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Waste apple wood: A safe and economical alternative substrate for the cultivation of *Pleurotus ostreatus* and *Lentinula edodes*

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ABSTRACT

The use of waste apple-wood as a source of sawdust to cultivate the mushrooms *Pleurotus ostreatus* and *Lentinula edodes* is a common practice, but it is imperative to ensure that the wood does not contain unsafe amounts of heavy-metals or pesticide residues. In this study, we sampled and investigated the pollution of heavy metals and pesticide residues in apple-wood from Yantai, Shandong, China and cultivated *P. ostreatus* and *L. edodes* using apple-wood as substrate. Heavy metals, pesticide residues, mineral elements, and biological efficiency were measured. Heavy metals were more commonly detected in the 73 apple-wood samples, but serious pollution was only an isolated phenomenon. No Pb was detected in *P. ostreatus* and *L. edodes*. The contents of Hg, As, Cd, and Cr were at safe levels. The contents of Ni were equivalent to those of wild mushrooms. Most notably, chlorpyrifos was detected in all the apple-wood tested. However, chlorpyrifos was only detected in *L. edodes* cultivated with apple sawdust. No other pesticide residues were detected in the other mushroom samples. The biological efficiency of *P. ostreatus* cultivated by apple sawdust was 89%, which was 80% of the control. The biological efficiency of *L. edodes* cultivated with apple sawdust was 81%, which did not differ significantly from the control. Apple-wood can replace wild oak as the material for *L. edodes* cultivation, but producers should ensure that the raw materials are safe. The main materials chosen to cultivate *P. ostreatus* should balance the two factors of raw material price and biological efficiency.

Keywords: apple wood, biological efficiency, heavy metals, mineral elements, pesticide residues

INTRODUCTION

Shandong Province is an important area of production for the export of apples in China, comprising one-third of the country's export volume. Yantai City in Shandong Province is also known as "Apple City". By 2019, the total area of apple cultivation in Yantai had reached 129,500 ha, and production accounted for 60% of the province (http://tjj.shandong.gov.cn/tjnj/nj2020/zk/indexce.htm). The existing apple trees in Shandong Province were primarily planted in the late 1980s and 1990s, and the problem of the orchards ageing has become a serious issue. The ageing of orchards in Qixia City of Yantai is more serious, as 76.55% of the orchards are >16 years old. The economic value of apple trees is primarily reflected in their fruit. As the

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apple trees age, the fruit quality will decline, and the old trees will be eliminated. The cycle of renewal of apple trees is typically approximately 10 years. With the transformation and upgrading of old apple orchards, the improvement in quality and efficiency and the annual pruning of apple trees, the main apple producing areas have accumulated many resources of apple wood. Many old apple trees have been felled in recent years in Yantai City. When calculated using an elimination rate of 10%, the annual felled apple trees and pruned apple branches amounted to 1.7 million tons. Felled old apple wood and pruned apple branches are currently primarily burned as fuel, and their consumption is limited. Owing to the difficulty of transportation, the apple branches are piled up and left to naturally rot, which easily causes the proliferation of diseases and insect pests. It also pollutes the environment, resulting in a substantial waste of resources. It is urgent to seek a higher value for apple wood and its large-scale utilisation. However, apple tree resources differ from wild forest resources. Fertiliser application and pesticide use in the growing areas for >10 years have acidified a large amount of soil and activated metal ions in the soil, leading to metal poisoning of some trees that can be highly serious (Zhanling et al., 2020), such as manganese. The contents of copper ions in the environment increased yearly after Bordeaux mixture was used (Norman, 1920; Li et al., 2005), and this also accumulates in the apple tree. Heavy metals are not degradable and easily accumulate in organisms through the food chain (Kopp et al., 2018). The evaluation of heavy metal enrichment and pesticide residues in discarded apple wood is a key factor to consider about recycling apple wood resources.

Currently, waste apple wood is used to make charcoal and composite materials (Jindo et al., 2014; Kowaluk et al., 2019; Najibeh et al., 2020). A broader use of these materials is the cultivation of edible mushrooms. As early as 1996, Jo et al. (1996) added 15% of apple wood chips to the medium for *Flammulina velutipes* and enhanced the yield by 9%; the economic efficiency increased by 29%. Angela et al. (2021) also trimmed fruit trees, such as peach, pear and apple, and mixed them with hay as a matrix to cultivate *Pleurotus ostreatus*. Li et al. (2001) used apple sawdust to cultivate *Lentinula edodes*, and the yield and quality of the fruit bodies were better than those cultivated on oak (*Quercus glandulifera* BL) wood. Of course, in addition to the use of waste fruit wood to cultivate edible mushrooms, many other types of industrial and agricultural wastes are used to cultivate edible mushrooms, such as the cultivation of *Pleurotus eryngii* on cotton waste and that of *P. ostreatus* on tea or paper waste (Kulshreshtha et al., 2013; Yang et al., 2016; Sardar et al., 2017). Whether cultivating edible mushrooms with abandoned apple wood or other industrial and agricultural waste, most researchers focus on the biological efficiency and the nutrients and economic benefits, rather than on the safety of waste itself.

Mushrooms can become enriched with heavy metals, and there are some health risks associated with the excessive consumption of mushrooms that contain large quantities of heavy metals (Mohsen et al., 2021). Li et al. (2011) found that by artificially adding Pd to the medium, the mycelia of *Panus gigantea* could reach the hyperaccumulation level of Pd (1,125.56 mg · kg^{-1} dry weight). Oskar et al. (2021) collected wild mushrooms from seven different areas in Poland and found that the Cd and Cu in edible mushrooms generally exceeded the standard level. Yu et al. (2021) analysed the heavy metal content of artificially cultivated *L. edodes* from 27 counties of 14 provinces in China and found that the contents of As and Cd in some samples exceeded the standard level. Furthermore, they studied the content of Cd in the substrate in more detail and found that the content of Cd in the substrate had an important impact on the fruiting bodies. Based on the accumulation of heavy metals in mushrooms, it is particularly important to examine edible mushrooms cultivated on industrial and agricultural wastes to ensure that they are safe.

