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Hydrogen Sulfide Emission Properties from Two Large Landfills in New York State

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Abstract: Landfills are a source of malodors, greenhouse gases, harmful pollutants, pests, noise, and litter. To reduce their impact on neighboring communities, landfill facilities and the policies they follow must reduce emissions of trace gases such as hydrogen sulfide (H_2S) and methane (CH_4). However, a comprehensive understanding of the spatial variability of both pollutants at landfills should first be established to obtain a clear picture of emissions at landfills. This study measured the mixing ratios of H_2S and CH_4 at two landfills in New York State (Fresh Kills Landfill and Seneca Meadows Landfill) in November 2021 using laser-based methods deployed in a mobile lab. H_2S emission fluxes were estimated based on a mass balance calculation. The highest mixing ratios of both H_2S and CH_4 were measured at Fresh Kills Landfill, at up to 7 parts per billion (ppb) and ~140 parts per million (ppm), respectively, yet these values resulted in a low $\Delta H_2S/\Delta CH_4$ ratio, at approximately $5.2 \pm 2.6 \times 10^{-5}$ mol mol^{-1} and a H_2S emission flux of 0.02 ± 0.01 mg m^{-2} day^{-1}. The highest $\Delta H_2S/\Delta CH_4$ ratio was observed at the Seneca Meadows Landfill at $8.6 \pm 4.3 \times 10^{-4}$ mol mol^{-1} and yielded a H_2S emission flux estimate of 17.7 ± 12.9 mg m^{-2}-day^{-1}. The variability in mixing ratios and $\Delta H_2S/\Delta CH_4$ ratios measured at the landfills can be attributed to various factors, including facility operations and design, landfill age, meteorology, types of waste, and pH levels, but further multiday measurements are needed at each landfill to improve emission estimates and determine a more accurate and resolute reasoning behind these variations.

Keywords: hydrogen sulfide; methane; odor; landfill; greenhouse gas emissions; leachate

Citation: Catena, A.M.; Zhang, J.; Commane, R.; Murray, L.T.; Schwab, M.J.; Leibensperger, E.M.; Marto, J.; Smith, M.L.; Schwab, J.J. Hydrogen Sulfide Emission Properties from Two Large Landfills in New York State. *Atmosphere* **2022**, *13*, 1251. https://doi.org/10.3390/atmos13081251

Academic Editor: Roberto Bellasio

Received: 1 July 2022
Accepted: 23 July 2022
Published: 6 August 2022

1. Introduction

Waste is an inevitable part of life, and how societies deal with it and its byproducts helps determine quality of life and environmental impact. The deposited waste emits gases of various compositions over time, all of which are collectively identified as landfill gas (LFG). The initial phase under aerobic conditions generates nitrogen (N_2), oxygen (O_2), and some carbon dioxide (CO_2), but as time goes on and oxygen is depleted, conditions become anaerobic, and the landfill gas changes composition to primarily CO_2 and methane (CH_4) [1,2]. Decomposition of organic material under anaerobic conditions due to microbial activity generates reduced species of LFGs—most notably methane and hydrogen sulfide (H_2S), which are the interest of this study [2–4]. LFG is a cause for concern for landfill-neighboring communities and facility operators due to its toxicity, climate impacts, and odor [4–8]. There has been a significant increase in global waste production over the last several decades; the amount of municipal solid waste (MSW) generated each year in the United States increased from 79.9 million metric tons (mt) in 1960 to 265.3 million mt in

2018 [9]. This increase has been burdensome on neighboring communities in that it has led to odor, health, noise, and pest problems [4]. Landfills are also an environmental justice issue since landfills and MSW facilities tend to be disproportionally located in communities of lower economic status and higher populations of people of color [10].

One important LFG component is H_2S—the primary contributor to LFG odor [11]. Other pollutants in LFG include CH_4, CO_2, methyl mercaptan (CH_3SH), dimethyl sulfide (CH_3SCH_3), and ammonia (NH_3) [5,12–15]. H_2S is a colorless, poisonous, and flammable gas that has a pungent odor of rotten eggs [8]. It is produced either when sulfur-containing landfilled material (e.g., gypsum drywall) breaks down via anaerobic decomposition, and/or when sulfate-reducing bacteria (SRB) oxidize organic substrates and use sulfate as the terminal electron acceptor [8,16]. Gypsum drywall is primarily composed of the gypsum mineral, which is made of calcium sulfate and water [8,16]. Since the reduction of sulfate is the main driver behind H_2S generation, and gypsum drywall is primarily disposed of at construction and demolition (C&D) landfills, H_2S emissions are generally much higher at C&D landfills as compared to MSW landfills [5,16].

Odor detection thresholds of ambient H_2S vary from person to person and can range from 0.5 to 300 parts per billion (ppb) [17]. Eye irritation can begin at H_2S mixing ratios (MR) as low as 25 ppb [8], while acute exposure ranging from 100–500 parts per million (ppm) has been reported to cause eye, throat, and lung irritation, asthma, fatigue, loss of appetite, headache, nausea, and olfactory paralysis [4,16,18]. Molar fractions over 500 ppm are often lethal within minutes, but are usually only observed in enclosed spaces, such as sewers, silos, manure pits, etc. [18,19]. Figure 1 depicts the corresponding health impacts from exposure to various levels of H_2S. The exceptionally low odor detection threshold of ~0.5 ppb means that H_2S has been a prevailing nuisance in neighboring communities. In addition to the negative physical health effects mentioned above, malodor from landfills has also been associated with alteration of outdoor activities and negative psychological effects such as stress, anger, anxiousness, exhaustion, and unhappiness [4]. Malodor is damaging and disruptive to the mental and physical wellbeing of neighboring communities.

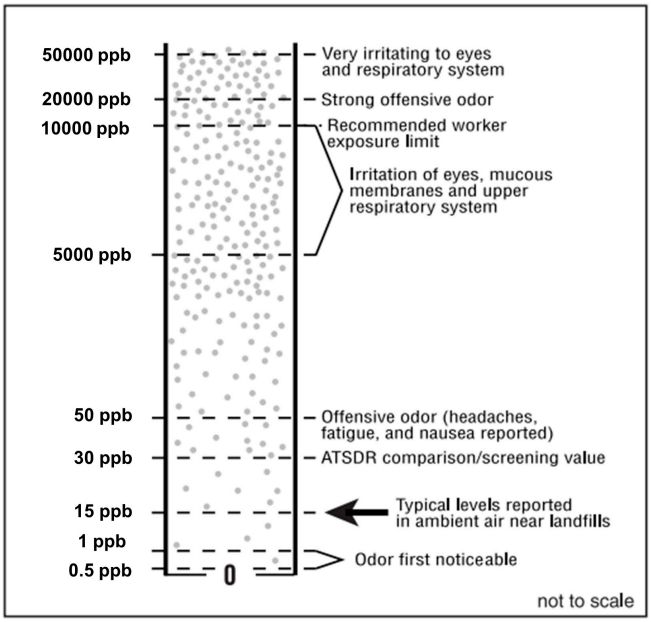


Figure 1. Ambient hydrogen sulfide mixing ratios and corresponding health effects from exposure (source: Centers for Disease Control and Prevention (CDC) Agency for Toxic Substances and Disease Registry (ATSDR) [20]). This material does not constitute its endorsement or recommendation by the U.S. Government, Department of Health and Human Services, or CDC. The material is available on the agency website for no charge.

