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Determination of Pesticide Residues of Organic Wheat Flours and Some Quality Criteria of Breads

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In this research, normal and sourdough breads manufactured with organic whole wheat flour, type 650 and the mixture (50 %, 50 %) of whole wheat flour and type 650 (w/w), were investigated. 258 Different pesticides including organic chlorinated, organic phosphorous, organic chlorinated-phosphorous, strobuline group fungicides, *n*-methyl carbamited, synthetic peritroit group, benzimidazole group and pyridylmethalamine group pesticides were searched as the indicator for organic flours and none of these pesticide residues were found in defined limitations. Moisture, ash, sedimentation, falling number, pharinograph, extensograph, wet and dry gluten values of organic flours were determined. Normal and sourdough breads were prepared with these flours and inner pore structure and shelf life of these breads were examined. It is determined that the bread produced with whole wheat flour was softer and the colour of this bread was dark than the others'. It is also observed that the shelf life of the bread produced with normal dough was shorter than that of sourdough bread. Additionally there was a significant break-down on the upper side of breads that produced with normal dough.

Key Words: Organic bread, Sourdough bread, Whole grain flour, Extensograph, Pharinograph.

INTRODUCTION

Organic agriculture is a kind of controlled and certified agricultural production made without using any chemicals[1]. As a result of food supply problems with rapid increase in world population after the middle of the 20th century, it was come into question to obtain more yields per unit area. For this reason, new policies were developed which stipulated "using rich inputs in production with high yielded varieties" which was named as Green Revolution in 1970s. However the outcomes of this policy were serious environmental pollution and destruction of natural balance. Against these negative effects, environmentalist producers in European countries had been begun to seek alternative production methods to produce healthy products

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which had no toxic effect to humans and could be produced without environmental pollution and destruction of the natural balance. Eventually organic bread plays an important role and takes an important place in human nutrition as a result of these policies.

Organic bread is a kind of bread manufactured according to the organic production legislations with using organic flour types from organic wheat grinded in suitable mills without any synthetic additive.

Bread has long been known as a food source throughout the world. Bread, indispensable food in human nutrition, has a good carrier property for the other foods according to its unique neutral aroma. It's easier to find and cheaper to buy than the other foods. It has a big importance in human nutrition as a source of carbohydrate and protein. It is mostly consumed in Turkey as the primary food source[2].

In this research, some properties of normal and sourdough breads manufactured with organic whole wheat flour, type 650 and the mixture of whole wheat flour and type 650 (50 %, 50 %) which had been analyzed for additives and residues were investigated.

EXPERIMENTAL

The research materials are normal and sourdough breads, coded as A, B, C, D, E and F, that were produced with whole (I), type 650 (II) and the mixture of whole and type 650 (50 %, % 50) (III) flours. Industrial sourdough provided from Ozmaya A.S. (Istanbul) and organic sourdough were used as sourdough. Rock salt from private company and refined and decontaminated tap water with hardness of 7.5 (^{o}fH) were used in the production.

Additive and residue analyses in flours were made with Luke Extraction method in GC/MS (Gas Cromotography/Mass Spectrophotometry)[3,4]. Moisture content, ash content, wet and dry gluten values, pharinograph, extensograph, falling number and sedimentation values were analyzed according to Ozkaya and Kahveci[5] and Uluoz[6]. Packaged bread samples were stored in laboratory conditions to determine the shelf life and the breads were monitored for mould formation and physical retrogradation. Homogeneity of inner pore structure, inner toughness and inner colour of bread were determined.

RESULTS AND DISCUSSION

Using of organic flour from organic wheat is the most important necessity in organic bread production. The analyses made for this reason showed that the flours that used as raw material in organic bread production include none of the pesticide residues (Table-1). These results indicated that the flours were suitable in the meaning of pesticide residue limits which are the most important indicators of organic agriculture.

