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Use of an Unmanned Aircraft System to Quantify NO_x Emissions from a Natural Gas Boiler

Brian Gullett[1], Johanna Aurell[2], William Mitchell[1], Jennifer Richardson[3]

[1]US Environmental Protection Agency, Office of Research and Development, Research Triangle Park, North Carolina, 27711, USA; [2]University of Dayton Research Institute, Dayton, Ohio, 45469-7532, USA; [3]The Dow Chemical Company, Midland, Michigan, 48667, USA.

Correspondence to: Brian Gullett (gullett.brian@epa.gov)

Abstract

Aerial emission sampling of four natural gas boiler stack plumes was conducted using an unmanned aerial system (UAS) equipped with a light-weight sensor/sampling system (the "Kolibri") for measurement of nitrogen oxide (NO), and nitrogen dioxide (NO_2), carbon dioxide (CO_2), and carbon monoxide (CO). Flights (n = 22) ranged from 11 to 24 minutes duration at two different sites. The UAS was maneuvered into the plumes with the aid of real-time CO_2 telemetry to the ground operators and, at one location, a second UAS equipped with an infrared/visible camera. Concentrations were collected and recorded at 1 Hz. The maximum CO_2, CO, NO, and NO_2 concentrations in the plume measured were 10,000 ppm, 7 ppm, 27 ppm, and 1.5 ppm, respectively. Comparison of the NO_x emissions between the stack continuous emission monitoring systems and the UAS/Kolibri for three boiler sets showed an average of 5.6 % and 3.5 % relative percent difference for the run-weighted and carbon-weighted average emissions, respectively.

Keywords: Emissions, natural gas, boiler, unmanned aircraft system, drone, continuous emission monitoring

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1 Introduction

Aerial measurement of plume concentrations is a new field made possible by advances in Unmanned Aircraft Systems (UAS, or "drones"), miniature sensors, computers, and small batteries. The use of a UAS platform for environmental sampling has significant advantages in many scenarios in which access to environmental samples are limited by location or accessibility. Hazards to equipment and personnel can also be minimized by the mobility of the UAS as well as their ability to be remotely operated away from hazardous sources. UAS-based emission samplers have been used for measurement of area source gases (Neumann et al., 2013; Rosser et al., 2015; Chang et al., 2016; Li et al., 2018), point source gases (Villa et al., 2016), aerosols (Brady et al., 2016), black carbon particles (Craft, 2014), volcanic pollutants (Mori et al., 2016), particle mass (Peng et al., 2015), and particle number concentrations (Villa et al., 2016).

UAS-based emission measurements are particularly suited for area source measurements of fires and can be used to determine emission factors, or the mass amount of a pollutant per unit of source operation, such as mass of particulate matter (PM) per mass of fuel (e.g., biomass) burned. These values can be converted into emission rates, such as mass of pollutant per unit of energy (e.g., g NO_x kJ^{-1}). These determinations typically rely on the carbon balance method in which the target pollutant is co-sampled with the major carbon species present and, with knowledge of the source's fuel (carbon) composition, the pollutant to fuel ratio or an emission rate/factor, can be calculated.

For internal combustion sources that have a process emission stack, downwind plume sampling can use the same method. When combined with the source fuel supply rate and stack flow rates (to determine the dilution rate), measurements comparable to extractive stack sampling may be possible. To our knowledge, determination of emission factors from a stack plume using a UAS-borne sampling system has not previously been demonstrated.

The feasibility of downwind plume sampling using a sensor-equipped UAS was tested on industrial boilers at the Dow Chemical Company (Dow) facilities in Midland, Michigan (MI) and St. Charles, Louisiana (LA). The sensor system was designed and built by the EPA's Office of Research and Development and the UAS was owned and flown by the Dow Corporate Aviation Group. To determine the comparative accuracy of the measurements, the UAS-based emission factor was compared with the stack continuous emission monitoring systems (CEMS). The target pollutants were nitrogen oxide (NO) and nitrogen dioxide (NO_2) to mimic the stack CEMS measurement methods. Carbon as carbon dioxide (CO_2) and carbon monoxide (CO) were measured on the UAS for the carbon balance method.