This study investigated the resources of waste apple wood in Yantai, Shandong Province, China's main area for the production of apples and examined the contents of heavy metals and pesticide residue. Waste apple wood as the main raw material enables the cultivation of two wood-rotting edible mushrooms – *P. ostreatus* and *L. edodes* – that are grown on a large scale. Pesticide residues, heavy metal elements and other mineral elements in *P. ostreatus* and *L. edodes* were also detected. The purpose of this study is to produce safe and reliable edible mushroom products while recycling waste apple wood resources.

MATERIALS AND METHODS

Strains and materials

P. ostreatus (LD0003) and *L. edodes* (LD0040) strains were preserved in the Key Laboratory of Edible Fungus Technology of College of Agriculture, Ludong University (Yantai, China).

The materials for the cultivation of edible mushrooms, including apple sawdust, oak sawdust, corn cob, cottonseed husk and wheat bran, were all purchased from QixiaXurui Biological Technology Co., Ltd (Yantai, China).

Collection and pretreatment of apple wood samples

A total of 21 sampling areas were established in the main apple producing areas (Qixia, Penglai, Haiyang and Muping) in Yantai, Shandong Province, including 13 sampling areas in Qixia, 3 in Penglai, 2 in Haiyang and 3 in Muping (Figure 1). A total of 73 apple wood samples were collected. The collected samples were left to dry in the sun and then cut into thin pieces with a profile cutting machine. A few pieces were randomly selected based on the size of the apple wood. The small

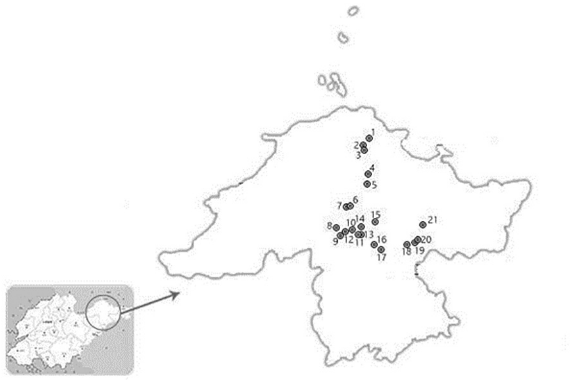

Figure 1. The global positioning system (GPS) localisation map of the sampling region.

branches were cut short by pruning. The sliced apple wood or short branches were placed in a Petri dish in a 60°C-blast dryer fan and baked until a constant weight, crushed and then sifted through a 0.5 mm sieve.

Cultivation of P. ostreatus and L. edodes

P. ostreatus and *L. edodes* were cultivated with apple wood sawdust as the main materials (Table 1). Formulae 2 and 4 were the best formulae for *P. ostreatus* and *L. edodes* cultivated with apple sawdust. *P. ostreatus* was also cultivated with cottonseed husk as the control (Formula 1), while *L. edodes* was cultivated with oak (*Quercus mongolica*) wood sawdust (Formula 3) as the control. Three replicates were established for each recipe (50 bags per group). The biological efficiency of each formula (fresh weight of fruiting body/dry weight of culture material) was calculated.

Table 1. Formulations for *Pleurotus ostreatus* and *Lentinus edodes*.

Number	Cultivated species	Formula
1 (CK)	*P. ostreatus*	Cottonseed husk 80%, corn cob 3%, wheat bran 15%, gypsum 2% and water content 60%
2	*P. ostreatus*	Apple sawdust 50%, cotton seed husk 20%, corn cob 20%, wheat bran 8%, gypsum 2% and water content 60%
3 (CK)	*L. edodes*	Oak sawdust 80%, wheat bran 18%, gypsum 2% and water content 60%
4	*L. edodes*	Apple sawdust 80%, wheat bran 18%, gypsum 2% and water content 60%

CK, control check.

Detection of pesticide residues, heavy metals and other mineral elements

The microwave digestion method was used to pretreat samples to detect heavy metals and other mineral elements (Siwulski et al., 2017; Falandysz and Treu, 2019). The digestion tube was cleaned according to the manufacturer's instructions (Multiwave3000; Austria Antompa Co., Ltd, Shanghai, China). A total of 0.3 g sample (0.3 g apple wood and cultivated raw materials and 0.2 g *P. ostreatus* and *L. edodes* powder) was added to the digestion tube; 6 ml (*P. ostreatus* and *L. edodes*, 4 ml) of concentrated HNO_3 was added; the cover was opened and the sample was soaked in the fume cupboard overnight. Next, it was placed in the microwave digestion instrument for digestion, heated to 130 °C over a 10-min period, incubated at this temperature for 5 min and then heated to 180 °C over a 10-min period, at this temperature for 30 min, and then the temperature was lowered to 60 °C. After the digestion, the digestion tube was opened and placed into an acid discharge meter (140 °C). After it had drained completely, 1 ml of concentrated HNO_3 was added to dissolve it, and then the volume was fixed to 50 ml with pure water. A total of 8–9 ml of supernatant was placed in a centrifugal tube and measured using a Prodigy full-spectrum direct-read inductively coupled plasma (ICP) emission spectrometer (Teledyne Leeman Labs, Hudson, NH, USA). The content of Hg was measured using 'GB5009.17-2014 National Food Safety Standard – Determination of Total Mercury and Organic Mercury in Food', while that of As was measured using 'GB5009.11-2014 National Food Safety Standard–Determination of Total Arsenic and Inorganic Arsenic in Food'. Se was determined as described by 'GB5009.93-2017 National Food Safety Standard – Determination of Selenium in Food'.

