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The aim was to compare the levels of contamination in organic and conventional raw materials. To this end, the level of contamination by heavy metals (lead, cadmium, arsenic, mercury), nitrates and nitrites, and some mycotoxins were monitored. Fifteen products were tested in their organic and conventional forms, including meat, milk, eggs, vegetables and cereals. The median levels of contamination were calculated and compared with the recommended or regulated maximum levels. The maximum levels were exceeded for lead in organic carrots and buckwheat, and in conventional wheat; for cadmium, in both organic and conventional buckwheat; for nitrates, in organic spinach; and for patulin in organic apples. Moreover, contamination of both conventional and organic wheat by deoxynivalenol was observed with a higher level in organic products. However, the health risk for consumers might be real only for the contamination by mycotoxins as the contaminated foods (apples, wheat) are the main contributors to total exposure.

Introduction

Organic products are understood to be all those raw commodities that are produced without synthetic pesticides, but natural pesticide uses are authorized (EC 1991). The first notion of organic farming in European countries was born in the 1920s. For a long time limited to small groups of producers, processors and consumers, the organic food market is currently booming in France with a 20% per year supply increase (Sylvander 1999). In 1999, organic farming in France represented 7500 farms and 270 000 ha, and roughly 1% of the total consumption of foods (Observatoire National de l'Agriculture Biologique 1999). Faced with the threat of pesticides exposure, other food safety problems ('mad cow disease', dioxins in milk, etc.) and environmental issues, many consumers are turning to organic foods in the hope of finding a healthy and environmentally friendly alternative (Fischer 1999, PNUD 2000). However, the increasing productivity of organic farming currently raises the question of food safety and only a few studies have addressed scientifically the advantages or drawbacks of organic products from this point of view. In a French study, the level of pesticides residues in organic products has been monitored (15 000 analyses) over 3 years (1976–78) in which only 2% contained residues (Puisais 1980). Although other studies have dealt with the nitrate contamination of organic versus conventional products (Lecerf 1995), the contamination by heavy metals or by mycotoxins has been little investigated.

The aim of the present study was to make a comparison of contamination between organic and conventional products. There are three ways of undertaking studies to compare conventionally and organically produced foods (Vetter *et al.* 1987): market-orientated supply studies (samples are taken from conventional and alternative shops), surveys (samples are products from selected farms with different forms of cultivation) and cultivation tests (samples are taken from experimental farms).

In the present case, a survey was conducted. Fifteen kinds of products, which included meat, milk, eggs, vegetables and cereals, were tested in their organic and conventional forms, and 192 samples were

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analysed (98 conventional products, 94 organic ones). Samples were tested for all the contaminants likely to be found in organic and conventional production: heavy metals such as lead (Pb), cadmium (Cd), arsenic (As) and mercury (Hg), nitrates and nitrites in vegetable samples, mycotoxins such as deoxynivalenol (DON), 3-acetyl DON, 15-acetyl DON, nivalenol (NIV), T-2 toxin, HT-2 toxin, diascetoxyscirpenol, patulin, fumonizin B_1, ochratoxin A, and zearalenone. In all, some 3924 analyses were performed.

Material and methods

Sample collection

Raw products in the form of several samples were selected from different farms representative of the cultivation forms that were to be compared (organic and conventional). Environmental factors such as climate and soil conditions were made as comparable as possible by purchasing samples to be compared at the same time and in selected neighbouring farms.

For animal products, the following were analysed.

- Six conventional and six organic samples of beef (1 kg/sample), pork (1 kg/sample), poultry (1 kg/sample) and eggs (12 eggs/sample).
- Eleven conventional and 11 organic samples of milk (1 litre/sample).

For vegetable products, the following were analysed.

- Six conventional and six organic samples of lettuce (0.5 kg/sample), tomato (0.5 kg/sample), carrot (2 kg/sample) and apple (2 kg/sample).
- Six conventional and five organic samples of spinach (0.2 kg/sample) and French beans (1 kg/sample).
- Five conventional and five organic samples of buckwheat (0.5 kg/sample) and barley (1 kg/sample).
- Eleven conventional and 11 organic samples of wheat (1 kg/sample).