In addition to H_2S, CH_4 is another leading component in LFG, as it comprises about 50% of LFG [21]. Methane is generated in landfills via the decomposition of organic material by methanogenic bacteria [22]. It has a high global warming potential of 84 times that of carbon dioxide on a 20-year time scale, and its ambient concentration has increased by 150% since preindustrial times [23]. The waste sector accounted for 31% of total methane emissions in New York State (NYS) in 2019, and MSW landfills accounted for 14.5% of total 2020 methane emissions in the United States [2,24]. Under ECL § 75-0107 and 6 NYCRR Part 496, NYS must reduce greenhouse gas (GHG) levels by 40% of 1990 levels by 2030, and 85% by 2050. To satisfy this requirement, there must first be a clear understanding of methane sources and hotspots in NYS before further research and policy can be developed to mitigate GHG levels. Since both H_2S and CH_4 are leading components of LFG, they are often indicative of one another at landfills and, in some situations, can be used as proxies for one another to determine landfill gas as a possible source. Both pollutants are oxidized in the atmosphere but given that H_2S has a lifetime of about 1–42 days while methane has a lifetime of 9–12 years and since sampling is immediately downwind of the source, degradation due to atmospheric oxidation loss can be neglected [25]. Given the obligation and urgency to reduce both odor and GHG emissions, it is important to first assess the spatial variability of both pollutants and determine how they differ between landfills. One approach is to use mobile measurements of both pollutants to quantify the molar fractions around the landfill and between different landfills.

The main objective of this paper is to (1) measure H_2S and CH_4 molar fractions sampled around two different landfills, (2) calculate ratios between H_2S and CH_4 to establish any relationship between the two pollutants, and (3) estimate the H_2S emission flux at both landfills to determine an emission rate and compare it with the results of previous studies. The H_2S flux is estimated using a novel approach by utilization of a simultaneous airborne CH_4 flux estimate together with the ratio of the enhancements of H_2S to CH_4 using mobile measurements. This methodology provides a valuable snapshot of H_2S emission flux estimates at landfills and can be developed further to be used at other facilities or locations. The results from this study will better equip the scientific community to determine the causes of differences between facilities, which operators and policymakers can use to improve operations and regulation. This will have an end goal of improving the quality of life of neighboring communities and reducing GHG emissions.

2. Materials and Methods

2.1. Measurements Period and Methods

Mobile observations of H_2S and CH_4 were collected in conjunction with a larger study aimed at improving estimates of methane emissions and spatial variations in MRs across NYS. The deployment occurred over 14 days in November 2021. Three days were spent at two different landfills in NYS, which are the focus of this study. The first landfill was Fresh Kills Landfill (40°33′54.8″ N 74°12′08.7″ W) on Staten Island, NY, with two days of measurement deployments; the second was Seneca Meadows Landfill (42°55′13.6″ N 76°50′29.6″ W) in Waterloo, NY, with one day of measurements. The site locations are shown in Figure 2. The landfills assessed in this study are categorized as MSW landfills. Samples were taken over the course of 2.5–5 h, starting about an hour before noon at each site. In addition, mobile ground measurements were supplemented with aircraft measurements during the campaign at the Seneca Meadows Landfill, allowing for estimation of a CH_4 emission flux.

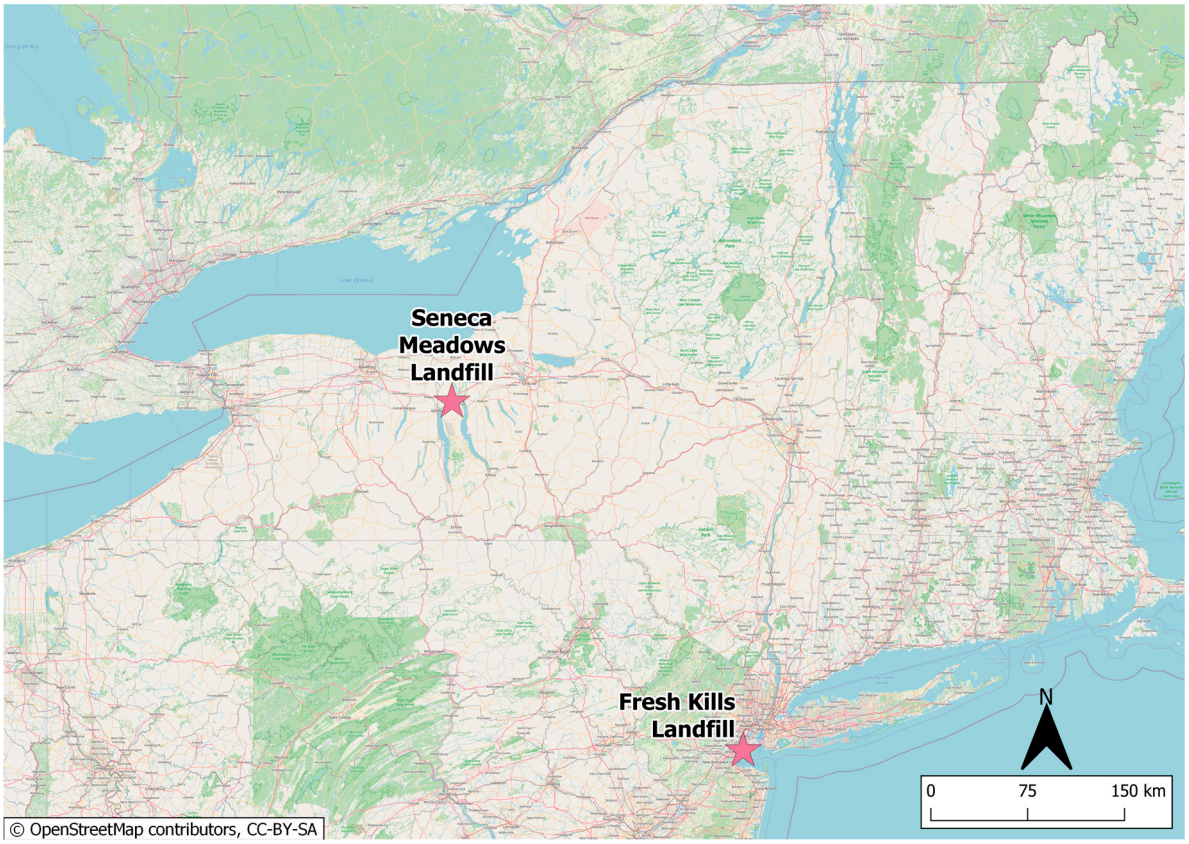


Figure 2. Site locations of Seneca Meadows Landfill and Fresh Kills Landfill using QGIS (https://qgis.org/, accessed on 20 July 2022).