A total of 179 pesticide residues were detected in 21 apple wood samples. The samples were pretreated as described by 'GB23200.113-2018 National Food Safety Standard – Determination of 208 Pesticides and Metabolites Residues in Foods of Plant Origin – Gas chromatography – Tandem Mass Spectrometry Method' and 'GB/T 20770-2008 Determination of 486 Pesticides and Related Chemical Residues in Grains – Liquid chromatography-tandem mass spectrometry (LC-MS-MS) Method'. Gas chromatography-mass spectrometry (GCMS-TQ8040 NX, Shimadzu, Tokyo, Japan) and liquid chromatography-tandem mass spectrometry (6460 Triple Quad LC/MS; Agilent Technologies, Santa Clara, CA, USA) were used to measure the pesticide residues.

Data analysis

Microsoft Excel 2007 (Redmond, WA, USA) and SPSS 17.0 (SPSS, Inc., Chicago, IL, USA) were used for data processing and statistical analysis, respectively. A Pearson's correlation coefficient was used for the correlation analysis. The contents of pesticide residues and heavy metals were calculated by dry weight ($mg \cdot kg^{-1}$ dw). The results represent the means of three replicates ± SD ($n = 3$). In the significance analysis, * and ** represent significant differences at $p < 0.05$ and $p < 0.01$, respectively.

RESULTS

Analysis of heavy metal contents in apple wood samples

The contents of 10 heavy metals (Pb, Cd, Hg, As, Cr, Ni, Zn, Fe, Mn and Cu) in the 73 apple wood samples collected were measured (Table 2).

Cd was not detected in 20 of the samples, and the detection rate was 72.60%. Six samples did not have detectable Hg, and its detection rate was 91.78%. Ten samples lacked detectable As, and the detection rate was 86.30%. Two samples lacked Cr, which was detected in 97.26% of the samples. All the samples contained Pb, Ni, Zn, Fe, Mn and Cu (Table 2).

The average contents of Pb, Cd, Hg, As, Cr, Ni, Zn, Fe, Mn and Cu were 2.37 mg · kg^{-1}, 0.20 mg · kg^{-1}, 0.057 mg · kg^{-1}, 0.60 mg · kg^{-1}, 3.49 mg · kg^{-1}, 4.73 mg · kg^{-1}, 18.55 mg · kg^{-1}, 486.61 mg · kg^{-1}, 84.73 mg · kg^{-1} and 34.38 mg · kg^{-1}, respectively. The inter-sample standard deviation and variation coefficient of each element were large, which reflected the large differences between samples (Table 2). The contents of heavy metals in the top five apple wood samples are also shown in Table 3. Some of the samples had extremely high levels of contamination, indicating that contamination is an individual phenomenon.

As shown in Table 4, the maximum positive correlation between the contents of Pb and Fe occurred in the 73 apple wood samples, with a similarity coefficient as high as 0.988. The contents of Pb, Fe and Cu positively correlate with each other and are not correlated with the contents of As, Cd and Ni, indicating the joint accumulation of Pb, Fe and Cu in apple wood.

Analysis of pesticide residues in apple wood samples

Among the 179 pesticide residues detected, five types, including thifensulfuron-methyl, rimsulfuron, nicosulfuron, iprodione and benfuracarb, were not reported because the quality control results did not meet the requirements. Among the remaining 174 pesticide residues, only 11 types of pesticide residues, including chlorpyrifos, cyhalothrin, cypermethrin, difenoconazole, tebuconazole, chlorbenzuron, carbendazim, imidacloprid, acetamiprid, cymoxanil and prochloraz were detected, while the remaining 163 pesticide residues were not detected (Table S1 in Supplementary Materials).

Chlorpyrifos was detected in all the apple wood samples, with the content ranging from 0.01 mg · kg^{-1} to 0.75 mg · kg^{-1}. Chlorpyrifos is an insecticide that is widely used in apple cultivation (Ho et al., 2020), which explains why all the samples contained detectable amounts of chlorpyrifos.

Carbendazim was detected in 19 samples and ranged from 0.015 mg · kg^{-1} to 4.86 mg · kg^{-1}, and the detection rate was 90.48%. Tebuconazole was detected in 16 samples ranging from 0.017 mg · kg^{-1} to 0.44 mg · kg^{-1}, and the detection rate was 76.19%. Imidacloprid was detected in 14 samples ranging from 0.031 mg · kg^{-1} to 0.32 mg · kg^{-1}, and the detection rate was 66.67%. Acetamiprid was detected in 13 samples ranging from 0.01 mg · kg^{-1} to 0.11 mg ·kg^{-1}, and the detection rate was 61.90%. Chlorbenzuron was detected in eight samples and ranged from 0.037 mg · kg^{-1} to 0.21 mg · kg$_{-}^{1}$ and the detection rate was 38.10%. Only cyhalothrin was detected in two samples at 0.14 mg · kg^{-1} and 0.19 mg ·kg^{-1}, respectively; cypermethrin was detected in two samples at 0.11 mg · kg^{-1} and 0.19 mg · kg^{-1}, respectively; difenoconazole was detected in two samples at concentrations of 0.034 mg · kg^{-1} and 0.083 mg · kg^{-1}, respectively, cymoxanil was detected in two samples at concentrations of 0.056 mg · kg^{-1} and 0.083 mg · kg^{-1}, respectively. Their detection rate was 9.52%. Prochloraz was detected in one sample at a content of 0.04 mg · kg^{-1}, and the detection rate was 4.76% (Table 5, Table S2 in Supplementary Materials).

