energy&fuels Article
pubs.acs.org/EF

Exposures to Carbon Monoxide from Off-Gassing of Bulk Stored Wood Pellets

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Supporting Information

ABSTRACT: There has been a significant increase in use of wood pellets in residential and commercial scale boiler systems within New York State, such an increase will lead to increased storage of bulk pellets in homes and buildings. Serious accidents in Europe have been reported over the past decade in which high concentrations of carbon monoxide (CO) have been found in bulk pellet storage bins. Thus, additional exposure data for CO in pellet bin storage areas are needed to assess the potential hazards. Using calibrated CO sensors, continuous CO measurements were made from the spring 2013 to spring 2014 in a number of wood pellet storage bins in New York State. The CO sensors, in some cases, in conjunction with sensors for CO_2, O_2, relative humidity, and temperature, were installed in a residential basement, an external storage silo, and several boiler room storage areas in schools and a museum. Peak concentrations in these pellet storage locations ranged from 14 ppm in the basement residence to 155 ppm inside the storage silo at a school. One-hour CO concentrations in the boiler rooms were typically 10−15 ppm. The measured concentrations were compared to regulatory standards of 50 ppm and recommended guidelines of 35 and 9 ppm for work and nonworking environments, respectively. The concentrations at the three locations in the middle school never exceeded the 35 ppm guideline. At the museum, the CO concentrations after pellets delivery did reach a maximum of 55 ppm for a 1-h average. However, high concentrations remained for only 4 days due to natural ventilation in this storage location. Storage areas for pellets must be considered confined spaces and require appropriate entry procedures. As the biomass heating with pellets becomes more prevalent, improved designs for storage bins must be considered to minimize the risk of exposure to CO to building occupants.

■ INTRODUCTION

The rise in fossil fuels costs, the need of energy security, and the desire for clean and renewable energy has stimulated the use of alternative heating fuels. Wood pellets are a biofuel that has gain popularity in many places, such as the Northeastern United States.[1,2] The New York State Energy Research and Development Authority (NYSERDA) has initiated multiple demonstration projects to introduce advanced wood-pellet fired boilers into New York State (NYS) including a European-built 150 kW (500 MBTU/h) wood pellet boiler installed at the Walker Center at Clarkson University and a 500 kW (1.7 MMBTU/h) installed at the Wild Center Museum in Tupper Lake, NY.

Storage of bulk wood pellets can involve a variety of storage bins. Many pellet bin configurations have been developed, including bins within the structure and bins external to the building. Supporting Information Figure S1 shows an external pellet silo. Interior bins can be configured in a number of ways as shown in Supporting Information Figures S2−S6. The most common storage bin designed for homes is the fabric storage bins (Supporting Information Figure S6). The best storage bin for a building depends on the budget, space available, among other factors. The bins can range in size according to the consumer needs from 3 tons to more than 100 tons.

The storage of wood pellets in confined spaces produce various gases including carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), and volatile organic carbons (VOCs)[3−6] that will accumulate in the air and can potentially reach toxic levels. Fatal accidents have been reported on maritime vessels providing bulk transport of wood pellets[7,8] and in large pellet storerooms of private households in Europe.[9] Thus, it is important to understand the potential health threat posed by stored pellets.

Elevated levels of CO can cause serious illness and death of exposed people. The adverse health effects associated with CO vary with its concentration and duration of exposure.[10,11] The most important health effects associated with exposure to CO are due to its strong bond with the hemoglobin molecule, forming carboxy-hemoglobin (COHb). COHb impairs the oxygen-carrying capacity of the blood, putting a strain on tissues with high oxygen demand, such as the heart and the brain.[10−13] Health effects observed from exposure to 10−2500 ppm of CO include early onset of cardiovascular disease, behavioral impairment; decreased exercise performance, reduced birth weight, sudden infant death syndrome, increase daily mortality rate, serious headache, dizziness, nauseas, unconscious, convulsions, and death, among others.[10,11,14] In order to protect peoples' health, a COHb level of 2.5% should not be exceeded.[10] Recommended guidelines for homes are 9 ppm of CO averaged over 8 h as proposed by American Society of Heating, Refrigerating, and Air-Conditioning Engineers (ASHRAE) and the World Health Organization (WHO). The National Institute of Occupational Safety and Health (NIOSH) guideline for work environments is 35 ppm over 8 h

Received: September 19, 2014
Revised: December 13, 2014
Published: December 16, 2014

DOI: 10.1021/ef5021186
Energy Fuels 2015, 29, 218−226

Table 1. Sites of the Pellets Bin in Use, Theirs Sizes, Time of Monitoring, Description of Place Being Monitor, and Types of Sensor Being Deployed to Monitor Pellet Emissions

No.	site	storage capacity (ton)	time of monitoring	sensor location	monitoring sensors
1	residential basement at Massena	10	March 5, 2013–April 18, 2014	basement near storage bag	CO
2	Malone Middle School, Malone	room capacity of 100, filled with 20	Sept. 27, 2013–April 23, 2014	air flow from room bin (air duct)	CO and PM
			Feb. 27, 2013–April 22, 2013; Oct. 2, 2013–April 23, 2014	hallway near bin	CO
			Feb. 27, 2013–April 22, 2013; Sept. 27, 2013–April 23, 2014	boiler room	CO
3	Saranac Lake Petrova Elementary School, Saranac Lake		March 27, 2013–April 22, 2013	boiler room	CO
		20	March 27, 2013–April 22, 2013; Aug. 27, 2013–April 22, 2012	silo	CO
4	Wild Center Museum, Tupper Lake	30	Sept. 27, 2013–Oct. 22, 2013	pellet bin	CO, CO_2, O_2, RH, and T
5	Walker Center, Clarkson University, Potsdam	10	Sept. 21, 2013–April 25, 2014	pellet bin	CO, CO_2, O_2, RH, and T
6	Energy Cabin, Clarkson University, Potsdam	3	Sept. 18, 2013–April 23, 2014	pellet bin	CO, CO_2, O_2, RH, and T

while the Occupational Safety and Health Administration (OSHA) has established a standard of 50 ppm over 8 h that is not to be exceeded. More detailed information on CO health effects and recommended maximum exposures is provided in the Supporting Information and Table S1.

