

Nickl, A.-L., Mertens, M., Roiger, A., Fix, A., Amediek, A., Fiehn, A., Gerbig, C., Galkowski, M., Kerkweg, A., Klausner, T., Eckl, M., and Jöckel, P. (2020). Hindcasting and forecasting of regional methane from coal mine emissions in the Upper Silesian Coal Basin using the online nested global regional chemistry–climate model MECO(n) (MESSy v2.53). Geoscientific Model Development, 13(4):1925–1943. 8, 80

Nisbet, E. G., Dlugokencky, E. J., Manning, M. R., Lowry, D., Fisher, R. E., France, J. L., Michel, S. E., Miller, J. B., White, J. W. C., Vaughn, B., Bousquet, P., Pyle, J. A., Warwick, N. J., Cain, M., Brownlow, R., Zazzeri, G., Lanoisellé, M., Manning, A. C., Gloor, E., Worthy, D. E. J., Brunke, E.-G., Labuschagne, C., Wolff, E. W., and Ganesan, A. L. (2016). Rising atmospheric methane: 2007–2014 growth and isotopic shift. Global Biogeochemical Cycles, 30(9):1356–1370. 107

Nisbet, E. G., Manning, M. R., Dlugokencky, E. J., Fisher, R. E., Lowry, D., Michel, S. E., Myhre, C. L., Platt, S. M., Allen, G., Bousquet, P., Brownlow, R., Cain, M., France, J. L., Hermansen, O., Hossaini, R., Jones, A. E., Levin, I., Manning, A. C., Myhre, G., Pyle, J. A., Vaughn, B. H., Warwick, N. J., and White, J. W. C. (2019). Very Strong Atmospheric Methane Growth in the 4 Years 2014–2017: Implications for the Paris Agreement. Global Biogeochemical Cycles, 33(3):318–342. 107

Pandey, S., Gautam, R., Houweling, S., van der Gon, H. D., Sadavarte, P., Borsdorff, T., Hasekamp, O., Landgraf, J., Tol, P., van Kempen, T., Hoogeveen, R., van Hees, R., Hamburg, S. P., Maasakkers, J. D., and Aben, I. (2019). Satellite observations reveal extreme methane leakage from a natural gas well blowout. Proceedings of the National Academy of Sciences, 116(52):26376–26381. 120

Pasquill, F. (1961). The estimation of the dispersion of windborne material. Meteor. Mag., 90(1161):33–49. 12, 13, 56

Paul, D., Chen, H., Been, H. A., Kivi, R., and Meijer, H. A. J. (2016). Radiocarbon analysis of stratospheric CO_2 retrieved from AirCore sampling. Atmospheric Measurement Techniques Discussions, 9(10):4997–5006. 19

Pavlov, A. A., Kasting, J. F., Brown, L. L., Rages, K. A., and Freedman, R. (2000). Greenhouse warming by CH_4 in the atmosphere of early Earth. Journal of Geophysical Research: Planets, 105(E5):11981–11990. 1

Peischl, J., Eilerman, S. J., Neuman, J. A., Aikin, K. C., de Gouw, J., Gilman, J. B., Herndon, S. C., Nadkarni, R., Trainer, M., Warneke, C., and Ryerson, T. B. (2018). Quantifying Methane and Ethane Emissions to the Atmosphere From Central and Western U.S. Oil and Natural Gas Production Regions. Journal of Geophysical Research: Atmospheres, 123(14):7725–7740. 8

Peischl, J., Karion, A., Sweeney, C., Kort, E. A., Smith, M. L., Brandt, A. R., Yeskoo, T., Aikin, K. C., Conley, S. A., Gvakharia, A., Trainer, M., Wolter, S., and Ryerson, T. B. (2016). Quantifying atmospheric methane emissions from oil and natural gas production in the Bakken shale region of North Dakota. Journal of Geophysical Research: Atmospheres, 121(10):6101–6111. 8