Analysis of heavy metal elements and other mineral elements in the raw materials

The main nutrient in apple sawdust is crude fibre, which has low contents of proteins and lipids and almost no carbohydrates. It can be used as a carbon source to cultivate wood rot edible mushrooms

Table 2. Analysis of the contents of heavy metal elements in apple wood samples.

Heavy metal	Detection rate (%)	Detection range (mg · kg^{-1})	Average (mg · kg^{-1})	Standard deviation	Coefficient of variation (%)
Pb	100	0.14–41.51	2.37	5.15	217.26
Cd	72.60	0.08–2.23	0.20	0.31	159.79
Hg	91.78	1.4–0.0031	0.057	0.17	292.96
As	86.30	0.17–6.14	0.60	0.75	125.91
Cr	97.26	0.12–16.12	3.49	2.75	78.70
Ni	100	1.30–21.82	4.73	3.29	69.45
Zn	100	1.12–115.15	18.55	19.48	105.02
Fe	100	107–8,709	486.61	1,081.55	222.26
Mn	100	20.3–204.62	84.73	43.93	51.85
Cu	100	5.97–235.93	34.38	34.80	101.21

Table 3. Top five contents of each heavy metal element in apple wood samples (mg · kg^{-1}).

Ranking	Pb		Cd		Hg		As		Cr		Ni		Zn		Fe		Mn		Cu	
	Sample number	Content	Sample number	Content	Sample number	Content	Sample number	Content	Sample number	Content	Sample number	Content	Sample number	Content	Sample number	Content	Sample number	Content	Sample number	Content
1	27	41.51	57	2.23	32	1.4	69	6.14	19	16.12	4	21.82	57	115.15	27	8,708.92	38	204.62	19	235.93
2	19	18.48	36	1.05	38	0.25	57	1.99	46	12.58	49	14.02	41	81.62	19	3,963.58	41	188.95	46	134.28
3	32	7.03	41	0.90	19	0.23	2	1.51	27	9.70	72	12.78	27	61.07	4	1,251.47	43	183.50	34	126.23
4	16	3.81	56	0.68	60	0.22	36	1.4	4	8.82	33	11.48	19	58.07	32	1,105.65	44	180.38	32	107.42
5	4	3.16	24	0.60	69	0.19	27	1.10	32	8.80	64	10.50	36	56.03	46	641.30	66	176.62	73	90.45

(unpublished). However, apple wood is agricultural waste and may have heavy metal pollution. Therefore, the raw materials were analysed for the presence of heavy metals and other mineral elements. The contents of As (0.43 ± 0.19 mg · kg^{-1}) and Hg (0.021 ± 0.0082 mg · kg^{-1}) in apple sawdust was higher than those in other cultivation materials, which could be caused by the accumulation of As and Hg in the tree owing to the long-term use of some pesticides in apple orchards (Ian et al., 1994). The Mn content in the sawdust of wild oak was as high as 198.80 ± 33.24 mg · kg^{-1}, and the content of Pb (0.90 ± 0.19 mg · kg^{-1}) was relatively high. The Ni content of corn cob (61.45 ± 10.58 mg · kg^{-1}) was significantly higher than that in other cultivation materials. In terms of other mineral elements, the content of P (11,736.02 ± 2,074.40 mg · kg^{-1}) in bran was much higher than that of sawdust, cottonseed husks and corn cobs, and the content of Ca in sawdust was relatively high (Table 6).

Analysis of pesticide residues in raw materials

A total of 179 pesticide residues were tested for each cultivation material. Chlorpyrifos (0.56 ± 0.075 mg · kg^{-1}), acetamiprid (0.16 ± 0.020 mg · kg^{-1}) and phoxim (0.053 ± 0.0047 mg · kg^{-1}) were only detected in apple sawdust. Phoxim (0.058 ± 0.0047 mg · kg^{-1}) was detected in bran, and pesticide residues were not detected in the other culture materials (Table 7).

Analysis of heavy metal elements and other mineral elements in P. ostreatus and L. edodes

The contents of heavy metals and other mineral elements (Pb, Cd, Hg, As, Cr, Ni, Zn, Fe, Mn, Cu, P, K, S, Ca, Mg and Se) in the fruiting body samples of *P. ostreatus* and *L. edodes* were analysed. Based on the 'GB 2762-2017National Food Safety Standard – Limits of Contaminants in Food' and the 'NY/T 749-2018 Green food – Edible Mushroom', Pb (fresh edible mushrooms 1.0 mg · kg^{-1}), Cd (fresh edible mushrooms 0.2 mg · kg^{-1}, *L. edodes* 0.5 mg · kg^{-1}), Hg (fresh edible mushrooms and its products 0.1 mg · kg^{-1}) and As (edible mushrooms and its products 0.5 mg · kg^{-1}) limited the contents as evaluation criteria. Pb was not detected in any sample. The contents of Cd, Hg and As were measured by their dry weight. Utilising the 'NY/T 749-2018', the contents of Cd, Hg and As in all the samples did not exceed the standard.

The contents of Zn and Fe in *P. ostreatus* cultivated in apple sawdust were higher than those of the control group, reaching 75.47 ± 5.03 mg · kg^{-1} and 92.35 ± 6.26 mg · kg^{-1} (Table 8). The World Health Organization (WHO) and the Food and Agricultural Organization (FAO) recommend a daily Reference Nutrient Intake (RNI) of zinc for men and women of 14 mg and 10 mg per capita per day, respectively. The requirements for teenagers are even higher. The reference intake of Fe for adult men is 13.7 mg · day^{-1}, and the reference intake for women is even higher, reaching 29.5 mg · day^{-1} (WHO and FAO, 2004). In addition, a lack of these mineral

Table 4. Correlation analysis of the content of each element in apple wood samples.

