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ORIGINAL PAPER

Migration of plasticizers from the gaskets of lids into oily food in glass jars: a European enforcement campaign

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Abstract The results of a joint European enforcement campaign are described. The two governmental food control laboratories in Stuttgart/Germany and Zürich/Switzerland analyzed the migration of the plasticizers from the gaskets of lids into food in glass jars for 411 products collected by the authorities of 21 European countries. Of these products, 308 contained free edible oil in contact with the gasket and were considered relevant for further evaluation. In 74 of the relevant products (24 %), either non-authorized plasticizers had been used or the migration exceeded the legal limits, in some cases for several parameters and by up to more than a factor of 10. Most of the products were 1–4 years from the end of their shelf life. Taking into account that migration proceeds throughout storage, sometimes even accelerates, limits are likely to be exceeded for many more products by the end of the shelf life. Polyadipate showed the lowest migration. Promising gaskets are either plasticized exclusively with a polyadipate or with combinations of substances. Joint enforcement is cost-effective, particularly when chemical analysis is demanding; accumulation of experience facilitates the evaluation of results, and joint measures are promising to be more effective.

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Introduction

The migration from the gaskets of lids into products in glass jars has a long history of non-compliances. In 1998, Hammarling et al. [1] showed that the migration of epoxidized soy bean oil (ESBO) from the gaskets into infant foods sometimes exceeded the tolerable daily intake (TDI) of 1 mg/kg body weight (bw) [2]. Surveys performed in 1999 and 2001 by the British and the EU authorities [3, 4] revealed no improvement. According to a Swiss survey, this problem was still not solved in 2004 [5]. ESBO is also used as a scavenger to react with HCl released from PVC during curing, but the safety of the resulting reaction products, primarily cyclic fatty acids containing chlorine [6–8], was not investigated. In 2003, the migration of semicarbazide was detected, a degradation byproduct of the blowing agent azodicarbonamide [9]. In 2004, the use of 2-ethyl hexanoic acid in the stabilizer soap was published [10].

In 2004, it was noted that the migration of ESBO into oily products in glass jars like sauces (pesto, tomato sauces, herbs and olive pastes) and products in oil (such as vegetables, garlic and fish) exceeded the legal limit, even though most products were years from the end of their shelf life. Among 86 products containing at least 3 % free oil in the food and ESBO as plasticizer in the gasket, merely 2 contained less than 60 mg/kg ESBO. The mean ESBO concentration in the jar content was 160 mg/kg, the maximum 580 mg/kg. When little or no ESBO migration was detectable, the gaskets were mostly plasticized with phthalates.

In 2005, 147 out of 158 European products from the Swiss market were non-compliant [11]. In the 91 products with ESBO in the gasket, the average ESBO concentration in the food was 216 mg/kg. The gaskets of 38 products were plasticized with phthalates, the migration of which reached 740 mg/kg. Later, products from outside Europe with 1,000–2,000 mg/kg phthalates in the food were detected, among which a product with diisononyl phthalate (DINP) in an absolute amount as high as half a gram in the food (300 g) and another with 1,130 mg/kg di(2-ethylhexyl) phthalate (DEHP) [12]. A child weighing 20 kg reached the TDI of DEHP (0.05 mg/kg bw) with less than 1 g of this product per day, which meant that the content of a single jar (300 g) was sufficient to bring exposure to the TDI for almost 1 year (provided other sources of DEHP were neglected).

As it turned out that not a single producer was able to provide compliant lids for products with free edible oil in contact with the lid, the Swiss authorities temporarily lifted the legal restrictions for epoxidized soy bean oil (ESBO) [13] in order to provide a way out for the food industry, but also to prevent evasion to plasticizers more toxic than ESBO. The EU legislators had to lift some of the restrictions up to 2009 (Commission Regulation 372/2007) in order to protect the lid manufacturers and to avoid that whole ranges of products had to be removed from the market. In summer 2005, IN.CAM. (Campegine, Italy) started the production of lids for pasteurized products with a PVC gasket exclusively plasticized with polyadipate (PA), and these lids proved to systematically comply with the European legal limits [14]. There were several efforts to replace the PVC gaskets by other materials, but so far without an important impact.

