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ORIGINAL ARTICLE – Forest Products Science and Technology

Evaluation of the Emission of Formaldehyde from Wood-Based Panels (MDFs and MDPs) in Brazil After Use

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Abstract

The rising popularity of various types of wood panels and the advances in manufacturing technology have increased the uses of wood. The bonding of fibers and particles to form panels, however, involves impregnation with resins, such as phenolic adhesives, which delivery desirable quality to the panels due to their insolubility in water, resistance against heat among others, but are also recognized as carcinogenic to humans (IARC, 2004). This study involved measuring the levels of formaldehyde, remaining in wood panels after use and disposal. It was initially evaluated new panels and then examined degraded ones. The results demonstrated significant volatilization of formaldehyde during the degradation process of the panels, with losses of 74.83% for MDF and 85.71% for medium density particleboards (MDP). The results demonstrated significant volatilization of formaldehyde during the degradation process of the panels, with losses of 74.83% for MDF and 85.71% for medium density particleboards (MDP).

Keywords: Wood-products, wood adhesives, wood degradation, volatilization of chemical compounds.

1. INTRODUCTION AND OBJECTIVES

In recent decades, the international market for wood products has been characterized by increasing demand for use of certified wood from planted forests, in line with the generally heightened concern over sustainability in all productive sectors. In this respect, Canada and the United States intend to obtain certification of their urban forests by 2022 as part of the Sustainable Forestry Initiative (SFI, 2021), while China, India, Indonesia and Holland have all sought evaluation of trees outside forests as part of the Programme for the Endorsement of Forest Certification (PEFC, 2021b).

On the matter of certification in the wood sector, the standouts are the Forest Stewardship Council (FSC) and the Programme for the Endorsement of Forest Certification (PEFC), both headquartered in the European Union and operating globally. Besides the impressive size of the certified forest areas worldwide, the obtainment of dual certification is increasing. In 2019 there was an 8% increase in these areas in relation to the previous year, corresponding to an additional 93 million hectares. Consumer demand and the corresponding behavior of the markets have certainly contributed both to dual certification and other inefficiencies, and relevant organizations and governments have been working together to reduce conflicts and form alliances that can effectively protect native forests and promote sustainability to assure human survival (Fernholz, 2021) In Indonesia and Malaysia, although the main certification systems (FSC and PEFC) are active, few if any dual certifications have been obtained (Depoorter & Marx, 2021)

The importance of forest certifications cannot be denied, but since they aggregate value to wood, caution is required regarding the possibility of corruption and abuse, especially greenwashing through seals of approval (Conniff, 2018).

Brazil, which is also an important site of forest products has achieved significant progress in certifying forest activities, but there is still much room for growth (Sanqueta, 2022). This certification has multiple advantages, including contributing to environmental education, stimulating respect for the laws and regulations, and promoting sustainable consumption, all of which are global trends (Almeida, 2008).

The quality seals and certificates of environmental responsibility are always welcome, because they indicate a search for excellence, but these conquests cannot be mistaken for the pursuit of easy profits or failure to consider the solid wastes generated from the use of wood.

Despite the development of new plastics and other building materials, there is still strong demand for wood in civil construction. This has motivated research and rapid development of new products, such as cross-laminated timber (CLT), also known as the "concrete of the future", which was introduced in Europe in the 1990s, but whose production in Brazil only began in 2012 (Oliveira, 2018). It is estimated that global capacity to produce this product, which has good acceptance in the market and helps reduce the amount of waste, will reach 4 million m^3 per year by 2025. At present, the main producers are located in Europe (48%), North America (43%), Oceania (6%) and Asia (3%) (Forest Business Network, 2020).

The market for wood is expanding. In particular, the total production of wood panels in Europe was 71.6 million m^3 in 2020, with the main use of the panels being to make furniture, accounting for 47% of output (UNECE/FAO, 2021). In Brazil, the sale and consumption of reconstituted wood panels in the same year reached 7.2 million m^3 (IBÁ, 2021).

