## Chapter 4 Gluten Content Change Over the Two Last Decades

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## Abbreviations

CD	Celiac disease
GF	Gluten-free

ppm parts per million

In recent years, the gluten-free (GF) food industry has become one of the most thriving industries. According to the Mintel Report on GF foods, in the U.S., the production of these foods has risen 136% between 2013 and 2105, with sales reaching \$11,609,000,000 [1]. However, the most remarkable aspect of this report is its GF market forecast of \$14,175,000,000 for 2018 in the worst-case scenario, while the best predictions could be around \$31,128,000,000 [1]. Reports by other companies portend a similar projection in Europe for 2015–2020 [2]. In the case of Canada, its government reported that 15.5% of newly launched food products in 2013 were GF [3]. According to the registered data, among GF food, snacks, bakery products, sauces and seasonings were the top categories in GF food launches. In addition to foodstuffs, around 200 new GF beverages were put on the market in Canada between 2007 and 2013 [3].

A clear example of this GF product expansion is easy to observe. While a few years ago, selling GF products was limited to specialty shops, today these products fill the shelves of grocery stores and supermarkets everywhere. However, this upsurge in the production and variety of GF products is not accompanied by an increase in the prevalence or awareness of celiac disease (CD). As Reilly [4] describes, searches for comments related to CD by Google Trend in the last decade remained constant, whereas those related to GF foods increased exponentially. Indeed, nowadays the search ratio is 1:8; that is, for each search for information on CD, there have been eight for gluten-free products [4].

Most American consumers of these products are not people who suffer from CD [5]. In these particular cases, their interest in GF foods appears to be similar to organic food conventionalization [6]. GF food for people without symptoms associated with gluten is motivated by a lifestyle based on cultural, ecological, civic, historical, ethical or health-related interest [6]. The use of ancient GF grains, for instance, may contribute to a product's enchanting appeal.

Along these lines, a worldwide survey carried out in 2015 among 30,000 adults in 60 countries indicated that 21% of the interviewees found the term "GF" to be very important in their choice of buying food [7]. However, it should not be forgotten that these types of products are more expensive than their counterparts that contain gluten, which also affects the purchasing decision [8]; this has a great impact on the age of the potential buyers of gluten-free food. The same survey revealed how around a third of respondents under age 34 are willing to buy food without gluten, despite the higher price, while only 12% of the over-65s would do so [7].

As mentioned in the chapter of this book concerning legislation, there are many countries that have accepted the definition of GF and the threshold of 20 mg of gluten per kg of food (or parts per million: ppm), proposed by the Codex Alimentarius to establish the foods that can be labeled "GF" (Chap. 2). It is important to understand that many consumers rely on the labeling or claims of GF when making their purchases; nevertheless, gluten can be unintentionally introduced into the food through direct contact with raw materials containing gluten (wheat, barley, and rye). This gluten contamination can happen to product development beforehand (during harvest, transport, or storage of raw materials), during processing (adding meat sauces, beans or soups that are intended to improve the sensory and technological characteristics of the dish), or after product embellishment (e.g., a serving dish that has been cross-contaminated with gluten-containing products). Although for some GF product consumers there would not be any repercussions, it can lead to reactions and severe symptoms in patients with CD or related pathologies. For this reason, a precise and routine control of gluten content in food is necessary to ensure its safety in people who can have a reaction resulting from the intake of this protein.

Several studies have been carried out concerning gluten contamination in GF-labeled products (Table 4.1). In 2010, two interesting Brazilian researches revealed that around 13% of gluten-free samples contained more than 20 mg/kg of gluten [9, 10]. Altogether, both studies analyzed 185 GF products, among them bread, flours, dough, sauce, cereal bars, and cereal-snack food groups.

Since 2010, the USA has been the country where most of the studies evaluating gluten content in GF labeled products have been carried out. Between 2011 and 2015, Thompson and Simpson (2015) collected 158 samples to analyze their gluten content [11]. The results concluded that gluten was detected in almost 13% of the samples (>5 mg/kg of gluten). Furthermore, it is important to point out that 5.1% of the samples had over 20 mg/kg. Other research conducted by the same group indicated a similar ratio of samples over the pre-set threshold (4:112) [12].