The EU regulation for plastics, Regulation 10/2011, applied from May 1, 2011, provides a positive list of additives permitted in gaskets, that is, the plasticizers used must be from this list. Several plasticizers have a specific migration limit (SML). For those without, Article 11 provides a generic-specific migration limit of 60 mg/kg. Among these are compounds of low toxicity, but also a few with a TDI of 1 mg/kg bw, from which an SML of 60 mg/kg can be derived. In the past, such SMLs were not listed, as they coincided with the overall migration limit (OML). It is a relevant difference, however, whether the exceedance of the 60 mg/kg limit refers to a health concern or merely to a contamination of the food. 1,2-Cyclohexanedicarboxylic acid diisononyl ester (DINCH) is an example: It has a TDI of 1 mg/kg (EFSA [15]), which means that the generic limit is also an SML, though not listed in Regulation 10/2011.

According to Regulation 10/2011, the overall migration is to be determined in food simulants. Simulation, however, was not possible, not only because no unused lids were available, but also since there are no agreed conditions with regard to contact for lids [16]. In recognizing this, Regulation 10/2011 provides a group restriction SML(T)32 of 60 mg/kg to a list of plasticizers measured in food, that is, the sum of the migration of all these plasticizers must remain below 60 mg/kg. The SML(T)32 allows a higher migration than the OML, since it does not cover other migrants, particularly not the slip agents (which commonly migrate at 10–20 mg/kg). Hence, products exceeding the SML(T)32 for the new Regulation 10/2011 would have even further exceeded the OML in food provided by the previous legislation, where the OML referred to either food or simulants. This was of importance as it was not clear to which products the old or the new legislation was applicable.

The severe violation of legal requirements was probably related to the absence of enforcement by authorities. This lack, in turn, was linked with the demanding analytical methodology: Four methods were required to cover all plasticizers. First the composition of the plasticizers in the gasket had to be analyzed in order to identify the substances present and for which migration in the food was to be controlled. These data were also used to support that the plasticizers found in food had migrated from the lid. This method had to ensure that all plasticizers present in a gasket were detected, which was achieved by a combination of direct analysis with an analysis after transesterification [17]. Transesterification was necessary for the detection of polyadipates and ESBO, but also confirmed the identity of the other plasticizers by the reaction products.

For the measurement of migration into food, three methods were needed. For the routine determination of ESBO in food, an online high-performance liquid chromatography-gas chromatography-flame ionization detection (HPLC-GC-FID) method was devised involving transesterification directly in the homogenized jar content and measurement of the methyl esters of the diepoxy linoleic acids [18]: these esters were isolated by HPLC and measured by GC-FID. For the analysis of the monomeric plasticizers, such as phthalates and acetylated monoglycerides, a GC method with detection by mass spectrometry (MS) was developed. The solutes of interest were separated from the sample matrix (mainly oil) using injector-internal thermal desorption in a programed temperature vaporizing (PTV) injector [19]. The most difficult task was the determination of polyadipates, as these consist of mixtures varying in the alcohol moiety, the molecular mass distribution and the end capping. Furthermore, the products used in the gaskets were not available for calibration. A GC–MS method was developed involving transesterification for measuring adipic acid as butyl ester [20]. A second method was needed for the characterization of the particular

polyadipate used to determine the conversion factor for the translation of the measured concentration of adipic acid into a concentration of migrated polyadipate [21].

The European Reference Laboratory (EURL, Ispra, Italy) brought the control of plasticizers from the gaskets of lids onto the agenda and promoted the establishment of the related methods in the national reference laboratories (NRLs) [22]. However, this only partially succeeded, as it went beyond the resources of many enforcement authorities. Furthermore, it did not seem efficient to perform such intricate control separately in each EU Member State. This led to the concept of a coordinated enforcement campaign, with the analysis of all samples being performed in two laboratories. The findings are reported in this paper.