The CO generation in bulk storage bins, such as the ones in this study, is the result of an oxidation process of the wood pellets that produces CO, CO_2, CH_4, and VOCs.[4,5] The nature of this process still needs to be determined. Prior studies found the process to be mass, temperature, moisture content, and volume dependent.[5−7] In addition, it has been hypothesized that the mechanism of formation of VOCs is due to the oxidation of fatty/resin acids by observing the emitted amount of aldehydes and ketones decreased by 45% during storage.[15] However, that study does not relate any of these carbonyl concentrations to the CO emissions, raising the question of how the CO is formed. Other research has related the CO emissions to microbial activities in the wood pellets and wood chips.[9,16] However, wood pellets are dried at around 500 °C in the production process and have a low water content (<7%) that creates unfavorable conditions for microbiological activity.[9]

In an attempt to understand the emission rates of stored wood pellets and be able to predict indoor concentrations, Fan and Bi studied the kinetics of CO off-gassed from stored wood pellets in small containers and developed a model that predicts the CO emissions per unit mass of stored pellets.[17] While Fan and Bi's model provided a framework for understanding the emission rates, validation of the model is still necessary particularly since the only data they employed was for softwood pellets. Models can never account for all aspects of an in situ situation (actual environments). Hence, field sampling is necessary to characterize the actual airborne CO concentrations in in-use systems. The purpose of this study is to characterize the potential exposures of CO from bulk storage pellets used in different typical U.S. storage systems, including in-use bins in schools, work places, and a residence.

■ EXPERIMENTAL DETAILS

Sampling Sites. Six different pellet bins were monitored during this study: the Walker Center and Energy Cabin at Clarkson University, the Wild Center Museum in Tupper Lake, NY, Malone Middle School in Malone, NY, Petrova Elementary School in Saranac Lake, NY, and a residential basement in Massena, NY. In several cases, multiple monitors were deployed to assess leakage of CO from the storage bins into normally occupied spaces. Table 1 summarizes the specific sites together with the monitoring system deployed.

Monitoring Systems. Sensor systems were constructed from commercially available gas sensors as described in Table 2 and the Supporting Information for this report. Two types of systems were developed. One system included only a CO sensor and in the other, CO, CO_2 and O_2 as well as temperature (T) and relative humidity (RH) were measured.

The systems were then deployed into the various locations monitored. At the Petrova Elementary School, the pellet storage system is a silo external to the building with 20 tons capacity. The sensor was connected at the end of the pneumatic delivery tube as shown in Supporting Information Figure S1.

For Malone Middle School, the CO and particulate matter (PM) sensors were placed in an air duct constructed to ventilate the storage bin at this site. The air in the duct should be representative of the concentrations within the bin. Supporting Information Figure S2 shows the CO and PM monitor connected into the side of the duct.

The more complete sensor systems were placed into the Walker Center pellet bin with 10 ton capacity (Supporting Information Figure S3), the container that serves as the pellet bin at the Wild Center with 30 ton capacity (Supporting Information Figure S4) and into the Energy Cabin pellet bin at Clarkson University with 3 tons capacity (Supporting Information Figure S5).

A monitor was placed in the basement of a home heated with a wood pellet boiler (Supporting Information Figure S6). The pellets are stored in a 10 ton gas permeable bag storage system. This bag system is a commercially available storage system that has been commonly installed in homes although more commonly as a 3 ton system rather than this 10 ton facility.

The monitoring systems came online at various times. The Massena, Saranac Lake, and Malone sites started in spring 2013. Most locations consumed their pellets by the end of April 2013 so there were little or no pellets in most of the storage facilities over the summer, so no monitoring was performed. New pellets were obtained at the end of the summer 2013 with different locations staring their monitoring (Tupper Lake and Potsdam sites) for the first time during summer 2013 and getting deliveries at different times until April 2014. The sampling dates are provided in Table 1. All of the CO concentration presented here are 1-h averages in order to understand the dynamics of the CO build up in storage bins. However, the 8-h guidance values are shown for reference since they provide conservative values. The 8-h average values that exceed the guidance levels are discussed since most of the sampling sites are occupational spaces. Only the Massena residence and the Energy Cabin at Clarkson University are nonworking environments.

