University of Groningen

Quantifying Local to Regional Emissions of Methane Using UAV-based Atmospheric Concentration Measurements
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DOI:
10.33612/diss.190478126

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2021

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Discussion and Outlook

This chapter gives an overview of the most important findings of this PhD thesis. The findings are discussed and this chapter then provides an outlook based on the findings and the discussion. The relevance of the results are discussed in relation to using unmanned aerial vehicles in combination with air sampling techniques to quantify methane sources, or other greenhouse gas (GHG) emitters, with the primary focus of this thesis being CH$_4$ emitted from coal mines. The discussion also extends beyond this concept, discussing available methane sensors for drone applications, as well as the current demand for coal around the world and how Covid-19 has impacted the world’s coal market.

5.1 The importance of methane source quantification

According to Saunois et al. (2020), the global methane emissions for 2008 - 2017 were estimated from a top-down approach (atmospheric inversions) to be 576 [Tg CH$_4$ year$^{-1}$], where approximately 60 [%] is attributed to anthropogenic sources. It is estimated that the current anthropogenic methane emissions trajectory is somewhere between the two warmest IPCC scenarios (RCP8.5 and RCP6.0) (Nisbet et al., 2016, 2019). These scenarios estimate that the mean average temperature increase above 3 [°C] by the end of the century. To reach the targeted Paris agreement goal of limiting global warming to be within 1.5 - 2.0 [°C], methane emissions will require a significant reduction. Methane is not only a potent GHG, but is also directly linked to the production of ozone. Changes in the temporal variations and magnitude of methane sinks and sources have however been characterized by large uncertainties in the last decades (Kirschke et al., 2013; Saunois et al., 2017; Turner et al., 2019; Saunois et al., 2020). This makes methane very important in the big climate picture, and an important GHG to monitor. Due to the poorly determined amount of emitted methane to the atmosphere from coal mining operations, and the lacking verification of emission estimates from individual shafts, CH$_4$ quantification
from these sources is vitally important in reducing the uncertainties of these emissions.

5.2 Atmospheric methane measurements

5.2.1 The active AirCore system

In chapter 2 of this thesis the UAV-based active AirCore system is presented and tested. The active AirCore system is based on the stratospheric AirCore sampler (Karion et al., 2010), but instead uses a small KNF 020L micropump to pull air from the inlet of the active AirCore into the ∼ 50 [m] long coiled-up stainless steel tube. A small pinhole orifice with a pinhole diameter of 45 [μm] is located between the pump and the AirCore to obtain a vacuum (< 1/2 × ambient pressure) downstream the orifice. This ensures conditions for critical flow, so that the active AirCore is filled with a flowrate that only depends on the ambient upstream pressure. The inlet of the active AirCore system had a drying-tube mounted upfront, a small tube filled with magnesium perchlorate that would absorb water from the air as the air was being sampled. Ambient pressure, ambient temperature, ambient relative humidity, and temperature within the carbon fiber box housing was monitored by Honeywell TruStability HSC pressure sensors, a DHT22 relative humidity sensor, and PT100 element temperature thermistors through an Arduino MEGA 2650-based datalogger. GPS positioning was also monitored through the datalogger by a ATM2.5 NEO-6M GPS sensor, as well as on the drone itself. The whole system has a weight of ∼1.1 [kg] and a maximum sampling time of 16 minutes. The maximum sampling time does however depend on the type of UAV being used, as well as the sampling flowrate and volume of the active AirCore system.

The active AirCore system was first tested in laboratory conditions, where air was simultaneously sampled with the AirCore and analyzed on Cavity Ringdown Spectrometer (CRDS) model G2401 (Crosson, 2008) for CO\textsubscript{2}, CH\textsubscript{4}, CO, and H\textsubscript{2}O. The air from the AirCore was then analyzed on the same CRDS and R-squared values between the two data sets were found to be > 0.97 for CO\textsubscript{2}, CH\textsubscript{4}, and CO. The active AirCore system was attached to a DJI Inspire pro 1 UAV, and was field-tested at a coastal site close to the Wadden sea. It was here demonstrated that the build-up of the nocturnal boundary layer could clearly be observed using vertical profiles up to 500 [m] over 3 consecutive flights. A clear enhancements in both CH\textsubscript{4} and CO\textsubscript{2} was also captured during a low-altitude horizontal transect over the wetlands north of the Wadden sea dike, showcasing the active AirCore’s ability to capture samples both horizontally and vertically. Having the UAV-based active AirCore system hover next to the inlet of a CRDS at 60 [m], it was found that the correlation between the continuous CRDS measurements and the sampled air from the AirCore had an R-squared value > 0.95 for both CH\textsubscript{4} and CO\textsubscript{2}. The mean difference was found to be 0.138 ± 0.362 [ppm] for CO\textsubscript{2} and 5.9 ± 3.9 [ppb] for CH\textsubscript{4}. The AirCore signal is seen as a smoothed-out signal of the continuous measurements, which can be explained by the smearing effects the CRDS analyzer has on the sample during the analysis process, as well as the diffusion working on the sample within the AirCore.