TABLE-1
PESTICIDE RESIDUES IN ORGANIC WHEAT FLOUR WITH RECOVERY DATA

Pesticide	Apparatus	Recovery (%)	Detection limit (mg kg^{-1})	Result
2-4-Dichlorophenoxyacetic acid	LC/MS-MS	92	0.006	nd
2-4 DDE	GC/ECD-FPD/MS	97	0.003	nd
2-4 DDT	GC/ECD-FPD/MS	71	0.006	nd
3-Hydroxycarbofuran	LC/MS-MS	97	0.050	nd
4-4 DDD	GC/ECD-FPD/MS	107	0.009	nd
4-4 DDE	GC/ECD-FPD/MS	100	0.012	nd
4-4 DDT	GC/ECD-FPD/MS	60	0.007	nd
Acephate	GC/ECD-FPD/MS	67	0.050	nd
Acetamiprid	LC/MS-MS	86	0.015	nd
Alachlor	GC/ECD-FPD/MS	95	0.020	nd
Aldicarb	LC/MS-MS	89	0.050	nd
Aldicarb sulfone	LC/MS-MS	92	0.050	nd
Aldicarb sulfoxide	LC/MS-MS	98	0.050	nd
Aldrin	GC/ECD-FPD/MS	51	0.048	nd
α-BHC	GC/ECD-FPD/MS	104	0.017	nd
α-Cypermethrin	GC-ECD/GC-MS	92	0.020	nd
α-Endosulfan	GC/ECD-FPD/MS	92	0.054	nd
Amitraz	LC/MS-MS	118	0.050	nd
Atrazine	LC/MS-MS	83	0.013	nd
Azinphos methyl	GC/ECD-FPD/MS	68	0.050	nd
Azobenzene	GC/ECD-FPD/MS	99	0.050	nd
Azoxystrobin	LC/MS-MS	67	0.019	nd
Benfurocarb	LC/MS-MS	77	0.020	nd
Benomyl	LC/MS-MS	83	0.013	nd
β-BHC	GC/ECD-FPD/MS	87	0.018	nd
β-Cyfluthrin	GC-ECD/GC-MS	89	0.012	nd
β-Endosulfan	GC/ECD-FPD/MS	96	0.031	nd
Bifenthrin	GC-ECD/GC-MS	79	0.009	nd
Boscalid	LC/MS-MS	106	0.035	nd
Bromophos ethyl	GC/ECD-FPD/MS	59	0.041	nd
Bromophos methyl	GC/ECD-FPD/MS	78	0.038	nd
Bromopropylate	GC/ECD-FPD/MS	100	0.009	nd
Bupirimate	GC/ECD-FPD/MS	87	0.050	nd
Buprofezin	GC/ECD-FPD/MS	61	0.050	nd
Butocarboxim	LC/MS-MS	56	0.050	nd
Captan	GC/ECD-FPD/MS	95	0.117	nd
Carbaryl	LC/MS-MS	98	0.023	nd
Carbendazim	LC/MS-MS	74	0.013	nd
Carbofuran	LC/MS-MS	97	0.030	nd
Carbosulfan	LC/MS-MS	83	0.020	nd
Chlorfenapyr	GC/ECD-FPD/MS	65	0.050	nd
Chlorfenson	GC/ECD-FPD/MS	92	0.016	nd
Chlorfenvinfos	LC/MS-MS	95	0.015	nd

Pesticide	Apparatus	Recovery (%)	Detection limit (mg kg^{-1})	Result
Chlorfluazuron	LC/MS-MS	86	0.038	nd
Chlorpropham	GC/ECD-FPD/MS	83	0.050	nd
Chlorpyriphos ethyl	GC/ECD-FPD/MS	101	0.004	nd
Chlorpyriphos methyl	GC/ECD-FPD/MS	86	0.013	nd
Chlorthalonil	GC/ECD-FPD/MS	80	0.038	nd
cis-Chlordane (alpha)	GC/ECD-FPD/MS	77	0.015	nd
cis-Heptachloroepoxide	GC/ECD-FPD/MS	60	0.050	nd
Clofentezine	LC/MS-MS	103	0.018	nd
Coumaphos (Asuntol)	GC/ECD-FPD/MS	76	0.050	nd
Cycloate	LC/MS-MS	112	0.014	nd
Cyfluthrin	GC-ECD/GC-MS	78	0.008	nd
Cymoxanil	LC/MS-MS	90	0.050	nd
Cypermethrin	GC-ECD/GC-MS	89	0.007	nd
Cyproconazole	LC/MS-MS	79	0.018	nd
Cyprodinil	LC/MS-MS	98	0.013	nd
δ-HCH	GC/ECD-FPD/MS	97	0.045	nd
Deltamethrin	GC-ECD/GC-MS	87	0.008	nd
Demethon-S-methyl	GC/ECD-FPD/MS	84	0.043	nd
Diazinon	LC/MS-MS	72	0.014	nd
Dichlofluanid	GC/ECD-FPD/MS	92	0.042	nd
Dichlorvos	GC/ECD-FPD/MS	99	0.019	nd
Dicofol	GC/ECD-FPD/MS	97	0.018	nd
Dicrotophos	LC/MS-MS	41	0.016	nd
Dieldrin	GC/ECD-FPD/MS	86	0.027	nd
Difeconazole	LC/MS-MS	74	0.016	nd
Dimefox	GC/ECD-FPD/MS	55	0.017	nd
Dimethoate	LC/MS-MS	43	0.020	nd
Dimethomorph	LC/MS-MS	78	0.019	nd
Dinobuton	GC/ECD-FPD/MS	92	0.100	nd
Dinocap	LC/MS-MS	89	0.025	nd
Disulfoton sulfone	GC/ECD-FPD/MS	68	0.050	nd
Disulfoton sulfoxide	GC/ECD-FPD/MS	70	0.050	nd
Ditalimfos	GC/ECD-FPD/MS	99	0.050	nd
Dithianon	LC/MS-MS	103	0.021	nd
Endosulfan sulphate	LC/MS-MS	161	0.010	nd
Endrin	GC/ECD-FPD/MS	91	0.044	nd
Endrin aldehit	GC/ECD-FPD/MS	105	0.100	nd
Endrin ketone	GC/ECD-FPD/MS	128	0.066	nd
Epoxiconazole	LC/MS-MS	112	0.016	nd
Esfenvalerate	GC-ECD/GC-MS	88	0.007	nd
Ethiofencarb	GC/ECD-FPD/MS	89	0.050	nd
Ethion	GC/ECD-FPD/MS	112	0.013	nd
Ethofumesate	GC/ECD-FPD/MS	102	0.050	nd
Ethoprophos	GC/ECD-FPD/MS	45	0.050	nd
Etrimfos	LC/MS-MS	86	0.010	nd
Famoxadone	LC/MS-MS	75	0.016	nd

