FORMALDEHYDE EXPOSURE AND THE INDOOR ENVIRONMENT

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ABSTRACT

The average American spends the majority of their time indoors. Most people are aware of outdoor pollutants and the impact on their health, but many are not aware of the potential dangers in their own homes. The indoor environment is not only influenced by outdoor pollutants migrating into the indoor atmosphere, but also pollutants created indoors. Once we acknowledge that indoor air is just as important as outdoor air, we can begin to look at ways to improve guidelines, detection methods, and assessments of indoor air. The purpose of this essay is to look at formaldehyde and the indoor environment. Areas to be covered are chemical properties, applications, exposure and toxicity, health affects, indoor sources, detections methods, indoor concentrations, guidelines and remediation process of formaldehyde in the indoor environment. The public health significance of formaldehyde in the indoor environment is to protect the public from the dangers inside their homes, to improve the way formaldehyde is detected and develop prevention methods to control formaldehyde emission into our indoor environment.

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1.0 INTRODUCTION

The average American spends the majority of their time indoors. Most people are aware of outdoor pollutants and the impact on their health, but many are not aware of the potential dangers in their own homes. The Environmental Protection Agency (EPA) has indicated that indoor air pollutants can be found at higher concentration indoors then outdoors. In addition, the ability to assess and detect indoor air pollutants has lagged behind the assessment of outdoor air pollutants. The indoor environment is not only influenced by outdoor pollutants migrating into the indoor atmosphere, but also pollutants created inside the home. These pollutants are created by space heaters, cooking, or from products found inside the home. Once we acknowledge that indoor air is just as important as outdoor air, we can begin to look at ways to improve guidelines, detection methods, and assessments of indoor air. Now that we have acknowledged, that indoor air quality plays a role in our health, we can begin to look at a volatile constituent that has a sufficient impact on our indoor environment.

Formaldehyde is appropriate because it is a dominant indoor air constituent. Exposure to high concentrations of formaldehyde in the indoor environment, through inhalation, can lead to health effects. There is significant evidence that formaldehyde exposure can lead to cancer. Formaldehyde is an organic chemical that is very prevalent in our environment. It is a colorless but strong smelling gas that is commonly used in medical laboratories and mortuaries. However, formaldehyde is also found in other products such as manufactured building materials and numerous household products. Exposure to formaldehyde can affect people differently. Some people are very sensitive while others do not see a noticeable reaction to the same level of formaldehyde. As long as formaldehyde is present in the air at levels above 0.1 ppm (parts per million), acute health affects can occur.

The public health significance of formaldehyde in the indoor environment is to protect the public from the dangers inside there homes. To improve the way formaldehyde is detected and prevention methods to control formaldehyde emission into our indoor environment. The purpose of this essay is to look at formaldehyde in the indoor environment. This paper evaluated studies, peer-reviewed articles, indoor air standards and regulations to assess formaldehyde in the indoor environment. Areas to be covered are chemical properties, applications, exposure and toxicity, health affects, indoor sources, detections methods, indoor concentrations, guidelines and remediation process of formaldehyde in the indoor environment.

1.1 CHEMICAL PROPERTIES

Formaldehyde is an organic compound with the formula CH_2O, see Figure 1, and classified as a volatile organic compound (VOC). Formaldehyde is found everywhere in the environment. This is because formaldehyde is being formed by a number of human and natural sources. Natural sources include biomass combustion or decomposition, and human sources include on-site industrial emissions and fuel combustion. In addition, formaldehyde is used worldwide in many consumer products.