It was found that the smear effect was the dominant factor in estimating the spatial resolution of the measurements, together with uncertainties in the GPS signal, molecular
diffusion, and Taylor dispersion within the AirCore during sampling, storage, and analysis. The effective spatial resolution was found to be between 24.1 - 46.0 [m] for CO$_2$ at typical cruising speeds of 1.5 - 2.5 [m/s], and 24.7 - 48.9 [m] for CH$_4$. An improvement in the spatial resolution would be possible by using a CRDS analyzer with a lower cavity pressure, such as 80 [Torr] (106 [hPa]). The lower cavity pressure would reduce the response time of the analyzer and in turn lead to a better spatial resolution. A further reduction of the cavity pressure would, however, only go so far. At a cavity pressure of 55 [Torr] (73 [hPa]), the molecular diffusion and smear effect contributes equally to the spatial resolution. A further reduction would mean the molecular diffusion takes over as the dominant factor of the spatial resolution, and thus a further reduction of the cavity pressure would not have as big an impact on the improvement of the spatial resolution. An 80 [Torr] analyzer was used together with the active AirCore system in Vinkovic et al. (2021), where they quantified emitted methane from a dairy farm in the Netherlands. The measurements done for chapters 3 and 4 did not have a low pressure cavity analyzer, but instead used a modified 140 [Torr] high-methane mode due to the high mole-fractions (signals reaching > 200 [ppm CH$_4$]).

It was found that the speed of the drone had a large impact on the spatial resolution, showcasing the importance of the flight planning. Due to the spatial resolution being proportional to the drone speed, a slow speed would be preferred. On the other hand, too slow would hinder the active AirCore system in being able to sample sources as much possible. Therefore a comprise has to be found to attempt to satisfy both cases. It would be beneficial to automate the flights to fly at a consistent speed of around 2 [m/s] to ensure that the spatial resolution remains as small as possible, while being able to cover the distance to make several passes downwind a plume. This was later applied in some of the data gathered for chapter 3 and 4.

### 5.2.2 Available methane sensors

The active AirCore system presented in chapter 2 gathers an air sample along the trajectory of the drone. This sample of air is then analyzed on a cavity ringdown spectrometer on the ground for methane mole-fractions and other specific GHG constituents (CO$_2$, CO, and H$_2$O). Although the UAV-based active AirCore system can provide accurate measurements of GHGs, it does not give in-flight information about the concentrations. The use of UAVs have become increasingly more popular in recent years, and when it comes to methane detection using UAVs, this is no different. As the second-most abundant anthropogenic GHG, there are many different sensors and analyzers capable of measuring the atmospheric mole fractions of methane. This section discusses different available sensors and analyzers for accurate methane detection that have good synergy with UAV applications. Table (5.1) presents a table with an overview over payload, type of analyzer, and methane precision for each study.

Using optical instruments are a common way of measuring atmospheric methane, and work on the principle of light intensity absorption as light interacts with the gas. Instruments such as the Cavity RingDown Spectrometers (CRDS) from Picarro and Los Gatos Research Inc. work on this principle, and are widely used by global (WMO, 2020a) and regional (ICOS, 2020; Walter et al., 2020) monitoring networks to obtain high precision
and accuracy measurements. Although these instruments can provide very accurate measurements, they often have the drawback of being large, heavy, and requiring significant power to run. To be able to fly these instruments on a drone, a very heavy-duty drone would be required, like in Berman et al. (2012). They flew a near-infrared Off-axis Integrated Cavity Output Spectrometer (Off-axis ICOS), weighing 19.5 [kg], using NASA’s Sensor Integrated Environmental Remote Research Aircraft (SIERRA). This is a fixed-wing drone with a wingspan of 6.1 [m] which can carry payloads up to 40 [kg]. They were able to measure CH$_4$ with a precision of 2.0 [ppb] at 1 [Hz], and CO$_2$ with a precision of 0.6 [ppm] at 1 [Hz]. The cost and time investment with such big UAVs are often large, and due to more strict policies being implemented on drone use, smaller drones and, in turn, smaller instrumentation is becoming increasingly necessary. Nathan et al. (2015) used a smaller fixed-wing drone equipped with a laser-based open path methane sensor, where the payload was 3.1 [kg]. They were able to measure methane with a precision of 0.1 [ppm] at 1 [Hz]. Another fixed-wing drone was used by Golston et al. (2017), where a open-path wavelength modulation spectrometer (payload weight 4.6 [kg]) was used to measure methane with a precision of 5 [ppb] at 1 [Hz].

The fixed-wing drones have the benefit of being able to (generally) carry a larger payload and fulfill larger power requirements, which can result in more accurate instrumentation. The distance which they can cover is also larger than that of rotary drones, and they naturally fly at larger speeds. Rotary drones are more flexible, fly with lower velocities, and offer the potential to hover. They are often easier to operate and require less permits for operation. Due to their smaller size, they are however limited in their carry capacity. One solution to this is to have an on-ground analyzer and tether a tube from the analyzer to the UAV. This method has the advantage of maintaining accurate measurements of CH$_4$ through the high-accuracy on-ground analyzer, but lacks flexibility due to the attached tube, which can potentially limit spatial sampling. Rotary tethered sampling of methane was done by Allen et al. (2019) as well as Shah et al. (2020). Allen et al. (2019) measured both CO$_2$ and CH$_4$ with a precision of 0.22 [ppm CO$_2$] and 1.08 [ppb CH$_4$] at 1 [Hz], and Shah et al. (2020) measured CH$_4$ with a precision of 0.7 [ppb] at 1 [Hz].