Peischl, J., Ryerson, T. B., Brioude, J., Aikin, K. C., Andrews, A. E., Atlas, E., Blake, D., Daube, B. C., de Gouw, J. A., Dlugokencky, E., Frost, G. J., Gentner, D. R., Gilman, J. B., Goldstein, A. H., Harley, R. A., Holloway, J. S., Kofler, J., Kuster, W. C., Lang, P. M., Novelli, P. C., Santoni, G. W., Trainer, M., Wofsy, S. C., and Parrish, D. D. (2013). Quantifying sources of methane using light alkanes in the Los Angeles basin, California. Journal of Geophysical Research: Atmospheres, 118(10):4974–4990. 118

Pitt, J. R., Allen, G., Bauguitte, S. J.-B., Gallagher, M. W., Lee, J. D., Drysdale, W., Nelson, B., Manning, A. J., and Palmer, P. I. (2019). Assessing london co_2, ch_4 and co emissions using aircraft measurements and dispersion modelling. Atmospheric Chemistry and Physics, 19(13):8931–8945. 58

Quay, P., Stutsman, J., Wilbur, D., Snover, A., Dlugokencky, E., and Brown, T. (1999). The isotopic composition of atmospheric methane. Global Biogeochemical Cycles, 13(2):445–461. 86

Ravikumar, A. P., Wang, J., and Brandt, A. R. (2017). Are Optical Gas Imaging Technologies Effective For Methane Leak Detection?,. Environmental Science & Technology, 51(1):718–724. 119

Rawlings, J., Pantula, S., and Dickey, D. (1998). Applied regression analysis. Springer texts in statistics. Springer, New York, NY [u.a.], 2nd edition. 53

Rella, C. W., Chen, H., Andrews, A. E., Filges, A., Gerbig, C., Hatakka, J., Karion, A., Miles, N. L., Richardson, S. J., Steinbacher, M., Sweeney, C., Wastine, B., and

Zellweger, C. (2013). High accuracy measurements of dry mole fractions of carbon dioxide and methane in humid air. Atmospheric Measurement Techniques, 6(3):837–860. 19, 23

Riddick, S. N., Mauzerall, D. L., Celia, M., Allen, G., Pitt, J., Kang, M., and Riddick, J. C. (2020). The calibration and deployment of a low-cost methane sensor. Atmospheric Environment, 230:117440. 119

Salesky, S. T. and Chamecki, M. (2012). Random Errors in Turbulence Measurements in the Atmospheric Surface Layer: Implications for Monin–Obukhov Similarity Theory. Journal of the Atmospheric Sciences, 69(12):3700 – 3714. 113

Satar, E., Berhanu, T. A., Brunner, D., Henne, S., and Leuenberger, M. (2016). Continuous $CO_2/CH_4/CO$ measurements (2012–2014) at Beromünster tall tower station in Switzerland. Biogeosciences, 13(9):2623–2635. 8, 79

Saunois, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J. G., Dlugokencky, E. J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F. N., Castaldi, S., Jackson, R. B., Alexe, M., Arora, V. K., Beerling, D. J., Bergamaschi, P., Blake, D. R., Brailsford, G., Brovkin, V., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry, C., Frankenberg, C., Gedney, N., Höglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H. S., Kleinen, T., Krummel, P., Lamarque, J. F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K. C., Marshall, J., Melton, J. R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F. J. W., Patra, P. K., Peng, C., Peng, S., Peters, G. P., Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W. J., Saito, M., Santini, M., Schroeder, R., Simpson, I. J., Spahni, R., Steele, P., Takizawa, A., Thornton, B. F., Tian, H., Tohjima, Y., Viovy, N., Voulgarakis, A., van Weele, M., van der Werf, G. R., Weiss, R., Wiedinmyer, C., Wilton, D. J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X., Yoshida, Y., Zhang, B., Zhang, Z., and Zhu, Q. (2016a). The global methane budget 2000–2012. Earth Syst. Sci. Data, 8(2):697–751. 3, 4, 79, 111, 112