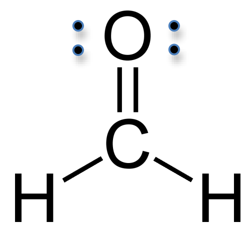

Figure 1 Structure of Formaldehyde

At room temperature, formaldehyde is a colorless, flammable gas. It has a pungent, distinct odor, and may cause burning sensation to the eyes, nose, and lungs at high concentrations. Chemical and physical properties are shown in Table 1. Formaldehyde is most commonly found in the air and is quickly broken down within hours. Formaldehyde is also found in soil and water, but the primary source of exposure is breathing air-containing formaldehyde. Indoor air usually contains higher levels of formaldehyde than outdoor air. (ASTDR, 2008)

Table 1. Chemical Properties of Formaldehyde

Chemical	Formaldehyde
Caste Number	50-00-0
Molecular Weight (g/mol)	30
Boiling Point (°C)	-19.1
Melting Point (°C)	-92
Water Solubility (mg/L)	400,000
Vapor Pressure (mm Hg)	3,890
Diffusivity in Water (cm^2/s)	1.74×10^{-5}
Diffusivity in Air (cm^2/s)	0.167
Octanol water Partition Coefficient	2.24
Henry Law Constant	3.4×10^{-7}

(RAIS, 2013)

1.2 APPLICATIONS OF FORMALDEHYDE

Formaldehyde is a chemical feedstock for many industrial processes. These feedstocks include urea-formaldehyde (UF), phenol-formaldehyde (PF), and melamine formaldehyde (MF). Figure 2 shows the industrial applications and products of formaldehyde. UF adhesives are the most commonly used in the manufacturing of wood based materials and furniture, commonly found in construction materials. About 95% of UF resin is used as a binder or adhesive in particleboard and medium fiberboard due to its low cost (Kowatsh, 2010). PF adhesive is very stable and water resistant and have a high adherence to wood. (Salthammer, Matese, & Maruzky, 2010) Approximately 60% of the PF resins are used for applications such as insulation binder, wood products, and laminates. MF resin is found in construction materials in the form of laminates and surface coatings. MF resins products are more costly and less appealing to consumers. (Kowatsh, 2010)

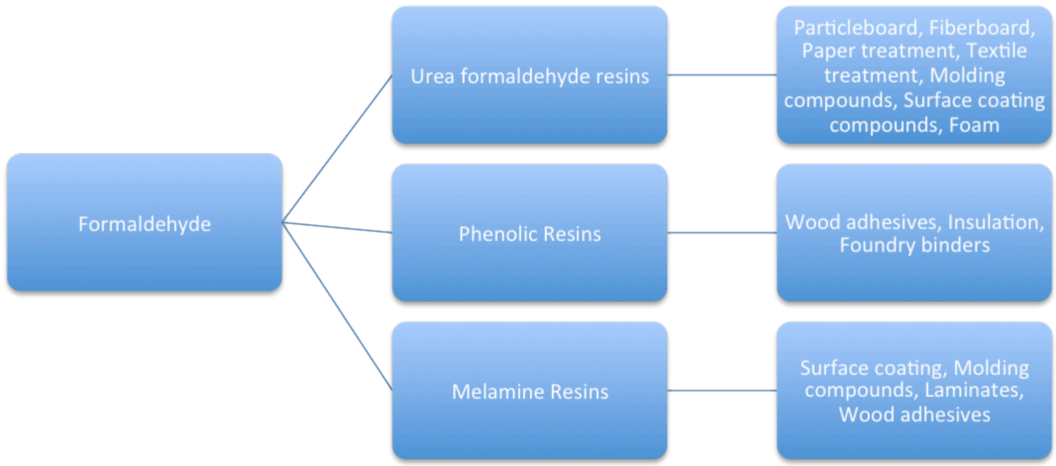

Figure 2. Industrial Applications and Products of Formaldehyde

1.2.1 INDOOR SOURCES

There are numerous sources of formaldehyde found in the indoor environment, but it is most commonly found in wood products. These pressed wood products such as particleboard, fiberboard, and plywood are major contributors to formaldehyde emissions. Emissions of formaldehyde are due to the presence of small amounts of free formaldehyde in the resin. Emissions from UF bonded particleboard may continue for months or even years, but the amount of formaldehyde emitted decreases with age. These wood products are widely used in homes for cabinetry, furniture and house construction. (Meyer & Hermanns, 1985) Flooring materials are another source of formaldehyde exposure. Formaldehyde emission from veneered and laminated wood-based products is mainly caused by adhesive and gluing.