Samples were transported from the farms to the laboratory and then stored at −18°C before analysis.

Analysis

All contaminants were analysed according to the SANCO guide (EC 2000). Accordingly, a validated analytical method for residues should achieve a mean recovery in the range 70–110%, with a relative SD < 20%. The testing of the performance of the method must be conducted on fortified samples, five at the limit of quantification (LOQ) level, and five at 10 times LOQ. Two blank samples are included in the set, and their value should not exceed 30% LOQ. This validation also includes testing of linearity and for demonstrating specificity. For the analysis of mycotoxins, certified reference materials were included (when existing) in each set of analyses.

Heavy metals analysis

For heavy metals analysis, samples were mixed, homogenized and dried at 80°C for 48 h. Organic matter was then destroyed with a microwave system, using maxidigest 3.6 Prolabo, for 5 min at 25% power. For Hg determinations, 5 ml nitric acid (68% Normapur) was added followed by 2 ml hydrogen peroxide (30% Normapur) for Pb, Cd and As determination. Pb, Cd and As were then determined using an atomic absorption spectrometer (AAS) with electrothermal atomization, with Zeeman effect correction (Varian spectrAA300). Hg was measured with cold vapour atomic absorption spectrometry. For Pb and Cd determination, $NH_4H_2PO_4$ (0.5%) was employed as a matrix modifier, and for As determination, the modifier was a palladium and magnesium mixture (50/50).

Mycotoxin analysis

Trichothecenes. A total of 5 g samples was extracted with 25 ml acetonitrilewater (14:86 v/v), purified on a MycoSep column, and derivatized with BSA/TMSI/TMCS (3:3:2). The determination was then carried out by gas chromatography/mass spectrometry (for DON only) (Trucksess *et al.* 1996). The ions measured were as given in table 1.

All trichothecenes were analysed simultaneously with only a single extraction and a single chromatographic run.

Table 1.

	Parent ions	Ions for quantification
DON–TMS	407	197, 317, 405
2-Acetyl DON	407	197
15-Acetyl DON	407	197, 259, 285
NIV-TMS	379	377, 289, 363
DAS-TMS	378	197, 255, 378
HT-2-TMS	185	138
T-2-TMS	185	138

Zearalenone. A sample (20 g) was ground and extracted with 100 ml acetonitrile–water (60:40) filtered and centrifuged at 2000*g* for 4 min. The mycotoxin was analysed with an immunoassay test kit, Ridascreen Biopharm®.

Fumonisin. A sample (5 g) was ground and extracted with 10 ml ethanol–water (75:25), filtered and centrifuged at 2000*g* for 15 min. The mycotoxin was analysed with an immunoassay test kit, Ridascreen Diffchamb.

Aflatoxin B_1. A sample (50 g) was extracted with 250 ml chloroform–water (91:9). One part of the extract was evaporated to dryness and redissolved in 60% methanol. Toxins were then eluted with methanol and determined by reversed-phase high-performance liquid chromatography (HPLC), post-column derivation with bromine and fluorescence detection of the separate components, with an excitation at 362 nm and emission at 434 nm.

Patulin. Patulin was extracted with ethyl acetate and toluene, and purified on a florizil column. Separation and detection was then carried out by HPLC with UV detection at 275 nm.

Ochratoxin A. A sample (50 g) was ground and extracted for 2 min with acetonitrile–water (60:40 v/v). A sample was then filtered and centrifuged at 2000*g* for 4 min. Of this filtrate, 4 ml was purified on an immunoaffinity column (Ochraprep®). Analysis was then carried out by reversed-phase HPLC, with fluorimetric detection with an excitation at 330 nm and emission at 465 nm.

Nitrates and nitrites analysis

Nitrites. Samples (50 g) were extracted with boiling water. After purification with acetonitrile, proteins were precipitated by potassium hexaferrocyanate and zinc acetate, and filtered. Sulphanilamide and dichromate were added and absorbance was measured at 538 nm.