DOI: 10.1021/ef5021186
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Table 2. Sensors and Their Respective Performance Used for This Field Study

sensors ID	measurement	detection limit	accuracy	reproducibility	range of detection
Vaisala Model HMT120[a]	temperature and relative humidity	%RH: 0 (4 mA) T (°C): −50	0−90%: ±3.0%RH 90−100%: ±4.0%RH −40 to +0, +40 to +80 °C: ± 0.4	±2%RH for 2 years. T (°C): not reported	%RH: 0−100 T (°C): −40 to 80
City Tech T3E/F[b]	CO (ppm)	4−20 mA d.c. 0.10 ± 0.02 μA/ppm	<9 ppm equiv.	linear range, 1 ppm	0−500
T7OXV CiTiceL[b]	% O_2	4−20 mA d.c, (0.0 ± 0.5)%, just accounting for the maximum error	maximum error in the 0−25% range is 0.5% at around 10% O_2	linear range, 0.1% vol	0−25%
IRceL Evaluation Kit[b]	%CO_2	0−1 V, 0%	−20 °C TO +50 °C: ± (0.1% vol CO_2 + 4% of concn.)	< ± 0.075% CO_2 (at all range) < ± 0.003% CO_2 (at zero)	0−5%
AEROCET 831 Aerosol Mass Monitor[c]	PM1, PM2.5, PM4 and PM10	0 μg/m^3	±10% calibration aerosol	0.5 μm/0.1 μg·m^3	0−1000 μg/m^3 ·1 min

[a]http://www.vaisala.com/. [b]http://www.citytech.com/. [c]http://www.metone.com/particulate-831.php/

RESULTS AND DISCUSSION

Massena Residence. The measurements in the Massena residence started on March 5, 2013 as shown in Figure 1a). Based on health guidelines presented in Supporting Information Table S1, 8-h exposure guidance levels of 9 and 35 ppm, respectively, are highlighted with dash−dot lines in all of the presented results. An initial spike of ~60 ppm of CO was observed. Subsequently, the values decreased to less than 9 ppm (ASHRAE threshold limit) for the rest of the spring 2013 heating season. In June, a rise in CO was observed although there was only a small quantity of pellets (~less than a ton) in storage as reported by the owner. Thus, the origin of this peak is not understood. The sensor was tested and recalibrated and reconnected on Aug. 2. Fresh softwood pellets were added to the storage bin in August. Figure 1b highlights this period. Concentrations above 9 ppm were observed, but as reported by the house owner, ventilation was applied because of the intense VOC odors emitted by the pellets. The concentration of CO decreased to below 9 ppm in less than 3 days and throughout the winter heating season with the measurements ending on April 18, 2014. Clearly, the ventilation (natural and/or mechanical) together with the difference in temperature (inside and outside the storage) reduced the concentration of CO as suggested by Emhofer.[18]

Petrova Elementary School. At the Petrova Elementary School in Saranac Lake, the storage bin is an outdoor silo (Supporting Information Figure S1) with a CO sensor at the bottom. In the spring of 2013, CO was monitored at the silo and at the containerized boiler room, and then, from the end of August 2013, only the silo was monitored. The monitoring at two sites was performed to check for leaks from the silo into the boiler room.

Figure 2 shows a 1-h CO value of approximately 20 ppm in the boiler room (maximum of 14 ppm for 1-h and 9 ppm for 8-h average), below the occupational guidelines, and up to approximately 155 ppm in the silo that is well above the guidelines and standards. By April 15, 2013, most of the pellets had been consumed and the concentrations returned to background conditions (<5 ppm). In the summer of 2013, the sensors were recalibrated and installed in just the silo. New pellets were delivered in October 2013. Concentrations at the silo rose to approximately 50 ppm in average, and only in one occasion reached to approximately 150 ppm. The values might be lower than previous heating season because of colder condition or pellets had an opportunity to age before they were delivered to the site. However, clearly enclosed silos such as this one need to be treated as confined spaces as per OSHA regulations if maintenance is required.

Malone Middle School. The Malone school uses the space in the basement that had been the coal bin for pellet storage. This bin had an active ventilation system producing a flow of 300 cubic feet meter. Three CO sensors were installed in this facility along with a particulate matter monitor (PM). One of the CO sensors was placed in hallway near the entrance door to the bin. A second sensor was deployed in the area around the two 1 MMBTU/h boilers. The third CO sensor and the PM monitor were installed in the outlet duct for the ventilation system (Supporting Information Figure S2). Data were collected in the spring of 2013 at the hallway and boiler room only. CO concentration increases as spikes between 10 and 15 ppm during the month of April for data averaged for 15 min and 1 h. The spikes lasted for less than an hour. It is likely

DOI: 10.1021/ef5021186
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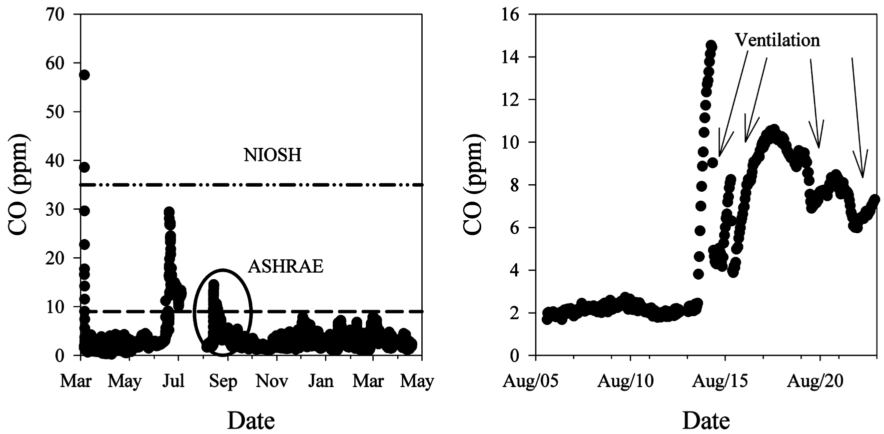

Figure 1. (a) CO measurement at the Massena Residence Basement (March 2013−April 2014). Dashed line refers to the ASHRAE limit (9 ppm of CO) and dotted line to NIOSH guidance level of 35 ppm of CO. (b) Time series (August 2−23, 2013) of CO measurement enlarged (inside the circle) from part a).