2 Materials and Method

Plume sampling tests were conducted on two natural-gas-fired industrial boilers located at Dow's Midland, Michigan and St. Charles, Louisiana facilities. The Midland boilers are firetube type boilers using low pressure utility supplied natural gas. They are equipped with low NO_x burners and utilize flue gas recirculation to reduce stack NO_x concentrations. The Midland facility burned natural gas with a higher heating value (HHV) of 9,697 kcal m^{-3} (1089 British Thermal Unit (BTU)/ft^{-3}). The two tested stacks are 14 m above ground level and 7 m apart. To avoid sampling overlapping plumes, only a single boiler was operating during the testing. The St. Charles boilers are D-type water package boilers using natural gas fuels (high pressure fuel gas (HPFG) and low pressure off-gas (LPOG)). They are equipped with low NO_x burners with flue gas recirculation to reduce stack NO_x concentrations. The boiler stacks are about 20 m apart and reach over 20 m in height above ground level. The St. Charles facility burned natural gas under steady state conditions with a composition of 77.12 % CH_4, 2.01 % C_2H_6, and 19.91 % H_2 and a HHV of 7,845 kcal m^{-3} (881 BTU ft^{-3}). Both boilers were operational during aerial sampling, but the wind direction and UAS proximity to the target stack precluded co-mingling of the plumes.

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Air sampling was accomplished with an EPA/ORD-developed sensor/sampler system termed the "Kolibri". The Kolibri consists of real-time gas sensors and pump samplers to characterize a broad range of gaseous and particle pollutants. This self-powered system has a transceiver for data transmission and pump control (Xbee S3B, Digi International, Inc., Minnetonka, MN, USA) from the ground-based operator. For this application, gas concentrations were measured using electrochemical cells for CO, NO, and NO_2 and a non-dispersive infrared (NDIR) cell for CO_2 (Table 1). All sensors were selected for their applicability to the anticipated operating conditions of concentration level and temperature as well as for their ability to rapidly respond to changing plume concentrations due to turbulence and entrainment of ambient air. Each sensor underwent extensive laboratory testing to verify performance and suitability prior to selection for the Kolibri. In anticipation of temperatures as low as 0°C at the Midland site, insulation was added to the Kolibri frame and the sampled gases were preheated prior to the sensor with the use of a heating element and micro fan inside the Kolibri.

Concentration data were stored by the Kolibri using a Teensy USB-based microcontroller board (Teensy 3.2, PJRC, LLC., Sherwood, OR, USA) with an Arduino-generated data program and SD data card. All four sensors underwent pre- and post-sampling two- or three-point calibration using gases (Calgasdirect Inc., Huntington Beach, CA, USA) traceable to National Institute of Standards and Technology (NIST) standards.

Table 1. UAS/Kolibri Target Analytes and Methods

Analyte	Instrument	Frequency	Cal. Gases (ppm) Midland	Cal Gases (ppm) LA
CO_2	SenseAir CO_2 Engine K30, NDIR[a]	Continuous, 1 Hz[b]	408, 990	392, 996, 5890
CO	E2v EC4-500-CO, Electrochemical cell	Continuous, 1 Hz	0[c], 9.67, 50.6	0, 9.9, 51.8
NO	NO-D4, Electrochemical cell	Continuous, 1 Hz	0, 2.1, 41.4	0, 2.1, 40.4
NO_2	NO2-D4, Electrochemical cell	Continuous, 1 Hz	0, 2.1, 10.4	0, 1.9, 10.4

[a]Non-dispersive infrared. [b]Hz – hertz. [c]Zero (0) cal gas = air.

The NO sensor (NO-D4) is an electrochemical gas sensor (Alphasense, Essex, UK) which measures concentration by changes in impedance. The sensor has a detection range of 0 to 100 ppm with resolution of < 0.1 RMS noise (ppm equivalent) and linearity within ±1.5 ppm error at full scale. The NO-D4 was tested to have a response time to 95 % of concentration ($T_{95\%}$) of 6.3±0.52 seconds and a noise level of 0.027 ppm. The temperature and relative humidity (RH) operating range is 0 to +50 °C and 15 to 90 % RH, respectively.