Although smaller size rotary drones are limited in their carrying capacity, several lightweight methane sensors have been developed to achieve high-accuracy measurements of methane measurements. For instance Shah et al. (2020) used a Off-axis ICOS on-board a rotary UAV, where the payload was 3.4 [kg], to measure methane at a precision of 2.2 [ppb] at 1 [Hz]. For larger-sized rotary drones, Martinez et al. (2020) developed an open-path CRDS methane sensor with a payload weight of 4.1 [kg], and were able to measure CH$_4$ with a precision of $\sim 10 - 30$ [ppb] at 1 [Hz]. Tuzson et al. (2020) developed a mid infrared laser absorption spectrometer for smaller-scale rotary drones and had a payload weight of 2.1 [kg]. They were able to measure methane with a precision of 1.1 [ppb] at 1 [Hz].

Like the $\sim 1.1$ [kg] active AirCore system presented in chapter 1, on-board sampling of air that can later be analyzed for atmospheric constituents is also a valid option for drone use. This method has the drawback of not being in-situ sampling, but has the advantage of being capable of analyzing the collected air sample for multiple GHG species on an on-ground high-accuracy analyzer. Besides the active AirCore system (Andersen et al.,
which gathers a continuous air sample along the trajectory of a drone for later analysis, Lowry et al. (2015); Brownlow et al. (2016), and Greatwood et al. (2017) use this principle, and gathered discrete bag samples of air at different altitude levels for later analysis. In the study of Lowry et al. (2015); Brownlow et al. (2016) and Greatwood et al. (2017), discrete 5L tedlar bag samples were collected up to a height of 2700 [m], and were analyzed in a lab for CH$_4$ mole fractions using a Picarro CRDS model 1301 with a precision of 0.5 [ppb], and later on for $\delta^{13}$CH$_4$. All these different drone systems have advantages and disadvantages, but there is no doubt that drones are, and will continue to be, a valuable platform for measuring atmospheric methane in the future.

Table 5.1. shows an overview of studies combining methane measurements and drones.

<table>
<thead>
<tr>
<th>Study</th>
<th>Payload weight (kg)</th>
<th>Type of analyzer</th>
<th>Type of drone</th>
<th>CH$_4$ precision at 1 [Hz] (ppb)</th>
<th>Other species precision at 1 [Hz]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Berman et al. (2012)</td>
<td>19.5</td>
<td>Near-infrared Off-Axis Integrated Cavity</td>
<td>Fixed wing</td>
<td>2.0</td>
<td>CO$_2$: 0.6 [ppm]</td>
</tr>
<tr>
<td>Nathan et al. (2015)</td>
<td>3.1</td>
<td>Laser-Based Open Path Integrated Cavity</td>
<td>Fixed wing</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>Golston et al. (2017)</td>
<td>4.6</td>
<td>Open-Path Wavelength Modulation Spectroscopy</td>
<td>Fixed wing + Rotary</td>
<td>5-10</td>
<td></td>
</tr>
<tr>
<td>Allen et al. (2019)</td>
<td>+0.6</td>
<td>Tethered Off-Axis Integrated Cavity</td>
<td>Rotary</td>
<td>1.08</td>
<td>CO$_2$: 0.22 [ppm]</td>
</tr>
<tr>
<td>Shah et al. (2020)</td>
<td>3.4</td>
<td>Off-Axis Integrated Cavity Output Spectroscopy</td>
<td>Rotary</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td>Shah et al. (2020)</td>
<td>4.8</td>
<td>Tethered Off-Axis Integrated Cavity</td>
<td>Rotary</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>Tuzson et al. (2020)</td>
<td>2.1</td>
<td>Mid Infrared Laser Absorption Spectrometer</td>
<td>Rotary</td>
<td>1.1</td>
<td></td>
</tr>
<tr>
<td>Martinez et al. (2020)</td>
<td>4.1</td>
<td>Open-path CRDS</td>
<td>Rotary</td>
<td>-10-30</td>
<td></td>
</tr>
<tr>
<td>Lowry et al. (2015)+ Brownlow et al. (2016)+ Greatwood et al. (2017)</td>
<td>&lt;0.5 5L Tedlar Bags + CRDS</td>
<td>Rotary</td>
<td>0.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Andersen et al. (2018)</td>
<td>1.1</td>
<td>Active AirCore + CRDS</td>
<td>Rotary</td>
<td>0.5</td>
<td>CO$_2$: 0.03 [ppm]</td>
</tr>
</tbody>
</table>

5.3 Coal mining

This thesis focuses on methane emissions from coal mine ventilation shafts, as presented in chapter 3 and chapter 4. Roughly 10 [%] of anthropogenic methane is estimated to come from underground coal mining complexes (Saunois et al., 2016a). During the coal mining process, most of the released methane is vented directly to the atmosphere through ventilation shafts on the ground. Air from the surface is also pumped down into the underground mines to ensure that the methane concentration does not reach levels above 0.5 [%], which could cause accidental ignition. The global estimate of emitted coal mining related methane is 29 - 61 [Tg CH$_4$ year$^{-1}$] between 2008 and 2017. The large uncertainty stems in part from poorly detailed data from all major coal producing countries (Saunois et al., 2020). Silesia, a region in southern Poland, had in 2017 an estimated emission of
447 [kt CH$_4$/year], about 27.3 [%] of Europe’s total CH$_4$ emissions (E-PRTR, 2017). This region’s large contribution to Europe’s total methane emission makes this region a prime candidate for study.