Nitrates. Samples (50 g) were extracted with boiling water and then purified with acetonitrile. The determination was carried by ionic chromatography WATERS and detection at 210 nm.

The limits of quantification (LOQ) for heavy metals, nitrates and mycotoxins are given in table 2.

Calculation method

Concentrations are expressed in mg kg^{-1} fresh material for nitrates and nitrites, and in µg kg^{-1} fresh

Table 2. Limits of quantification (LOQ) for heavy metals, nitrates and mycotoxins.

Contaminants	Unit	LOQ
Lead	mg kg^{-1}	0.005–0.05
Cadmium	mg kg^{-1}	0.002–0.01
Arsenic	mg kg^{-1}	0.05–0.5
Mercury	mg kg^{-1}	0.005–0.02
Nitrates	mg kg^{-1}	1
Patulin	µg kg^{-1}	5
DON, 3-acetyl DON, 15-acetyl DON, nivalenol	µg kg^{-1}	10
T-2 toxin, HT-2 toxin, diacetoxyscirpenol	µg kg^{-1}	50
Fumonisin B1	µg kg^{-1}	200
Ochratoxin A	µg kg^{-1}	1.5
Zearalenone	µg kg^{-1}	1.25

material for heavy metals and mycotoxins. Even if there is a guide for calculating the median of low-level contamination of food taking into account the number of samples below the LOQ (GEMS/Food-EURO 1995), it has been assumed that results less than LOQ were equal to LOQ and the medians of contamination were calculated on this basis.

A non-parametric Mann–Whitney (Wilcoxon) test statistic compared contamination of organic versus conventional grown raw materials using SYSTAT software.

Results

Contamination by heavy metals

Pb, Cd, Hg and As have been detected in both organic and conventional samples. The levels of Pb or Cd found in samples were compared with the maximum levels for certain contaminants in foodstuffs proposed by the EC and under discussion for a future directive. For Hg and As, there are neither maximum levels proposed by the EC nor any kind of recommendation.

Lead

Pb was found at a level more than LOQ in both conventional and organic carrots, spinach, wheat, barley, buckwheat and milk. It was found at a level more than LOQ, but only in organic apples.

Pb levels are above the maximum levels commonly accepted in the EU in organic carrots and buckwheat, and in conventional wheat (table 3). The contamination by Pb of organic carrots is significantly higher than that for conventional carrots ($p < 0.05$). However, whereas the contamination by Pb of organic barley, spinach, buckwheat and conventional lettuces is higher than that of conventional barley, spinach, buckwheat and organic lettuces, the differences were not significant.

Levels of Pb more than LOQ have not been found in the other foodstuffs.

Cadmium

Cd was found at a level more than LOQ in both conventional and organic apples, carrots, spinach, lettuces, wheat, barley, buckwheat and milk. Cd was

Table 3. Contamination of both conventional and organic products by lead.

Products	Maximum level, ($\mu g\ kg^{-1}$)[a]	Median ($\mu g\ kg^{-1}$)	Maximum ($\mu g\ kg^{-1}$)	Number of samples > LOQ	Number of samples > recommendations
Organic apples	100	10	20	1/6	0
Organic carrots		530[b]	1800	6/6	4/6
Conventional carrots		15	40	3/6	0
Organic spinach	300	70	100	3/5	0
Conventional spinach		10	10	2/6	0
Organic lettuces		20	40	5/6	0
Conventional lettuces		115	300	5/6	0
Organic wheat	200	50	160	5/11	0
Conventional wheat		50	310	5/11	1/11
Organic barley		100	160	4/5	0
Conventional barley		50	150	2/5	0
Organic buckwheat		60	420	2/5	1/5
Conventional buckwheat		50	150	2/5	0
Organic milk	20	5	16	3/11	0
Conventional milk		5	17	1/11	0

[a] Maximum levels for certain contaminants in foodstuffs, proposed by the EC and under discussion for a future directive.
[b] Significant difference between the mean concentration of the organically and conventionally produced raw materials ($p < 0.05$).