The NO_2 sensor (NO2-D4) is an electrochemical gas sensor (Alphasense, Essex, UK) which likewise measures by impedance changes. It has a NO_2 detection range of 0-10 ppm with resolution of 0.1 RMS noise (ppm equivalent) and linearity error of 0 to 0.6 ppm at full scale. Its $T_{95\%}$ was measured as 32.3±3.8 seconds with a noise level of 0.015 ppm. The temperature and RH operating range is 0 to +50 °C and 15 to 90 % RH, respectively.

The CO_2 sensor (CO_2 Engine® K30 Fast Response, SenseAir, Delsbo, Sweden) is a NDIR gas sensor and the voltage output is linear from 400 to 10,000 ppm. The temperature and RH operating range is 0 to +50 °C and 0 to 90 % RH, respectively. The CO_2-K30 sensor was measured to have a $T_{95\%}$ response time at 6000 ppm CO_2 of 9.0 ± 0.0 seconds and having a noise level of 1.6 ppm.

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The CO sensor (e2V EC4-500-CO, SGX Sensortech Ltd, High Wycombe, Buckinghamshire UK) is described more fully elsewhere (Aurell et al., 2017; Zhou et al., 2017). Variations of the Kolibri sampling system allow for measurement of additional target pollutants including particulate matter (PM), polycyclic aromatic hydrocarbons (PAHs), volatile organic compounds (VOCs) including carbonyls, energetics, chlorinated organics, and perchlorate (Aurell et al., 2017; Zhou et al., 2017).

At both facilities the aviation team from Dow flew their DJI Matrice 600 UAS, a six-motor multicopter (hexacopter), into the plumes with EPA/ORD's Kolibri sensor/sampler system attached to the undercarriage (Figure 1). In this configuration of sensors, the Kolibri system weighed 2.4 kg. Typical flight elevations at Midland and St. Charles were 21 and 32 m above ground level (AGL), respectively, and flight durations ranged from 9 to 24 min. At the St. Charles location, the UAS pilot was approximately 100 m from the center point of the two stacks, easily allowing for line of sight operation. A telemetry system on the Kolibri provided real time CO_2 concentration and temperature data to the Kolibri operator who in turn advised the pilot on the optimum UAS location.

CEMS on the boiler stacks produced a continuous record of NO_x emission and O_2 concentrations. Stack and CEMS types located at the Midland and St. Charles facilities are shown in Table 2. The stack NO_x analyzer is capable of split concentration range operation: Low (0-180 ppm) and High (0-500 ppm).

Figure 1. Dow UAS with Kolibri attached to the undercarriage.

Table 2. CEMS Instruments at both Dow locations.

Gas Measured	Midland CEMS	St. Charles CEMS
O_2	Gaus Model 4705	ABB/Magnos 106
NO_x	Thermo Model 42i-HL	ABB/Limas 11

The plant CEMS undergo annual relative accuracy audit testing (NSPS Subpart Db, Part 70) using EPA Method 7E (2014) for NO_x and Method 3A (2017a) for O_2. Calculation of NO_x emissions use the appropriate F factor, a value

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that relates the required combustion gas volume to fuel energy input, as described in EPA Method 19 (2017b). Flue gas analysis for O_2 and CO_2 are performed in accordance with Method 3A (2017a) using an infrared analyzer to allow for calculation of the flue gas dry molecular weight.

The CEMS and UAS/Kolibri data were reduced to a common basis for comparison of results. Emission factors, or mass of NO_x per mass of fuel carbon burned, and emission rates, or mass of NO_x per energy content of the fuel, were calculated from the sample results. The determination of emission factors, mass of pollutant per mass of fuel burned, depends upon foreknowledge of the fuel composition, specifically its carbon concentration, and its supply rate. The carbon in the fuel is presumed for calculation purposes to proceed to either CO_2 or CO, with the minor carbon mass in hydrocarbons and PM ignored for this source type. Concurrent emission measurements of pollutant mass and carbon mass (as CO_2 + CO) can be used to calculate total emissions of the pollutant from the fuel using its carbon concentration and fuel burn rate.