### 5.3.1 Methane emission quantification from a coal mine ventilation shaft

Chapter 3 investigates a single coal mine ventilation shaft in the USCB, using the UAV-based active AirCore system presented in chapter 2. The emission from a single coal mine ventilation shaft can vary drastically from shaft to shaft. The E-PRTR inventory (E-PRTR, 2017) lists a spread of 1 - 68 [kt CH$_4$/year] for the complete coal mines located in the Silesia region, and an internal Carbon Dioxide and Methane Mission (CoMet) inventory, which divides the coal mine’s emissions by the number of active ventilation shafts for a specific coal mine, sees a spread of 0.03 - 25.9 [kt CH$_4$/year] between individual coal mine ventilation shafts. A large variation between shafts was observed by Varon et al. (2020), who used satellite measurements to quantify methane emissions from three different ventilation shafts in the United States, Australia, and China, and found emissions of 20.3 ± 9.2 to 51.2 ± 20.4 [kt CH$_4$/year] between the shafts. In the study of Luther et al. (2019), variations between different ventilation shafts, as well as variable temporal emissions for a single shaft, were observed. Emission rates of 6 ± 1 to 10 ± 1 [kt CH$_4$/year] were found for a single shaft a few hours apart, and another shaft was quantified to be 17 ± 3 [kt CH$_4$/year]. According to Saunois et al. (2016a), the emitted methane from these mining facilities is strongly influenced by which type of mining takes place, where underground mining typically emits up to 10 times more than surface mining. The emitted methane is also very region specific and can vary strongly between regions. The quality of the extracted coal also influences the emitted methane, with brown coal emitting more than hard coal.

As part of the 2017 CoMet mission, we used the UAV-based active AirCore system to sample methane concentrations from a coal mine ventilation shaft, and developed a methodology to quantify the CH$_4$ emissions. We flew the drone downwind the shaft and sampled air in a plane (or ‘curtain’) perpendicular to the wind direction. The sampled CH$_4$ was used together with a mass balance and inverse Gaussian approach to get an estimate of the source strength. Of the 15 flights performed during the campaign, 8 flights spanning two separate days passed the list of criteria set to ensure good sampling condition, and were further used for CH$_4$ quantification. The daily estimates on day 1 were 4.9 ± 1.4 [kt CH$_4$/year] for the inverse Gaussian approach, and 3.7 ± 2.3 [kt CH$_4$/year] for the mass balance approach. For day 2, the daily emissions were 4.3 ± 3.1 [kt CH$_4$/year] for the inverse Gaussian approach and 6.9 ± 5.0 [kt CH$_4$/year] for the mass balance approach. We saw a strong variability in the estimated emissions, not only on a day-to-day basis, but on flight-by-flight basis. We believe this is caused by actual variations in the ventilation shaft’s emission rate, and this is also reflected in the hourly inventory emission data from the ventilation shaft presented in chapter 4. Luther et al. (2019) had a similar finding, where they quantified emissions from a single shaft at two different times during a day, and found that at the in the morning the emission rate was 6 ± 1 [kt CH$_4$/year], while a few hours later the emission rate had changed to 10 ± 1 [kt CH$_4$/year]. Krings et al. (2013) quantified the emissions from two coal mine ventilation shafts in western Germany.
using airborne remote sensing data, and found emission rates of 43.1 ± 1.0 to 31.8 ± 5.2 [kt CH₄/year] for the first shaft, where the higher emission stemmed from a flight track closer to the ventilation shaft. The quantified emission for the other shaft was 12.4 ± 0.4 [kt CH₄/year].

A similar inverse Gaussian approach to the one used in chapter 3 and 4 of this thesis was also used by Shah et al. (2019, 2020) to do near-field flux quantification of methane from a point source downwind a controlled methane release using a tethered UAV, as well as a UAV carrying a near-infrared laser spectroscopy instrument. Shah et al. (2020) quantified methane fluxes with uncertainty bounds between 17 ± 10 (1σ) [%] to 227 ± 98 (1σ) [%]. The dominant part of the uncertainty stemmed from the wind. Allen et al. (2019) used a mass balance approach with a fixed-wing drone, complimented by a rotary UAV, to quantify landfill emissions, and found that the uncertainty was dominated by variability in the background mole-fraction and wind measurement variability. Nathan et al. (2015) used both a mass balance approach and an inverse Gaussian approach to quantify methane emissions from a compressor station in the Barnett Shale, Texas, by flying a fixed-wing drone downwind the compressor station. Emission rates over a one-week period and 22 flights were 14 ± 8 [g CH₄ s⁻¹], with significant temporal variations. The uncertainty stemmed primarily from the kriging variance (56 [%]) and wind speed (21 [%]), with other contributions being CH₄ mixing (10 [%]), grid resolution (4 [%]) and interpolation scheme (5 [%]). Common for all these approaches is the large contribution to the uncertainty caused by the wind, which is also reflected in our study. Stable wind conditions is vital for these type of quantification strategies, and is for the active AirCore technique one of the biggest obstacles.