Element	Pb	Cd	Hg	As	Cr	Ni	Zn	Fe	Mn
Cd	0.031								
Hg	0.156	0.080							
As	0.059	0.386**	0.133						
Cr	0.530**	0.124	0.336**	−0.001					
Ni	0.056	−0.090	0.054	−0.076	0.378**				
Zn	0.361**	0.733**	0.162	0.207	0.446**	0.203			
Fe	0.988**	0.050	0.118	0.065	0.560**	0.112	0.394**		
Mn	0.249*	0.125	0.235*	0.211	0.171	0.050	0.216	0.243*	
Cu	0.346**	0.104	0.452**	−0.027	0.626**	0.025	0.292*	0.342**	0.250*

*$p < 0.05$ (two-tailed); **$p < 0.01$ (two-tailed).

Table 5. Analysis of the contents of pesticide residues in apple wood samples.

Pesticide residues	Detection rate (%)	Detection range (mg · kg^{-1})	Average (mg · kg^{-1})	Standard deviation	Coefficient of variation (%)
Chlorpyrifos	100	0.01–0.75	0.16	0.19	119.64
Cyhalothrin	9.52	0.14–0.19	0.17	0.04	21.43
Cypermethrin	9.52	0.11–0.19	0.15	0.06	37.71
Chlorbenzuron	38.10	0.037–0.21	0.12	0.07	59.87
Carbendazim	90.48	0.015–4.86	0.70	1.12	157.31
Imidacloprid	66.67	0.031–0.32	0.10	0.08	76.69
Acetamiprid	61.90	0.01–0.11	0.04	0.03	76.88
Difenoconazole	9.52	0.034–0.083	0.06	0.03	59.23
Tebuconazole	76.19	0.017–0.44	0.13	0.13	97.43
Cymoxanil	9.52	0.056–0.083	0.07	0.02	27.47
Prochloraz	4.76	0.04	0.04		

Table 6. The content of heavy metal elements and other mineral elements in culture material samples (mg · kg^{-1} dry weight).

Element	Apple sawdust	Oak sawdust	Cottonseed husk	Corn cob	Wheat bran
Pb	ND	0.90 ± 0.19	ND	0.15 ± 0.05	ND
Cd	0.26 ± 0.20	0.23 ± 0.04	0.04 ± 0.01	0.07 ± 0.02	0.05 ± 0.02
Hg	0.021 ± 0.0082	0.014 ± 0.0045	0.0038 ± 0.00035	0.019 ± 0.003	0.0054 ± 0.0016
As	0.43 ± 0.19	0.16 ± 0.025	0.24 ± 0.046	0.27 ± 0.11	0.16 ± 0.037
Cr	2.00 ± 0.71	3.20 ± 0.26	1.89 ± 0.19	1.82 ± 0.12	1.99 ± 1.02
Ni	11.42 ± 1.22	19.02 ± 0.34	3.40 ± 1.30	61.45 ± 10.58	11.27 ± 4.23
Zn	36.56 ± 2.76	12.17 ± 1.50	29.17 ± 1.40	46.34 ± 13.76	95.55 ± 15.54
Fe	469.52 ± 47.26	534.98 ± 46.71	133.18 ± 2.36	707.34 ± 26.48	389.38 ± 117.62
Mn	56.45 ± 10.76	198.80 ± 33.24	43.43 ± 2.88	70.44 ± 6.63	151.94 ± 21.69
Cu	42.37 ± 12.00	30.76 ± 2.18	22.04 ± 2.08	102.79 ± 29.97	30.02 ± 7.20
P	285.89 ± 74.33	421.20 ± 11.30	965.73 ± 194.83	1,892.39 ± 245.57	11,736.02 ± 2,074.40
K	2,976.74 ± 491.72	996.00 ± 63.02	9,184.94 ± 514.20	7,001.17 ± 1,652.27	12,811.04 ± 1,902.13
S	1,011.27 ± 202.44	1,375.46 ± 34.10	1,453.87 ± 163.89	2,933.34 ± 272.59	4,044.84 ± 441.75
Ca	11,220.94 ± 2,918.93	18,263.25 ± 445.73	3,370.71 ± 263.35	3,097.61 ± 599.40	1,476.53 ± 264.57
Mg	1,120.56 ± 169.58	1,241.33 ± 282.32	1,721.86 ± 376.95	1,166.53 ± 252.63	4,234.88 ± 492.44
Se	0.030 ± 0.0058	ND	0.043 ± 0.0087	0.032 ± 0.0036	0.0364 ± 0.011

ND: not detected.

elements is recognised as "hidden hunger" (Gregory et al., 2017). *P. ostreatus* and *L. edodes* cultivated with apple sawdust can be used as good dietary sources of Zn and Fe. Therefore, based on the contents of heavy metals and other mineral elements, discarded apple sawdust can completely replace wild oak wood and cotton seed husks (Table 8).