The UAS/Kolibri emission factors were calculated from the mass ratio of NO + NO_2 with the mass of CO + CO_2 resulting in a value with units of mg NO_x kg^{-1} C. CO_2 concentrations were corrected for upwind background concentrations. CEMS values of O_2 and fuel flowrate were used to calculate stack flowrate using US EPA Method 19 (2017b). This Method requires the fuel higher heating value and an F factor (gas volume per fuel energy content, e.g., m^3 $kcal^{-1}$ (ft^3 BTU^{-1})) to complete this calculation. For natural gas, the F factor is 967 m^3 10^{-6} kcal (8,710 ft^3 10^{-6} BTU) (Table 19-2, EPA Method 19 (2017b)). The concentration, stack flowrate, and fuel flowrate data allow determination of NO_x and C emission rates.

3 Results and Discussion

The UAS/Kolibri team easily found the stack plumes at both locations using the wind direction and CO_2 telemetry data transmitted to the ground operator. Use of an infra-red (IR)/visible camera on a second UAS at St. Charles for some of the flights aided more rapid location of the plume and positioning of the UAS/Kolibri. Gas concentration fluctuations were rapid and of high magnitude as observed in a representative trace in Figure 2. CO_2 concentrations to 10,000 ppm were observed; the relatively lower average CO_2 concentrations reflect the rapid mixing and entrainment of ambient air causing dilution.

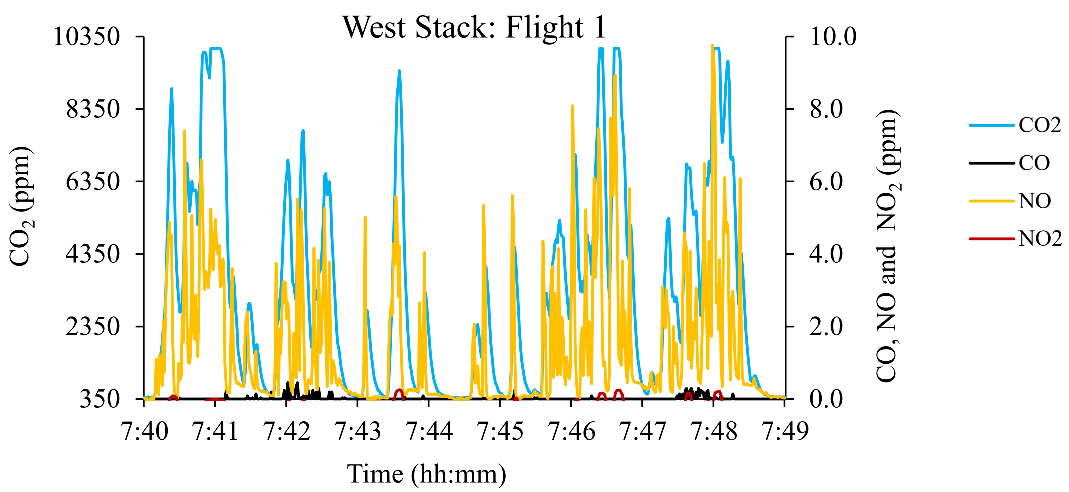

Figure 2. Example of UAS/Kolibri-measured plume concentrations from the St. Charles West Boiler.

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Sampling data and emission factors from the UAS/Kolibri are shown in Tables 3, 4, and 5 for the Midland, St. Charles east stack, and St. Charles west stack, respectively. Eight sampling flights were conducted at the Midland site, five on the St. Charles East boiler, and nine on the St. Charles West boiler. Both boilers at the Midland site were operated under the same conditions, so their results have been presented together. Flights averaged 14 min (10 % relative standard deviation (RSD)) at the Midland facility and just over 20 min (10 % RSD) at the St. Charles facility. The shorter flight times in Midland were due to lower UAS battery capacity caused by colder temperatures (the sampling temperatures in the plume averaged 10±3°C). Average plume NO_x concentrations were 0.88±0.32 ppm at Midland and 1.22 ppm and 2.41 ppm at the two St. Charles boilers with an average RSD of 37 %, 36 %, and 12 %, respectively. The NO emission factor was typically 97 % of the total NO_x, with the NO_2 providing the minor balance.

Table 3. Midland UAS/Kolibri Sampling Data and Emission Factors.