Among wood products, reconstituted wood panels stand out, with use in many industrial segments, especially MDF (Mantanis et al., 2018). In 2020, global production of 367 million m^3 of wood was allocated to make panels of various types (FAO, 2020).

The production of wood panels uses raw materials in different stages of disaggregation (veneers, fibers or particles), which are bonded with resins and other components, according to the case, under high temperature and pressure. In this context, Brazil stands out for its plentiful supply of wood from its forests due to the favorable characteristics of its ecosystems (Valverde, 2012). This is especially the case of planted forests of pine and eucalyptus in the South and Southeast regions (Freire et al., 2015).

When resins based on urea-formaldehyde, phenol-formaldehyde, melamine-formaldehyde or urea-formaldehyde-melamine which have a wide industrial interest are used to produce panels, the wastes cannot be employed as fertilizer, poultry bedding or energy generation (Pereira, 2005).

As a biomaterial with minimal influence on climate change, wood is a preferred raw material to compose many new products, such as cross laminated timber (CLT), as demonstrated by the large number of research articles published on the theme. The generation of residues has attracted commensurate attention from public authorities in Brazil, leading to the issuance of the technical standards ABNT NBR 15.316–2 (2019), regarding medium density fiberboards, and ABNT NBR 14.810–2 (2018), regarding medium density particleboards, with part 2 respectively specifying the testing requirements and methods and establishing limits for emission of formaldehyde from panels without coating (interval between 8 mg/100 g and 20 mg/100 g).

In Europe, EN 120-1 covers the emission of formaldehyde from MDF, classified in two levels, the lower one being up to 9 mg/100 g (BS EN, 1992), while the corresponding German standard establishes limits of 6.5 mg/100 g for particleboards and 7 mg/100 g for fiberboards (Roffael, 2006). These are lower than the limits in Brazil and the rest of Europe, but all countries' regulations allow the use of formaldehyde in the resins utilized to produce wood-based panels.

Although the World Health Organization has indicated that exposure to formaldehyde occurs in many indoor workplaces (Who, 2010) and there is no US legislation classifying formaldehyde as a carcinogen, starting in 2016 many Italian companies began keeping track of workers' exposure to that volatile component, paying heed to the IARC classification and the decision of the National Toxicology Commission (Scarselli, 2017).

In Brazil, a regulatory standard issued by the Labor Ministry deals with unhealthy working conditions and activities in general. It establishes a maximum working period of 48 hours a week for employees exposed to formaldehyde at a level up to 1.6 p.p.m. or 2.3 mg/m^3, with exceeding hours being paid at a higher wage rate (NR 15, 1978).

According to Trein (2015), formaldehyde is classified as a component with class I danger, with potential risks to the environment and public health, so that wastes from wood-based panels must be treated properly before disposal.

Resins based on formaldehyde are widely used for production of wood-based panels, mainly because of their low cost and good performance, with rapid curing. Nevertheless, consideration should be given to the existing regulatory standards (IARC, 2004 and NBR 10.004 – 2004 from ABNT). The second document deals with the classification of solid wastes in Brazil and classifies formaldehyde as hazardous, also stipulating that any solid residue must be classified according to its most damaging substance.

The European Union has established a program for ecological labeling of furniture produced with panels, establishing formaldehyde limits for medium density particleboards (MDP) of 62 mg/m^3 and for medium density fiberboards (MDF) of 81 mg/m^3 (European Commission, 2016).

In Brazil, where this research was developed, the law 12.305/2010 established the National Policy on Solid Wastes. Its Article 30 covers the shared responsibility of the life cycle of products, attributing it individually and in series to manufacturers, importers, distributors, merchants, consumers and providers of public services for urban cleaning and management of solid wastes. There is academic engagement in the search for sustainable adhesives due to environmental restrictions, given that lignin has proven to be a promising raw material, although there are still challenges for its use on an industrial scale. (SOUZA et al., 2020)

The fortuitous discovery of "green glue", whose formula does not contain solvents and/or other compounds prejudicial to health (Gouveia, 2019); the use of citric acid as binder, contributing to reduce environmental impacts (Lee et al., 2020); and the use of wood wastes as sources of bioresins to produce circular construction materials (Silva, et al., 2021), indicate the efforts undertaken to find sustainable solutions for the woodworking industry.