Saunois, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J. G., Dlugokencky, E. J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F. N., Castaldi, S., Jackson, R. B., Alexe, M., Arora, V. K., Beerling, D. J., Bergamaschi, P., Blake, D. R., Brailsford, G., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Frankenberg, C., Gedney, N., Höglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H. S., Kleinen, T., Krummel, P., Lamarque, J. F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., Melton, J. R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F.-J. W., Patra, P. K., Peng, C., Peng, S., Peters, G. P., Pison, I., Prinn, R., Ramonet, M., Riley, W. J., Saito, M., Santini, M., Schroeder, R., Simpson, I. J., Spahni, R., Takizawa, A., Thornton, B. F., Tian, H., Tohjima, Y., Viovy, N., Voulgarakis, A., Weiss, R., Wilton, D. J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X., Yoshida, Y., Zhang, B., Zhang, Z., and Zhu, Q. (2017). Variability and quasi-decadal changes in the methane budget over the period 2000–2012. Atmospheric Chemistry and Physics, 17(18):11135–11161. 4, 79, 107

Saunois, M., Jackson, R. B., Bousquet, P., Poulter, B., and Canadell, J. G. (2016b). The growing role of methane in anthropogenic climate change. Environmental Research Letters, 11(12):120207. 3, 4, 19, 79

Saunois, M., Stavert, A. R., Poulter, B., Bousquet, P., Canadell, J. G., Jackson, R. B., Raymond, P. A., Dlugokencky, E. J., Houweling, S., Patra, P. K., Ciais, P., Arora, V. K., Bastviken, D., Bergamaschi, P., Blake, D. R., Brailsford, G., Bruhwiler, L., Carlson, K. M., Carrol, M., Castaldi, S., Chandra, N., Crevoisier, C., Crill, P. M., Covey, K., Curry, C. L., Etiope, G., Frankenberg, C., Gedney, N., Hegglin, M. I., Höglund-Isaksson, L., Hugelius, G., Ishizawa, M., Ito, A., Janssens-Maenhout, G., Jensen, K. M., Joos, F., Kleinen, T., Krummel, P. B., Langenfelds, R. L., Laruelle, G. G., Liu, L., Machida, T., Maksyutov, S., McDonald, K. C., McNorton, J., Miller, P. A., Melton, J. R., Morino, I., Müller, J., Murguia-Flores, F., Naik, V., Niwa, Y., Noce, S., O'Doherty, S., Parker, R. J., Peng, C., Peng, S., Peters, G. P., Prigent, C., Prinn, R., Ramonet, M., Regnier, P., Riley, W. J., Rosentreter, J. A., Segers, A., Simpson, I. J., Shi, H., Smith, S. J., Steele, L. P., Thornton, B. F., Tian, H., Tohjima, Y., Tubiello, F. N., Tsuruta, A., Viovy, N., Voulgarakis, A., Weber, T. S., van Weele, M., van der Werf, G. R., Weiss, R. F., Worthy, D., Wunch, D., Yin, Y., Yoshida, Y., Zhang, W., Zhang, Z., Zhao, Y., Zheng, B., Zhu, Q., Zhu, Q., and Zhuang, Q. (2020). The Global Methane Budget 2000–2017. Earth System Science Data, 12(3):1561–1623. 4, 44, 45, 79, 107, 111, 117, 118

Schwietzke, S., Pétron, G., Conley, S., Pickering, C., Mielke-Maday, I., Dlugokencky, E. J., Tans, P. P., Vaughn, T., Bell, C., Zimmerle, D., Wolter, S., King, C. W., White, A. B., Coleman, T., Bianco, L., and Schnell, R. C. (2017). Improved Mechanistic Understanding of Natural Gas Methane Emissions from Spatially Resolved Aircraft Measurements. Environmental Science & Technology, 51(12):7286–7294. 8

Schwietzke, S., Sherwood, O. A., Bruhwiler, L. M. P., Miller, J. B., Etiope, G., Dlugokencky, E. J., Michel, S. E., Arling, V. A., Vaughn, B. H., White, J. W. C., and Tans, P. P. (2016). Upward revision of global fossil fuel methane emissions based on isotope database. Nature, 538(7623):88–91. 5

Seinfeld, J. H. and Pandis, S. N. (1998). Atmospheric chemistry and physics: from air pollution to climate change. John Wiley & Sons, Inc., Hoboken, New Jersey, U.S.A., 1st edition edition. 113