Date	Flight	Flight time (hh:mm:ss)			NO_2	NO	NO_x	Avg. CO_2 ppm
	#	Up	Down	Total	mg kg^{-1} C	mg kg^{-1} C	mg kg^{-1} C	
11/14/2018	1	10:29:00	10:43:00	00:14:00	201	618	819	1213
11/14/2018	2	11:13:04	11:28:28	00:15:24	186	624	810	1138
11/14/2018	3	12:54:17	13:08:47	00:14:30	230	659	889	2948
11/14/2018	5	13:27:40	13:42:05	00:14:25	99	570	669	4658
11/15/2018	6	10:24:20	10:39:30	00:15:10	61	394	454	3703
11/15/2018	7	10:41:36	10:52:40	00:11:04	84	397	481	3983
11/15/2018	8	10:55:10	11:10:10	00:15:00	126	398	524	4781
Average				**00:14:13**	**141**	**523**	**664**	**3203**
Stand. Dev.				**00:01:28**	**65**	**121**	**179**	**1514**
RSD (%)				**10**	**46**	**23**	**27**	**47**

Flight # 4 excluded from calculations as CO was observed, which originated from a cycling second boiler.

Table 4. St. Charles East Stack UAS/Kolibri Sampling Data and Emission Factors.

Date	Flight	Flight time (hh:mm:ss)			NO_2	NO	NO_x	Avg. CO_2 ppm
	#	Up	Down	Total	mg kg^{-1} C	mg kg^{-1} C	mg kg^{-1} C	
07/23/2019	1	09:49:00	10:07:00	00:18:00	1	1442	1442	2305
07/23/2019	2	10:12:00	10:34:00	00:22:00	15	1461	1476	2526
07/23/2019	3	10:45:00	11:08:00	00:23:00	5	1534	1539	785
07/23/2019	4	11:11:00	11:31:00	00:20:00	101	1684	1785	1082
07/23/2019	5	11:52:00	12:01:00	00:09:00	107	2110	2217	1923
Average				**00:20:45**	**30**	**1530**	**1560**	**1675**
Stand. Dev.				**00:02:13**	**47**	**110**	**155**	**869**
RSD (%)				**11**	**155**	**7.2**	**9.9**	**52**

Flight # 5 was not included in the average as elevated CO concentrations were detected, likely from other sources in the facility.

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Table 5. St. Charles West Stack UAS/Kolibri Sampling Data and Emission Factors.

Date	Flight #	Flight time (hh:mm:ss) Up	Down	Total	NO_2 mg kg^{-1} C	NO mg kg^{-1} C	NO_x mg kg^{-1} C	Avg. CO_2 ppm
07/24/2019	1	07:31:00	07:49:00	00:18:00	25	1366	3221	1490
07/24/2019	2	07:52:00	08:16:00	00:24:00	49	1263	3503	1397
07/24/2019	3	08:19:00	08:38:00	00:19:00	87	1420	3415	1611
07/24/2019	4	09:23:00	09:46:00	00:23:00	65	1341	4509	1525
07/24/2019	5	09:49:00	10:11:00	00:22:00	47	1296	4813	1463
07/24/2019	6	10:16:00	10:36:00	00:20:00	52	1299	3773	1449
07/24/2019	7	10:38:00	11:00:00	00:22:00	53	1316	4194	1482
07/24/2019	8	11:51:00	12:13:00	00:22:00	90	1460	3129	1662
07/24/2019	9	13:17:00	13:39:00	00:22:00	47	1464	3606	1645
Average				**00:21:20**	**57**	**1358**	**1416**	**3796**
Stand. Dev.				**00:01:56**	**21**	**74**	**86**	**586**
RSD (%)				**9**	**36**	**5.5**	**6.0**	**15**

Table 6 presents the average O_2 and NO_x measurement results and the fuel supply rate at both locations. Values for natural gas supply, adjusted for the C_2H_6 and H_2 composition of the St. Charles fuel, were used to calculate the fuel carbon supply rate. These data allow calculation of the emission factor, mass of NO_x to the mass of carbon, reported in Table 7.