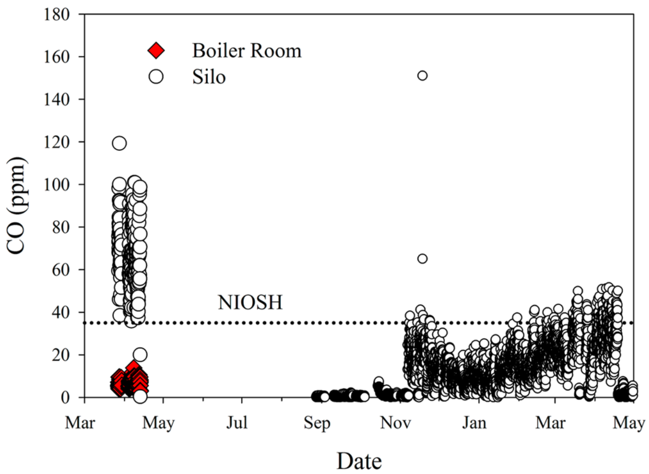

Figure 2. Time series of CO measurements inside the boiler room and pellet silo located outside of the Petrova Elementary School.

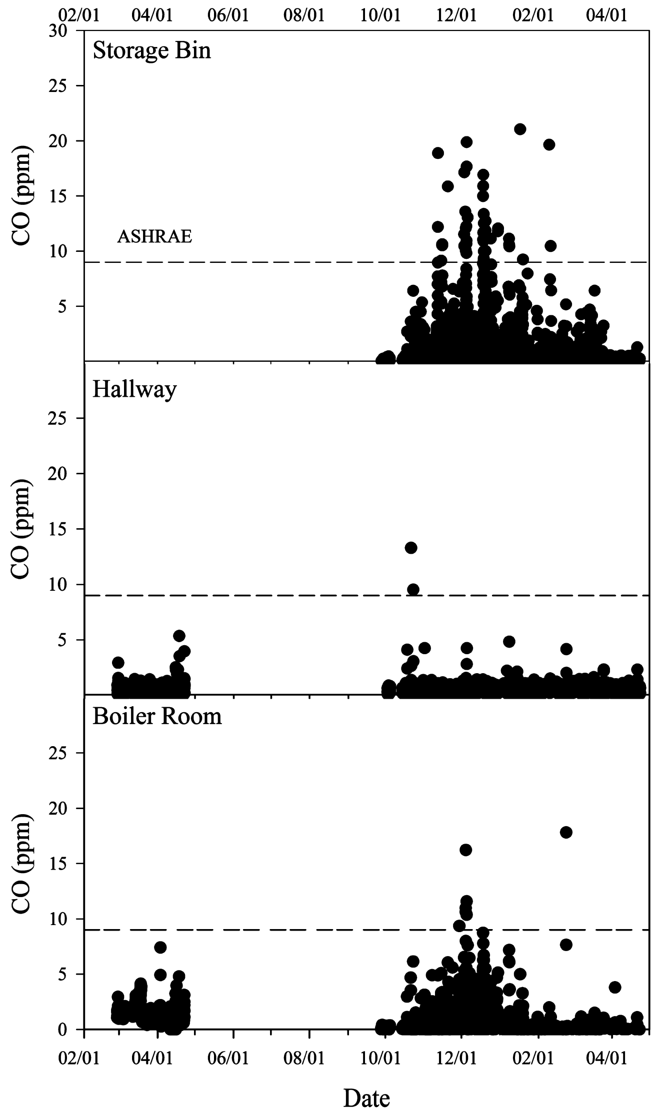

Figure 3. Time series of CO measurements at three locations (storage bin, hallway, and boiler room) in Malone Middle School.

that the high CO concentrations were due to other causes (gasoline powered equipment brought in from outdoors and still operating) and not due to the storing of wood pellets.

The sensors were returned to the laboratory for calibration and reinstall at the end of September and beginning of October 2013. Figure 3 shows the 1-h average CO concentrations at the three locations in the school basement. Peaks of CO were observed in the month of October that were reported as a result of storing and operating various gasoline-powered grounds keeping equipment in the hallway where the motors might be running as they moved them in from outside the building.

There were no pellets in the bin until November fourth when the first delivery of the season was made. At the hallway door, there are 6 peaks that correlate with high concentrations of CO in the air duct ventilation, but they were only spikes for short periods of less than 5 ppm. The pattern in boiler room from November until December suggest that the two boilers were leaking CO or that there might be a leak from storage bin to the boiler room. However, concentrations never exceeded the NIOSH recommendation (35 ppm) and OSHA limits (50 ppm) for occupational space. From the end of January, these concentrations diminished to well below guideline levels.

