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# Dynamics of Carbon Monoxide (CO) and Volatile Organic Concentrations in Wood Pellet Storage Bins

## Final Report

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# Dynamics of Carbon Monoxide (CO) and Volatile Organic Concentrations in Wood Pellet Storage Bins

***Final Report***

**Prepared for:**

**New York State Energy Research and Development Authority**

Albany, NY

Ellen Burkhard, Ph.D.
Senior Project Manager

Prepared by:

**Clarkson University**

Potsdam, NY

Philip K. Hopke, Ph.D.
Alan Rossner, Ph.D.
Lydia Soto-Garcia, Ph.D.

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# Notice

# Abstract

Emissions of carbon monoxide (CO) and volatile organic compounds (VOCs) arising from stored wood pellets have been previously reported. In some instances, the reported CO concentrations were extremely high. Thus, this study has been conducted of the concentrations of CO observed in in-use pellet bins and in the basement of a residence with a large pellet storage bin container. Concentrations in the larger bins associated with commercial scale wood pellet boiler systems have been found to exceed the Occupational Safety and Health Administration (OSHA) regulatory level of 50 parts per million (ppm) over an 8-hour period. Thus, the pellet bins are required to be considered as confined spaces. In the residence, 8-hour average concentrations up to 44 ppm CO have been observed. These values are well in excess of the guidance recommendation of the American Society of Heating, Refrigeration, and Air Conditioning Engineers (ASHRAE) of 9 ppm for indoor air quality. To better understand the off-gassing of CO and VOCs, laboratory studies of stored wood pellets were conducted. This work suggested that the off-gassing is a surface oxidation phenomenon that is limited by the total amount of pellet surface area. An improved kinetic model has been developed that includes this surface area limiting factor. The rate of CO off-gassing from pellets was tested for a variety of pellet and environmental conditions, including pellet composition (i.e. softwood, hardwood or blended), headspace volume of the container, humidity, pellet surface/volume ratio, and temperature. CO and VOC production differed by pellet composition in the following order: softwood>blended>hardwood. Softwood pellets produce more CO and VOCs than blended pellets than do hardwood pellets. Increasing the pellet storage temperature increases the rate at which the CO is emitted. Pellets stored under high relative humidity had a faster and higher CO emission rate. For nearly water-saturated storage conditions, the final CO concentration was about three times as high as that for a normal humidity condition. Dried wood pellets were less active in CO off-gassing compared to as received wood pellets and tested without additional drying in the laboratory. Aging the pellets by storing them for 90 days even at relatively low temperatures (6-8 ºC) reduces their emissions by 20 to 30%. Higher concentration, but lower emission rates were found from softwood followed by blended and hardwood pellets, respectively. The field testing of storage bins and laboratory testing of pellets in small volume containers did not observe CO concentrations as high as were reported in the European incidents involving fatalities, but the CO concentrations observed in our laboratory and field work are sufficient to be of health concern. The off-gassing of CO documented in this study and its potential hazards should be managed through proper storage and appropriate ventilation of the storage facilities.

# Keywords

Carbon monoxide, off gassing, outgassing, wood pellets, pellet bins

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# Acronyms and Abbreviations

| | |
|---|---|
| ACGIH | American Council of Governmental Industrial Hygienists |
| ASHRAE | American Society of Heating, Refrigerating, and Air-Conditioning Engineers |
| CHP | combined heat and power |
| hr | hour |
| CO | carbon monoxide |
| $CO_2$ | carbon dioxide |
| EPA | United States Environmental Protection Agency |
| FTIR | Fourier transform infrared spectroscopy |
| kg | kilogram |
| kW | kilowatt |
| $m^3$ | meter |
| MMBtu/h | million British thermal units per hour |
| NAAQS | National Ambient Air Quality Standards |
| NIOSH | National Institute of Occupational Safety and Health |
| NYS | New York State |
| NYSERDA | New York State Energy Research and Development Authority |
| $O_2$ | oxygen |
| OSHA | Occupational Safety and Health Administration |
| PM | particulate matter |
| ppm | parts per million |
| RH | relative humidity |
| SUNY-ESF | SUNY College of Environmental Science and Forestry |
| T | temperature |
| VOC | volatile organic compound |