The objective of this study was to analyze the volatilization behavior of formaldehyde, used Besides new panels (in large scale to make bonding resins for medium density fiberboards (MDF) and medium density particleboards (MDP) (Lessmann, 2008; Ribaski, 2018). No studies were found that measured the release of formaldehyde levels throughout the degradation process, similarly to what was proposed by this research, denoting the pioneering nature of this initiative and the importance of the results. Commercial reference, it was analyzed panels undergoing degradation, illegally discarded elsewhere, since formaldehyde is considered a hazardous volatile component according to international standards such as EN 13986:2004, Wood-based panels for use in construction - Characteristics, evaluation of conformity and marking; ANSI 208.1:2016 from the American National Standards Institute, and the Japanese standard JIS A 5908 & 5905.

2. MATERIALS AND METHODS

2.1. Study area

This study was conducted in the city of Rio de Janeiro, Brazil's second most populous, which has two bays, two coves, five lagoons, 240 rivers and streams, a long sand spit and a large urban forest (IBGE, 2022).

As samples, it was obtained degraded wood panels from the municipal solid waste collection system, at the treatment or transfer units, as well as used panels illegally dumped in Campo Grande, the district of the city with the largest area and population.

2.2. Specification of the new and used panels and the collection points

The two types of panels evaluated are widely used by industries in the Brazilian wood segment and in the international market, mainly in the furniture industry in general. Medium density fiberboard (MDF) is a panel of wood fibers from fast-growing species, obtained from reforestation with fast production cycles. The raw material is made up of a set of physically and mechanically disaggregated wood fibers, generating excellent use of the wood, discarding the tree bark, leaves, branches and roots, this yield being much higher than the production of species native sawn wood. The medium density particleboard (MDP) panel has the particularity of being formed by larger "particles" than those used in the MDF panel. MDP is a technological development of particleboard panels used a few decades ago. Both types of panels require adhesives and pressing in industrial plants and can be covered with melamine coatings that provide different aesthetic and finishing aspects, colored or resembling patterns of wood species "in natura", which is very well accepted by the consumer market. Both MDF and MDP, in conditions of internal use, isolated from humidity or from contact with water, can have great durability, over 20 years, but in conditions of high humidity, or exposure to the external environment, the degradation of these panels is accelerated, and its durability reduced.

New medium density fiberboard (MDF) and medium density particleboard (MDP) panels, the two types consumed the most, were obtained from stores and served as controls in the experiments.

Regarding the used and degraded/discarded panels, we collected three samples of each of the two types of panels studied (MDF and MDP), it is not possible to know the exposure time or the degradation conditions to which the panels were subjected. These samples were collected at four different points. Two were in the district of Campo Grande (Av. Brasil point 1 – beside the road in front of number 41.100, and Av. Brasil point 2 – on the corner with Estrada do Tingui, along the margin of Canal do Melo). The third and fourth points were respectively in the district of Bangu (Bangu Waste Transfer Station– ETR Bangu) and Santa Cruz (Santa Cruz Waste Transfer Station – ETR Santa Cruz). Each sample collected, with different shapes and number of pieces, was sufficient to fill a plastic bag with capacity of 30 liters.

From each location/sample, it was generated 20 subsamples with dimensions 25 mm in length, 25 mm in width and thickness of 8 mm.

It was not rare to find multiple samples of the same type of panel at the same place. Because of the impossibility of determining the date of manufacture, the time at the disposal point and the weather conditions to which the materials were exposed, we applied a preliminary visual classification, subject to posterior measurement of the levels of formaldehyde in the laboratory to corroborate or belie the initial classification. Then it was adopted the following designations of the materials collected according to the visual classification of the level of degradation: new material - control (L0); low degradation level (L1); medium degradation level (L2); and high degradation level (L3). Figures 1 and 2 show samples of the specimens from the evaluated panels.