Since the delivery of the wood pellets generate combustible wood dust, it was imperative to measure the concentration of particles generated at this site. One of the major contributors to the risk of an explosion is the size of the dust particles. In general, the finer the combustible particle is the higher the risk

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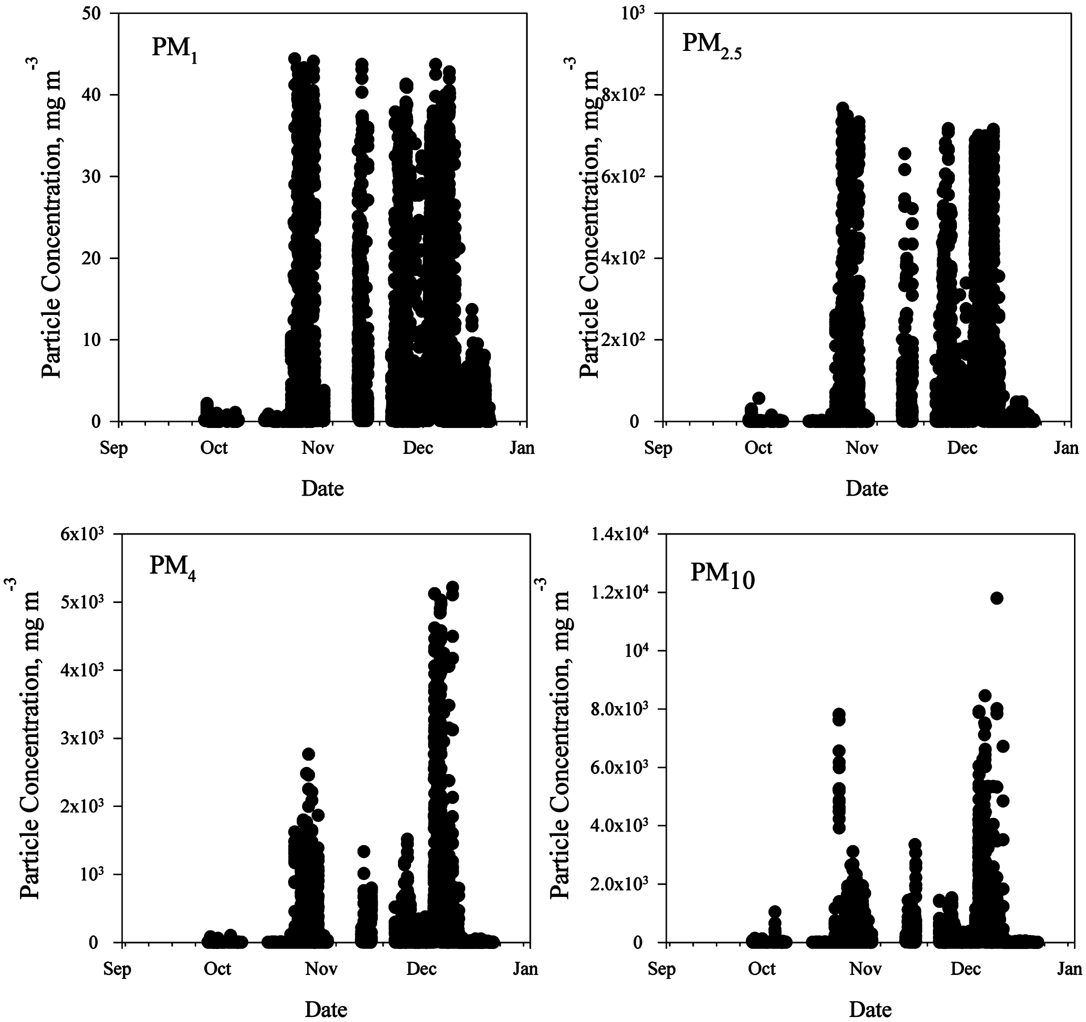

Figure 4. Particle concentration in the air duct exhaust at the storage bin in Malone Middle School.

of explosion. The National Fire Protection Association (NFPA) defines wood dust as a deflagrable (combustible dust) wood particulate with a median diameter of 420 μm or smaller, having a moisture content of less than 25%. The level of explosion limit for wood dust suspended on air is 40 g m^{-3} (4000 mg m^{-3}). Results of concentration of particles are shown in Figure 4. The sensor started having problems after mid-December, so these results are not presented. The data from November until mid-December clearly shows that the concentration of any of the particle sizes measured do not exceeds the concentration of airborne particles in the safety standards established (Max: PM10 ~ 12 000 μg m^{-3}; PM4 ~ 5000 μg m^{-3}; PM2.5 ~ 750 μg m^{-3}; and PM1 ~ 45 μg m^{-3}).

Wild Center. Background sampling started at the beginning of September, but there were unexpected spikes on all sensors that suggested an interference with the signal. The first pellets delivery was on Sept. 29, 2013. Data presented here is ranges Oct. 1−22, 2013; after this period, the system continues with interferences. Figure 5 shows the results of the five sensors for about 20 days in Oct. 2013. CO concentrations reached a maximum 1-h average of around 55 ppm. The 8 h average value reached 35 ppm that is the NIOSH guideline, but it did not exceed the OSHA regulatory standard of 50 ppm. The elevated concentrations only persist until Oct. 4 (around 5 days). These data show a relationship among temperature, CO, and CO_2 that suggests that with an increase in temperature CO and CO_2 will increase the rate of off-gassing from the pellets. The maximum peaks of temperature match with the minimum peaks of %RH and %O_2, suggesting that the off-gassing occurs with a depletion of oxygen and lower RH. This result has been observed in a previous field study of larger scale storage bins (silo) were higher temperatures shows higher emission factors of CO and lower concentration of O_2.[19] The occasionally high temperature in the pellets bin appears to be a leak of heat from the solar-thermal panels on the side of the container. Nevertheless, the O_2 (21%−20.5%) and CO_2 (700−450 ppm) concentrations are quite constant over the rest of the sample period (Oct. 5−22). Thus, the limited decomposition of the pellets and the natural ventilation into this bin resulted in relatively constant atmospheric compositions.