Experimental

Early in 2011, the participating 21 authorities (see "Results") collected about 20 samples each from their market. They had been instructed to select products with free edible oil in the food which may get in contact with the gasket of the lid, to store the samples in upright position at ambient temperature up to the end of August 2011 and to shake them occasionally. Then two jars of each product were sent to Stuttgart for analysis. The gasket of the lid of the first jar was analyzed for the composition of the plasticizers, from which it was deduced which plasticizer should be measured in the homogenized jar content. Plasticizers detected in the gasket at a concentration of at least 0.5 g/100 g were analyzed in food in Zurich. The methods applied were mentioned in the Introduction [17–21].

When the results were above the legal limit or within the measurement uncertainty at the limit, the second jar was analyzed; results were averaged. The uncertainty of the final result was calculated as contributions from the uncertainties of the involved methods: 25 % for the monomeric plasticizers, 20 % for ESBO and 25 % for polyadipates. At the end of November 2011, the results were reported to the authorities having sent in the samples. It was left up to them what measures to take.

Results

Compliance of a product should be tested at the end of the shelf life (up to which a product has to meet all the regulatory requirements). However, with a few exceptions, the products tested had shelf lives with expiry dates in the years 2012–2015. The production date was unknown, but it was assumed that most samples were young. By collecting the products early in the year and analyzing them starting in August, approximately half a year of age was added (by the time of analysis, no sample was at its expiration date). Nevertheless, since the migration from the gasket of lids increases continuously, sometimes even accelerating after many months [23, 24], it must be assumed that the migration at the end of the shelf life had been substantially higher than measured in this survey.

Participating countries

Nineteen EU member states as well as Norway and Switzerland sent in a total of 415 products in glass jars, usually two jars each. Table 1 lists the countries and the number of samples they delivered. Four samples were lost or leaked upon arrival. For one product, it was noticed that one of the jars was with a lid mainly containing di-(2-ethylhexyl)-phthalate (DEHP), whereas the other contained the corresponding terephthalate. The samples had the same lot number, and the lids looked exactly the same. They were counted as two samples.

Some countries focused on products of their own manufacturers, others more on the products found on their market, which resulted in good coverage of the products available in Europe (center columns of the Table 1).

As expected (and not avoidable), some samples did not fulfill the criteria set out, that is, there was no free edible oil in contact with the gasket. Some foods consisted of pastes of a consistency preventing contact with the lid. Others contained substantial amounts of fat, but the fat was inside the food, such as in sausages or olives, whereas the surrounding liquid was essentially aqueous. Finally, there were oil-in-water emulsions preventing contact of the fat with the gasket, such as mayonnaise and similar sauces.

The 103 products considered uncritical (not relevant according to the goal of the campaign) were analyzed to check whether migration was as low as expected. They were not considered for further evaluation, as it was assumed that the producers knew that these products were uncritical in this respect and had no reasons to select lids accordingly. In cases of doubt, products were classified as "relevant." In this way, 308 samples were further evaluated (second column in Table 1).

Table 1 also lists the number of samples found to be non-compliant or "uncertain." The term "uncertain" was used for products with a migration within the measurement uncertainty from one or several applicable legal limit. It is assumed that at least for most of the samples further storage would have increased the migration above the limit. The highest number of non-compliant samples was collected by Spain (8), followed by Poland (7), whereas all samples from Germany were compliant.

Table 1 Participating countries; number of samples sent in; number of samples with free oil contacting the lid ("relevant"); distinction between domestic, European (EU, Norway or Switzerland) and products from outside Europe; number of non-compliant products and of uncertain results

Country providing sample	Nr. samples	Relevant samples	Declared country of production			Non-compliant samples	Uncertain samples
			Domestic	EU, NO or CH	Others		
Austria	21	17	2	17	2	3	1
Belgium	20	13	6	9	5	2	0
Cyprus	20	10	0	20	0	3	2
Denmark	19	16	1	12	6	4	0
Finland	22	6	5	11	6	1	2
France	20	19	2	10	8	6	1
Germany	20	14	11	6	3	0	2
Greece	4	3	4	0	0	1	0
Ireland	21	12	0	11	10	6	3
Italy	20	16	12	3	5	2	5
Luxembourg	20	17	1	19	0	6	2
Malta	20	20	3	17	0	1	0
The Netherlands	20	18	7	4	9	3	1
Norway	20	15	0	15	5	2	2
Poland	20	19	8	10	2	7	1
Portugal	20	15	6	11	3	6	0
Slovenia	20	19	5	12	3	6	3
Spain	20	19	17	3	0	8	1
Sweden	20	11	6	12	2	2	1
Switzerland	19	15	0	0	19	4	0
United Kingdom	24	13	13	8	3	1	0
Sum	410	307	109	210	91	74	27