Urea Formaldehyde Foam Insulation (UFFI) was used in the construction of homes. This thermal insulation is a product of mixing urea-formaldehyde based resin with a foaming agent (Fox, 1985). Urea-modified phenol-form-aldehyde resins are used as binders and produce lower emissions of formaldehyde. Insulation made of UF foam has higher emissions of formaldehyde. The United States banned UFFI in 1982 and is not widely used today. (Salthammer, Matese, & Maruzky, 2010)

Combustion sources found in the indoor environment are known to be strong sources of formaldehyde exposure. About half of the world's populations, especially developing countries, rely on traditional fuels (such as biomass) for their source of domestic energy. Formaldehyde is one of the primary components emitted from biomass fuels. Other indoor combustion sources include unventilated fuel burning appliances, such as gas appliances, wood stoves, and kerosene space heaters. In addition, cigarette smoke is another primary combustion source found in the indoor environment. (Salthammer, Matese, & Maruzky, Formaldehyde in the Indoor Environment, 2010)

Other indoor related sources of formaldehyde exposure are "permanent press" fabrics. These fabrics include T-shirts, pants, and shirts. This fabric has been treated with formaldehyde resins and is in close proximity to the breathing zone. Over the years, technology has improved and the amount of emissions has decreased since this fabric was introduced in the 1960s. Formaldehyde can also be found in electronic equipment, paper, fabric dyes, inks, and cosmetics can contribute to emissions. (Salthammer, Matese, & Maruzky, 2010) Table 2 summarizes sources of formaldehyde exposure in the indoor environment.

Table 2. Formaldehyde and Common Indoor Sources

Products	Examples
Permanent Press Fabrics	Clothing, lines, draperies
Flooring Materials	Floor covering (rugs linoleum, varnishes, plastics), carpet adhesive binder
Insulation	Urea formaldehyde foam insulation (UFFI)
Combustion Devices	Wood stoves, gas appliances, kerosene stoves, cigarettes
Pressed-wood products	Pressed–wood products, fiberboard, plywood, particle board, and decorative paneling
Other	Cosmetics papers, electronic equipment, fabric dyes, ink

1.2.2 HEALTH AFFECTS AND TOXICITY

Formaldehyde is a hazardous air pollutant that is found in residential buildings (California Environmental Protection Agency, 2004). Inhaling formaldehyde can cause both short-term and long-term health effects. Formaldehyde exposure arises from either inhalation or dermal contact. However, the effects of formaldehyde depend on the length of time you are exposed and an individual's sensitivity.

The primary targets of formaldehyde exposure are the lungs, eyes, noses, and mouth. Acute effects are discomfort due to odor, sensory irritation to the eyes, nose, and throat (*i.e.* via inhalation). The levels at which people can detect formaldehyde by odor varies between individuals, from 50 to 500 ppb (California Environmental Protection Agency, 2004). Moderate exposure levels of formaldehyde can cause symptoms such as, burning and itching of the eyes or nose, stuffy nose and sore throat. Chronic exposure of formaldehyde exposure can irritate the lung passageways and cause respiratory symptoms. These symptoms include coughing, wheezing, chest pains, bronchitis, and asthma. In addition, formaldehyde is a strong sensitizer, which can cause allergic reactions. (California Environmental Protection Agency, 2004) Even though, inhalation is the dominant exposure pathway, dermal contact with formaldehyde can result in skin irritation and allergic reaction.