Walker Center. This location has the first imported high-efficiency wood pellet boiler in the Northeastern U.S.A. Walker had a pellet delivery (softwood) on Nov. 4, 2013. The measured values are shown in Figure 6. CO concentrations rose to approximately 40 ppm. Concentrations remained above 10 ppm for 7 days (Nov. 4−11), and after Nov. 17, the concentration dropped to background conditions (<2 ppm). During these 7 days, the temperature showed a weak relationship with the CO and CO_2 off-gassing, in contrast with the observations from the Wild Center (Figure 5).

DOI: 10.1021/ef5021186
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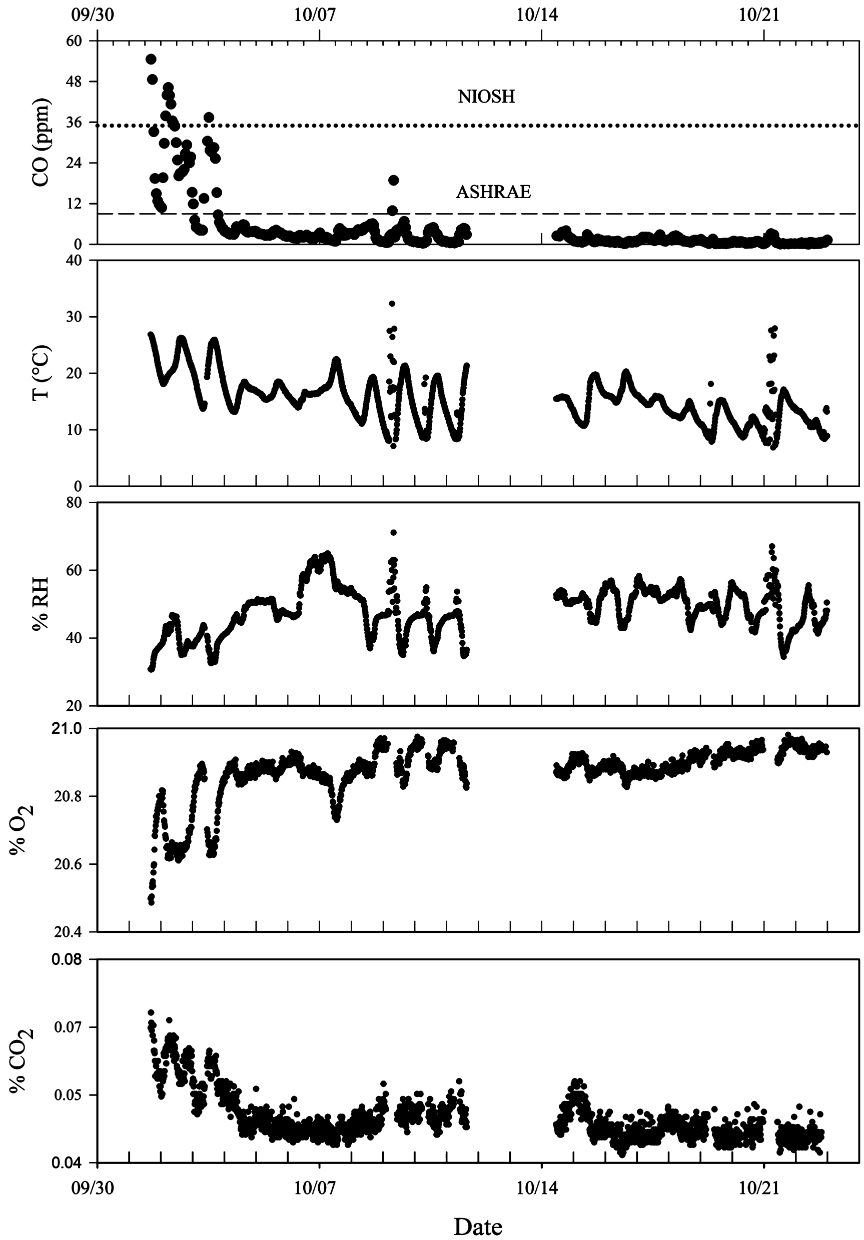

Figure 5. Time series for CO, temperature (T), relative humidity (%RH), $\%O_2$, and $\%CO_2$ in the Wild Center Museum in Tupper Lake, NY.

However, the lower values of RH (from 55 to 25%) and $\%O_2$ (from 20.0 to 19.8%) align with the peaks of CO and CO_2 (600−700 ppm). For O_2, this behavior can be due to the oxidation of the wood pellets. The water vapor variations may be attributed to the infiltration or ventilation in the building, while CO and CO_2 are being produced within the storage bin. The observed results do not suggest that water vapor reduction is attributable to water uptake by the pellets based on a moisture content analysis of the pellets in this storage bin after 4 months that showed a value of 4.5% (manufacturer's specifications for fresh pellets <5%).

During the winter, mid-December to the end of January, all sensors responded with ordinary spikes or off sets, especially the temperature sensor (range: 20−60 °C). This behavior might be the result of a malfunctioning room heater inside the bin that activates at low temperatures to ensure that the pellets do not freeze. In addition, there is an increase in CO (<10 ppm) and CO_2 (0.12%) concentrations at the end of February until the beginning of March that correspond to the starting of the boiler during this limited period. Because of modifications to the boiler system that were being performed, the boiler was not operated during this heating season at any other time. The results presented in the Walker Center shows that this site is not as well ventilated as the Wild Center resulting in a longer period for the CO to dilute. However, the fresh pellets delivered in November generated sufficient CO to exceed the occupational guideline value. Therefore, this storage bin has been defined as a confined place.