Observations were collected aboard a mobile lab modified from a 2007 Dodge Sprinter van [26,27]. The mobile lab measured H_2S and CH_4 using a Picarro G2204 cavity ring-down spectrometer manufactured in Santa Clara, CA, USA. The Picarro G2204 measures pollutants by passing air samples through an optical cavity and reporting their absorption spectra using a near-infrared laser. As noted on the Picarro G2204 fact sheet, H_2S measurements have a measurement range from 0 to 300 ppm, with a 1-σ precision of 1 ppb. The measurement range for CH_4 spans 0–20 ppm, with a 1-σ precision of 2 ppb [28]. Calibrations for CH_4 and H_2S were performed using certified calibration standards. A 13.25 kWh eight-battery power supply system supplied the analyzers with approximately 6–9 h of power, depending on the air conditioning use in the van.

Global Positioning System (GPS) coordinates were recorded every second using a GlobalSat BU-353-S4 USB GPS manufactured in New Taipei City, Taiwan. The van was also equipped with an Airmar 220WX multiparameter meteorological sensor (manufactured in Milford, NH, USA), which was intended to measure ambient temperature, pressure, and wind speed and direction. Due to instrument malfunction, the wind parameter measurements could not be used for analysis. Instead, supplementary meteorological observations from the NYS Mesonet were used from nearby sites, as described below. All analyzers were connected to an external inlet installed horizontally on the upper edge between the roof and the windshield, just above the driver's seat. Observations were averaged to a 1-minute timescale prior to analysis.

2.2. Measurement Locations (Landfills)

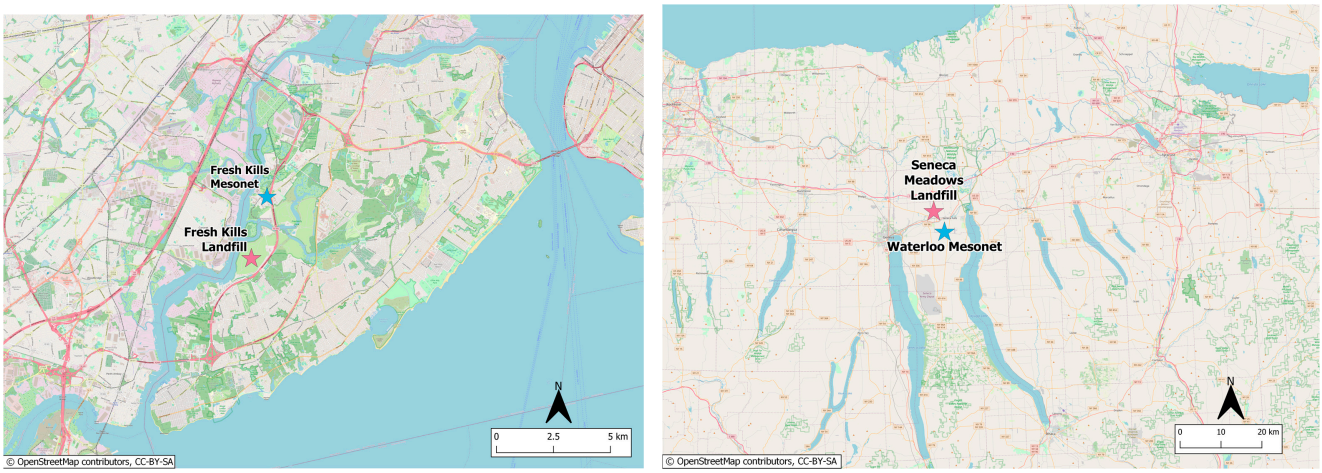
Fresh Kills Landfill is a closed but not fully capped 890-hectare landfill located on the southeastern corner of Staten Island, NY. The landfill operated from the late 1940s until its closure in 2001. While it was operational, it was recognized as the largest landfill in

the world [29]. There are four main cells of the landfill. Three of the cells are fully capped and inactive, and the fourth cell, at the time of this study, was under construction to be fully capped. This fourth cell is Section 1/9, which is the only section of the landfill where measurements were taken. Section 1/9 covers 176 hectares of land along the Arthur Kill Strait [30]. The landfill has six LFG flares, a Selexol LFG recovery plant, a yard-waste composting facility, and two on-site leachate treatment plants. The leachate treatment plants have a combined capacity of 4,773,395 L per day using methods such as sequencing batch reactors, clarifiers, pH adjustment, and sand filters. The Selexol LFG recovery plant has one thermal oxidizer [31]. This was the only landfill where the team accessed the interior roads of the landfill.

Seneca Meadows (Seneca) Landfill is a 363-hectare site located in Waterloo, NY. There is an on-site leachate management system along with gas control equipment, stormwater ponds, a tire recycling facility, and LFG flares (three enclosed, one open, and two thermal oxidizers). The leachate management system consists of three above-ground storage tanks, sulfide controls, and a reverse osmosis system. The pretreated leachate is then sent off-site. The LFG is processed at a renewable gas processing plant across the street on the eastern side of the landfill, which is where the flares are located [32]. Seneca is currently an active and open landfill, and accepted 1,962,506 mt of non-hazardous waste in 2018, making it the largest landfill in NYS [33].

Both landfills are required to submit a non-methane organic compound emissions report to the NYS Department of Environmental Conservation and, if emissions exceed the 37 ton/year limit, must install an LFG collection system (40 CFR 60 Subpart Cf and 6 NYCRR Part 208). They are also required to accept non-hazardous waste only and have a composite liner, leachate collection system, methane capture system, and a daily covering of new waste (known as Subtitle D) [4,34].

The wind speed and direction data used for this study were retrieved from the NYS Mesonet Network sites, including Fresh Kills (SIFKL) and Waterloo (WATE) [35]. The Fresh Kills Mesonet site (Figure 3) is located approximately 2.5 km north of the Fresh Kills Landfill. The Waterloo Mesonet site (Figure 3) is ~5 km southeast of the Seneca Meadows Landfill.