Pesticide residue detection in P. ostreatus and L. edodes

Fruiting body samples of *P. ostreatus* and *L. edodes* were analysed for 179 pesticide residues. Chlorpyrifos was only detected in *L. edodes* of formula 4, and the national standard does not limit the amount of residues in edible mushrooms. However, the European Union's limit of chlorpyrifos in mushrooms is 0.01 mg · kg^{-1} fresh weight (EU, 2021). In this study, the chlorpyrifos content of *L. edodes* cultivated in the formula 4 was 0.12 ± 0.025 mg · kg^{-1} dry weight, which merits our attention. No pesticide residues were detected in other samples. The detection of chlorpyrifos may be related to residues in the apple sawdust. Chlorpyrifos primarily controls *Eriosoma lanigerum*. Owing to serious infestations of *E. lanigerum*, all fruit farmers use chlorpyrifos. The amount of apple sawdust in formula 4 reached 81%. Therefore, the pesticide residues in the apple wood itself may have a greater impact on the pesticide residues in the fruit bodies of *L. edodes*.

Comparison of the biological efficiency of edible mushrooms cultivated with apple sawdust and other materials

Apple sawdust and oak sawdust were used as the main materials to cultivate *L. edodes*. The biological efficiency of the three flushes of mushroom cultivation was approximately 80%, and the difference in yield was not significant (Table 9). However, the use of apple sawdust and cottonseed husk as the main materials to cultivate *P. ostreatus* resulted in significantly different yields. Among them, the biological efficiency of *P. ostreatus* cultivated with cottonseed husk can reach 112%, while the biological efficiency of *P. ostreatus* with apple sawdust was only 90% (Table 9). Therefore,

Table 7. Pesticide residue contents of culture raw material samples (mg · kg^{-1} dry weight).

Pesticide residues	Apple sawdust	Oak sawdust	Cottonseed husk	Corn cobs	Wheat bran
Chlorpyrifos	0.56 ± 0.075	ND	ND	ND	ND
Acetamiprid	0.16 ± 0.020	ND	ND	ND	ND
Phoxim	0.053 ± 0.0047	ND	ND	ND	0.058 ± 0.0047

ND, not detected.

Table 8. Contents of heavy metal elements and other mineral elements in *Pleurotus ostreatus* and *Lentinus edodes* (mg · kg^{-1} dry weight).

	P. ostreatus		*L. edodes*	
	Formula 1 (CK)	Formula 2	Formula 3 (CK)	Formula 4
Pb	ND	ND	ND	ND
Cd	0.11 ± 0.04	0.17 ± 0.02	1.06 ± 0.12	0.19 ± 0.03
Hg	0.12 ± 0.03	0.033 ± 0.0024	0.018 ± 0.0028	0.028667 ± 0.0045
As	0.52 ± 0.035	0.059 ± 0.010	0.48 ± 0.035	0.72 ± 0.094
Cr	ND	0.60 ± 0.06	0.86 ± 0.19	1.34 ± 0.27
Ni	0.16 ± 0.05	1.62 ± 0.24	3.00 ± 0.36	9.29 ± 0.78
Zn	65.95 ± 8.50	75.47 ± 5.03	108.43 ± 7.97	90.22 ± 8.63
Fe	69.18 ± 16.13	92.35 ± 6.26	95.47 ± 10.40	51.51 ± 7.86
Mn	8.03 ± 0.30	7.25 ± 1.42	16.83 ± 3.15	14.89 ± 2.64
Cu	12.63 ± 0.86	8.47 ± 2.20	26.03 ± 4.69	13.35 ± 2.44
P	9,244.25 ± 1,133.32	9,275.98 ± 697.41	9,737.44 ± 1,277.03	9,470.33 ± 658.03
K	16,654.00 ± 2,968.71	18,212.00 ± 936.16	18,507.33 ± 2,236.68	11,271.36 ± 2,829.37
S	4,037.00 ± 249.92	4,186.00 ± 75.45	24,796.33 ± 4,636.61	16,388.44 ± 2,085.69
Ca	178.24 ± 17.15	132.01 ± 10.57	908.50 ± 61.21	851.67 ± 31.13
Mg	1,115.00 ± 148.58	1,463.33 ± 97.01	1,479.67 ± 320.01	889.33 ± 38.44
Se	0.028 ± 0.0061	0.162 ± 0.034	0.096 ± 0.013	0.0605 ± 0.012

ND, not detected.

Table 9. Comparison of the biological efficiency of edible mushrooms cultivated with apple sawdust and other materials.

	Pleurotus ostreatus		*Lentinus edodes*	
	Formula 1 (CK)	Formula 2	Formula 3 (CK)	Formula 4
Biological efficiency %	112.17 ± 4.23**	89.70 ± 2.04	80.79 ± 1.89	81.62 ± 1.40

CK, control check.
**$p < 0.01$ (two-tailed).

the choice of ingredients to cultivate *P. ostreatus* must balance the two factors of price and yield.

DISCUSSION

Previous studies on the use of agricultural waste to cultivate edible mushrooms focused on yield or nutritional content. For example, Patricia et al. (2014) used the invasive aquatic plant water hyacinth (*Eichhornia crassipes*) to cultivate *P. ostreatus*. Elahe et al. (2016) used different agricultural wastes to cultivate *P. eryngii* and examined the fresh and dry weight, protein content and other indicators of mushroom. Most researchers who study pesticide residues focus on those used in mushroom cultivation, such as famoxadone, trifloxystrobin, beta-cypermethrin, pyriproxyfen, avermectin and diflubenzuron. However, less attention has been paid to whether pesticide residues from the raw materials used to cultivate them accumulate in the mushrooms (Coward et al., 2006; Du et al., 2018). In this study, the apple wood in the sampling area was tested for heavy metals and pesticide residues, and there was indeed some degree of heavy metal and pesticide residue pollution in the apple wood. However, overall, the pollution was not serious, and there were only a few cases in which the pollution was serious. Therefore, during the process of using apple wood or other industrial and agricultural wastes to produce edible mushrooms, the safety of the culture material must be evaluated first. Pollution of edible mushrooms owing to heavy metal and pesticide residue pollution must be curbed from the source.