Energy Cabin. The Energy Cabin is the smallest bin in this study. The delivery of pellets is performed manually from 40 pounds bags that had been stored and placed outside the cabin generally for more than 2 weeks of storage. Figure 7 shows very low values of CO (<5 ppm) during the entire sampling period except for April when values reached approximately 10 ppm, above the 9 ppm of ASHRAE guideline. This CO off-gassing is followed by an increase in temperature in the bin that coincides

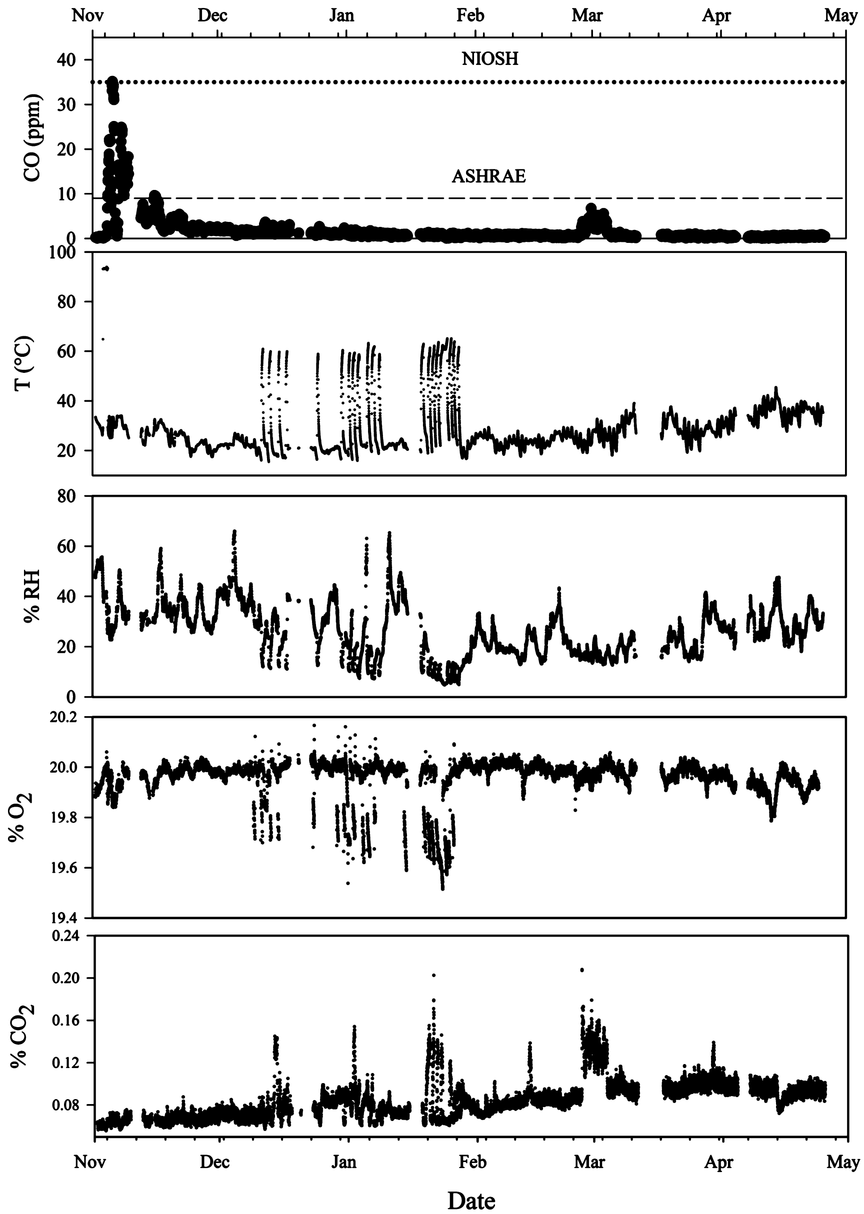

Figure 6. Time series for CO, temperature, relative humidity, $\%O_2$, and $\%CO_2$ in the Walker Center at Clarkson University in Potsdam, NY.

with the minimum RH and O_2 values during this period. In general, there is a weak relation between the CO off-gassing and the temperature, % RH, % O_2, and CO_2 over the entire period at this site. CO (<10 ppm), O_2 (21%), and CO_2 (450 ppm) concentrations are relatively constant over the entire period probably because the wood pellets had an opportunity to age before they were placed inside the bin.

The results presented here for the six different small-scale pellet storage bins show, in general, that the concentration were not as high as reported by Gauthier[9] and Emhofer[18] with all of the concentrations being below 200 ppm. This behavior might be in part due to the intrinsic ventilation of all the sites under study and the chemical composition and quality of the wood pellets manufacturer here (low ash and moisture content). Prior studies showed that commercial wood pellets produce less CO and PM than other solid fuels.[20] Also, the data show that CO levels increased only during the days immediately following the delivery of fresh pellets. It then decreases slowly to background values. Emhofer[18] concludes that storage rooms should not be entered within 4 weeks after pellet deliver, but our data suggest a significantly shorter time, more on the order ~1 week, may be acceptable. However, measurements of the CO concentrations should be made before entry.