(**a**) (**b**)

Figure 3. (**a**) Locations of Fresh Kills Landfill on Staten Island, NY, in relation to the New York State Fresh Kills Mesonet Station. The Mesonet site is stationed about 2.5 km north of the landfill. (**b**) Site location of Seneca Meadows Landfill in Waterloo, NY. The New York State Waterloo Mesonet site is located about 5 km southeast of the landfill.

2.3. Data Analysis

The raw H_2S data contains variability between negative and positive values, but clear, useful signals of plumes are evident. Monteiro et al. (2022) [36] observed similar H_2S molar fractions measured from an equivalent Picarro G2204. A background H_2S molar

fraction was calculated for each location and used to calculate an enhancement above the background (dH_2S). The background-corrected dH_2S was smoothed using a median filter from the SciPy package in Python. A median filter was chosen to inhibit the influence of outliers in the dataset. Due to instrument noise, some instances of negative values were present after the correction but were within the minimum detection limit of 1 ppb. $\Delta H_2S/\Delta CH_4$ ratios were estimated using the slope from a reduced major axis (RMA) regression between the two pollutants for the entire period of measurements at both sites to account for uncertainty in both observational variables.

The CH_4 emission flux at Seneca Meadows was measured from aircraft measurements by Scientific Aviation (SA). The determination involved observing CH_4 from instruments onboard an aircraft while circling the facility at different altitudes, with a ~1 km radius. Gauss' theorem was applied to calculate the flux divergence through this virtual cylinder around the facility using observed MRs and the horizontal wind field. The aircraft circled the facility 16 times from as low to the ground as possible at 126 m to above the plume at 429 m above ground level. To calculate the flux contribution from the facility, the outward horizontal flux was integrated along all points on a cylindrical path around the facility, as described by Conley et al. (2017) and Schwietzke et al. (2019) [37,38].

Due to flight restrictions, airborne measurements of methane were not obtained for Fresh Kills Landfill. To estimate the flux for this location, the most recent methane emission estimate for Fresh Kills from the EPA's self-reported 2020 Greenhouse Gas Reporting Program (GHGRP) Inventory was used [39]. For comparison, the self-reported methane flux for Seneca Meadows was also used to calculate a second H_2S flux estimate for Seneca Meadows. The H_2S flux estimate at Fresh Kills using the EPA's GHGRP-reported CH_4 flux value must be solely used as a qualitative comparison, as it represents a technically different estimate than the H_2S flux calculated using the airborne CH_4 flux. The EPA's self-reported methane flux estimate was modeled and calculated using a variety of inputs, including landfill size and depth, quantity of landfilled waste, degradable organic carbon content of the landfilled material, a CH_4 generation rate constant, and several additional inputs for facilities capturing LFG, including hours of operation for gas collection, transport to destruction devices, and monitored CH_4 concentrations, flow rates, pressures, temperatures, and moisture contents of the captured LFG [40,41]. The airborne measurements were direct measurements of CH_4 taken on the same day as the ground-level mobile measurements taken by the mobile lab and calculated based on the method outlined by Conley et al. (2017) [37] and as described above. Given the major difference in the CH_4 flux estimates between the EPA's self-reported value and the airborne estimate, the estimated H_2S flux from Fresh Kills determined for this paper only represents a qualitative look at emissions from the landfill. While these emission estimates provide a valuable snapshot of emissions at these landfills, these emission rates are only representative of a short period of measurements (2.5–5 h) at these two sites.

The H_2S emission flux was estimated using the $\Delta H_2S/\Delta CH_4$ ratio and methane flux in the following equation:

$$F(H_2S) = F(CH_4) \cdot \frac{\Delta H_2S}{\Delta CH_4}, \quad (1)$$

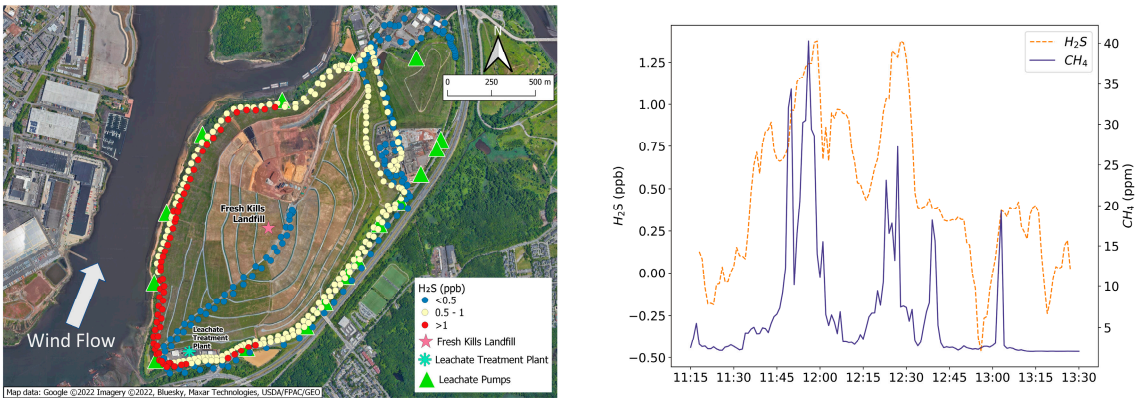
where $F(H_2S)$ is the calculated flux rate of H_2S; $F(CH_4)$ is the methane flux rate, calculated either from the airborne SA measurements or self-reported to the EPA; and $\frac{\Delta H_2S}{\Delta CH_4}$ is the ratio of H_2S to CH_4 enhancements above the background.

3. Results and Discussion

3.1. Landfill-Dependent H_2S Mixing Ratio, $\Delta H_2S/\Delta CH_4$ Ratio, and Emission Flux at Fresh Kills Landfill

The first two deployments of the study took place at Fresh Kills Landfill on 2 November 2021 and 3 November 2021. Measurements on the first day at Fresh Kills Landfill (FK1) observed H_2S mixing ratios up to ~1.3 ppb (Figure 4). Simultaneous enhanced signals of both H_2S and CH_4 were measured on the southern and western sides of the landfill, as

shown in Figure 4. The time-series plot indicates elevated MRs of CH_4 throughout the entire course of measurements at FK1, while the MRs of H_2S peaked sporadically just slightly above the detection limit of 1 ppb. Since there were only a few of these instances when the H_2S MRs were above the detection limit, there was insufficient H_2S to calculate a valid $\Delta H_2S/\Delta CH_4$ ratio for FK1.