# 1 Introduction

When fuel oil prices rose sharply in the fall of 2008, many institutions that rely on fuel oil for heat began to seriously consider wood combustion as a cost-effective alternative to provide building heating. However, the range of wood-fired boilers available at that time was limited to those with relatively low thermal efficiencies and significant pollution emissions. Thus, NYSERDA initiated demonstration projects to introduce advanced European technologies into New York State (NYS). The initial demonstration project was a European-built 150-kilowatt (kW; 500-Million Btu (MMBtu)/hr) wood pellet boiler installed at the Walker Center at Clarkson University in Potsdam, NY. Subsequently, the technology was licensed for construction fabrication in NYS and the first of these locally fabricated boilers, a 500-kW (1.7 MMBtu/hr) boiler was installed at The Wild Center in Tupper Lake, NY. Following these initial demonstrations projects, a number of other systems were deployed installed at schools in the region. An 9.6-MMBtu/hr wood pellet boiler, combined heat and power system to burn wood pellets was installed in the Gateway Building on the campus of SUNY’s College of Environmental Science and Forestry in Syracuse and a number of additional installations are planned or under construction in NYS. Thus, a substantial increase in the use of wood pellets as a heating fuel within NYS has led to the need to store the pellets on-site to provide the fuel for these systems.

Reports of high carbon monoxide (CO) concentrations in pellet storage areas in Europe with serious safety consequences have recently been published. Thus, it is important to better understand the potential health and safety threat posed by stored pellets. Gauthier et al. (2012) noted several deaths arising from CO poisoning in large pellet storage areas and reviewed previously reported incidents in marine vessels and commercial and residential wood pellet and wood chip storage facilities in Europe. In the laboratory, Fan and Bi (2013) measured the kinetics of CO production in pellets held in small containers to develop a kinetic model for the CO emissions. Field and laboratory studies conducted in the U.S. Northeast have further characterized the concentration and formation kinetics of CO, and provided information about potential safety issues related to the bulk storage of pellets. Pellets made in the Northeastern U.S. can differ significantly in their composition from those pellets made in Europe or British Columbia (Canada) due to the greater abundance of hardwood in the Northeast. Thus, this study has examined off-gassing from hardwood, softwood, and hardwood/softwood blended pellets through a combination of monitoring in functioning storage bins as well as additional laboratory studies.

This work addressed the following objectives:

- Continuous seasonal monitoring of carbon monoxide (CO), carbon dioxide ($CO_2$), oxygen ($O_2$), relative humidity (RH), and temperature (T) for three bulk pellet storage bins covering a range of 1 ton to 20 tons.
- Continuous seasonal monitoring of carbon monoxide (CO) in four pellet bins in a variety of locations including schools and a residence and determine if CO is leaking into occupied areas of one of the schools.
- Continuous seasonal monitoring of dust in the pellet bins at two schools.
- Daily monitoring of CO, $CO_2$, $O_2$, RH, T, and methane ($CH_4$) in a laboratory-based study of pellet storage for soft wood, hardwood, and blended (hardwood and softwood) pellets over a one-month period.

# 2 Prior Off-Gassing Studies

Over the past 15 years, there have been a limited number of reports of the formation of carbon monoxide (CO) from stored wood products, particularly wood pellets. The first known incident appears to be the death of one person and injury to two others in Rotterdam, the Netherlands during the unloading of wood pellets (Melin 2010). This incident alerted the shipping industry to the potential danger of large quantities of stored pellets. However, a second shipping incident occurred during the unloading of a ship in the port of Helsingborg, Sweden where the subsequent incident review found that the crew and dockhands had failed to follow the prescribed procedures for dealing with the unventilated cargo hold (SMSI 2007). Gauthier et al. (2012) made the first reports of deaths in wood pellet storerooms of private households. In January 2010, an engineer died after entering a pellet bunker having a storage capacity of approximately 155 tons of pellets. The bunker stored wood pellets for a district boiler system supplying heat to about 700 households. In February 2011, a woman was found dead in an 82 cubic meter pellet storeroom that provided fuel to a wood pellet heating system that supplied 60 households in Switzerland.