The UAV-based active AirCore system is not an instantaneous sampling method, and samples the emitted methane plume over the course of 10 - 12 minutes. Given unstable wind conditions, this gives time for the plume to meander and possible be picked up several times during the same flight, or it can be that the plume meanders to the point where it completely avoids the drone flight track all together. Examples of this can be seen in figures (3.6) and (3.6) a, d, g, and j, where the horizontal dispersion coefficients, σᵧ, are significantly larger than the vertical dispersion coefficient, σᵧ (see table (3.6) for values). The observed plume is often seen as having a wide horizontal distribution while the vertical distribution is small and narrow. The observed stretched-out plume is not only caused by the wind, but also in part due to the non-instantaneous sampling of the active AirCore system, which is affected by molecular diffusion and analyzer cavity effects. From the start of the sampling to the end of the active AirCore analysis, 20 - 30 minutes will have passed. This leads to a smoothed analyzed mole-fraction signal, as discussed in chapter 2 of this thesis. This smoothing will thus lead to wider-looking plumes. For the inverse Gaussian approach in chapter 3 and 4, this smoothing has been accounted for by convolving the observed signal with that of the analyzers response function, as explained in Winderlich et al. (2010). It was found that using a moving average of 33 - 34 seconds of the sampled signal would yield the observed signal.

The inverse Gaussian methodology from chapter 3 and 4 assumes a perfect Gaussian shape of the data, which is the case given sufficient averaging time (Seinfeld and Pandis, 1998; Fritz et al., 2005; Salesky and Chamecki, 2012). The required averaging timescales can
however be compensated for by having multiple transects in mobile van measurements (Caulton et al., 2018). The methodology in chapter 3 uses single-pass transects at different altitudes, and does therefore not have sufficient averaging time to warrant the assumption of a perfect Gaussian, which adds significant uncertainty to the quantified estimation. However, the observations from not only one, but multiple transects spread out over several different heights provide additional constraints on the plume’s location, as well as it’s shape. The multiple transects allows the center-height of the plume and the plume’s vertical and horizontal dispersion to be constrained.

Using pseudo data, the two approaches were investigated on performance, and it was found that the inverse Gaussian approach performed better than the mass balance approach. For the inverse Gaussian approach, retrieved source strengths were within 4 [%] of the true source strength with an uncertainty of 8 - 16 [%], compared to the mass balance which retrieved source strengths within 10 [%] of the true source strength with an uncertainty ranging from 26 - 51 [%]. Through the pseudo data experiments, we found that the flight planning was of utmost importance. The pseudo data experiments showed that multiple transects at different altitude levels was key in obtaining a solid quantification, and to provide sufficient constraints on the parameters of the inverse Gaussian function. The vertical spacing between transects, as well as the distance between the center-height of the plume and the middle flight transect, was found to yield the best performance when the vertical spacing and distance was smaller than 2.5 times the vertical distribution of the plume ($\sigma_z$). Such a planning could be achieved by estimating the vertical distribution of the plume prior to the flight, by measuring the turbulence intensity and the standard deviation of the wind direction, as described by Thoma and Squier (2014).

Due to the active AirCore’s sampling method, the air samples were also analyzed for CO$_2$ and CO mole fractions. The final finding of chapter 3 was the correlation of emitted CH$_4$ with CO$_2$, with all flights having an R-squared value larger than 0.69 ($N > 124$), as shown in figure (3.8). The slope ranged from $3.54 \pm 0.25$ [ppm CH$_4$/ppm CO$_2$] to $5.27 \pm 0.07$ [ppm CH$_4$/ppm CO$_2$]. This corresponded to an emitted CO$_2$ flux of $2.9 \pm 0.4$ [kt CO$_2$/year] on day 1, and $2.9 \pm 2.1$ [kt CO$_2$/year] on day 2 for the inverse Gaussian approach. The mass balance approach found the emitted fluxes to be $2.0 \pm 1.1$ [kt CO$_2$/year] and $4.6 \pm 1.1$ [kt CO$_2$/year] for the same respective days. No correlation was found between CO and CH$_4$ or CO$_2$. This work shows that the UAV-based active AirCore can not only benefit methane quantification, but also other GHG sources, due to the flexible sample-taking and drone flexibility. This was also shown by Vinkovic et al. (2021), which used the active AirCore system to not only measure CH$_4$, CO$_2$, and CO, but also N$_2$O and COS. This was done by analyzing the active AirCore samples on a Picarro CRDS as well as with an Aerodyne Quantum Cascade Laser Spectrometer (QCLS). The ability to measure multiple different tracers is one of the big advantages of the active AirCore system. This gives the possibility of doing tracer release experiments while simultaneously measuring other related tracers.
5.3.2 Estimate of regional methane emissions

In chapter 4, we used the methodology developed for the active AirCore system in chapter 3, and found an estimate to the total regional emissions from the USCB area. In May to June 2018, a larger CoMet campaign took place in the USCB, combining aircraft measurements, drone measurements, and mobile van measurements. We performed 59 UAV-based active AirCore flights downwind of 5 different ventilation shafts, and quantified the emitted methane using the methodology developed in chapter 3. For the 5 shafts, the quantified methane emissions ranged from 1.2 - 15.0 [kt CH\textsubscript{4}/year], with a mean average emission of 5.5 ± 2.6 [kt CH\textsubscript{4}/year] for the inverse Gaussian approach. The mass balance quantified methane emissions ranging from 0.3 - 19.3 [kt CH\textsubscript{4}/year], with a mean average of 5.4 ± 3.2 [kt CH\textsubscript{4}/year]. The intra-shaft variability is large, as well as variability within each shaft. The large variability within each shaft is reflected in the shaft’s hourly inventories, and it is therefore clear that comparisons between the quantified UAV-based active AirCore flights with annually averaged inventories will not be possible. A comparison yields a negligible R-squared value of 0.06. A better correlation was found by comparing daily-averaged fluxes using the hourly inventory data, and an R-squared value of 0.23 was found. The best obtainable correlation was found to be shaft-averaged emissions, where we found an R-squared value of 0.86 for the inverse Gaussian quantified flights and 0.68 for the mass balance approach. The conclusion that snapshot-like quantification, like our 10-12-minute long active AirCore flights, can only provide a very rough comparison with annual inventories was also concluded by Luther et al. (2019).