Publication	Used kit	Sample number	Sample classification	Gluten detection	Positive samples	Country	Period
Valdes et al. (2003) [22]	R5	3088	Not classified by food group	3.2–20 ppm, 628 samples (20.3%) 20–100 ppm, 754 samples (24.4%) 100–200 ppm, 120 samples (3.9%) >200 ppm, 197 samples (3.9%)	1	Spain and other European counties	2003ª
Gelinas et al. (2008) [19]	R5	77	Variety of food groups not defined	>20 ppm, 16 sample (20.8%)	Breakfast cereals, cookies, pancake, flour and sauce	Canada	2008 <sup>a</sup>
Laureano (2010) [9]	R5	70	Bread, flours, dough, sauce, cereal bars, and snacks	5–20 ppm, 11 samples (15.7%) >20 ppm, 9 sample (12.9%)	Bread, Flours, dough, sauce, cereal bars, and snacks	Brazil	2010 <sup>a</sup>
Plaza-Silva (2010) [10]	R5	115	Variety of food groups	>20 ppm, 15 sample (13.0%)	I	Brazil	2010 <sup>a</sup>
Daniewski et al. (2010) [21]	R5	22	Variety of food groups	≥20 ppm, 6 samples (27.3%)	Pasta, bread, biscuits, bakery,	Poland	2010ª
Agakidis et al. (2011) [20]	Skerritt	26	Flours, dairy, sweets, miscellaneous	>20 ppm, 2 sample (7.7%)	Flours, dairy	Greece	2012 <sup>a</sup>
Thompson and Simpson (2015) [11]	R5	158	1	5 to ≤10 ppm, 6 samples (3.8%) >10 to <20 ppm, 7 samples (4.43%) ≥20 ppm, 8 samples (5.1%)	1	USA	2011– 2014
Gibert et al. (2013) [13]	R5	205	Bread, pasta, pastry, biscuits, pizza and breakfast cereals	>5 ppm, 16 samples (6%) >20 ppm, 1 sample (0.5%)	Bread, pasta, pastry, biscuits, and breakfast cereals	Italy, Spain, Germany and Norway	2013ª

Table 4.1 Summary of publications related to gluten-free labeled products

(continued)

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Publication	Used kit	Sample	Sample classification	Gluten detection	Positive samples	Country	Period
Thompson and Grace (2013) [12]	R5	112		≥ 20 ppm, 4 samples (3.6%)		USA	2013 <sup>a</sup>
Sharma et al. (2015) [23]	R5	275	Grains/seeds/nuts/ legumes, condiments/ sauces, curry/soup/soup mixes, baking mixes, baked foods, pasta products, breakfast cereals, snack foods, granola/bars/ energy bars, beverages/ice-creams/ frozen desserts, meat/meat substitutes/refrigerated or frozen foods and others	>5 ppm, 10 samples (3.6%) >20 ppm, 3 sample (1.1%)	Grains/seeds/nuts/ legumes, condiments/ sauces, pasta products, breakfast cereals, snack foods and meat/meat substitutes/ refrigerated or frozen foods	USA	20154
Bustamante et al. (2017)	R5Skerritt	1652	Flours, breakfast cereals/ bars, bakery, pastry/ dough, bread, pasta, cereal snack and yeast	5-10 ppm, 25 samples (1.5%) 11-20 ppm, 32 samples (1.9%) 21-100 ppm, 39 samples (2.4%) 100-200 ppm, 5 samples (0.3%) >200 ppm, 9 samples (0.5%)	Flours, breakfast cereals/bars, bakery, pastry/dough, bread, pasta, cereal snack	Spain	2004– 2016
<sup>a</sup> Corresponds to the	e publication	year, due tu	"Corresponds to the publication year, due to the lack of information about when the study took place	it when the study took place			

Unfortunately, none of the authors gave information on the food groups studied or positive samples classification.

With regard to Europe, during this same time period, an analysis of bread, pasta, pastry, biscuits, pizzas, and breakfast cereals from Italy, Spain, Germany and Norway was conducted, in which the researchers noted minimal gluten contamination [13]. Although some traces were detected [17] in food samples (>5 mg/kg), only one sample (of pastry) from 205 GF products showed gluten levels over 20 mg/kg.

Some customers attribute the GF assumption of a product to reading the list of ingredients. As a consequence, research in which GF products were not labeled but appeared to be free of gluten-containing ingredients (no wheat, barley, or rye) have been carried out (Table 4.2). In the case of these kinds of foodstuffs, it can be said that Thompson promoted their analysis when she advised them about oats contamination with gluten in the USA [14]. In almost one-third of 12 studied oats, gluten levels were over 20 mg/kg. Later on, two studies conducted in two different countries (Brazil and Poland), declared similar percentages of gluten contamination above the standard limit (of 9.3 and 10.5%, respectively). The same year, Thompson et al (2010) published a study with 22 grains, seeds and flours [15]; according to the findings, millet flour and grain, white rice flour, buckwheat flour, sorghum flour, and soy flour were the most-contaminated raw materials [15].