Shah, A., Allen, G., Pitt, J. R., Ricketts, H., Williams, P. I., Helmore, J., Finlayson, A., Robinson, R., Kabbabe, K., Hollingsworth, P., Rees-White, T. C., Beaven, R., Scheutz, C., and Bourn, M. (2019). A Near-Field Gaussian Plume Inversion Flux Quantification Method, Applied to Unmanned Aerial Vehicle Sampling. Atmosphere, 10(7):1–25. 10, 45, 113

Shah, A., Pitt, J. R., Ricketts, H., Leen, J. B., Williams, P. I., Kabbabe, K., Gallagher, M. W., and Allen, G. (2020). Testing the near-field Gaussian plume inversion flux quantification technique using unmanned aerial vehicle sampling. Atmospheric Measurement Techniques, 13(3):1467–1484. 9, 10, 45, 58, 79, 110, 111, 113

Sharan, M., Yadav, A. K., and Singh, M. P. (1995). Comparison of sigma schemes for estimation of air pollutant dispersion in low winds. Atmospheric Environment, 29(16):2051–2059. 14, 15

Shindell, D., Kuylenstierna, J. C. I., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z., Anenberg, S. C., Muller, N., Janssens-Maenhout, G., Raes, F., Schwartz, J., Faluvegi, G., Pozzoli, L., Kupiainen, K., Höglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V., Hicks, K., Oanh, N. T. K., Milly, G., Williams, M., Demkine, V., and Fowler, D. (2012). Simultaneously Mitigating Near-Term Climate Change and Improving Human Health and Food Security. Science, 335(6065):183–189. 4

Shukla, P., Skea, J., Slade, R., van Diemen, R., Haughey, E., Malley, J., Pathak, M., and Portugal, P. J. (2019). Technical Summary, 2019. In: Climate Change and Land: an IPCC special report on climate change, desertification, land degradation, sustainable land management, food security, and greenhouse gas fluxes in terrestrial ecosystems. Technical report, in press. 2

Simma, M., Mjøen, H., and Boström, T. (2020). Measuring Wind Speed Using the Internal Stabilization System of a Quadrotor Drone. Drones, 4(2):1–10. 118

Stanisavljevic, M. (2021). Isotopic signatures from coal mining shafts in the Upper Silesia Coal Basin. In preparation, 1(1):1–1. 86

Stockie, J. M. (2011). The Mathematics of Atmospheric Dispersion Modeling. SIAM Review, 53(2):349–372. 15

Stull, R. B. (1988). Introduction to Boundary Layer Meteorology. Springer, Dordrecht, Netherlands, 1 edition. 35

Sweeney, C., Karion, A., Wolter, S., Newberger, T., Guenther, D., Higgs, J. A., Andrews, A. E., Lang, P. M., Neff, D., Dlugokencky, E., Miller, J. B., Montzka, S. A., Miller, B. R., Masarie, K. A., Biraud, S. C., Novelli, P. C., Crotwell, M., Crotwell, A. M., Thoning, K., and Tans, P. P. (2015). Seasonal climatology of CO_2 across North America from aircraft measurements in the NOAA/ESRL Global Greenhouse Gas Reference Network. Journal of Geophysical Research: Atmospheres, 120(10):5155–5190. 8

Swolkień, J. (2020). Polish underground coal mines as point sources of methane emission to the atmosphere. International Journal of Greenhouse Gas Control, 94:102921. 45, 46, 64, 79, 80, 84, 85

Tans, P. and Keeling, R. (2017). Trends in Atmospheric Carbon Dioxide. National Oceanic and Atmospheric Administration, Earth System Research Laboratory, Global Monitoring Division, available at: http://www.esrl.noaa.gov/gmd/ccgg/trends/ (last access: April 2017). 19

Tans, P. P. (2009). System and method for providing vertical profile measurements of atmospheric gases. US Patent Office. 19

TCCON (2020). Total Carbon Column Observing Network, Home page. https://tccon-wiki.caltech.edu, Accessed: 2020-09-23. 7

The MathWorks, I. (2019). Optimization Toolbox™. https://nl.mathworks.com/help/optim/ug/fmincon.html, Accessed: 2019-08-03. 53