One of the findings from chapter 4 is that although a single flight may not yield a correct estimation, the average of multiple flights will bring the estimate closer to those of the inventories, as can be seen in the statistical distribution in figure (4.9). We also conclude that more than a single flight is required to give a good estimation of a shafts emission rate. From figure (4.9), we also see a consistent bias comparing the quantified results with those of inventory emissions, and if the inventory emissions are to be treated as the truth, we consistently underestimate the methane flux. Estimating the regional USCB CH\textsubscript{4} and CO\textsubscript{2} emissions therefore require us to up-scale the average quantified shaft emission.

The highly correlated shaft-averaged inventory and quantified emissions provided us with a linear curve which we used to up-scale the average shaft emissions from the five quantified shafts to a regional emission of the whole USCB. The USCB has, according to an internal CoMet inventory, 59 active coal mine ventilation shafts. We obtained our regional CH\textsubscript{4} and CO\textsubscript{2} estimate by assuming that every ventilation shaft emitted an average of the five ventilation shafts we had quantified, and multiplied this average with the number of active ventilation shaft. We estimate the regional emissions of the USCB using three approaches; scaling the E-PRTR annual inventory, the quantified shaft-averaged emission rate, and the shaft averaged emission rate that are derived from the hourly emission inventory. The first approach obtained a regional estimate of 332.6 [kt CH\textsubscript{4}/year] using the Inverse Gaussian and 268.2 [kt CH\textsubscript{4}/year] using the mass balance. The second approach obtained regional estimates of 324.5 ± 175.5 [kt CH\textsubscript{4}/year] using the inverse Gaussian approach, and 318.6 ± 188.8 [kt CH\textsubscript{4}/year] using the mass balance approach. The final approach estimated a regional emission rate of 446.9 ± 133.2 [kt CH\textsubscript{4}/year] using the inverse Gaussian, and
318.6 ± 103.4 [kt CH$_4$/year] using the mass balance. These results, especially the last approach using the inverse Gaussian, overlap with the E-PRTR (2017) inventory of 447.9 [kt CH$_4$/year], and also compares well with regional estimates of the USCB conducted by Fiehn et al. (2020). They found the regional estimate on two different days to be 437.6 ± 114.2 [kt CH$_4$/year] and 478.8 ± 95.1 [kt CH$_4$/year], and were estimated using a mass balance approach. Using the same up-scaling approach, the regional emission of CO$_2$ was estimated to be 0.2 - 0.3 [Mt CO$_2$/year] for both the mass balance and inverse Gaussian approach. This is ≈ 1 [%] of the annual E-PRTR (2017) CO$_2$ inventory for the USCB (35.3 [Mt CO$_2$/year]) the estimates from Fiehn et al. (2020) of 38.2 ± 22.7 [Mt CO$_2$/year] and 35.3 ± 11.7 [Mt CO$_2$/year]. Since Fiehn et al. (2020) obtained their regional estimate from aircraft flights around the region, and the E-PRTR (2017) inventory is not even listing coal mining as a potential source of CO$_2$, we conclude that we indeed see that coal mine ventilation shafts are not a major CO$_2$ source in the USCB. It does however indicate that the inventory is missing a CO$_2$ source of ≈ 1 [%].

5.3.3 The global demand of coal

Global demand for coal started to decline in 2014, but was in 2019 still the largest energy source worldwide and was responsible for 36 [%] of the world’s electricity production. The worldwide coal demand dropped in 2019 by 1.8 [%], the primary reasons being low gas prices and a smaller demand on electricity (IEA, 2020a). Europe and the United States have been steadily decreasing their coal production over the last decade. 2019 saw the largest drop in coal-fired power production ever, primarily due to a strong investment in renewable-based energy production and coal-to-natural gas switching. Some countries have also announced coal phase-outs, like Germany, and plan to be coal-free by 2038 (IEA, 2020b). The drop in the United States was mainly due to a large coal-to-natural gas switching and cheap gas prices. India, for the first time in four generations, also saw a decrease in the demand for coal-fired power production. China, the largest coal producer and coal importer in the world, as well as other countries in south-east Asia, saw a significant increase in coal-demand. However, this increase was not large enough to offset the overall decrease in demand worldwide.