Unrelated to these incidents, Svedberg and Galle (2001) had investigated complaints of eye irritation and odor in a pellet factory in Sweden. They identified the presence of hexanal, pentanal, methanol, acetone and carbon monoxide using Fourier-transform infrared spectroscopy (FTIR). A subsequent study (Svedberg et al. 2004) investigated and described the presence and formation of CO and VOCs including hexanal in wood pellet storage facilities. They also monitored the emissions from kiln drying of wood and determine that the emissions were not specific to wood pellet production but were more general in nature. They concluded that "high levels of hexanal and carbon monoxide were strongly associated with storage of wood pellets and may constitute an occupational and domestic health hazard." However, this warning clearly was not widely disseminated and thus, measures were not taken that might have precluded he deaths noted previously.

Additional laboratory studies of CO off-gassing were undertaken by Kuang et al. (2008, 2009a, and 2009b). They examined the effects of storage temperature (T), headspace volume, and relative humidity (RH) on the extent of CO off-gassing. Increased temperature produced higher CO concentrations. These studies focused on softwood pellets as commonly produced in western Canada. There are no comparable studies on the emissions from hardwood or softwood/hardwood blended pellets.

Arshadi et al. (2009) examined the VOC emissions from pellets made of blends of pine and spruce. The fatty and resin acid concentrations were measured using gas chromatography-mass spectrometry for newly produced pellets and those aged for two and four weeks. The concentrations of aldehydes and ketones were determined by high performance liquid chromatography (HPLC) at zero, two and four weeks post production to explore the relationships between the acid content and the result carbonyl emissions over time. Drying temperature also influenced the aldehyde and ketone emissions of fresh pellets. They found a strong correlation between the pine fraction in the pellets and the initial fatty/resin acid content but the correlation decreased over storage time. The fatty and resin acid concentrations decreased by 40% during the four-week storage period. The drying temperature also influenced the aldehyde and ketone emissions of fresh pellets. The amounts of emitted aldehydes and ketones generally decreased by 45% during storage as the fatty/resin acids oxidized. Thus, they conclude that it is the oxidation of the fatty acids/resin that is the primarily mechanism for the observed off-gassing of VOCs. However, they did not relate VOC emission patterns to CO emissions.

Alternatively, Ferrero et al. (2011) measured the temperature and gas concentrations changes in a newly established pine wood chip pile (20 meter [m] × 15 m × 6 m containing approximately 400 tons of fresh weight material) for 150 days. The temperature was measured at 10 positions within the pile, and concentrations of $CO_2$, $O_2$, CO, and $CH_4$ were made at four locations. There was a substantial rise in temperature within the first 10–12 days of storage. The analysis of the gas samples collected in this period suggests that the temperature increase was caused by aerobic microbial processes.

# 3 Field Studies

The initial design of this current project was to monitor CO, $CO_2$, $O_2$, RH, and T in operating storage pellet containers on the campus of Clarkson University (Walker Center and Energy Cabin) and at The Wild Center. Subsequently, additional monitoring for CO was requested for several locations in New York State:

- Pellet bins at Malone Middle School in Malone.
- Pellet silo at the Petrova Elementary School in Saranac Lake.
- Pellet bin and boiler room area in the Gateway Building on the campus of the SUNY College of Environmental Science and Forestry (SUNY-ESF) in Syracuse.
- 10-ton pellet bag container in a residential basement in Massena.

The locations and monitors are listed in Table 1.

In the pellet bins at the Malone Middle School and at SUNY-ESF, monitors for airborne particulate matter (PM) were also installed. Soto-Garcia et al. (2015a) presents the results of these studies as an open access publication (http://pubs.acs.org/doi/abs/10.1021/ef5021186).