Cr and Ni are not specified in the Chinese national standard. However, the European Food Safety Authority proposed a maximum daily intake of 0.0028 mg · kg^{-1} body weight for Ni (Benford et al., 2015; Oskar et al., 2021). Thus, the maximum weekly intake of Ni was 0.0196 mg · kg^{-1} body weight. Calculated based on an average weight of 60 kg, the safe content of Ni per person per week is 1.176 mg. The content of Ni in this study was calculated based on the dry weight of the mushrooms and assuming an average moisture content of 90%. The mushrooms with the highest amount of Ni (9.29 ± 0.78 mg · kg^{-1} dry weight) when fresh were approximately 0.929 mg · kg^{-1}. Based on data from the National Bureau of Statistics of China (http://www.stats.gov.cn/tjsj/ndsj/2020/indexch.htm), the average annual per capita intake of vegetables and edible mushrooms in China is 98.6 kg, which is 1.89 kg · week^{-1}. The consumption of *L. edodes* in China is far less than half of the total amount of vegetables and edible mushrooms. Therefore, the content of Ni is also safe. The maximum recommended daily intake of Cr is not provided, but the content of Cr (0.6–1.34 mg · kg^{-1}) in this study is similar to that reported in wild edible mushrooms (0.9–1.4 mg · kg^{-1}) (Giannaccini et al., 2012). Compared with the prices of other main materials for edible mushroom cultivation, apple sawdust is highly cost-effective. The price of apple sawdust (approximately 94 USD per ton) is lower than that of oak sawdust (approximately 125 USD per ton). In addition, in recent years, there has been a shortage of resources, and the price of cultivation materials for edible mushrooms has been increasing. For example, the price of cottonseed husk can reach 376 USD per ton depending on the season. When *P. ostreatus* and *L. edodes* are cultivated with apple sawdust as the main material, their pesticide residues and heavy metal contents do not exceed the standard. There is no significant difference in the biological efficiency of *L. edodes* cultivated with apple sawdust and oak sawdust. Apple sawdust can be used instead of oak sawdust for *L. edodes* to produce fruiting bodies. Ivarsson et al. (2021) cultivated *P. ostreatus* with faba bean (*Vicia faba* L.) hulls as the main raw material, and the biological efficiency reached 109 ± 28%. Sorina et al. (2016) cultivated *P. ostreatus* using coffee grounds, and the biological efficiency reached 97%. Although the biological efficiency of *P. ostreatus* cultivated with apple sawdust is only 81% of that of cottonseed husk cultivation, considering the cost of raw materials, apple sawdust can be used to partially replace cottonseed husk during cultivation. The addition of apple wood resources stabilises the price system of the main raw materials for edible mushroom cultivation while also stabilising the costs of edible mushroom cultivation. It can not only recycle agricultural waste but also protect wild oak wood resources.

This study only used waste apple sawdust to cultivate two edible mushrooms, *P. ostreatus* and *L. edodes*, and did not cultivate more types of edible mushrooms. Chlorpyrifos was detected in *L. edodes*, which merits special attention. It is worth noting that mushrooms are known to specifically accumulate heavy metals. For example, *Agaricus* can accumulate higher concentrations of Cd, *F. velutipes* can specifically accumulate Pb and *Ganoderma lingzhi* can accumulate As and Cd (Stijve and Besson, 1976; Wong et al., 2007; Huang et al., 2008; Zhu et al., 2014). They further show that special attention should be paid to evaluating the safety of raw materials when using waste apple wood to cultivate edible mushrooms.

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AUTHOR CONTRIBUTIONS

G.Y. collected and analysed the data and drafted the manuscript. X.L. collected samples and analysed the data. S.Z. and S.S. collected the apple wood and analysed contents of pesticide residues and heavy metals. Y.Y. collected the cultivated raw materials and analysed contents of pesticide residues and heavy metals. J.C. collected *Pleurotus ostreatus* and *Lentinus edodes* samples and analysed contents of pesticide residues and heavy metals. W.L. and X.C. designed the study, guided the research work in each step, and wrote and finalised the manuscript.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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SUPPLEMENTARY MATERIALS

Table S1. List of pesticide residues not detected.

Numbers	Pesticide residues	Numbers	Pesticide residues
1	Atrazine	85	Fenpropimorph
2	Azinphos-methyl	86	Fenthion
3	Azoxystrobin	87	Fenvalerate
4	Benalaxyl	88	Fipronil
5	Bendiocarb	89	Flucythrinate
6	Benfluralin	90	Flufenoxuron
7	Benfuracarb	91	Flusilazole
8	Benoxacor	92	Fluvalinate
9	Bifenthrin	93	Furathiocarb
10	Boscalid	94	HCH-gamma
11	Bromopropylate	95	Imazalil
12	Buprofezin	96	Indoxacarb
13	Butachlor	97	Isocarbophos
14	Cadusafos	98	Isofenphos
15	Carbofuran	99	Isoprocarb
16	Chlorfenvinphos	100	Isoprothiolane
17	Chlordane	101	Kresoxim Methy l
18	Chlorfenapyr	102	Malathion
19	Chlorpropham	103	Metalaxyl
20	Chlorpyrifos Methyl	104	Metamitron
21	Cyanazine	105	Methamidophos
22	Cyfluthrin	106	Tolclofos-methyl
23	Cyprodinil	107	Triadimefon
24	DDD (p,p’)	108	Triadimenol
25	DDE (p,p’)	109	Triazophos
26	DDT (o,p’)	110	Triflumizole
27	DDT (p,p’)	111	Trifluralin
28	Deltamethrin	112	Vamidothion
29	Diazinon	113	Vinclozolin
30	Dichlorvos	114	Hexythiazox
31	Dicloran	115	Linuron
32	Dicofol	116	2-phenyl-phenol
33	Diethofencarb	117	Bupirimate
34	Methidathion	118	DDD (o,p’)
35	Metolachlor	119	DDE (o,p’)
36	Mevinphos	120	Isoproturon
37	Myclobutanil	121	Quintozane
38	Napropamide	122	Tetrachlorvinphos
39	Nitrothal-isopropyl	123	Tetradifon
40	Oxadixyl	124	Tolylfluanid
41	Oxadiazon	125	Butocarboxim
42	Paclobutrazol	126	Heptenophos
43	Parathion	127	Acephate
44	Penconazole	128	Aldicarb