Pesticide	Apparatus	Recovery (%)	Detection limit (mg kg^{-1})	Result
Fenamiphos	GC/ECD-FPD/MS	58	0.050	nd
Fenarimol	GC/ECD-FPD/MS	58	0.050	nd
Fenazaquin	LC/MS-MS	96	0.002	nd
Fenchlorphos	GC/ECD-FPD/MS	62	0.016	nd
Fenitrothion	GC/ECD-FPD/MS	97	0.058	nd
Fenoxycarb	LC/MS-MS	82	0.010	nd
Fenpropathrin	GC/ECD-FPD/MS	79	0.008	nd
Fenson	GC/ECD-FPD/MS	72	0.024	nd
Fensulfothion	LC/MS-MS	96	0.058	nd
Fenthion	GC/ECD-FPD/MS	67	0.022	nd
Fenvalerate	GC-ECD/GC-MS	91	0.009	nd
Flamproph methyl	GC/ECD-FPD/MS	78	0.050	nd
Flucythrinate	GC-ECD/GC-MS	92	0.009	nd
Fludioxonyl	LC/MS-MS	88	0.005	nd
Fluvalinate	GC-ECD/GC-MS	99	0.008	nd
Folpet	GC/ECD-FPD/MS	71	0.117	nd
Fonofos	LC/MS-MS	98	0.035	nd
Formothion	GC/ECD-FPD/MS	87	0.032	nd
Furathiocarb	LC/MS-MS	110	0.023	nd
Heptachlor	GC/ECD-FPD/MS	94	0.049	nd
Heptachlor endoepoxide (isomer A)	GC/ECD-FPD/MS	68	0.041	nd
Heptachlor exoepoxide (isomer B)	LC/MS-MS	74	0.024	nd
Heptenophos	GC/ECD-FPD/MS	89	0.016	nd
Hexachlorobenzene	GC/ECD-FPD/MS	62	0.050	nd
Hexaconazole	LC/MS-MS	92	0.027	nd
Hexythiazox	LC/MS-MS	82	0.019	nd
Imazalil	LC/MS-MS	71	0.021	nd
Imidacloprid	GC/ECD-FPD/MS	90	0.050	nd
Iodofenphos	LC/MS-MS	78	0.010	nd
Ioxynil	LC/MS-MS	84	0.025	nd
Iprodione	GC/ECD-FPD/MS	52	0.050	nd
Kresoxim-methyl	GC-ECD/GC-MS	84	0.009	nd
λ-Cyhalothrin	GC/ECD-FPD/MS	106	0.017	nd
Lindane (G-HCH)	GC/ECD-FPD/MS	88	0.050	nd
Linuron	LC/MS-MS	94	0.005	nd
Lufenuron	LC/MS-MS	92	0.015	nd
Malaoxon	LC/MS-MS	87	0.030	nd
Malathion	LC/MS-MS	98	0.013	nd
Mecarbam	LC/MS-MS	87	0.010	nd
Metalaxyl	GC/ECD-FPD/MS	83	0.047	nd
Methamidophos	LC/MS-MS	128	0.037	nd
Methidathion	LC/MS-MS	100	0.050	nd
Methiocarb	LC/MS-MS	99	0.050	nd
Methomyl	GC/ECD-FPD/MS	94	0.035	nd
Methoxychlor	LC/MS-MS	90	0.010	nd
Metolachlor	LC/MS-MS	83	0.020	nd