Unanticipated construction problems and delays with the boiler installation at SUNY-ESF prevented CO measurements at this site. At the other locations the highest concentrations of up to 155 parts per million (ppm) were observed in the 20-ton pellet silo adjacent to the Saranac Lake Petrova Elementary School, a storage bin with a low natural ventilation rate. In the Malone Middle School, where the pellets are stored in what used to be the coal bin, active ventilation had been installed to ensure generally low particulate matter concentrations. However, even with active ventilation a CO value of 45 ppm was measured in exhaust air from the bin following the introduction of a large shipment of pellets (20 tons), At Clarkson, following the delivery of 10 tons of pellets in early November to the Walker Center pellet storage bin, CO concentrations rose to 60 ppm and declined over the next month. At The Wild Center, a maximum value of 200 ppm CO was measured after a pellet delivery.

**Table 1. Location and Types of Monitors Deployed to Monitor Pellet Emissions**

| Location | Monitors |
|---|---|
| Walker Center pellet bin | CO, CO2, O2, RH, and T |
| Energy Cabin pellet bin | CO, CO2, O2, RH, and T |
| The Wild Center pellet bin | CO, CO2, O2, RH, and T |
| Malone Middle School pellet bin | CO and PM |
| Malone Middle School hallway near bin | CO |
| Malone Middle School boiler room | CO |
| Saranac Lake Petrova Elementary School | CO |
| SUNY-ESF CHP pellet bin | CO and PM |
| SUNY-ESF CHP boiler room | CO |
| Basement of residence with 10-ton pellet storage bag | CO |

In the 3-ton capacity bin for the Energy Cabin on the Clarkson campus, the maximum observed value was less than four ppm. This bin is filled manually with bagged pellets purchased locally. However, the age of these bagged pellets is unknown. Finally, in the basement of the residence in Massena, there was a high initial value of approximately 70 ppm observed when the CO sensor was first installed in March 2013. The concentrations declined to below four ppm until June when there was an unexplained rise to approximately 30 ppm with a subsequent decline to a relatively steady value of 14 ppm. The sensor was checked and found to be performing correctly. After reinstallation of the sensor, values were low. In early August, the bin was filled with 10 tons of softwood pellets, and the CO concentration increased to 15 ppm. Because of the odor of the VOCs that off-gassed from the softwood pellets, the owner provided ventilation resulting in a decrease in measured CO during each active ventilation period.

Additional measurements have been made since the publication of the initial results. Figure 1a shows the extended time series of 8-hour average measurements made in the Massena basement. Figure 1b shows an enlargement of the period in expanded view August 2013 using 15 minute average values. Fresh softwood pellets were placed into the bin during this period. The emission of VOCs was sufficient to induce the homeowner to ventilate the space, and the figure shows the effects of the ventilation on the CO concentrations. In the more recent measurements, values again exceeded the ASHRAE 8-hour guidance concentration of 9 ppm CO for indoor air quality, which is the same value as the health-based National Ambient Air Quality Standards set by the EPA. Thus, storage of pellets within a house without adequate active ventilation can lead to indoor CO concentrations that exceed the existing guidelines for residential indoor air.

**Figure 1. Time series of measurements in the basement of the Massena, NY residence**

a) Complete time series of 8 hour values; b) expanded view of August 2013 using 15 minute values.