The International Agency for Research on Cancer (IARC) recently concluded, based on human exposure in the workplace and based on animal studies that formaldehyde could cause nasopharyngeal cancer. Occupational studies have noted statistically significant association between exposure to formaldehyde and incidence of lung and nasopharyngeal cancer (EPA, 2012). This evidence is considered “limited” then “sufficient” because of the possibility of other agents contributing to the cancer risk (EPA, 2000). Formaldehyde is also known to cause tumors in the nasal cavity in rats and higher airborne concentration should increase the damage. However, due to formaldehyde being metabolized rapidly, the focus of formaldehyde epidemiology has been on cancers of the upper respiratory tract. More then 90% of inhaled formaldehyde gas is absorbed and metabolized in the upper respiratory tract (WHO, 2010). Formaldehyde exposure and links to leukemia have been reported, but lack sufficient evidence for a causal relationship between them (California Environmental Protection Agency, 2004). Based on this evidence the EPA has classified formaldehyde as a probable human carcinogen, (Group B1) (Hun, Corsi, Morandi, & Siegel, 2010). Table 3 and 4 shows toxicity values and toxic affects of formaldehyde.

Table 3. Toxicity Values for Formaldehyde

Chemical	Oral slope Factor	Inhalation Unit Risk	Chronic Oral Reference Dose	Chronic Inhalation Reference Dose	Sub chronic oral Reference Dose	Sub chronic Inhalation Reference Dose
	(mg/kg-day)$^{-1}$	(ug/m^3)$^{-1}$	mg/kg-day	mg/m^3	mg/kg-day	mg/m^3
Formaldehyde	---	1.30×10^{-5}	0.20	9.83×10^{-3}	0.30	3.68×10^{-2}

"---" – not available
(EPA, 2014) (RAIS 2013)

The reference dose (RfD) for formaldehyde, 0.2 mg/kg-day, is an estimate of a daily oral exposure in the human populations (EPA, 2014). It is not a direct estimate of risk but rather a reference point to gauge the potential for health effects. Lifetime exposure above the RfD does not mean that a health effect will occur (EPA 2012). The agency for Toxic Substance and Disease Registry (ASTDR) has established a chronic minimal risk level (MRL) of 0.003 ppm, which is based on respiratory effects in humans (EPA 2012). A MRL is an estimate of the daily human exposure that is likely to be without risk of noncancerous health effects.

The EPA estimated an inhalation unit risk of 1.3×10^{-5} (μg/m^3)$^{-1}$ (EPA, 2014). If an individual continuously breathes air contaminated with formaldehyde at an average of 0.08 μg/m^3 over their lifetime, there chance of developing cancer is one in a million (EPA, 2012).

Table 4. Cancer and Chronic Affects of Formaldehyde

Cancer Affect:	
Oral	**Inhalation**
--- [1]	Squamous cell carcinoma
Chronic Affect:	
Oral	**Inhalation**
Reduced weight gain, histopathology in rats	Increases lesions in nasal epithelium; respiratory system

(RAIS, 2013)
(EPA, 2014)
[1] Not available

2.0 DETECTION METHODS

Air sampling strategies for measuring room air components can be subdivided into short-term and long-term. Active sampling is suitable for short-term and long-term provided the airflow rates have been reduced. Air is passed through a sampling device using a pump and the air column is accurately determined. Passive sampling is used for measurements over a long period of time. This type of sampling uses a device, which is capable of taking air samples at a rate controlled by diffusion through a membrane. (Salthammer, Matese, & Maruzky, 2010)

Measuring devices for formaldehyde should be easily transported and calibrated. Derivatization methods, DNPH, chromotropic, and acetylacetone methods, are the best for the analysis of indoor air and are the most common. All have drawbacks of having long sampling times between 0.5-2 hours. (Salthammer T. , 2013)