Pesticide	Apparatus	Recovery (%)	Detection limit (mg kg^{-1})	Result
Metribuzin	GC/ECD-FPD/MS	81	0.032	nd
Mevinphos	LC/MS-MS	92	0.025	nd
Molinate	LC/MS-MS	110	0.022	nd
Monocrotophos	LC/MS-MS	50	0.027	nd
Monolinuron	LC/MS-MS	105	0.017	nd
Myclobutanil	GC/ECD-FPD/MS	50	0.050	nd
Nuarimol	GC/ECD-FPD/MS	85	0.050	nd
Omethoate	LC/MS-MS	125	0.017	nd
Oxamyl	GC/ECD-FPD/MS	94	0.050	nd
Oxidixyl	GC/ECD-FPD/MS	71	0.050	nd
Oxy-chlordane	GC/ECD-FPD/MS	102	0.029	nd
Oxyfluorfen	LC/MS-MS	104	0.025	nd
Paraoxon ethyl	LC/MS-MS	100	0.024	nd
Parathion ethyl	LC/MS-MS	115	0.016	nd
Parathion methyl	GC/ECD-FPD/MS	82	0.050	nd
Penconazole	GC/ECD-FPD/MS	99	0.050	nd
Pendimethalin	GC/ECD-FPD/MS	66	0.024	nd
Pentachloraniline	GC-ECD/GC-MS	77	0.007	nd
Permethrin	LC/MS-MS	92	0.033	nd
Phentoate	LC/MS-MS	96	0.029	nd
Phorate	LC/MS-MS	88	0.021	nd
Phosalone	LC/MS-MS	118	0.015	nd
Phosmet	LC/MS-MS	87	0.030	nd
Phosphamidon	GC/MS	97	0.050	nd
Piperonyl butoxide	LC/MS-MS	82	0.020	nd
Primicarb	GC/ECD-FPD/MS	91	0.024	nd
Primiphos-ethyl	LC/MS-MS	73	0.015	nd
Primiphos-methyl	LC/MS-MS	109	0.017	nd
Procymidone	GC/ECD-FPD/MS	88	0.010	nd
Profenophos	LC/MS-MS	85	0.023	nd
Promecarb	LC/MS-MS	99	0.027	nd
Propamocarb	LC/MS-MS	84	0.050	nd
Propargite	GC/ECD-FPD/MS	100	0.050	nd
Propiconazole	LC/MS-MS	94	0.015	nd
Propoxur	LC/MS-MS	92	0.050	nd
Propyzamide	LC/MS-MS	58	0.018	nd
Prothiophos	LC/MS-MS	87	0.050	nd
Pyrethrins	GC-ECD/GC-MS	84	0.020	nd
Pyridaben	LC/MS-MS	73	0.021	nd
Pyridaphenthion	LC/MS-MS	102	0.023	nd
Pyrimethanil	GC/ECD-FPD/MS	78	0.020	nd
Pyriproxyfen	LC/MS-MS	74	0.013	nd
Pyrazophos	LC/MS-MS	107	0.017	nd
Quinalphos	GC/ECD-FPD/MS	92	0.050	nd
Quinomethionate	GC/ECD-FPD/MS	85	0.027	nd
Quintozene (PCNB)	GC/ECD-FPD/MS	87	0.014	nd

Pesticide	Apparatus	Recovery (%)	Detection limit (mg kg^{-1})	Result
Resmethrin	GC/ECD-FPD/MS	71	0.050	nd
Simazine	GC/ECD-FPD/MS	69	0.008	nd
Sulfotep	LC/MS-MS	84	0.046	nd
Sulprofos	GC/ECD-FPD/MS	96	0.050	nd
Taufluvalinate	GC-ECD/GC-MS	101	0.008	nd
Tebuconazole (Raxil)	GC/ECD-FPD/MS	71	0.050	nd
Tecnazene	GC/ECD-FPD/MS	82	0.021	nd
Terbutryn	LC/MS-MS	95	0.013	nd
Tetraconazole	GC/ECD-FPD/MS	90	0.050	nd
Tetradifon	GC/ECD-FPD/MS	91	0.017	nd
Tetrasul	GC/ECD-FPD/MS	85	0.016	nd
Thiamethoxam	LC/MS-MS	72	0.028	nd
Thiobencarb (benthiocarb)	GC/ECD-FPD/MS	87	0.050	nd
Thiobendazole	LC/MS-MS	82	0.050	nd
Thiophonate-methyl	LC/MS-MS	96	0.019	nd
Tolclofos methyl	GC/ECD-FPD/MS	84	0.050	nd
Tolyfluanide	LC/MS-MS	74	0.042	nd
trans-Chlordane (gamma)	GC/ECD-FPD/MS	80	0.015	nd
Triadimefon	LC/MS-MS	88	0.021	nd
Triadimenol	LC/MS-MS	90	0.020	nd
Triallate	LC/MS-MS	92	0.031	nd
Triazophos	LC/MS-MS	61	0.018	nd
Trichlorfon	GC/ECD-FPD/MS	74	0.033	nd
Trifloxystrobin	LC/MS-MS	71	0.017	nd
Triflumizole	LC/MS-MS	83	0.014	nd
Trifluralin	GC/ECD-FPD/MS	89	0.003	nd
Vinclozolin	GC/ECD-FPD/MS	104	0.016	nd