Table 4. Contamination of both conventional and organic products by cadmium.

Products	Maximum level, (μg kg^{-1})	Median (μg kg^{-1})	Maximum (μg kg^{-1})	Number of samples > LOQ	Number of samples > recommendations
Organic apples	50[a]	5	7	1/6	0
Conventional apples		5	6	1/6	0
Organic carrots		25.5	81	6/6	0
Conventional carrots		9	16	5/6	0
Conventional French bean	50[a]	5	6	1/6	0
Organic tomatoes		5	11	3/6	0
Conventional spinach	200[a]	35.5[c]	44	6/6	0
Organic spinach		85	156	5/5	0
Organic lettuces		14	67	6/6	0
Conventional lettuces		21	54	6/6	0
Organic wheat	100[a]	20	100	8/11	0
Conventional wheat		30	40	11/11	0
Organic barley		20	60	3/5	0
Conventional barley		20	30	3/5	0
Organic buckwheat		40	110	5/5	1/5
Conventional buckwheat		70	140	5/5	1/5
Organic milk	100[b]	1	3	5/11	0
Conventional milk		1	3	3/11	0
Organic eggs		2	4	1/6	0
Conventional poultry	50[b]	10	10	1/6	0

[a] Maximum levels for certain contaminants in foodstuffs, proposed by the EC and under discussion for a future directive.
[b] Conseil Supérieur d'Hygiène Publique de France (1996).
[c] Significant difference between the mean concentration of the organically and conventionally produced raw materials ($p < 0.05$).

found at a level more than LOQ, but only in conventional French beans and poultry, and in organic tomatoes and eggs. Cd levels were above the maximum levels proposed by the EC in organic and conventional buckwheat (table 4). The contamination by Cd of organic spinach was significantly higher than that of conventional spinach ($p < 0.05$). However, whereas the contamination by Cd of organic barley, carrots and conventional lettuces, wheat and buckwheat was higher than the contamination of conventional barley, carrots, and organic lettuces, wheat and buckwheat, the differences were not significant.

Levels of Cd more than LOQ have not been found in the other foodstuffs.

Arsenic and mercury

As was found at level more than LOQ in both conventional and organic carrots, and only in organic spinach. One sample of organic carrot was strongly contaminated with 1270 μg kg^{-1} As. Hg was found at a level more than LOQ in conventional carrots (table 5).

Table 5. Contamination of both conventional and organic products by mercury and arsenic.

Products	Contaminants	Maximum levels (μg kg^{-1})	Median (μg kg^{-1})	Maximum (μg kg^{-1})	Number of samples > LOQ
Organic carrots	arsenic	no	100	1270	1/6
Conventional carrots			100	120	1/6
Organic spinach			100	100	1/5
Conventional carrots	mercury	no	5	110	1/6

Levels of As or Hg more than LOQ have not been found in the other foodstuffs.

Contamination by nitrates and nitrites

Nitrates were found at level more than LOQ in both conventional and organic spinach, carrots, lettuces, French beans and tomatoes. Conventional tomatoes were significantly more contaminated by nitrates than organic ones ($p < 0.05$). Conventional spinach, French beans and organic carrots, and lettuces are more contaminated than organic spinach, French beans and conventional carrots and lettuces, but the differences failed to reach significance.

Nitrates levels are above the maximum levels (JECFA 1995) only in organic spinach (table 6).

Nitrites have also been found at a level more than LOQ only in conventional spinach. The difference between conventional and organic productions is significant ($p < 0.05$), but only one sample of conventional spinach was highly contaminated with 10.1 mg kg^{-1} nitrites (table 6).

Levels of nitrates or nitrites more than LOQ have not been found in the other foodstuffs.

Contamination by mycotoxins

Patulin was found at a level more than LOQ in organic apples, and its levels were above the maximum regulated level in one of six samples analysed (Codex Alimentarius 1999) (table 7). Organic apple were more contaminated by patulin than conventional ones, but the difference was not significant.