With regard to bakery products, in research conducted in Brazil, positive samples were found in 6.1% of the products [16], and another study showed a cross-contamination by beans served in restaurants there [17]. Their results showed that 16% of the samples and 45% of the restaurants suffered from gluten contamination. A recent study performed in the USA, involving 101 non-gluten-free-labeled samples concluded that five samples (breakfast cereals, spices, snacks, seasoning mixes, and oat fiber) contained more than 20 mg/kg of gluten [18].

In view of the above-mentioned results, as well as according to logic, it seems that gluten level control has been higher in GF labeled foods than in those that are apparently GF (by checking food labels or composition). In this regard, research conducted simultaneously in Canada and Greece with GF-labeled and non-labeled foods (but apparently GF) showed similar percentages of samples above 20 mg/kg gluten in both groups (about 20% in Canada and 10% in Greece) [19, 20]. It is important to note that the sample sizes of both studies did not exceed 150 food-stuffs. On the contrary, in another study with a moderate sample size, but that took place in Poland, there was a higher ratio of positive samples in GF-labeled samples than in those that were apparently GF [21].

The first large-scale study evaluating gluten content not only in GF-labeled products but also in non-labeled ones, was carried out by Valdes et al (2003) [22] in which they analyzed 3,088 GF-labeled samples from several European countries. Their results indicated a high gluten detection (1,699 samples with >3.2 mg/kg) in these kinds of products, with almost one-third of the total samples having gluten over 20 mg/kg. For non-GF-labeled samples, this analysis found that in 1,366 samples, 66% were contaminated with gluten (>3.2 mg/kg of gluten) and 570 with more than 20 mg/kg. Moreover, in the samples studied, the authors observed that maize was the raw material most contaminated with gluten; therefore, it is possible to

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Publication	Used kit	Sample number	Sample classification	Gluten detection	Positive samples	Country	Period
Valdes et al. (2003) [22]	R5	1366	Not classified by food group	3.2–20 ppm, 341 samples (25.0%) 20–100 ppm, 325 samples (23.8%) 100–200 ppm, 95 samples (7.0%) >200 ppm, 150 samples (1.1%)	1	Spain and other European counties	2003*
Thompson (2004) [14]	R5	12	Rolled or steel-cut oats	>20 ppm, 9 sample (32%)	Rolled or steel-cut oats	USA	2004
Gelinas et al. (2008) [19]	R5	71	Variety of food group not defined	> 20 ppm, 16 sample (22.5%)	Cereals bars, flours, chips, breakfast cereals, bran, bean mix, oatmeal	Canada	2008*
Plaza-Silva (2010) [10]	R5	86	Variety of food group	>20 ppm, 8 sample (9.3%)	1	Brazil	2010*
Daniewski et al. (2010) [21]	R5	19	Variety of food group	≥ 20 ppm, 2 samples (10.5%)	Flakes, flour	Poland	2010*
Thompson et al. (2010) [15]	R5	22	Grains, seeds and flours	>5 ppm, 9 samples (41%) >20 ppm, 7 sample (32%)	Millet flour and grain, White rice flour, buckwheat flour, sorghum flour and soy flour.	USA	2010*
Agakidis et al. (2011) [20]	Skerritt	15	Flours, dairy, sweets, miscellaneous	>20 ppm, 2 sample (13.3%)	Flours	Greece	2012*
Oliveira et al. (2014) [17]	Skerritt	60	Beans	>20 ppm, 10 sample (16.7%)	Beans	Brazil	2014*
Farage et al. (2017) [16]	R5	130	Bakery products	>20 ppm, 8 sample (6.1%)	Bakery	Brazil	2014

 Table 4.2
 Summary of publications related to non-gluten-free labeled products

Sharma et al. (2015) [23]	R5	186	Grains/seeds/nuts/ legumes, condiments/ sauces, curry/soup/soup mixes, pasta products, breakfast cereals, snack foods, granola/bars/ energy bars, beverages/ ice-creams/frozen desserts, meat/meat substitutes/ refrigerated or frozen foods, and others	>5 ppm, 48 samples (25.8%) >20 ppm, 36 sample (19.3%)	Grains/seeds/nuts/legumes, curry/soup/soup mixes, pasta products, breakfast cereals, snack foods, granola/bars/ energy bars, beverages/ ice-creams/frozen desserts, meat/meat substitutes/ refrigerated or frozen foods, and others	USA	2015*
Thompson et al. R5 (2016) [18]	R5	101	I	>5 to<20 ppm, 9 samples(8.9% ≥20 ppm, 5 samples (4.9%)	Breakfast cereal, spices, snacks, seasoning mix, green tea leaves, oat cereal, legume, oat fiber	USA	2016*
Bustamante et al. (2017) [27]	R5 Skerritt	962	Flours, breakfast cereals/ bars, bakery, pastry/ dough, bread, pasta, cereal snack and yeast	5–10 ppm, 28 samples (3.1%) 11–20 ppm, 17 samples (1.9%) 21–100 ppm, 26 samples (2.7%) 100–200 ppm, 13 samples (1.4%) >200 ppm, 38 samples (4.0%)	Flours, breakfast cereals/ bars, bakery, pastry/dough, bread, pasta, cereal snacks and yeast	Spain	2004-2016