The following year saw difficulties for many people and industries alike, and the impact that the Covid-19 pandemic had on coal demand was not spared from the same difficulties. 2020 experienced the largest drop in coal demand since the second world war, and dropped worldwide by 5 [%] from the 2019 levels. The large impact of the Covid-19 pandemic, primarily in the first half on 2020, severely crippled the industrial output worldwide. China, due to a robust economic recovery in 2020, was not as affected as elsewhere (IEA, 2020a). The large drop in coal-demand was caused by an unusual drop in electricity demand around the world, likely triggered by the temporary shutdown of many industrial sites. The use of coal for power generation was affected even further due to low gas prices, leading to the overall large drop in coal demand. According to the International Energy Agency (IEA), the world’s coal demand has been estimated to have dropped by a massive 7 [%] between 2018 and 2020. A two-year decrease this large is completely unprecedented in the IEA records, which started in 1971 (IEA, 2020a).

Following the initial impact of the Covid-19 pandemic in 2020, the IEA’s prediction is that
the industrial output and electricity demand is expected to increase in 2021, an increase which will be led by China, India, and south-east Asia. The higher electricity demand, as well as higher natural gas prices, are expected to slow the trend of decreasing coal demand in Europe and the United states. This may lead to a growth in the trend for Europe and the United States for the first time in nearly a decade (IEA, 2020a).

Towards 2025, the demand for coal is again expected to flatten out, although trends are expected to vary region by region. Following the 2021 increase in coal demand in both Europe and the United states, the coal demand in these regions are again expected to decrease. China remains the largest coal producer, importer, and largest influence on the coal demand, but it is forecast that a plateau in China’s coal demand will be reached towards 2025. China announced in late 2020 that they pledge to achieve carbon neutrality by 2060, which will without a doubt have an impact on the future coal demand. India, as well as other countries in the south-east Asia region are also forecast to increase coal use through 2025. This is due to new coal-fired power plants being built, and expanding industrial production.

Although the demand for coal is linked to the emitted methane, there are many factors that play a role in how much methane is being emitted. The manner of how the coal is extracted makes a large difference, where underground mining emits up to 10 times more methane than surface mining (Saunois et al., 2020). The quality of the extracted coal adds another layer, where brown coal emits more methane than hard coal. The geological underground structure, which is very region specific, and history of the area (basin uplift) also influences the amount of emitted methane (Saunois et al., 2020). Therefore, a declining demand for coal does not necessarily mean an equal reduction in emitted methane. On top of the aforementioned reasons, the percentage of on-site methane recycling would also have an impact on the total emitted methane. The large volumetric flowrate and methane concentrations makes it challenging to capture the methane and to utilize it cost effectively, but projects are currently being developed and tested to make this more viable (EPA, 2019). If large quantities of the ventilated methane can be captured and re-purposed at coal mines around the world, this would have a strong impact on the overall emitted methane.

In conclusion, despite an overall trend of decreasing coal production and coal demand in the US and Europe, methane emissions from coal will still be a major player in years to come, in particular in Asia. With expected increase in demand and production of coal in China and Southeast Asia, and since emitted methane from coal production remains one of the more uncertain parts in the global methane budget, it is vitally important to develop and maintain good equipment and methodologies to accurately monitor the impact coal-related methane emissions will have on the climate. The UAV active AirCore system presented in chapter 2 is well-suited for this purpose, by utilizing the methodology presented in chapter 3 to quantify and help monitor individual coal mine ventilation shaft emission rates.
5.4 Perspectives and recommendations

Drone-based atmospheric measurements of GHGs is a relatively new way to obtain information about the climate. This thesis focuses on the active AirCore sampling tool to accurately measure the atmospheric mole-fractions of CO₂, CH₄, and CO, and was used primarily to quantify CH₄ emissions from coal mining shafts. The methodology developed in this thesis focuses on point source emissions, but the UAV-based active AirCore system is not limited to sampling GHGs from only these source-types, as shown in chapter 2. Other sources of particular interest would be the quantification of CH₄ emissions from coal mining shafts, which is currently being studied by Vinkovic et al. (2021). Livestock methane emissions from enteric fermentation and manure are major anthropogenic emitters, and their emissions often lack independent verification from atmospheric measurements. Saunois et al. (2020) states that for the period 2008 to 2017, enteric fermentation and manure management contributed about one-third of the global anthropogenic CH₄ emissions. Landfills are also of interest, as it is a major anthropogenic methane source which, according to Saunois et al. (2020), contributed to about 12 [%] of the global anthropogenic emissions between 2008 and 2017. Landfills has in recent years been studied by combining aircraft measurements with a mass balance approach (Peischl et al., 2013; Cambaliza et al., 2017). Aircraft have an advantage over UAVs when studying landfills when measurements are performed further downwind of landfill, due to the emitted plume being more uniform and less changeable. This is particularly useful for modeling of the plume, since measured concentrations can easier be assumed to be constant, as opposed to when closer to the landfill where local wind turbulence and variations in the plume emissions will have a larger impact. The drawback is however that the further away the measurements are made, the risk of introducing possible errors, such as reduced plume concentrations and an increased risk of concentrations being influenced by other nearby sources, increases (Mønster et al., 2019). This is the advantage UAVs have over aircraft when it comes to studying landfill emissions. An example of a UAV landfill study is that of Allen et al. (2019). They used a fixed-wing drone in combination with a tethered rotary drone to a Los Gatos Research Ultra-portable GHG Analyzer (LGR-UGGA) to determine the methane flux from two landfill sites in England, using proxy measurements of CO₂ mole-fractions and wind data. The UAV-based active AirCore could here provide an additional technique to quantify landfill CH₄ emissions.