Deoxynivalenol (DON) was found at a level more than LOQ in both conventional and organic wheat and barley (table 7). Organic wheat and barley were more highly contaminated by DON than conventional ones, but the differences were not significant.

3-acetyl DON was found at a level more than LOQ in organic wheat.

Nivalenol was found at a level more than LOQ in both conventional and organic barley and in organic wheat (table 7). Organic wheat and barley were more higher contaminated by NIV than conventional ones, but the differences were not significant.

HT-2 toxin was found at a level more than LOQ in both organic wheat and barley (table 7).

Zearalenone was found at a level more than LOQ in conventional barley (table 7).

None of the other mycotoxins analysed were found in the selected foodstuff.

Table 6. Contamination of both conventional and organic foodstuffs by nitrates and nitrites.

Products	Contaminants	Maximum levels (mg kg^{-1})[a]	Median (mg kg^{-1})	Maximum (mg kg^{-1})	Number of samples > LOQ	Number of samples > Maximum levels
Conventional spinach	nitrates	3000	1591	1901	6/6	0
Organic spinach			1135	3923	5/5	1/5
Conventional carrots			113	198	6/6	0
Organic carrots			394	1010	6/6	0
Conventional French bean			711.5	985	6/6	0
Organic French bean			561	880	3/3	0
Conventional tomatoes			19[b]	26	6/6	0
Organic tomatoes			1	4	2/6	0
Conventional lettuces			804.5	1442	6/6	0
Organic lettuces			1221	2103	6/6	0
Conventional spinach	nitrites	no	7.5[b]	10.1	6/6	–
Organic spinach			0.2	0.2	0/5	–

[a] Joint Expert Committee on Food Additives (1995).
[b] There is a significant difference between the mean concentration of the organically and conventionally produced raw materials ($p < 0.05$).

Table 7. Contamination of both conventional and organic foodstuffs by mycotoxins.

Products	Mycotoxins	Maximum levels (μg kg^{-1})	Median (μg kg^{-1})	Maximum (μg kg^{-1})	Number of samples > LOQ	Number of samples > maximum levels
Organic apples	Patulin	50[a]	5	1240	2/6	1/6
Conventional wheat	DON	no	55	215	10/11	–
Organic wheat			106	494	6/11	–
Conventional barley			41	73	4/5	–
Organic barley			69	209	3/5	–
Organic wheat	3 acetyl DON	no	10	17	1/11	–
Organic barley	NIV	no	83	301	4/5	–
Conventional barley			10	57	1/5	–
Organic wheat			10	98	10/11	–
Organic wheat	HT-2	no	50	456	1/11	–
Organic barley			50	183	2/5	–
Conventional barley	Zearalenone	no	1.25	3.4	2/5	

[a] Codex Alimentarius (1999).

Discussion

Heavy metals

Lead. Pb above the maximum levels proposed by the EC was found in organic carrots, conventional wheat and organic barley. There was significantly more Pb in organic carrots than in conventional ones (530 versus 15 μg kg^{-1}, median, $p < 0.05$). Two French surveys carried out in 1992 and 1996 showed that the average contamination by Pb of root vegetables was 415 μg kg^{-1} (Direction Générale de la Santé 1992) and from 50 to 100 μg kg^{-1} (Conseil Supérieur d'Hygiène Publique de France 1996a). If the contamination by Pb in conventional carrots is in the margin of the ones highlighted in previous papers, the contamination of organic carrots is far higher. However, in a review, Woese *et al.* (1997) deduced that there were no major differences with respect to the levels of heavy metals between vegetables from organic and conventional production. The practices of organic farming cannot explain the difference in heavy metal contamination. The difference observed in the present study is, therefore, possibly linked to a selective and sporadic contamination (e.g. atmospheric) rather than to the organic production itself.