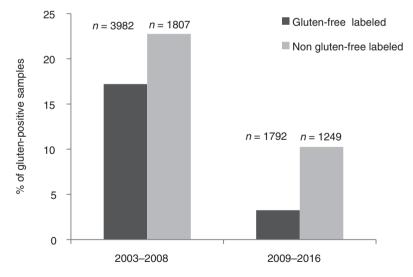
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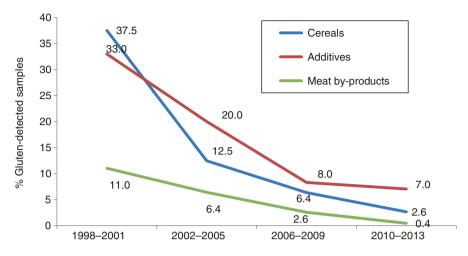
conclude that according to Valdes et al (2003), around 30% of analyzed samples were over 20 mg/kg in GF-labeled products, as well as non-labeled ones.

By contrast, two recently published studies conducted on Spanish (Bustamante et al. 2017 [27]) and North American [23] on GF claimed products indicated that highly gluten-contaminated samples were more common in non-GF-labeled products. Indeed, the positive samples percentage of apparently GF products doubles the percentage of GF-labeled ones (around 10% vs. around 5%). The explanation that could justify the discrepancy between these two studies and that carried out by Valdes et al (2003) [22] could be the Codex Alimentarius revision, that was done in 2008. Since that year, the threshold proposal of 20 mg/kg or ppm of gluten to declare a food as GF was implemented by various government regulations. This guided the food industry toward stricter hazard analysis and critical control point implementation and, probably, to the development of new composition formulas. Figure 4.1 was created by the data collected from the gluten analysis of GF research previously mentioned.

Tables 4.1 and 4.2 show how before 2008 there was a marked reduction in glutenpositive samples (over 20 ppm of gluten). It is true that the decrease was even greater for GF- labeled products (reaching 3% of samples over 20 ppm of gluten) – which is logical, taking into account that the food industry must guarantee the GF claim. However, it must be emphasized that the "apparently GF" products (those without any gluten-containing ingredient shown on the label) have lowered the glutenpositive samples to half from 2008 until 2016. With regard to gluten-contaminated samples, an overview (from 1998 to 2013) of nearly 10,000 food products [24] sold in Spain, confirmed the presupposition that commercially GF-rendered food groups



**Fig. 4.1** Evolution of gluten-positive samples (>20 mg/kg) sorted by 2003–2008 and 2009–2016 time periods for gluten-free labeled and non-labeled products. Data are expressed as the percentage of the total sample analyzed (n) in each period



**Fig. 4.2** Evolution of gluten-detected samples, expressed as % of the total analyzed, in three food groups: cereals, additives, and meat derivatives (Modified from Bustamante et al. [24])

represented by cereal-based foods, was the more contaminated with this prolamin than naturally gluten-free food in other food categories, represented by meat by-products) [24] (Fig. 4.2). Taking into account the positive samples' description of Table 4.2, this was also seen in other research. Apart from gluten contamination, it is well known that commercially GF-rendered products were poor from an organo-leptic point of view, as well as extremely expensive when compared with homologous gluten-containing breads or baked products. It is worth noting that another frequently contaminated food group is that of additives, such as paprika, cinnamon, or curcumin powder, among others.

The different standards and regulations led the food industry toward stricter hazard analysis and critical control point implementation and, probably, to new composition formulas. After the Codex Alimentarius's proposed threshold [25], detected gluten was significantly down (Fig. 3.1). In that sense, gluten traces containing cereal-based foods were reduced to around 3% and then safer GF-labelled products were manufactured. Moreover, after the EU No. 1169/2011 standard was enforced, apparently GF, but not GF-labeled products, also became less contaminated.

The achievements attained have been essential to promote safe food consumption for celiac patients and groups with gluten-related disorders. Nevertheless, work must continue in this direction. Data shows that wheat consumption per capita has increased [26], which may contribute to gluten cross-contamination of "probably safe" foods (GF-labeled products and products apparently not containing gluten). In this sense, it is important to properly train food handlers, because sometimes their knowledge of the subject is not adequate enough. Moreover, it should keep on controlling the gluten content of the "apparently" GF food, because owing to economical aspects, many people include these products in their diet instead of GF-labeled ones.

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