Natural ventilation has been recommended by Emhofer,[18] such as utilizing caps on the filling tubes that allow air exchange between the storeroom and the surroundings to reduce CO concentrations. However, caution should be taken with respect to cross ventilation and differences in temperatures between the storeroom and surroundings. Also, as shown in our results at Massena residence, by opening windows (natural ventilation) and installing an exhaust fan (mechanical ventilation); the CO and VOCs (odors) were readily reduced.

■ CONCLUSIONS

This field study demonstrated that there is off-gassing of sufficient CO from stored pellets to represent a hazard that needs to be adequately addressed. Although no concentrations that would directly produce short-term extreme health effects

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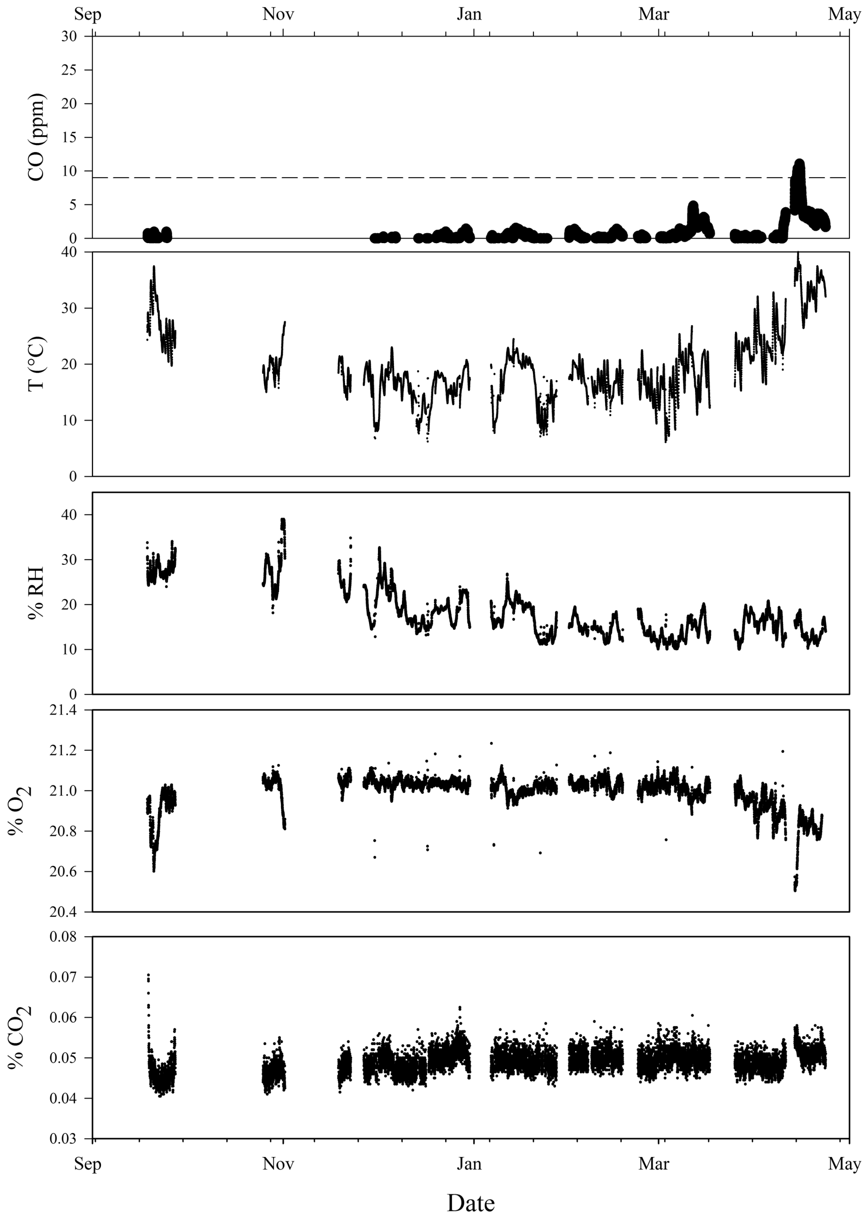

Figure 7. Time series for CO, temperature, relative humidity, $\%O_2$, and $\%CO_2$ in the Energy Cabin at Clarkson University in Potsdam, NY.

were observed, concentrations above levels set as exposure guidelines of 35 ppm in occupational settings and 9 ppm in homes were exceeded, especially immediately after pellet delivery. The concern is that as biomass boilers become more widely used, a broad array of homes with varying levels of natural ventilation will install pellet heating systems with inside the structure storage bins. In energy efficient (low air exchange rates) homes, this situation could produce unacceptable CO concentrations. These results raise a safety question regarding how pellets storage bins are designed and sited. Active and natural ventilation clearly reduces the average concentrations, although higher values were still occasionally observed. Pellet aging clearly reduced the amount of observed CO. There was a clear positive relationship with the off-gassing of CO and CO_2 and temperature where higher temperatures produce higher off-gassing production. Thus, bin temperatures need to be considered when designing or choosing a pellet storage bin.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Additional information regarding the health effect and exposure limits of CO, and a detail description of the experimental setup with respect to the sensor systems and sampling sites. This material is available free of charge via the Internet at http://pubs.acs.org/.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We want to acknowledge NYSERDA for their financial support and to all the people who allowed us to make measurements at their home, schools, and museum.

DOI: 10.1021/ef5021186
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DOI: 10.1021/ef5021186
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