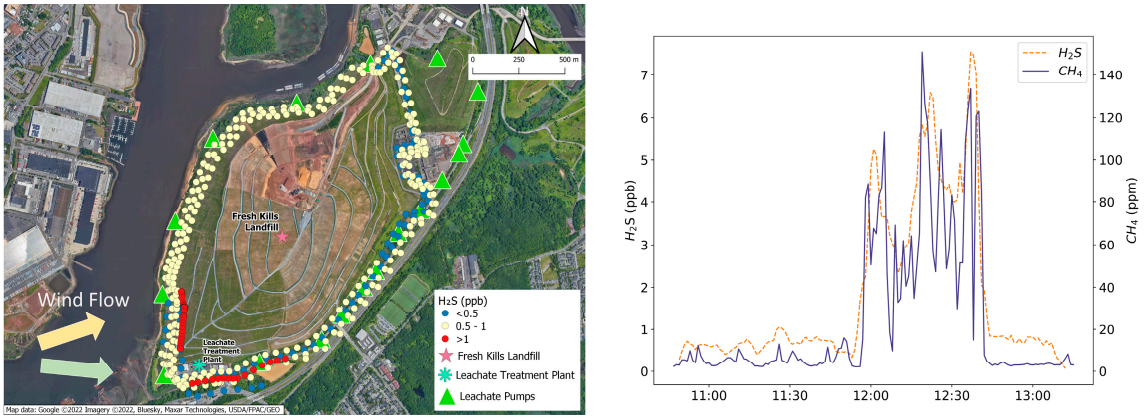
(**a**) (**b**)

Figure 4. Fresh Kills Day 1, 2 November 2021: (**a**) Loops driven around Fresh Kills and the associated H_2S measurement MRs, binned and indicated by color. Each loop was slightly offset to the right and below the prior loop to prevent overplotting. The wind flow vector in the bottom left depicts the average wind direction for the period of measurements. The wind direction was consistent for the duration of the measurements. (**b**) The 1-minute time-series plot of both H_2S (ppb) and CH_4 (ppm) for the entire period of measurements.

The H_2S MRs were much higher during the second day at Fresh Kills (FK2)—up to ~7 ppb (Figure 5). In addition to higher H_2S MRs, exceptionally high measurements of CH_4 were also recorded at about ~140 ppm on the southwestern and western sides of the landfill. The van drove in three loops around the landfill prior to parking on the western side at 11:45 EST for over an hour. Figure 5 shows that the bulk of these higher MRs for both pollutants were measured on this western side. All of the elevated H_2S MRs were observed when the van was parked near the leachate pump, indicating that the observed plumes are quite limited in extent, and may not be well captured from a moving vehicle.

Although the increased MRs for both pollutants from FK2 provided higher confidence in the ratio calculation for that day, there was still considerable variability in the ratio, which had a slope of $5.2 \pm 2.6 \times 10^{-5}$ mol mol^{-1} and R^2 of 0.7318 (Figure 6). The statistical error from the RMA regression at FK2 was about 9%, and the estimated total uncertainty was about 50%, given the high detection limit and noise level of the H_2S signals. High MRs of both pollutants were collocated with the leachate pumps, but there was large variability in CH_4 within the plume and the timing of the H_2S and CH_4 peaks was slightly out of sync. Moreover, given the proximity to the leachate pumps during the recorded high signals, it is uncertain whether this ratio is indicative of the entire landfill. However, these values are comparable to previous measurements from a 1995 study by the United States Environmental Protection Agency, Region 2, New York City, NY, USA (EPA), which yielded ~1×10^{-5} mol mol^{-1} for Section 1/9 of the landfill and ~2×10^{-5} mol mol^{-1} for the entire landfill when measuring emissions from the landfill surface and passive vents [30]. While both of these estimates are smaller than what was measured in this study, they are still comparable to the $5.2 \pm 2.6 \times 10^{-5}$ mol mol^{-1} ratio calculated for FK2. This also brings up the point that the EPA (1995) [30] study was conducted over 25 years ago, and the landfill had been closed for over 20 years at the point of this study (apart from accepting debris from the aftermath of the 11 September 2001 attacks [29]), indicating that landfills will continue to emit both H_2S and CH_4 for a substantial amount of time (+20 years) after

closure. Previous studies by Kim et al. (2006) and Parker et al. (2002) measured and compared mixing ratios between old and new landfills and determined that older landfills emit less H_2S as compared to new landfills, due to depletion of SRB within the landfill over time [8,14,42]. However, Parker et al. (2002) [42] determined that the older landfill (~17 years old) surveyed in their study emitted high amounts of oxygenated organics, suggesting that microbial activity does not always depend on the age of the landfill. In addition, the Fresh Kills Landfill is significantly larger in size than the older landfill studied in Kim et al. (2006) [14], indicating that due to its large size, landfilled material at Fresh Kills may take longer to break down, resulting in continued emissions of H_2S and CH_4.



(**a**) (**b**)

Figure 5. Fresh Kills Day 2, 3 November 2021: (**a**) Aerial image of the loops driven around Fresh Kills and the associated H_2S measurement MRs, binned and indicated by color. Each loop was slightly offset to the right and below the prior loop to prevent overplotting. The wind flow vectors in the bottom left depict the average wind direction for the first (upper yellow arrow) and the second halves (lower green arrow) of the measurements. (**b**) The 1-minute time-series plot of H_2S (ppb) and CH_4 (ppm) for the measurement period at Fresh Kills.

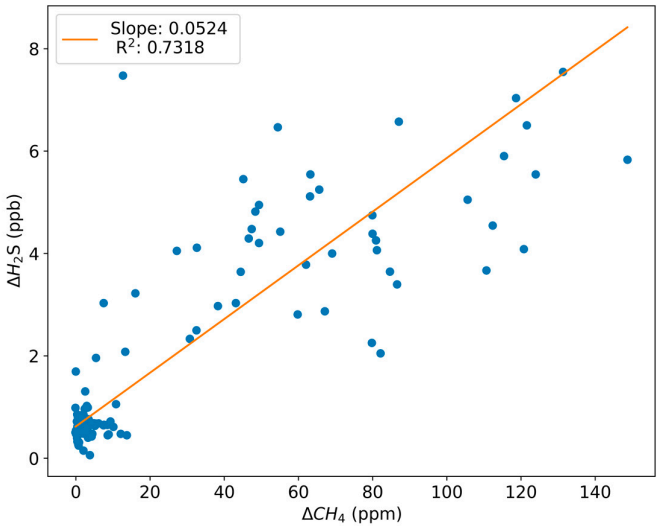


Figure 6. One-minute ΔH_2S to ΔCH_4 MRs from Fresh Kills Day 2 on 3 November 2021: The slope of 0.0524 is used as the ratio between the enhancements of H_2S and CH_4, resulting in a ratio of $5.2 \pm 2.6 \times 10^{-5}$ mol mol^{-1}. Although the ratio is low, the R^2 value of 0.7318 (n = 144) indicates that the pollutants are strongly correlated.