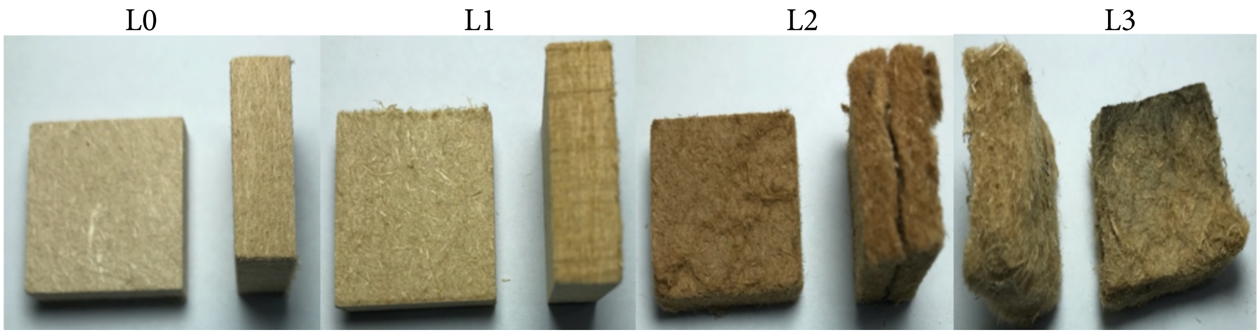

Figure 1. Samples of MDF medium density fiberboards evaluated: new and degradation levels 1, 2 and 3.

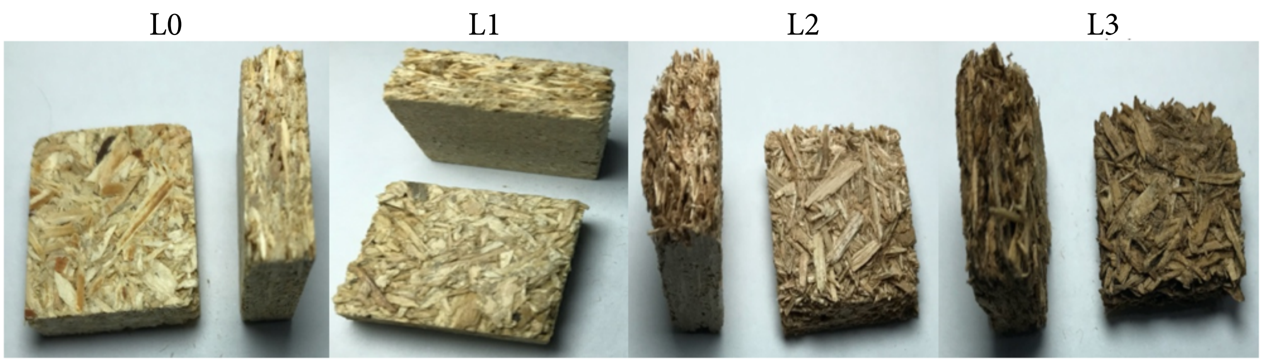

Figure 2. Samples of MDP medium density particleboards evaluated: new and degradation levels 1, 2 and 3.

2.3. Laboratory tests

The subsamples were prepared in the laboratory according to the European Standard EN 717-3:1997. The 20 subsamples of each material considered generated repetitions of the tests, characterizing them as "compound samples". After standardization, the subsamples were placed in plastic bags, hermetically sealed, labeled and stored in the laboratory. A total of 20 grams of each material was used in the assays to detect emission of formaldehyde, utilizing 7 to 12 subsamples of each material, to characterize the repetitions of each evaluation.

All the subsamples were kept for 15 days in an outside covered area at ambient temperature for acclimation, and then they were weighed. After this, the subsamples were placed in a forced-air oven at constant temperature of 105° C for 6 hours. The dried subsamples were again weighed to obtain the average moisture content of each type/place, according to the formula [(moist weight – dry weight) *100/ dry weight].