Plasticizers in the gaskets of the lids

Totally, 14 plasticizers were identified in the gaskets, listed in Table 2 with the substance number in the EU Regulation 10/2011 and the abbreviation used here. Apparently, industry changed over from a few substances applied as sole plasticizer in 2004 (mainly ESBO, DEHP and DINP/DIDP) to a broad variety and combinations.

For the products considered relevant, Table 2 also lists the number of lids containing the given plasticizer, the percentage of these lids containing it and the average concentration (% in terms of mg/100 mg) in the gasket. The plasticizers acPG, Ehol and Mesamoll were not quantitatively determined in the lids (nq). Triacetin, a rarely used additive, listed in Regulation 10/2011, but not included in the SML(T)32, was not analyzed.

The plasticizers are sorted by frequency of use. Of the 308 lids from products considered relevant, 73 % contained ESBO, some of them as the virtually only plasticizer, in many others as one of several and in some gaskets in minor concentration, probably serving as stabilizer (HCl scavenger). Polyadipates in 66 % and ATBC in 55 % of the gaskets followed. DBS was present in many gaskets, but only in small amounts, presumably added to lower the viscosity of the plastisol applied to the lid during manufacturing.

Several plasticizers were used illegally. Ehol, a mixture of 2-ethyl hexyl palmitate and stearate, is neither listed in Directive 2002/72 or Regulation 10/2011, nor has it been evaluated by EFSA. 2,2,4-Trimethyl-1,3-pentanediol diisobutyrate (TXIB) was evaluated by EFSA in 2006 [15], but the authorization in Regulation 10/2011 with an SML of 5 mg/kg was restricted to the use in single-use gloves.

Alkyl (C_{10}–C_{21}) sulfonic acid, esters with phenol (Mesamoll), was evaluated by the EFSA at the end of 2009 [25]. EU Regulation 10/2011 provides an SML of 0.05 mg/kg food and excludes its use in articles for contact with fatty foods (such as the oily products in jars). It consists of a mixture of mono-, di- and trimers, but in this survey, only the monomer was determined in food, which means that only a fraction of the migration was measured.

Most of the gaskets contained several plasticizers: The average was over 3. As shown in Fig. 1, many contained 4 or 5 plasticizers, which underlines the importance of analyzing them all. With combinations, it seems possible to reduce the migration without excessively increasing the viscosity of the plastisol, for example, resulting from the use of polyadipates (viscosity of the plastisol is limited for standard machinery).

Table 3 characterizes the plasticizers and their combinations in the gaskets of the lids, excluding those reported

Table 2 Plasticizers identified in the gaskets

Substance nr. Reg. 10/2011	Plasticizer	Abbreviation	Nr. of lids	% of lids	Average conc. (%)
532	Epoxidized soybean oil	ESBO	226	73	17
73, 797	Polyesters of hexanedioic acid with polyols, polyadipates	PA	203	66	18
138	Acetyl tributyl citrate	ATBC	169	55	12
242	Di-*n*-butyl sebacate	DBS	147	48	1.4
8	Acetylated mono- and di-glycerides	acPG	142	46	nq
783	Acetylated and hydrogenated castor oil mono-glyceride (Danisco Grindsted Soft-N-Safe)	ARMG	49	16	8.5
775	Diisononyl cyclohexane-1,2-dicarboxylate	DINCH	31	10	12
798	Bis(2-ethylhexyl) terephthalate	DEHT	6	2	22
283	Bis(2-ethylhexyl) phthalate	DEHP	4	1.3	21
207	Bis(2-ethylhexyl) adipate	DEHA	4	1.3	12
Not listed	Ester of C16:0 and C18:0 (1:1) fatty acids with 2-ethylhexylalcohol	Ehol	3	1.0	nq
728 + 729	Diisononyl + diisodecyl phthalate	DIDP/DINP	1	0.3	32
497	2,2,4-trimethyl-1,3-pentanediol diisobutyrate	TXIB	1	0.3	9.5
884	Alkyl(C10-C21)sulfonic acid, esters with phenol (Mesamoll)		1	0.3	nq