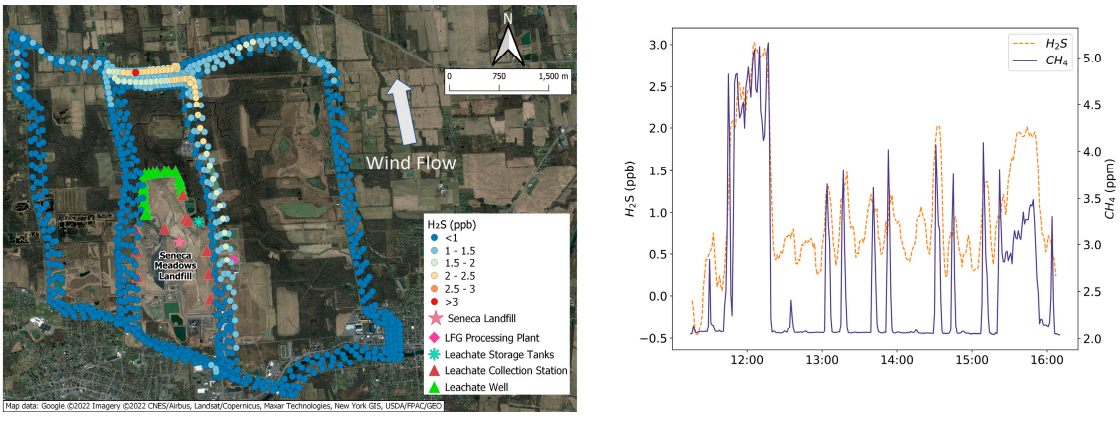
During the first day at Fresh Kills, the leachate treatment plant located on the southern tip of the landfill was hypothesized to be a major source of emissions (see Figures 4 and 5).

Leachate is the resultant liquid byproduct of waste, which is known to be highly contaminated by various pollutants and heavy metals—including hydrogen sulfide and methane—and is a major source of both emissions [8,43,44]. Since the measured winds were south–southwesterly, the van was located upwind of the plant on the southern side, indicating that the leachate treatment plant was not likely to have been the source of emissions. The higher MRs measured on the southern and western sides coupled with the southwesterly winds shown in Figure 4 suggest that the plumes were emitted from the leachate exhaust pumps. These pumps are connected to the leachate collection and treatment system and are located about every 410–480 m around the landfill's perimeter. As was the case with the previous day, the higher MRs seen on the second day at Fresh Kills on the western side of the landfill were most likely also emitted from the leachate exhaust pumps, given the westerly winds that day, which can be seen in the aerial image from Figure 5.

The H_2S emission flux was estimated to be 0.02 ± 0.01 mg m^{-2} day^{-1} using the 2020 EPA-reported GHGRP CH_4 emission flux for Fresh Kills Landfill of 351.3 mg m^{-2} day^{-1} over the entire landfill area of 8.9×10^6 m^2 [39]. This estimate acts as a more qualitative look at the H_2S emission flux due to different sections of the landfill involved in the calculated EPA GHGRP CH_4 emission flux value vs. the measured $\Delta H_2S/\Delta CH_4$ ratio from this study. The ratio calculated from this study involves measurements from just Section 1/9 of the landfill, while the EPA GHGRP CH_4 emission estimate involves all sections from the landfill. Consequently, this gives a relative magnitude of H_2S emissions at Fresh Kills Landfill.

3.2. Landfill-Dependent H_2S Concentration, $\Delta H_2S/\Delta CH_4$ Ratio, and Emission Flux at Seneca Meadows Landfill

Observations from Seneca also indicated a moderately strong correlation between ΔH_2S and ΔCH_4. Although Figure 7 shows that both MRs were observed to be lower than the second day at Fresh Kills (H_2S ~ 3 ppb, CH_4 ~ 5 ppm), the slope of the linear regression was much higher, at 0.8646, with a statistical error of about 6.5%. The simultaneous plumes of both pollutants throughout the measurement period can be seen in the time-series plot in Figure 7b. Figure 8 shows the high correlation of the pollutants in concentration above background. An estimated $\Delta H_2S/\Delta CH_4$ ratio of $8.6 \pm 4.3 \times 10^{-4}$ mol mol^{-1} was determined for Seneca Meadows Landfill, given the total uncertainty of about 50%.



(**a**) (**b**)

Figure 7. Seneca plots: (**a**) The outer and inner loops driven around Seneca Meadows Landfill, and the H_2S measurement MRs associated with the south–southeasterly wind vector in the top right, which was sustained throughout the period of measurements. Each loop was slightly offset to the right and below the prior loop to prevent overplotting. (**b**) The 1-minute time-series plot of H_2S (ppb) and CH_4 (ppm) for the entire period of measurements.

The locations of the higher molar fractions recorded at Seneca also indicate a different source than previously thought. Given the south–southeasterly winds, the enhanced mixing

ratios on the eastern side of the landfill could be attributed to emissions from the LFG processing plant across the street, rather than the landfill itself.

Since there were no flight restrictions at Seneca Meadows Landfill, an H_2S emission flux rate was determined using the airborne measurements of CH_4 for this location only. From Equation (1), the H_2S emission flux at Seneca was calculated to be 17.7 ± 12.9 mg m^{-2} day^{-1}, using the airborne CH_4 emission flux estimate of 20,463 ± 4673 mg m^{-2} day^{-1} over the landfill area of 3.634 × 10^6 m^2. Due to both the high ratio and correlation between H_2S and CH_4, there is high confidence in this H_2S emission flux estimate.

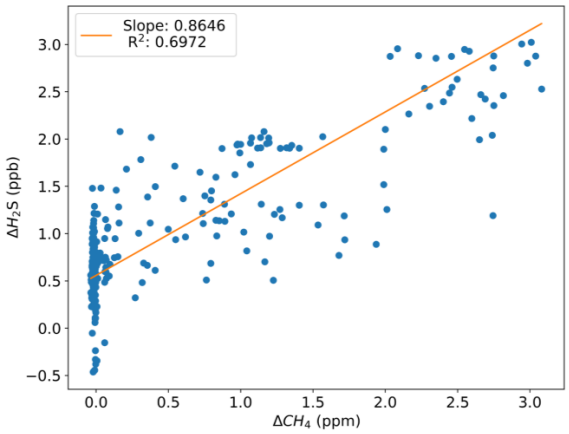


Figure 8. One-minute ΔH_2S to ΔCH_4 MRs from Seneca on 17 November 2021. The slope of 0.8646 and R^2 value of 0.6972 (n = 292) together show that the ratio between the two pollutants was significant at Seneca.

For comparisons to the estimated H_2S flux at FK2, the 2020 Seneca Meadows EPA GHGRP self-reported CH_4 emission flux of 5976.8 mg m^{-2} day^{-1} [39] was also used to determine the H_2S flux at Seneca, yielding a much lower value of 5.1 mg m^{-2} day^{-1}. The CH_4 emission flux measured by the aircraft was 3.4 times greater than the EPA-estimated value. This could suggest that the EPA's self-reported estimate may be undercounting the total methane emissions at Seneca, or that the airborne estimates are overcounting. Previous studies support the former suggestion [45–49].