The release of formaldehyde was measured by the "flask method", according to the European Standard EN 717-3:1997, which establishes that test specimens with known mass of up to 20 grams will be secured approximately 40 mm above water in a hermetically sealed flask, kept at constant temperature. The formaldehyde released is absorbed by the water and subsequently measured by acetyl acetone extraction spectrophotometry, with the result expressed as mg/kg of dry panel material.

In the assays, it was used 11 polypropylene flasks with height of 15 cm and capacity of 500 ml, with lids of the same material. The mouth of each flask had an opening that allowed passage of the set of samples, with a base measuring 25 mm x 25 mm.

The lids of the flasks had holes drilled in the center with a 1/8" bit, permitting fixation of a hook with length of 5 cm made of stainless steel with equal caliber, threaded for secure attachment to the flasks, with two stainless steel washers and two rubber rings, to provide the necessary water tightness during the tests.

To measure the release of formaldehyde, we used deionized water, ammonium acetate, glass stirring rods, analytic scale, 500 ml beakers, 1000 ml round-bottom volumetric flasks with lids, 1000 ml amber glass flasks with lids and toggles, 500 ml Erlenmeyer flasks with 10 ml pipettes, and aluminum foil to reduce clarity. It was used a UV spectrophotometer to measure the absorbance values of each mixture designated in a control spreadsheet, permitting calculation of the concentration of formaldehyde in the mixture, representing the formaldehyde volatilized from the samples of each type of degraded panel.

2.4. Statistical analysis

Aiming the data analysis, it was compared the mean levels of formaldehyde emissions according to the types of panels, levels of degradation and places of sampling.

In the statistical interpretation did not consider the results found for new panels. These data were used as control, with discrepant values in relation to degraded panels, as described in the following items. The data were analyzed by applying the Scott-Knott test for comparison of the means, carried out with the Assistat version 7.7 beta statistical package (Silva & Azevedo, 2016). The Scott-Knott test is a test for univariate analysis, therefore, more indicated when there is only one variable under study, and the analyzed variable was formaldehyde volatilization; the great advantage of this test is the absence of ambiguity present in multiple comparison procedures.

3. RESULTS AND DISCUSSION

The results of formaldehyde release are presented in Tables 1 to 3 and depicted graphically in Figure 3, regarding the averages of the samples of each type of panel, level of degradation and place of collection/sampling, as well as their interactions.

Table 1. Results obtained from the evaluations of formaldehyde emission from MDF panels*.

Emission of Formaldehyde (mg/kg dry panel)	L0 (test.)	L1	L2	L3
General average by level of degradation	10.37 (0,007)	2.87 (2,651)	2.92 (1,709)	2.04 (0,862)
Emission of Formaldehyde (mg/kg dry panel)	**Av. Brasil 1**	**Av. Brasil 2**	**ETR Bangu**	**ETR Santa Cruz**
General average by place of collection	1.96 (0,781)	4.99 (1,760)	2.10 (1,095)	1.25 (0,326)
Average emission of formaldehyde by place of collection and level of apparent degradation	**Av. Brasil 1**	**Av. Brasil 2**	**ETR Bangu**	**ETR Santa Cruz**
L1	2.72	6.67	1.17	0.94
L2	2.01	5.14	3.31	1.23
L3	1.16	3.16	1.84	1.59

* Values in parentheses indicate the standard deviation of the data. In the data from the intersection between "location" and "level of degradation", some data presented a single measurement, thus, in these, the standard deviation has not been calculated.

Table 2. Results obtained from the evaluations of formaldehyde emission from MDP panels**.