(*Continued*)

Table S1. Continued.

Numbers	Pesticide residues	Numbers	Pesticide residues
45	Pendimethalin	129	Aldicarb-sulfoxide
46	Permethrin	130	Aldoxycarb
47	Phenthoate	131	Bensulfuron-methyl
48	Phorate	132	Captan
49	Phosalone	133	Carbaryl
50	Phosmet	134	Carbofuran-3-hydroxy
51	Phoxim	135	Clethodim
52	Pirimicarb	136	Cyromazine
53	Pirimiphos-methyl	137	Dichlofluanid
54	Procymidone	138	Emamectin benzoate(Et)
55	Profenofos	139	Ethiofencarb
56	Promecarb	140	Fenhexamid
57	Prometryn	141	Fluazifop-p-butyl
58	Propamocarb	142	Iprodione
59	Propargite	143	Iprovalicarb
60	Propham	144	Isofenphos-methyl
61	Propiconazole	145	Methiocarb
62	Propoxur	146	Methomyl
63	Propyzamide	147	Methoxyfenozide
64	Pyrazophos	148	Monocrotophos
65	Pyridaben	149	Nicosulfuron
66	Pyridaphenthion	150	Omethoate
67	Pyrimethanil	151	Oxydemeton-methyl
68	Quinalphos	152	Pirimiphos-ethy l
69	Simazine	153	Pymetrozin
70	S421	154	Quizalofop-ethyl
71	Dimethoate	155	Rimsulfuron
72	Edifenphos	156	Spinosad
73	Endosulfan (alpha isomer)	157	Spiroxamine
74	Endosulfan (beta isomer)	158	Tebufenozide
75	Endosulfan sulfate	159	Thiabendazole
76	Ethion	160	Thiacloprid
77	Ethoprophos	161	Thiamethoxam
78	Etofenprox	162	Hifensulfuron-methyl
79	Etrimfos	163	Thiodicarb
80	Fenarimol	164	Triasulfuron
81	Fenitrothion	165	Trichlorfon
82	Fenobucarb	166	Triflusulfuron-methyl
83	Fenoxycarb	167	Thiofanox-sulfone
84	Fenpropathrin	168	Thiofanox-sulfoxid

DDD, Dichlorodiphenyldichloroethane; DDE, Dichlorodiphenyldichloroethylene; DDT, Dichlorodiphenyltrichloroethane; HCH, Hexachlorocyclohexane.

Table S2. Pesticide residue test results of apple wood (mg · kg^{-1} dry weight).

Apple Wood Sample No	Chlorpyrifos	Cyhalothrin	Cypermethrin	Difenoconazole	Tebuconazole	Chlorbenzuron	Carbendazim	Imidacloprid	Acetamiprid	Cymoxanil	Prochloraz
1	0.010	ND	ND	ND	ND	ND	0.015	ND	ND	ND	ND
2	0.45	ND	0.19	0.083	0.16	0.21	1.43	0.19	0.038	0.083	0.040
3	0.13	ND	ND	ND	ND	ND	0.40	0.10	0.023	ND	ND
4	0.04	ND	ND	ND	0.052	0.037	ND	ND	ND	ND	ND
5	0.14	ND	ND	ND	0.10	ND	0.35	0.075	0.010	ND	ND
6	0.035	ND	ND	ND	0.090	0.16	0.40	0.037	0.021	ND	ND
7	0.037	ND	ND	ND	0.044	ND	0.12	0.036	0.015	ND	ND
8	0.22	ND	0.11	ND	0.031	ND	ND	ND	ND	ND	ND
9	0.14	ND	ND	ND	0.027	ND	0.028	ND	ND	ND	ND
10	0.094	ND	ND	ND	0.083	ND	1.22	0.095	0.048	ND	ND
11	0.10	ND	ND	ND	0.11	0.13	4.86	0.11	0.030	ND	ND
12	0.043	ND	ND	ND	0.060	ND	1.25	0.14	0.013	ND	ND
13	0.010	ND	ND	ND	0.017	ND	0.047	ND	0.11	ND	ND
14	0.18	ND	ND	ND	0.44	0.21	0.62	0.32	0.077	ND	ND
15	0.31	0.14	ND	ND	0.41	ND	0.62	0.11	0.072	ND	ND
16	0.029	ND	ND	ND	ND	0.052	0.086	0.045	ND	ND	ND
17	0.066	ND	ND	0.034	0.16	ND	0.25	ND	ND	ND	ND
18	0.017	ND	ND	ND	ND	ND	0.04	ND	ND	ND	ND
19	0.067	ND	ND	ND	0.098	ND	0.12	0.047	ND	ND	ND
20	0.47	0.19	ND	ND	0.18	0.093	1.3	0.065	0.053	0.056	ND
21	0.75	ND	ND	ND	ND	0.051	0.13	0.031	0.025	ND	ND

ND, not detected.