Thoma, E. and Squier, B. (2014). OTM 33 geospatial measurement of air pollution, remote emissions quantification (GMAP-REQ) and OTM33A geospatial measurement of air pollution-remote emissions quantification. https://cfpub.epa.gov/si/si_public_record_Report.cfm?Lab=NRMRL&dirEntryId=309632, Accessed: 2021-06-03. 59, 67, 114

Turnbull, J. C., Keller, E. D., Baisden, T., Brailsford, G., Bromley, T., Norris, M., and van, A. Z. (2014). Atmospheric measurement of point source fossil CO_2 emissions. Atmospheric Chemistry and Physics, 14(10):5001–5014. 10, 19, 45, 79

Turner, A. J., Frankenberg, C., and Kort, E. A. (2019). Interpreting contemporary trends in atmospheric methane. Proceedings of the National Academy of Sciences of the United States of America, 116(8):2805–2813. 4, 79, 107

Turner, D. (1970). Workbook of atmospheric dispersion estimates. U.S. Department of Health, Education, and Welfare, 1st edition edition. 13

Tuzson, B., Graf, M., Ravelid, J., Scheidegger, P., Kupferschmid, A., Looser, H., Morales, R. P., and Emmenegger, L. (2020). A compact QCL spectrometer for mobile, high-precision methane sensing aboard drones. Atmospheric Measurement Techniques, 13(9):4715–4726. 10, 11, 45, 79, 110, 111

van den Bossche, M., Rose, N. T., and De Wekker, S. F. J. (2017). Potential of a low-cost gas sensor for atmospheric methane monitoring. Sensors and Actuators B: Chemical, 238:501–509. 119

van der Laan, S., Neubert, R. E. M., Meijer, H. A. J., and Simpson, W. R. (2009). A single gas chromatograph for accurate atmospheric mixing ratio measurements of CO_2, CH_4, N_2O, SF_6 and CO. Atmospheric Measurement Techniques, 2(2):549–559. 28

Van Dingenen, R., Crippa, M., Maenhout, G., Guizzardi, D., and Dentener, F. (2018). Global trends of methane emissions and their impacts on ozone concentrations. Technical report, EUR 29394 EN, Publications Office of the European Union, Luxembourg. ISBN: 978-92-79-96550-0, doi: 10.2760/820175. 3, 79

Varon, D. J., Jacob, D. J., Jervis, D., and McKeever, J. (2020). Quantifying Time-Averaged Methane Emissions from Individual Coal Mine Vents with GHGSat-D Satellite Observations. Environmental Science & Technology, 54(16):10246–10253. 112, 120

Villa, T. F., Gonzalez, F., Miljievic, B., Ristovski, Z. D., and Morawska, L. (2016). An Overview of Small Unmanned Aerial Vehicles for Air Quality Measurements: Present Applications and Future Prospectives. Sensors, 16(7):1–29. 10, 45, 79

Vinkovic, K., Andersen, T., Peters, W., de Vries, M., Hansen, A., and Chen, H. (2021). Quantification of methane from dairy cows in the Netherlands. In preparation, 0(0):0. 9, 55, 109, 114, 118

Walter, S., Röckmann, T., and the $MEMO^2$ team (2020). $MEMO^2$: MEthane goes MObile – MEasurements and MOdelling. EGU General Assembly 2020,, EGU2020-6760(Online, 4–8 May 2020):1. https://doi.org/10.5194/egusphere-egu2020-6760. 8, 109

Watai, T., Machida, T., Ishizaki, N., and Inoue, G. (2006). A Lightweight Observation System for Atmospheric Carbon Dioxide Concentration Using a Small Unmanned Aerial Vehicle. Journal of Atmospheric and Oceanic Technology, 23(5):700–710. 19

Werner, C., Davis, K., Bakwin, P., Yi, C., Hurst, D., and Lock, L. (2003). Regional-scale measurements of CH_4 exchange from a tall tower over a mixed temperate/boreal lowland and wetland forest. Global Change Biology, 9(9):1251–1261. 8, 79