Emission of Formaldehyde (mg/kg dry panel)	L0 (test.)	L1	L2	L3
General average by level of degradation	19.45 (1,605)	2.14 (1,043)	4.70 (2,151)	1.51 (0,682)
Emission of Formaldehyde (mg/kg dry panel)	**Av. Brasil 1**	**Av. Brasil 2**	**ETR Bangu**	**ETR Santa Cruz**
General average by place of collection	2.23 (1,148)	2.49 (2,699)	1.70 (0,891)	1.88 (0,646)
Average emission of formaldehyde by place of collection and level of apparent degradation	**Av. Brasil 1**	**Av. Brasil 2**	**ETR Bangu**	**ETR Santa Cruz**
L 1	3.17	0.76	2.66	1.98
L 2	2.57	5.60	0.90	1.19
L 3	0.95	1.11	1.54	2.47

** Values in parentheses indicate the standard deviation of the data. In the data from the intersection between "location" and "level of degradation", some data presented a single measurement, thus, in these, the standard deviation has not been calculated.

Table 3. Statistical analysis of comparison of the means by the Scott-Knott test, disregarding the values of control samples***.

Emission of Formaldehyde (mg/kg dry panel)			MDF	MDP
General average of panels sampled			2.58 a (1,772)	2.08 a (1,374)
Emission of Formaldehyde (mg/kg dry panel)		**L1 (low)**	**L2 (medium)**	**L3 (high)**
General average by level of degradation		2.51 a (1,905)	2.74 a (1,808)	1.73 a (0,754)
Emission of Formaldehyde (mg/kg dry panel)	**Av. Brasil 1**	**Av. Brasil 2**	**ETR Bangu**	**ETR Santa Cruz**
General average by place of collection	2.10 b (0,890)	3.74 a (2,726)	1.90 b (0,869)	1.57 b (0,572)

*** Means followed by the same letter do not differ statistically according to the Scott-Knott test at 5% probability. Values in parentheses indicate the standard deviation of the data.

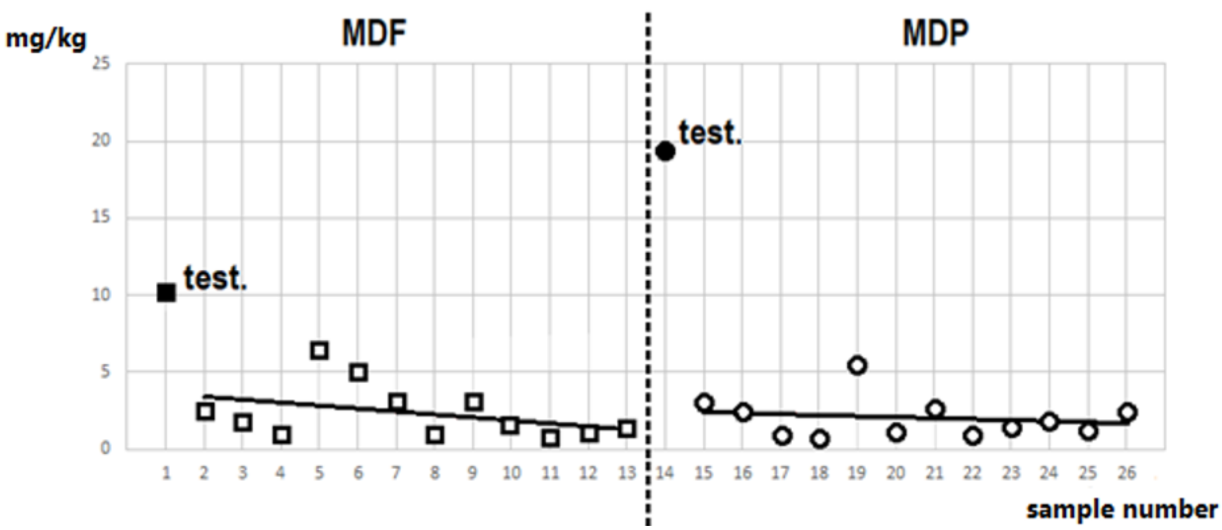

Figure 3. Dispersion values of formaldehyde emissions (mg/kg) found in the samples of MDF and MDP, including the equation of the trend line.