The acetylacetone method involved the cyclization of 2,4-pentanedione, ammonium acetate, and formaldehyde to form dihydropyrine 3,5-diacetyl-1, 4-dihydrolutidine (DDL) (Salthammer T., 2013). Passing air through an absorber, which traps the formaldehyde, carries out sampling. This reaction forms DDL which is completed within 10 minutes at 40 °C. Longer sampling times of 40 minutes can be a drawback of the acetylacetone method when formaldehyde concentrations rapidly change over time. This method is recommended in Europe and Japan for the determination of formaldehyde emissions from wood based materials. The acetylacetone method is easier to use and highly sensitive for formaldehyde when combined with fluorescence spectroscopy, making it convenient for online measuring devices. Today portable instruments are available and enable reliable *in situ* measurements of formaldehyde on a manageable time scale. (Salthammer, Matese, & Maruzky, 2010)

The DNPH method is used for the simultaneous analysis of formaldehyde and other aldehydes and ketones. In acidic solutions, hydrazones are formed from DNPH by nucleophilic addition to the carbonyl group, which is followed by the elimination of water. During sampling, air is pulled through cartridges typically containing silica, and coated with an acidic solution of DNPH. This method is described in U.S. EPA Method TO-11A and ASTM D 5197 and is accepted as an international standard by International Organizational for Standardization (ISO). (Salthammer, Matese, & Maruzky, 2010)

The Chromotropic Acid Method (CAM) consists of the reaction of formaldehyde in the presence of concentrated sulfuric acid and chromotropic acid. CAM for formaldehyde is widely used in air pollution studies, especially for formaldehyde analysis because it is highly selective. Phenols, some organic substances, and strong oxidizers can cause interference. A main disadvantage of this method is the low solubility of chromotropic acid in solutions. (Salthammer, Matese, & Maruzky, 2010) However, this method has been standardized in the USA by the National Institute for Occupational Safety and Health (NIOSH) and is still an international reference method of formaldehyde analysis because of its simplicity and sensitivity. (Dar, Shafique, Anwar, Zaman, & Naseer, 2012)

DNPH, acetylacetone, and CAM methods and are the most widely used but there are other detection methods available. The Pararosaniline method causes the formation of a magenta dye when formaldehyde and pararosaniline react in the presence of sodium sulfite. This method is limited and it is susceptible to inference. In the past, this method was a widely used technique for measuring formaldehyde concentrations. The MBTH method (3-methyl-2-benzothiazolinehydrazone) is a colorimetric method for the determination of aliphatic aldehydes of low molecular weight. MBTH reacts with aldehydes to give an azide. This reaction is less sensitive than the pararosaniline method.

2.1 INDOOR AIR CONCENTRATIONS AND GUIDELINES

Over 30 million metric tons of formaldehyde represents the global worldwide consumption of formaldehyde for a variety of products (Kowatsh, 2010). Formaldehyde is one pollutant that has been found in residential indoor air at levels, which exceed occupational standards. This is due to low air exchange rates in the indoor environment. At room temperature formaldehyde can exist as a gas or solid polymer. Formaldehyde will accumulate wherever water is present in the indoor environment due to its solubility in water. (Salthammer, Matese, & Maruzky, Formaldehyde in the Indoor Environment, 2010)

Over the years regulations have concentrated more on outdoor pollutants then indoor pollutants. Formaldehyde is an air pollutant that is a direct result of our society and the need to supply food, clothing, housing, and mobility to billions of people. In our urban environment the average formaldehyde concentration is up to 40 ppb indoors, and a 15 ppb concentration of formaldehyde outdoors can be considered normal. However these “normal” levels are not to be confused with safe or acceptable levels. Today air pollution controls and technology have already helped to lower formaldehyde concentrations in highly polluted cities, such as Los Angeles. (Salthammer T. , 2013)

Formaldehyde emission rates and indoor air quality are difficult to measure because both fluctuate. Pressed wood products with UF resin (i.e. particleboard, medium-density fiberboard, and hardwood plywood) are a major sources of formaldehyde emitters. The use of pressed wood products is not supposed to decline in the future.