From the point of view of health consumer, these levels of contamination above the maximum levels might not pose a problem as carrots and wheat are not the only contributors to dietary Pb exposure, and they are not strong contributors as the consumption of buckwheat is not very high.

Cadmium. There is significantly more Cd in organic spinach than in conventional spinach (85 versus 35.5 μg kg^{-1}, median, $p < 0.05$). If the median contamination of conventional spinach is in accordance with the average contamination reported by the French Ministry of Health for leafy vegetables (Direction Générale de la Santé 1992) (35.5 versus 20 μg kg^{-1}), the median contamination of organic spinach is higher (85 μg kg^{-1}). The Conseil Supérieur d'Hygiène Publique de France (1996b) showed contamination from 50 to 60 μg kg^{-1} in lettuces and spinach. However, similar to Pb in organic carrots, Woese *et al.* (1997) suggested that the difference cannot be attributed to the type of production, but it might be explained by a selective and sporadic contamination.

Cd above the maximum levels proposed by the EC were found in both conventional and organic buckwheat, but with no significant difference. If the recommended levels of contamination are exceeded, there is unlikely to be a health risk for consumers as the consumption of buckwheat is not very high and because buckwheat is not a strong contributor to dietary Cd exposure.

Arsenic and mercury. There is no significant difference in median contamination by As and Hg between vegetables produced either organically or conventionally. Even if there is no regulation for these two heavy metals, analyses have shown high levels of As in organic carrots and Hg in conventional carrots.

Nitrates and nitrites

Nitrates above the maximum levels (JECFA 1993) were found in organic spinach. However, the median contamination by nitrates of organic spinach is still around the average as observed by a French survey (1135 versus 1870 mg kg^{-1}) (Direction Générale de la Santé 1990). Even if the level of contamination did not exceed the maximum levels, conventional tomatoes are significantly more contaminated by nitrates than organic ones.

Concerning nitrites, even if there is no regulation, the level found in conventional spinach seems very high. There is also a significant difference of nitrites contamination between organic and conventional spinach (7.5 versus 0.2 mg kg^{-1}, median, $p < 0.05$). These conventional spinach could have been initially contaminated by nitrates, and poor storage conditions at the laboratory might have Pb at these high levels of nitrites.

Mycotoxins

Patulin, DON, 3-acetyl DON, 15-acetyl DON, NIV, HT-2 and zearalenone were at levels more than LOQ in three products, both organic and conventional. There is a proposed maximum level only for patulin in apple juice, and one of six samples of organic apple contained levels of patulin above this proposed maximum level (Codex Alimentarius 1999). Thus, there might be a risk to the health of the consumer as apple juice is the main contributor to patulin exposure.

Organic apples are on average no more contaminated than conventional ones, but not when considering contamination based on means (211.16 ± 504.02 versus 35.83 ± 75.52 µg kg^{-1}). The difference failed to be significant only because of one strongly contaminated sample of organic apples (1240 µg kg^{-1}). This very high contamination was possibly the result of some unfavourable climatic conditions during production and storage. Indeed, organic apples may be more prone to contamination by toxins produced by moulds than conventional ones because they are not treated to the same extent with antifungal agents.

Even if there is no regulation for the other mycotoxins in raw materials, analyses have shown high levels of DON in both organic and conventional cereals. As in France, barley consumption is not widespread, it is wheat contamination by DON that seems to be the most worrying for human health.

Conclusions

The study compared the levels of contaminants in organic and conventionally grown raw materials. Because of its design (analyses on raw materials, few number of samples, etc.), the results show no conclusive evidence whether conventional products are more or less safe than organic ones. In contrast, future studies are directed towards some combination of foodstuff contaminant that might pose a threat to health, real or probable, as maximum levels are exceeded, particularly with organic apples/patulin, and organic and conventional wheat/deoxynivalenol.

In these cases, the first exposure of consumers should be assessed. Second, future studies should target one combination and analyse more samples, which should include finished products, to assess the effects of industrial processing, thus providing the real conditions of exposure. Finally, future studies should also include cultivation tests to improve the quality of production.

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