nq not quantitated

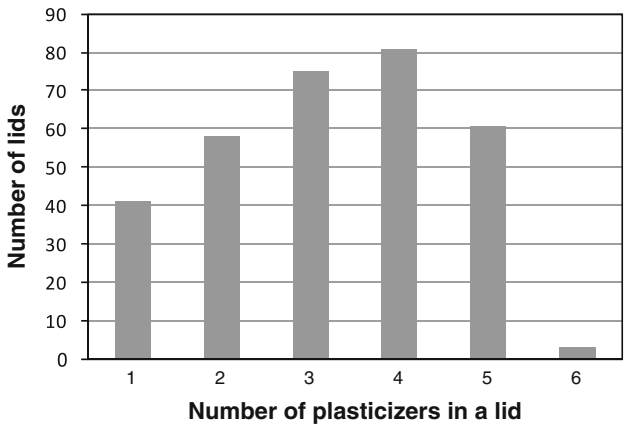

Fig. 1 Number of plasticizers detected in the gaskets of the 308 products considered relevant

in Table 4 involving non-authorized plasticizers. Components present at a concentration below 1 % related to the mass of the gasket were neglected (often DBS or plasticizers assumed to be impurities/carryovers). The first criterion applied was the number of plasticizers detected. The results show high complexity and variability.

In 20 lids of the 308 products considered relevant, gaskets were plasticized exclusively by ESBO. In another 41 lids, ESBO was combined with a minor portion of one of the other five plasticizers listed in the second line. The gaskets in 14 lids exclusively contained PA. Of the 15 lids with mainly ATBC, 14 also contained another plasticizer. A few gaskets were exclusively made with DINCH or ARMG.

In the gaskets with 3 plasticizers, either ESBO, ATBC or PA dominated. The two minor plasticizers either consisted of others of the same three, ARMG, DINCH or acPG. In the combinations with 4–6 plasticizers, the same substances were used in various proportions.

Non-authorized plasticizers were found in the 9 lids characterized in Table 4. Only 5 gaskets, all in Asian products, contained substantial amounts of phthalates. Considering that 42 products from Asia were checked, this corresponds to a strong improvement compared to a few years ago. Also, the products with Mesamoll and TXIB were from Asia, but two of the three with Ehol were labeled as Spanish and Danish, respectively.

Overall results

Figure 2 shows the sum of the migrated plasticizers included in the SML(T)32 of EU Regulation 10/2011 in terms of concentration in food (mg/kg) for the 308 samples considered relevant, sorted by increasing value. This sum exceeded 60 mg/kg for 54 % of the samples. For 34 samples, the migration was 5 times higher, for 8 samples more than 10 times, with a maximum of 1,314 mg/kg (tuna in oil).

EU Regulation 10/2011 requires recalculating migration to amounts per area of contact surface (mg/dm^2) for all packs with a content below 500 milliliters or grams (which was the case for 292 of the 308 products considered relevant), whereby the whole internal surface (i.e., including that of the glass) has to be measured. As the real ratio of contact surface to content is substantially higher than the

Table 3 Characterization of the plasticizers in the gaskets of the products considered relevant