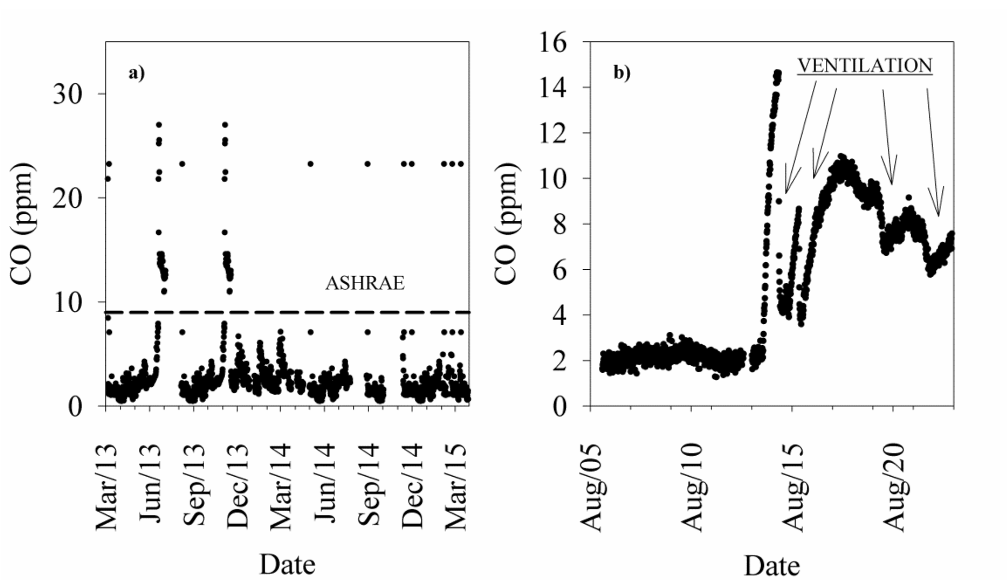
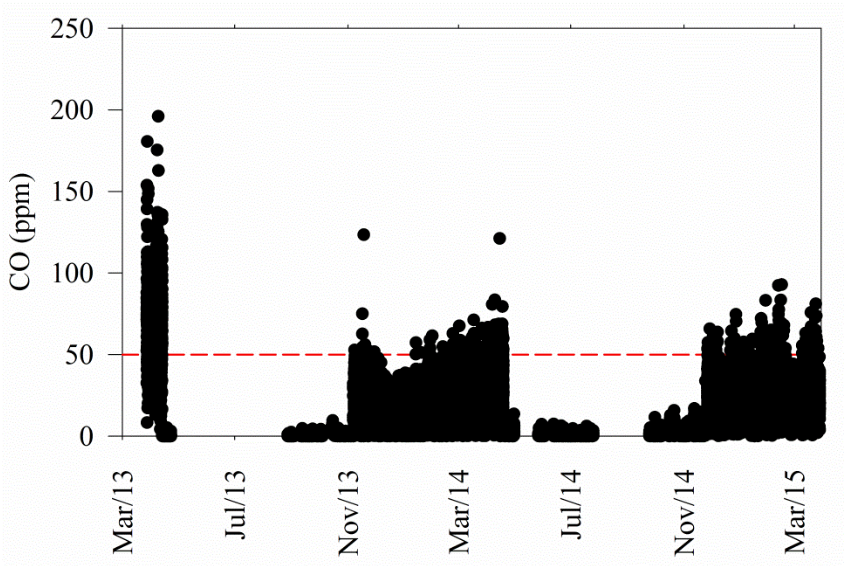



Figure 2 shows an extended time series of 8-hour values measured within the pellet storage silo at the Petrova Elementary School in Saranac Lake. Throughout the measurement period, CO concentrations often exceeded the regulatory limit of 50 ppm. Thus, from Figure 2, it can be seen that throughout the measurement period, CO concentrations often exceed the OSHA 8-hour workplace standard of 50 ppm. The CO measurements for this storage bin was classified as a confined space and appropriate measures have been taken to ensure worker safe entry into the bin whenever maintenance is required.

**Figure 2. Extended Time Series of 8-Hour Averaged Measurements in the Pellet Silo at Saranac Lake Elementary School**




Additional measurements were made in the pellet bin of the Malone Middle School through March 2015, although there were problems with the monitoring system resulting in the loss of a significant portion of data for the 2014-2015 heating season. This bin was originally the coal bin for the previous heating system. When the two pellet boilers were installed, active ventilation was installed in the pellet bin to prevent dust build-up. There was only a single period in which concentrations in excess of 50 ppm were measured. However, the indoor bin has now been replaced by an outdoor pellet storage silo similar to that used in Saranac Lake and the pellets are no longer stored inside the school building.

Overall, although concentrations did rise above ASHRAE and OSHA health-based, indoor air quality guidance levels and occupational standards of 9 and 50 ppm, respectively, none of the values rose to concentrations that presented an imminent threat of death such as the concentrations observed in Europe. It is clear that the off-gassing of CO from pellets could produce concentrations in excess of recommended limits for home occupancy or regulatory limits for worker exposure. Thus, pellet bins inside a dwelling do present a potential health threat and large capacity pellet bins in commercial applications such as schools or museums represent a worker safety issue that needs to be addressed through proper confined space procedures.