At present, there are no agreed upon standards for formaldehyde concentrations in residential settings. Many organizations have established occupational definitions and levels for formaldehyde. (CDC, 2009) The US Department of Housing and Urban Development (HUD) has regulated formaldehyde emissions from wood products. HUD has set the "maximum allowable concentration" at 300 ppb. WHO developed guidelines in non-occupational settings at 100 ppb for 30 minutes. The State of California Office of Health Hazard Assessment (OHHEA) established exposure levels for formaldehyde are less than typical ambient levels and they recommend a concentration of 23 ppb. (California Environmental Protection Agency, 2004)

Indoor air guidelines for formaldehyde are appropriate because indoor exposures are the dominant contributor to personal exposure (via inhalation). Indoor concentrations may be high enough to cause adverse health effects. The lowest concentration reported to cause sensory irritation of the eyes in humans is 0.36 mg/m^3 for four hours. Short-term exposure (i.e. 30 minutes) 0.1 mg/m^3 is recommended to prevent sensory irritation in the general population. This guideline will also prevent long-term health effects, including cancer. (WHO, 2010)

In older homes formaldehyde average concentrations without UFFI are well below 0.1ppm. In newer homes that have a significant amount of pressed wood products, levels can be greater than 0.3 ppm. (EPA, 2013)

Since formaldehyde can increase your risk of getting cancer, there is no known level of formaldehyde that is considered risk free. Therefore, it is recommended to reduce your formaldehyde concentration as much as possible. This will also decrease the likelihood of other irritant effects associated with long-term exposure to formaldehyde.

2.2 REMEDIATION

Ventilation is very important for controlling formaldehyde. Bringing outdoor air into your home can reduce your exposure to formaldehyde and other pollutants. This form of control is the most effective in tightly sealed buildings and is ineffective in buildings where ventilation rates are already 0.5 ACH (air exchange per hour) or higher. Formaldehyde concentrations in buildings are especially sensitive to increase in temperature. An increase in temperature of 40 degrees Fahrenheit can result in a doubling of formaldehyde concentrations. Keeping indoor temperatures moderate will help keep formaldehyde concentration low indoors. During summer months high humidity increases the emission rate of formaldehyde from pressed wood products. Keeping the humidity at about 40% to 50% indoors will also keep emission rates low.

Studies have shown a strong correlation between temperature and humidity and emission rates (Hun, Corsi, Morandi, & Siegel, 2010). Federal Emergency Management Agency (FEMA) and the Agency for Toxic Substances and Disease Registry (ATSDR) evaluated 96 unoccupied trailers. These trailers were similar to the trailers used to house people affected by Hurricane Katrina. Results showed that during baseline sampling formaldehyde levels averaged 1.04 ppm. In addition, running the air conditioner formaldehyde levels dropped to an average of 0.39 ppm and with open windows formaldehyde levels dropped to an average of 0.09 ppm. A summary of these results is shown in Table 5. These results show an association between room temperature and formaldehyde levels; formaldehyde levels increase as room temperature increase (*i.e*. seen in trailers with closed windows). (ASTDR, 2006)

Table 5. EPA Sampling of FEMA Temporary Housing Trailers

Test Condition	Mean (ppm)	Minimum (ppm)	Maximum (ppm)
Baseline	1.04	0.01	3.66
Air Conditioning	0.39	0.00	1.63
Windows Open	0.009	0.01	0.49

(ASTDR, 2006)

The most effective control method is to eliminate the emission source completely. Customer complaints have shown that levels of formaldehyde above 0.1 ppm are usually due to improperly installed or defective products. Sometimes, an isolated panel is usually the source for high emissions.

In the past, formaldehyde emission was tackled with remedial treatment of buildings with chemicals. One of these methods was fumigating homes with anhydrous ammonia or concentrated ammonia solutions. However, this method was not reliable because it was difficult to saturate an entire home with a toxic vapor. Other methods consist of painting wood products with vapor barrier paints or preventing emissions by controlling the wood at the manufacturing stage. Over the past few years progress has been made in the manufacturing of pressed wood products with reduced emissions. In 1984 more then 50% of the U.S manufacturing UF bonded wood products has decreased emissions in mobile homes (Meyer & Hermanns, 1985).