One of the difficulties with the point source quantification methodology discussed in this thesis is the complexity of the wind profiles. Dinger et al. (2018) showed observations of turbulent dispersion of SO₂ puffs, and found that under cloudy conditions, the direction and the height of the emitted puff can drastically change within a short time span. The flights from chapter 3 and 4 of this thesis were made under clear-sky conditions and with relatively steady wind conditions, however, strongly variable wind conditions were also observed throughout the campaigns. Wind direction and wind speed measurements were taken throughout the flights, using radiosonde balloon launches and a ground-based meteo-station, and an average was used for the quantification process. More wind measurements during the flight, such as using the drone’s own internal stabilization system, could add significant information to the behaviour of the wind at specific points throughout the flights. Horizontal wind estimates were made by Simma et al. (2020), who were
able to achieve RMSE values of the 10 [s] moving average filtered data of between 0.26 - 0.29 [m/s] for wind speed, and 4.1 - 4.9 [°] for wind direction. They used a small IRIS+ quad-copter from 3D robotics, and used on-board accelerometers, gyroscopes, magnetometer, and barometer, in combination with equations developed by Allibert et al. (2014) to obtain information about the wind. If this could be included in the active AirCore flights, wind measurements could be made every 15 - 25 [m] (every 10 [s] with an average cruise speed of 1.5 - 2.5 [m/s]). Combining the GHG measurements from the active AirCore and a more complex mapping of the wind throughout the flight could then be used to compliment point source modeling. Using multiple UAV-based active AirCore systems could also help in obtaining more information about the turbulent conditions. Having two (or more) drones following the same trajectory with a specific time interval between them (15 - 20 seconds) could observe how the plume changes on short time-scales. This would, however, only be possible with pre-programmed flight trajectories and build-in fail-safes to ensure the drones do not interfere with each other.

Another beneficial addition to the active AirCore system would be a small light-weight in-situ CH$_4$ sensor that would compliment the active AirCore sampling. Having a small in-situ sensor on-board the UAV would greatly help in finding the emitted CH$_4$ plume, which could then change the flight trajectory to optimally sample the plume with the active AirCore. The current active AirCore system relies on good up-front planning, and since it gathers a sample of air along the trajectory of the drone for later analysis, there is no way of knowing whether the emitted plume has been sampled or not until after the completion of the flight and analysis of the AirCore. The in-situ sensor would only be needed to give an indication of elevated CH$_4$ mole-fractions, while the sampling and quantification would be done using the active AirCore. Studies using small sensors such as the Figaro TGS 2611-E0 (van den Bossche et al., 2017), Figaro TGS 2600 (Collier-Oxandale et al., 2018; Riddick et al., 2020), and Figaro TGS 2602 (Collier-Oxandale et al., 2019) have been done over long-term periods to determine long-term trends, and would not be suitable for any form of quantification as an inclusion to the active AirCore system. It could however be useful in finding and indicating the plume’s location. Similarly to the small methane sensor, using a remote sensing camera such as in Dinger et al. (2018) could prove useful in identifying where the plume is located, and plan flights accordingly.

Optical gas imaging technology tends to focus on the oil and gas industry and leak detection, like in Ravikumar et al. (2017) and Lyman et al. (2019). A possible reason for this is the requirement from the U.S. Environmental Protection Agency (EPA) that requires all operators to use optical gas imaging as part of their repair- and leak detection program. Lyman et al. (2019) state that OGI cameras perform poorly during winter time, and can benefit from being complimented by other techniques that can help identifying leakages which will aid wintertime repair efforts. The UAV-based active AirCore system could be used to fill this role, with its unique sampling methodology, flexibility, and ability to access difficult-to-reach locations. On the other hand, OGI cameras could also prove useful in combination with the active AirCore in identifying the CH$_4$ plume emitted form a coal mine ventilation shaft, and used together with the model from Ravikumar et al. (2017) to simulate the emitted plume, which can be compared and verified using the active AirCore system.
Another future step would be to combine the UAV-based active AirCore measurements with satellite observations, whether for verification or quantification purposes. Pandey et al. (2019) estimated the CH$_4$ emissions from a natural gas well blowout using satellite observations. Of the 20 days the well was emitting large quantities of CH$_4$, only orbits from 1 day could be used for the emission quantification, which estimated the emissions to be 120 ± 32 [t CH$_4$/h]. Cusworth et al. (2021) also used multi-satellite imaging to quantify the CH$_4$ emissions from a gas well blowout. The UAV-based active AirCore system could in these cases be used to verify the emissions from the wells by flying downwind the wells during the time of the blowout. Cloud coverage is a big limitation for satellite quantification, like in Pandey et al. (2019) where 1 out of the 20 days the blowout lasted had favorable conditions for CH$_4$ quantification, and is something that does not limit quantification using the active AirCore system. Varon et al. (2020) quantified the emissions from several coal mine ventilation shafts using satellite observations over the span of 2 years. Due to the flexibility of the drone, difficult to reach areas can be accessed, and with the ease of deployment and operations, the active AirCore system could be a complimentary technique to verify the emission estimates made via satellite observations. Identification of large CH$_4$ emitting sources via satellites, or via airborne observations such as in Frankenberg et al. (2016) and Borchardt et al. (2021), can also work well when complimented by the UAV-based active AirCore system. As large CH$_4$ plumes are detected, the UAV-based active AirCore system can be deployed to quantify the emissions of those plumes. This opens up a new avenue for future research in both emission verification and emission determination.