# 4 Lab Studies

To obtain a better understanding of the processes that give rise to the off-gassing of CO, laboratory studies were conducted in which small drums were partially filled with pellets and CO concentrations in air above the pellets (the headspace) were measured over time. Measurements of volatile organic compounds (VOCs) off-gassed from the pellets were also measured along with VOC concentrations in the Massena residence and in several of the larger pellet storage bins. Emissions from samples of hardwood, softwood, and hardwood/softwood blend pellets were measured. Soto-Garcia et al. (2015b) reported the results of the CO studies in an open access article (http://pubs.acs.org/doi/abs/10.1021/acs.energyfuels.5b00347).
The results of the VOC measurements are presented in Soto-Garcia et al. (2015c) in a separate open access article (http://pubs.acs.org/doi/abs/10.1021/acs.energyfuels.5b01398).

Over time CO is emitted from the wood pellets stored in bulk form, eventually rising to a constant value with the final value and speed rate of CO production depending on the pellet content, temperature and the amount of surface area. This behavior is similar to what had been previously reported in other similar designed studies. The softwood and hardwood/softwood blend produced significantly more CO than the hardwood pellets. The CO concertation curves rise to a plateau with softwood and blends producing concentrations between 600 and 1,000 ppm of CO after 20 days and the hardwood pellet curve reaching 400 ppm at that same time.

VOC emissions were also greater from the softwood pellets, with respect to both concentrations and the variety of compounds that were detected. For the hardwood pellets, the major VOC compounds were hexanal, pentanal, pentane, and methanol. These compounds could be expected to be produced through the oxidation of fatty acids such as linoleic acid. From the softwood containing pellets, additional compounds such as α-pinene and β-pinene as well as many other ketones and aldehydes were detected. Although fatty acid oxidation is clearly occurring, fatty acid content alone it cannot explain the measured CO concentrations.

Soto-Garcia et al. (2015b) have modified the kinetic model of Fan and Bi (2013) to include a surface area term. The observed exponential rise to a constant maximum strongly supports the concept of a surface limited reaction. Other experiments on ground-up pellets presented in Soto-Garcia et al. (2015b) also support the hypothesis of a surface process producing the CO. The resulting model provided a better fit for the data than the Fan and Bi model. The modeling results are presented in Table 2 in terms of emissions per unit mass of pellets per day and the total emissions of CO per unit mass of pellets.

The results of this initial study verify what has been observed in prior studies; there is less off-gassing of CO from hardwood than from softwood pellets. Blended pellets produce intermediate values. However, we do not yet fully understand the off-gassing mechanism(s) and the various oxidative processes that are giving rise to the observed CO concentration. Further work is needed to fully understand the origin of the CO in terms of what is being oxidized at room temperatures at normal room temperatures.

**Table 2. Emission Parameters Estimated from Kinetic Analysis of the Laboratory Off-Gassing Data**

| | **Initial CO emission rate (~20 ˚C, 1 atm) (moles CO/kg pellets-day)** | **Potential CO emissions (moles CO/kg pellets)** |
|---|---|---|
| Softwood #1 | 1.15 × 10-5 | 1.27 × 10-4 |
| Softwood #2 | 1.25 × 10-5 | 1.31 × 10-4 |
| Softwood #3 | 1.40 × 10-5 | 1.21 × 10-4 |
| Softwood Old (4 ºC, ~90 days) | 7.27 × 10-6 | 1.22 × 10-45 |
| Hardwood #1 | 9.80 × 10-6 | 5.16 × 10-5 |
| Hardwood #2 | 9.71 × 10-6 | 5.16 × 10-5 |
| Hardwood #3 | 8.99 × 10-6 | 5.65 × 10-5 |
| Hardwood Old (4 ºC, ~90 days) | 3.73 × 10-6 | 5.86 × 10-5 |
| Blended pellets #1 | 1.29 × 10-5 | 1.01 × 10-4 |
| Blended pellets #2 | 1.42 × 10-5 | 1.01 × 10-4 |
| Blended pellets #3 | 1.52 × 10-5 | 1.12 × 10-4 |
| Blended Old (4 ºC, ~90 days) | 7.16 × 10-6 | 5.63 × 10-5 |