The results of the analysis of organic flours and breads produced from these flours are given in Table-2. Because of the high bran ratio in whole flour, ash and moisture content were found high while normal and exceeded sedimentation, wet and dry gluten values were lower. It's classified in mid gluten flour type[6] with having gluten value between 20 and 27. It's cited that the flours which have a falling number 150 and lower have high amylase activity, have a falling number between 200 and 250 have normal amylase activity and have a falling number 300 and higher have low amylase activity[5]. So all flours analyzed in this research have low amylase activity.

Because of the high bran ratio in whole flour, sedimentation, gluten, falling number, softening degree and extensograph values were low while water holding, maturing time and stability values were high. Various studies have shown that regular consumption of whole grains and whole grain products, which are rich in dietary fiber, vitamins, minerals and and phytochemicals including phenolics, carotenoids, vitamin E, lignans, β-glucan, inulin, resistant starch, sterols and phytates, is associated

TABLE-2
SOME CHARACTERISTICS OF ORGANIC DOUGH AND BREADS

	Flour types		
	Whole	Type 650	50 % whole + 50 % type 650 (w/w)
Humidity (%)	13.15	12.95	13.05
Normal sedimentation	20	23	22
Extended sedimentation	24	26	25
Moisture gluten (%)	23.5	28.7	25.2
Dry gluten (%)	8.2	9.9	86
Ash in dry matter (%)	1.806	0.805	1.296
Falling number	340	351	344
Farinograph			
Water absorption (%)	66.5	62	65
Development time (min)	6	4.5	6
Dough stability (min)	7.5	10.5	8
Degree of softening (FU)	60	70	70
Ekstensograph (135 min)			
(R_5) (EU)	200	270	260
($R_{m)}$ (EU)	220	280	260
Energy (cm^2)	31	50	35
Extensibility value (mm)	80	143	100

Bread types	Shelf life (day)	Humidity (%)	Appearance and pore microstructure
A	4	43.2	Densely pore and brown intrastructure
B	5	40.8	Rarely pore and light brown intrastructure
C	4	39.4	Densely pore and white intrastructure
D	5	36.9	Rarely pore and yellowish intrastructure
E	4	41.7	Densely pore and brown intrastructure
F	5	41.4	Rarely pore and light brown intrastructure

R_5 = Dough resistance, R_m = Maximum resistance, FU = Farinograph unit, EU = Ekstensograf unit, A = Whole flour + normal yeast, B = Whole flour + soured dough, C = Type 650 flour + normal yeast, D = Type 650 flour + soured dough, E = 5 0% whole flour + 50 % type 650 flour + normal yeast, F = 50 % whole flour + 50 % type 650 flour + soured dough.

with many physiological benefits related to reduced risks of various types of chronic diseases such as coronary heart disease, colon cancer and diabetes[7-13]. On the other hand, in bread made of whole grain flour, the existence of bran and germ deteriorates the rheological properties of dough, lowers of loaf volume, increases crumb firmness, darkens crumb appearance and gives different flavour profiles of whole wheat breads from those of white flour[7,8,11]. Therefore, because of the higher bran and germ in whole grain flour, the whole grain bread has not been attractive to consumers.

While all bread samples produced with sourdough have 5 days shelf life, it was 4 days for breads produced with normal dough. It is related with smaller inner pores of sourdough breads which retards the retrogradation of the product.

In conclusion, the demand of the organic products has increased tremendously in the last decades. Organic cereals and flour of these cereals are used in the production of the organic bread. The results of the researches indicate that there are some disadvantages of the organic bread such as low bread volume, crumb firmness and dark colour. However organic breads containing none of the chemical residues are considered healthier than normal breads.

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