Winderlich, J., Chen, H., Gerbig, C., Seifert, T., Kolle, O., Lavrič, J. V., Kaiser, C., Höfer, A., and Heimann, M. (2010). Continuous low-maintenance CO_2/CH_4/H_2O measurements at the Zotino Tall Tower Observatory (ZOTTO) in Central Siberia. Atmospheric Measurement Techniques, 3(4):1113–1128. 49, 113

WMO (2020a). Observations. https://public.wmo.int/en/our-mandate/what-we-do/observations, Accessed: 2020-09-09. 6, 109

WMO (2020b). What we do. https://public.wmo.int/en/our-mandate/what-we-do, Accessed: 2020-09-09. 6

Wolf, B., Chwala, C., Fersch, B., Garvelmann, J., Junkermann, W., Zeeman, M. J., Angerer, A., Adler, B., Beck, C., Brosy, C., Brugger, P., Emeis, S., Dannenmann, M., De Roo, F., Diaz-Pines, E., Haas, E., Hagen, M., Hajnsek, I., Jacobeit, J., Jagdhuber, T., Kalthoff, N., Kiese, R., Kunstmann, H., Kosak, O., Krieg, R., Malchow, C., Mauder, M., Merz, R., Notarnicola, C., Philipp, A., Reif, W., Reineke, S., Rödiger, T., Ruehr, N., Schäfer, K., Schrön, M., Senatore, A., Shupe, H., Völksch, I., Wanninger, C., Zacharias, S., and Schmid, H. P. (2017). The SCALEX Campaign: Scale-Crossing Land Surface and Boundary Layer Processes in the TERENO-preAlpine Observatory. Bulletin of the American Meteorological Society, 98(6):1217–1234. 10, 45

Zannetti, P. (1990). Gaussian Models, pages 141–183. Springer US, Boston, MA. 12

Zazzeri, G., Lowry, D., Fisher, R. E., France, J. L., Lanoisellé, M., Kelly, B. F. J., Necki, J. M., Iverach, C. P., Ginty, E., Zimnoch, M., Jasek, A., and Nisbet, E. G. (2016). Carbon isotopic signature of coal-derived methane emissions to the atmosphere: from coalification to alteration. Atmospheric Chemistry and Physics, 16(21):13669–13680. 5, 45, 79

Zhang, Y., Gautam, R., Pandey, S., Omara, M., Maasakkers, J. D., Sadavarte, P., Lyon, D., Nesser, H., Sulprizio, M. P., Varon, D. J., Zhang, R., Houweling, S., Zavala-Araiza, D., Alvarez, R. A., Lorente, A., Hamburg, S. P., Aben, I., and Jacob, D. J. (2020). Quantifying methane emissions from the largest oil-producing basin in the United States from space. Science Advances, 6(17):eaaz5120. 7

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P

List of publications

5. **Truls Andersen**, Marcel De Vries, Jarosław Nęcki, Justyna Swolkień, Wouter Peters, Huilin Chen: Regional methane emissions from the Upper Silesia Coal Basin (USCB) quantified using UAV-based atmospheric measurements, in preparation
4. Katarina Vinkovic, **Truls Andersen**, Wouter Peters, Marcel de Vries, Arjan Hansen, and Huilin Chen, Quantification of methane from dairy cows in the Netherlands, in preparation.
3. **Truls Andersen**, Katarina Vinkovic, Marcel De Vries, Bert Kers, Jarosław Nęcki, Justyna Swolkień , Anke Roiger, Wouter Peters, and Huilin Chen : Quantifying methane emissions from coal mining ventilation shafts using an Unmanned Aerial Vehicle (UAV)-based active AirCore system, Atmospheric Environment X, In review, 2021.
2. Adrià Salvador Palau, Sabrina Eder, **Truls Andersen**, Anders Komár Ravn, Gianangelo Bracco, and Bodil Holst, Center-line intensity of a supersonic helium beam, Physical Review A, 98, 6, 063611, https://doi.org/10.1103/PhysRevA.98.063611, 2018.
1. **Truls Andersen**, Bert Scheeren, Wouter Peters, and Huilin Chen, A UAV-based active AirCore system for measurements of greenhouse gases, Atmospheric Measurement Techniques, 11, p. 2683-2699, https://doi.org/10.5194/amt-11-2683-2018, 2018.