Table 6. Multi-Run Average Stack CEMS Data

	Midland	St. Charles	
	Both Boilers	East Boiler	West Boiler
O_2 (%)	8.2	4.9	4.5
NO_x (ppm)	15.7	50.4	42.9
Fuel rate	39.3 10^6 kJ h^{-1}	155.2 10^6 kJ h^{-1}	177.8 10^6 kJ h^{-1}

Table 7. Comparison of Average NOx Emission Factors from CEMS and UAS/Kolibri

	Run-Averaged NOx Emission Factor, mg NO_x kg^{-1} C (± 1 std dev)		
	Midland	St. Charles	
	Both Boilers	East Boiler	West Boiler
CEMS	612 ± 10	1555 ± 50	1303 ± 29
UAS/Kolibri	664 ± 179	1560 ± 155	1416 ± 86
RPD: CEM & UAS/Kolibri, %	8.2	0.3	8.3

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The UAS/Kolibri NO_x emission factor for Midland is 8 % higher than the simultaneous CEMS value. For the East and West boilers at St. Charles, the UAS/Kolibri NO_x emission factor value is <1 % and 8 % higher, respectively, than the CEMS values. The difference for the UAS/Kolibri in Midland may be attributed in part to the extremely cold temperature affecting the performance of the electrochemical sensors. The standard deviations for the CEMS data are based on the run-average NO_x values for each test. These values were calculated based on 10 sec averaging for the Midland tests, 60 sec averaging in St. Charles, and 1 sec averaging for the UAS/Kolibri. Higher standard deviations for the UAS/Kolibri are predictable given the rapidly changing values and wide range (~0-10 ppm) of NO_x data observed in Figure 2. Difference testing for the CEMS and UAS/Kolibri using $\alpha = 0.05$ and assumed unequal variances indicate that only the West Boiler and UAS/Kolibri are statistically distinct.

The emission rates calculated from the UAS/Kolibri data are 5.6 kg $NO_x \cdot 10^{-3}$ kJ, 14.6 kg $NO_x \cdot 10^{-3}$ kJ, and 13.3 kg $NO_x \cdot 10^{-3}$ kJ (0.013, 0.034, and 0.031 lbs $NO_x \cdot 10^{-6}$ BTU), respectively, for the Midland, East St. Charles, and West St. Charles boilers, below the regulatory standard of 15.5 kg $NO_x \cdot 10^{-3}$ kJ (0.036 lbs $NO_x \cdot 10^{-6}$ BTU). The emission factors were also calculated as carbon-weighted values to reflect potential differences in plume sampling efficiency between runs. The Midland, East St. Charles, and West St. Charles UAS/Kolibri emission factors were, respectively, 607, 1525, and 1409 mg NO_x kg^{-1} C. These amounted to relative percent differences of 0.8, 1.9, and 7.8 % between the CEM and UAS/Kolibri values, for an overall run-weighted average difference of 5.6 %. The difference between the CEM readings and those from the Kolibri weighted by the carbon collection amounts, reflecting the success at being within the higher plume concentrations, was 3.5 %.

4 Conclusions

This work reports, to our knowledge, the first known comparison of continuous emission monitoring measurements made in a stack with downwind plume measurements made using a UAS equipped with emission sensors.

The UAS/Kolibri system was easily able to find and take measurements from the downwind plume of a natural gas boiler despite lack of any visible plume signature. The telemetry system aboard the Kolibri system reported real time CO_2 concentrations to the operator on the ground, allowing the operator to provide immediate feedback to the UAS pilot on plume location. Comparison of the CEM data with the UAS/Kolibri data from field measurements at two locations showed agreement of NO_x emission factors within 5.6 % and 3.5 % for time-weighted and carbon-collection-weighted measurements, respectively.

Data availability. The tabular and figure data are available at the Environmental Dataset Gateway https://edg.epa.gov/metadata/catalog/main/home.page.

Author contributions. BG was the prime author of the paper and the project lead. JA conducted the Kolibri field testing and data analysis. WM designed the instrument electronics. JR led the UAS group and field test arrangements.

Competing interests. The authors declare that they have no conflict of interest.

Disclaimer. The views expressed in this article are those of the authors and do not necessarily represent the views or policies of the U.S. EPA.

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