# 5 Conclusions

The results of this study demonstrate that off-gassing of CO from stored pellets is sufficient to represent a hazard that needs to be adequately addressed. Although we did not measure CO at concentrations that would be immediately dangerous to life and health, or likely to produce significant short-term extreme effects in healthy adults, concentrations above the levels set as exposure guidelines in both homes and in occupational settings were clearly and repeatedly exceeded. These results do raise a real safety issue regarding in-building indoor storage of bulk pellets. Emhofer et al. (2014) investigated the temperature differential dependence of ventilation rates for a specific small-scale pellet storage system. Their work suggested that passive ventilation might be adequate in some cases. However, they recommended that during the four weeks immediately following the delivery of pellets, there be no entry into the pellet storage area. If necessary, entry should only be made after a sufficiently long period of forced ventilation to guarantee safe entry. Our recommendation is that safe entry can only be assured if the CO concentrations are measured before entry. Based on the Emhofer et al. (2014) results for a single bin design, further study will be required to assess the effectiveness of designed passive ventilation to prevent on the CO concentrations. However, active ventilation clearly reduces the average concentrations and can be achieved at relatively low costs in terms of equipment and operation.

The laboratory study shows that CO emission and $O_2$ depletion are related to a surface reaction on wood pellets, described here by the parameter of $w_{R\text{-}CO}$, fitting very well with the experimental measurements in drums, especially for CO.

Storage temperature is a stronger factor that affects the off-gassing from stored wood pellets more than the age of the pellets. Higher concentration and faster emission rates were observed with higher temperatures (more than 30 °C). For this reason, it is recommended to store pellets in a cool temperature space, or even better, at colder temperatures such as outdoor storage, which are common during the winter season in New York State. External storage facilities will always reduce the indoor exposure from off-gassed CO.

Pellets stored under high relative humidity had a faster and higher CO emission rate. For almost saturated storage conditions, the final CO concentration was about three times as high as that for a normal humidity condition. Dried wood pellets were less active in CO off-gassing compared to as-received wood pellets. This would support a recommendation that wood pellets be stored under low humidity conditions. In terms of types of pellets produced in the Northeastern U.S., hardwood pellets were observed to off gas less CO than softwood pellets, yet caution should be taken because hardwood pellets also have the tendency to absorb more moisture than softwood pellets further strengthening the recommendation for low-humidity storage. However, the off-gassing mechanism and the various oxidative processes that are giving rise to the observed CO concentrations is not yet understood. Further work is needed to fully understand the origin of the CO in terms of what is being oxidized at normal room temperatures.

The laboratory study also demonstrated that VOCs are off-gassed from stored wood pellets. The main VOCs produced due to the oxidation of stored wood pellets are: methanol, pentane, pentanal, and hexanal. Higher concentrations, but lower emission rates, were found from softwood followed by blended and hardwood pellets, respectively. Lower storage temperatures reduced the emission rates and the maximum concentrations of these gases. Also, aging of the pellets reduced the VOCs concentrations. Storage of pellets considering these factors (age and storage temperature) can be considered to reduce the emission of VOCs when the pellets are distributed to the end users.

VOC concentrations in large volume pellet storage at schools and occupational settings from this field work never exceeded appropriate Threshold Limit Value (TLV) standards, but the odor threshold for pentanal was exceeded after the fresh pellet delivery. Hexanal odor and sensory irritation levels were exceeded according to current toxicological studies, suggesting the need of improving ventilation in store rooms to mitigate short term impacts of VOCs, specifically aldehydes coming from stored wood pellets. As recommended by earlier studies, the use of ventilation, such as windows, fans, tubes that connect the store room with the outside air can reduce the concentration of these VOCs, but may not be adequate to maintain air concentrations of CO or VOCs. Further study needs to be done to examine temperature, cross-ventilation, type of storage, room volume, and other factors that will need to be considered to recommend a storage room design and proper ventilation in domestic and commercial buildings using pellet boilers.

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**toll free:** 866-NYSERDA
**local:** 518-862-1090
**fax:** 518-862-1091

info@nyserda.ny.gov
nyserda.ny.gov