The control samples of new panels, of the MDF and MDP types, class II, acquired in the market, at the end of the evaluation regarding the measurement of the emission of formaldehyde levels in mg/kg of dry panel, respectively indicated the values of 10.37 mg /kg and 19.45 mg/kg (Table 1 and 2). The Brazilian standards that establish the emission limits of formaldehyde levels, by the perforator method, for the types of panels, (ABNT NBR 15.316-2:2019 and ABNT NBR 14.810-2:2018) set the same emission limit for the class E I, (up to ≤8 mg/100g - corresponding to 80 mg/kg) as well as for Class E II panels. (>8 mg/100 g to ≤ 20 mg/100 - corresponding to >80 mg/kg to ≤ 200 mg/kg). There are few current international references for a comparison of results; in the work of the authors Chew and Ong (Che & Ong, 1989) for panels made in Malaysia, the authors found very variable values for particleboards, ranging from 45 to 106 mg/100g by the perforator method.

Another one is the work of He et al. (2012) which they also evaluated the emission of formaldehyde release, but at different production stages of wood-based panels, and they also reported that little information had been published in his respect. The authors described a direct correlation between the quantity of formaldehyde used to produce the panels (especially the use of resins based on urea-formaldehyde) and the rates of formaldehyde emission, finding a linear relationship.

In our study, the visual degradation of the MDF panels, classified as L2 (medium degradation level) and L3 (high degradation level), in the two sampling places, had higher volatilization of formaldehyde than those classified as L1 (low degradation level). However, it was assumed that local factors might have influenced the level of formaldehyde emission found in the panels. In any event, the values did not differ significantly from a statistical standpoint.

In contrast with the formaldehyde emission result found in the control sample (new MDF panels), of 10.37 mg/kg, the values for discarded MDF panels ranged from 0.94 mg/kg to 6.67 mg/kg. Therefore, formaldehyde is a substance that will be released from MDF panels along their lifetime. According to the data obtained in this study, the values can reach 9% of the total, which was the largest difference encountered between the control sample and the discarded material with lowest formaldehyde emission.

Of particular note is that even in situations of high degradation, the MDF panels still contained most of their original formaldehyde at the time of disposal, indicating concern over such disposal in sanitary landfills or reuse along with other materials.

With respect to the MDP panels, the control samples had higher average formaldehyde emission, 19.45 mg/kg, versus a range for discarded panels of 0.90 mg/kg to 5.60 mg/kg. Therefore, the emission of formaldehyde during the life cycle of MDP was smaller than the figure for MDF panels, meaning that the MDP panels retained more formaldehyde during their life cycle.

The results found were obtained from samples collected in conditions without reference data from discarded panels after use. As stated by Zhang et al. (2018), various factors can influence the emission of formaldehyde from wood-based panels. These factors go beyond the type of panel, and include aspects like wood species, type of adhesive, temperature, moisture and velocity of the wind to which panels are exposed, as well as the quantity of resins and variables of the production process.

4. CONCLUSIONS

The samples of new panels (commercial reference) of the MDF, class II, and MDP types submitted to laboratory testing by the flask method, contained levels of remaining formaldehyde lower than the maximum limits specified in the European Standard (EN 13986:2004) as well as the technical standards adopted in Brazil (MDF of 10.37 mg/kg and MDP of 19.45 mg/kg). However, it is necessary to study post-use alternatives that consider the residual formaldehyde in the panels, even after long use cycles and high degradation levels. Common practices for wood wastes, encouraged in some locales, such as composting, production of biomass and mulching, among others, can result in high toxicity due to the presence of residues in panels such as MDF and MDP.

Further research is necessary to evaluate the levels of formaldehyde in wood-based panels immediately after production, since formaldehyde volatilizes continuously during the life cycle, as found in in the various degraded materials examined in this study.

There is no doubt that all the formaldehyde employed in bonding resins will eventually be present in the human environment. For this reason, researchers and the public at large need to exert pressure for the substitution of hazardous bonding resins by sustainable and ecologically correct substances.

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