3.3. Comparisons with Previous Work at Landfills

The Seneca H_2S emission flux estimate using the airborne measurements aligns more with the EPA (1995) (Section 1/9 Cell estimate) [30] and Colledge et al. (2008) [50] studies, as shown in Table 1. The FK2 H_2S emission flux estimate of 0.02 ± 0.01 mg m^{-2}-day^{-1} was much lower than both Seneca estimates (5.1 ± 2.6 mg m^{-2}-day^{-1} using the EPA GHGRP CH_4 flux; and 17.7 ± 12.9 mg m^{-2}-day^{-1} using the airborne CH_4 flux), which was mostly due to the low ratio at FK2 and the smaller reported CH_4 flux estimate as compared to Seneca. While the FK2 H_2S flux estimate is lower than that from the previous work listed in Table 1, it is relatively comparable to the findings of Eun et al. (2007) [51] and Xu et al. (2014) [5], and most similar to the lower end of the EPA (1995) [30] range for the entire landfill.

Table 1. H_2S emission flux estimates from this study compared to previous work.

Study	Type of Landfill/Location	H_2S Emission Flux Estimate mg m^{-2} Day^{-1}
This study	MSW/Fresh Kills Landfill, NY, USA	0.02 ± 0.01 *
This study	MSW/Seneca Meadows Landfill, NY, USA	17.7 ± 12.9 ** 5.1 ± 2.6 *
EPA (1995) [30]	MSW/Fresh Kills Landfill, NY, USA	Entire landfill: 0.08–10497.6 Section 1/9 cell: 0.19–10.94
Eun et al. (2007) [51]	C&D/multiple landfills in Florida, USA	0.192–1.76
Xu et al. (2014) [5]	C&D/Orlando, Florida, USA	0.40–1.07
Colledge et al. (2008) [50]	C&D/Warren Recycling Landfill in Ohio, USA	19.01–210.82

* Using the EPA's GHGRP self-reported CH_4 flux emission rate. ** Using the simultaneous airborne CH_4 flux measurements.

There are a few discrepancies between the estimates listed in Table 1 and the fluxes estimated in this paper. The EPA (1995) [30], Eun et al. (2007) [51], and Xu et al. (2014) [5] studies calculated the H_2S flux using a flux chamber method. In contrast, the flux estimated for this paper was calculated using the estimated CH_4 emission flux, as described in Section 2.3 [5,30,37,50,51]. Colledge et al. (2008) [50] estimated the emission flux using AERMOD and a simple box model, while Eun et al. (2007) [51] used an inverse distance-weighted method to average the H_2S flux. In addition to different measurement methods, these studies also involved longer periods of measurements. The EPA (1995) [30] sampled hundreds of gas chambers for the entire landfill over a three-week period, the measurements of Xu et al. (2014) [5] were extended over 10 months, and Eun et al. (2007) [51] collected ~20 samples over 5 days for each site of sampling. The emission estimates calculated for this study only involved one day of sampling over 2.5–5 h. However, the estimates from this study are valuable because they provide direct measurements of the H_2S emission fluxes from each landfill, as compared to model, bottom-up, or top-down methods, which can be biased by invalidated input parameters and inaccurate assumptions [52].

Comparisons of measured H_2S mixing ratios from previous studies can be seen in Table 2 [4,8,50,53–55]. Past works presented in the table include studies of both MSW and C&D landfills.

Table 2. H_2S emission flux estimates from this study compared to previous works.

Study	Type of Landfill/Location	Sampling Month (s)	Measurement Method	Measured H_2S Mixing Ratios
This Study	MSW/NY, USA	November	Continuous mobile measurements	1.3–7 ppb
Heaney et al., 2011 [4]	Regional MSW/NC, USA	January–February, September–November	Continuous monitoring at fence line	0–14.86 ppb
EPA (1995) [30]	MSW/Fresh Kills Landfill, NY, USA	June and July	Passive vents, flux chamber measurements, and gas collection system measurements at Section 1/9 cell of the landfill	Entire landfill 0.11–220 ppm Section 1/9 5.3–35 ppm
Parker et al., 2002 [42]	MSW/co-disposal site, United Kingdom	September and March	LFG sampling from gas wells	1.2–5.4 ppm
Shon et al., 2005 [56]	MSW/Daegu landfill, S. Korea	January	Ambient air sampling at the entrance of landfill, along the border, and center	0.026–27.01 ppb
Fang et al., 2012 [53]	MSW/Shanghai, China	May	Ambient air sampling	2.9–109 ppb

Table 2. *Cont.*

Study	Type of Landfill/Location	Sampling Month (s)	Measurement Method	Measured H_2S Mixing Ratios
Vasarevicius, 2011 [57]	MSW/Jerubaiciai landfill, Plunge district, Lithuania	August, November, February, April	Ambient air sampling from equidistant points over the landfill	(Highest amounts) Aug—8.6 ppm Nov—8.1 ppm Feb—6.0 ppm Apr—7.3 ppm
Kim, 2006 [14]	MSW/Dong Hae, S. Korea	May, July, November, and December	LFG venting pipe	(Measured area <5 years old) May—2.6–124.4 ppm July—6.8–523.8 ppm Nov—0.44–281.0 ppm Dec—70.3–181.3 ppm (Measured area 5–23 years old) May—0.0002–0.002 ppm July—0.00055 ppm Nov—0.0013–0.005 ppm
de la Rosa et al., 2006 [58]	MSW/Mexico City, MX	September–November	Sampling from drilled holes in the cover and vent	~18–150 ppm
Font et al., 2010 [59]	Valencianne Community, Spain	July and November	July—3.6–16.3 ppb Nov—2.5–8.8 ppb	Ambient air sampling
Song et al., 2007 [54]	MSW/S. Korea	May, July, August, October, and December	Ambient air sampling	0.046–5.396 ppb
Lee et al., 2006 [55]	10 C&D Landfills/ Florida, USA	NA	Ambient air sampling	0.003–50 ppm
Colledge et al. (2008) [50]	C&D/Warren Recycling Landfill in Ohio, USA	October–September, June–August	Continuous ambient air sampling	0–178 ppb

The highest MRs observed in this study (FK1 ~1.3 ppb; FK2 ~7 ppb; Seneca ~3 ppb) were comparable to low-end MRs measured in previous landfill studies (Font et al., 2010, Song et al., 2007, and Heaney et al., 2011) [4,54,59], as shown in Table 2. Since the H_2S MRs measured from this study were above the odor threshold limit of ~0.5 ppb, this poses more of an odor and nuisance issue than a toxicity concern.