3.0 CONCLUSIONS

Among the many number of indoor pollutants, formaldehyde has always had a high position as an indoor air pollutant and has been extensively studied throughout the years. Formaldehyde is a highly reactive aldehyde, it is an important chemical in industry, and it can be found indoors and outdoors due to natural and human processes. Every few years' discussions on formaldehyde take place, for example in 2004 following the classification of formaldehyde as a Group 1 carcinogen. Also more recently, survivors of Hurricane Katrina suffered adverse health effects from poor indoor air quality from the trailers they were living in.

Many sources contribute to formaldehyde concentrations in residential homes because of the variety of building materials and insulation. Insulation with formaldehyde is not used in the United States and mostly found in other countries. Building materials such as wood materials, laminates, carpets, paints, and varnishes are common source of formaldehyde exposure. Newer homes have higher emission rates of formaldehyde then older homes from these products dues to emissions rates decreasing with age. Mobile homes are known for having a high formaldehyde concentration. This is usually due to these homes having more wood based materials and a lower exchange rate. This issue came to the forefront after Hurricane Katrina and caused an increase in studies looking at indoor air quality.

Health effects of indoor air pollution have received relatively little attention from the scientific community. Today the bulk of public concern continues to be directed to the health impacts of outdoor air. Most people perceive their risk for poor air quality is higher outdoors then indoors. However, the majority of people spend most of their time inside a building than outside. For example the average US resident spends 88% of their day inside a building, 7% inside a vehicle, and only 5% spent outside (Jones, 1999). Over the decades many changes have occurred in the way buildings are constructed. Modifications in building designs have been driven by the need to increase energy efficiency. Modern homes are better insulted and have lower air exchange rates. Other modifications are advances in construction technology have lead to a greater use of synthetic building materials. Therefore, buildings are more comfortable but have provided an environment in which airborne pollutants are produced and build up a higher concentration then before. (Jones, 1999) Advancing technologies have been developed for measuring outdoor constituents and are not suitable for indoor use.

The average American is spending more time inside then outside. More concentration needs to be on indoor air quality then outdoor air. Recent surveys have shown that emission rates have gone down over the years due to the progress made in indoor air quality.

As mentioned earlier, formaldehyde is classified as a Group 1 carcinogen by IARC. There are many discussions over this classification and resulted in other organizations re-evaluating the data. This has resulted in different countries and organizations having different guidelines and standards. Currently, many countries and organization have new and different ways to evaluate indoor air quality to create standards and guidelines. There are a number of indoor related formaldehyde guidelines worldwide but different organization apply different criteria. Most standards are based on epidemiological and toxicological studies. These values fall into one of two categories; occupational and residential environments. Formaldehyde has been linked to nasopharyngeal cancer and possibly leukemia. The World Health Organization

(WHO) recommends a short-term (*i.e.* 30 minute) guideline of 0.1 mg/m^3 for preventing sensory irritation in the general population.

Today, no one will doubt that formaldehyde is a relevant indoor air pollutant. Building materials play an important role in contributing to formaldehyde emissions indoors but we should consider other sources such as outdoor air, cooking, heaters, etc. Many studies have been conducted in many different countries, in all different types of indoor environments. Formaldehyde concentrations depend on the age of the building. New homes are often associated with new building materials. New building materials have a higher emission rate and decrease with age. It is possible to produce building materials that have a low emission rate but air exchange rates have decreased over the years. Reliable tools are available to measure formaldehyde concentrations even at low levels in rural environments. Evaluations of recent emission studies and indoor air surveys have shown that the situation has improved due to the advancements made over the decades. As improvements in formaldehyde emissions have improved, air exchanges rates in homes have decreased. For the time being formaldehyde and its impact on indoor air quality is still be up for discussion.

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