The $\Delta H_2S/\Delta CH_4$ ratios estimated for both landfills can be seen in Table 3. The results were highly variable between the landfills, which could be due to several factors, including site design, landfill age, meteorology, type of waste accepted, pH levels, etc.

Table 3. One-minute $\Delta H_2S/\Delta CH_4$ ratios and R^2 values for each landfill.

	Ratio (mol mol^{-1})	R^2
Fresh Kills Day 2	$5.2 \pm 2.6 \times 10^{-5}$	~0.7
Seneca	$8.6 \pm 4.3 \times 10^{-4}$	~0.7

Site design contributes to the variation in MRs and emission ratios. As mentioned above, Fresh Kills has leachate exhaust pumps stationed around the landfill's perimeter, which are the most plausible source of the observed emissions on both days. Although the highest MRs were seen north of the leachate wells at Seneca Meadows Landfill, it cannot be confirmed that these emissions were coming from the leachate wells, since the van did not have access to the interior of the landfill. The landfills have different leachate management systems, which could help explain the MR differences. The age of the landfill also impacts the H_2S emissions. Decomposition of waste over time changes the composition and strength of LFG emissions [8,14,60]. Previous works comparing H_2S emissions from old and new landfills have shown that older landfills emit lower amounts of H_2S as compared to newer landfills [8,14,42]. This could be due to decreased SRB over time due to reductive activity [8].

Meteorology also plays a role in H_2S emissions. Past studies have shown that warmer temperatures result in higher emissions of total H_2S [8,14,57,61]. Increased moisture from a recent rainstorm or elevated humidity greatly enhances sulfate-reducing bacterial activity, increasing H_2S formation [8,44]. Pressure differences also play a role, in that lower pressure results in higher emissions, while higher pressure results in lower emissions of H_2S [62]. Further study at each landfill could better address the influence of meteorology.

H_2S formation heavily depends on the type of waste in the landfill. As noted above, published work indicates that a major source of H_2S in landfills is the breakdown of gypsum drywall at C&D facilities [8]. However, in addition to gypsum drywall, sulfur-containing food waste such as dairy and meat products, along with paper waste, also results in H_2S formation [8]. Since the amount of these waste materials differs between landfills, the H_2S MR also varies, as seen during this study.

Another important factor in H_2S production is the pH level. H_2S forms most readily at a pH between 5 and 6 [8,63]. An increase in pH from 6 to 9 results in an almost 99% reduction in H_2S [44]. It is possible that the landfills have different acidity levels due to varying types of waste—a property that could be determined in future work.

A major limitation for the determination of H_2S emissions was the significant uncertainty in the measurements. The measured MRs at FK1 were slightly above the detection limit of 1 ppb, limiting the ability to determine a conclusive relationship between the H_2S and CH_4 pollutants for this day. However, considerably higher measured MRs were observed on the second day at Fresh Kills compared to the first day, giving greater confidence in the measurements taken on that day. This leaves an important question for further research: How much does the H_2S concentration change from day to day? Multiday measurements at each landfill would greatly enhance understanding of day-to-day variability.

4. Conclusions

Mobile measurements of hydrogen sulfide and methane MRs at two different landfills in NYS exhibited high variability in H_2S emissions between the landfills. Two different landfills were analyzed in this study: Fresh Kills Landfill on Staten Island, NY, and Seneca Meadows Landfill in Waterloo, NY. Measurements took place in November 2021, with two days at Fresh Kills and one day at Seneca Meadows. The highest MRs for both H_2S and CH_4 occurred at Fresh Kills on the second day. However, the lowest H_2S MRs were also recorded at Fresh Kills on the first day of measurements. The variability in MRs could be due to several factors, including site design/characteristics, landfill age, meteorology, types of waste accepted, and pH levels. In addition to the variability in MRs, there was also variability in the $\Delta H_2S/\Delta CH_4$ ratios. Seneca Meadows Landfill yielded a higher ratio at $8.6 \pm 4.3 \times 10^{-4}$ mol mol^{-1}, as compared to the $5.2 \pm 2.6 \times 10^{-5}$ mol mol^{-1} measured at FK2. Using the CH_4 emission flux estimate from the supplementary aircraft measurements at Seneca, the H_2S emission flux was estimated to be 17.7 ± 12.9 mg m^{-2}-day^{-1} for this location. Due to flight restrictions, no aircraft measurements were performed at Fresh Kills Landfill; thus, the 2020 self-reported EPA GHGRP CH_4 emission flux was used to estimate the H_2S emission flux of 0.02 ± 0.01 mg m^{-2} day^{-1} at Fresh Kills Landfill. The H_2S flux estimate calculated for Fresh Kills should only be used as a qualitative comparison, due to the different methodology used to estimate the methane flux. For comparison to this value, the self-reported EPA GHGRP CH_4 flux reported for Seneca resulted in a H_2S flux of 5.1 ± 2.6 mg m^{-2}-day^{-1}. To understand the differences in H_2S, multiday measurements need to be taken at each landfill to assess the influence of the various factors described above. Additional measurements will be taken in the summer of 2022. If the reasons for these variations can be determined, facility operators and policymakers can use this information to better improve conditions, operations, and regulations to reduce odor problems and limit GHG emissions.

Author Contributions: Conceptualization, A.M.C., J.J.S., J.Z., L.T.M., R.C. and E.M.L.; investigation, A.M.C., J.Z., J.J.S., M.J.S., J.M. and M.L.S.; data curation, A.M.C., J.J.S., J.Z., R.C., L.T.M. and M.L.S.; formal analysis, A.M.C., J.J.S., J.Z., R.C., L.T.M. and M.L.S.; software, A.M.C. and J.Z.; project

administration, J.J.S., R.C., L.T.M. and E.M.L.; methodology, A.M.C., J.J.S., J.Z., L.T.M., R.C., M.J.S., E.M.L. and M.L.S.; supervision, J.J.S., L.T.M., R.C. and E.M.L.; visualization, A.M.C.; writing—original draft preparation, A.M.C.; writing—review and editing, J.Z., J.J.S., L.T.M., R.C., E.M.L., J.M., M.J.S. and M.L.S.; validation, J.J.S. and J.Z.; funding acquisition, J.J.S., L.T.M., R.C. and E.M.L. All authors have read and agreed to the published version of the manuscript.

Funding: This study was funded by the New York State Energy Research and Development Authority (NYSERDA), Contract 156227.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Please refer to https://doi.org/10.6084/m9.figshare.20205515.v2 for all data pertaining to this paper (accessed on 27 July 2022).

Acknowledgments: We would like to thank Dirk Felton and Oliver Rattigan from New York State Department of Environmental Conservation for providing the Picarro G2204 for this study.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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