Number of plasticizers	Nr. of lids	Plasticizers identified
1-2	20	ESBO
	41	ESBO + minor acPG, DBS, DINCH, DEHA, DEHT
	14	PA
	2	PA + minor acPG
	1	ATBC
	14	ATBC + minor PA, acPG or DBS
	3	DINCH
	2	ARMG
	4	DEHT + minor ESBO
3	32	13-29 % ESBO + 2 other plasticizers, of which
		5 14-23 % ESBO + acPG + minor ATBC or DBS
		4 17 % ESBO + 10-17 % ARMG + 7-12 % PA
		9 15-22 % ESBO, 6-10 % DINCH, 1.5 % DBS
		3 17 % ESBO + 8 % PA + 8 % ARMG
	22	20-28 % ATBC + 11-13 % PA + acPG
	15	18-35 % PA + 2 other plasticizers, of which
		6 27-34 % PA + 5 % ATBC + 1 % DBS
		5 20 % PA + 13 % ESBO + 1 % DBS
	1	20 % DINCH + 9 % DEHA + 2 % ATBC
	1	20 % DINCH + 10 % DEHT + 9 % ESBO
4	12	ESBO + PA, ARMG, ATBC, DINCH, DBS
	41	PA + ATBC, ESBO, acPG, DINCH, DBS
5	16	ESBO + PA, ATBC, ARMG + minor contributions
	44	PA + ESBO, ATBC, DINCH, acPG + minor components

conversion factor assumed by legislation (6 dm^2/kg food), evaluation by mg/dm^2 is less severe, for products in glass jars commonly increasing tolerance by a factor of roughly 2.

The internal surfaces of the packs (lids and jars) were measured and the absolute amounts of migrated plasticizers divided by this value for all samples of less than 500 ml or g content. As shown in Fig. 3, 72 (23 %) of the products considered relevant exceeded the 10 mg/dm^2 limit, 16 of them at least 5 times, 6 more than 10 times, with a maximum at 119 mg/dm^2.

Table 5 lists the non-compliances by the causes (specifying the plasticizer of concern or sum of plasticizers), the number of cases for a given cause, the legal limit in terms of mg/dm^2 (in bracket when the application was outside that authorized), the maximum migration measured in terms of mg/dm^2, the factor by which the maximum migration was above the legal limit and the mean migration in the samples exceeding the restriction.

Totally, 108 non-compliances were noted in 74 products. The most extreme case was a Thai product containing 8.1 mg/dm^2 Ehol (not authorized), 8.6 mg/dm^2 DEHP (34 times the limit, though illegal for this application), 22 mg/dm^2 DEHA (7.3 times the limit) and 10.7 mg/dm^2 ESBO (just at the limit).

ESBO was the most frequent reason for non-compliance and also responsible for the highest migration. Of the 29 products with at least 35 % ESBO in the gasket (ESBO being the exclusive or virtually exclusive plasticizer), 24 exceeded the limit. Those 5 complying were sauces with emulsified oil or stiff pastes of which it was equivocal whether they should have been considered relevant for this campaign. This confirms earlier findings that gaskets exclusively plasticized with ESBO tend to exceed the legal limit [5].

All 3 lids with high ARMG concentrations in the gasket (35–40 %, two exclusively with ARMG) and all 3 exclusively with DINCH far exceeded the SMLs. Only 4 of the 9 products with at least 30 % ATBC in the gasket exceeded the limit. However, for 3 of the 5 others, the migration was close to the limit. They had shelf lives up to the years 2014 or 2015, by which they would almost certainly have exceeded the limit. The remaining two respecting the limit were pastes with low fat content and again of unclear pertinence for this evaluation. This suggests that also ATBC is inadequate as the only plasticizer for oily products.

The migration of acPG and DBS was never above the limit because of low concentrations in the gaskets.

The migration of PA is generally low, since a large part of these mixtures is of such a high molecular mass that migration is extremely slow. All polyadipates encountered corresponded to substance number 73 in EU Regulation 10/2011 (none to substance 72). For the 14 lids exclusively with PA in the gasket, the mean migration was only 1.6 mg/dm^2. Two products were at the limit (SML(T)31) of 5 mg/dm^2 (one within the measurement uncertainty above the limit: 5.2 mg/dm^2), both with large lids on low jars, that is, with unfavorable ratio of lid size to jar content.

Table 4 Characterization of the gaskets containing non-authorized plasticizers or plasticizers not compliant for the application (in bold)

Number of lids	Composition plasticizers	Country of production
2	DEHP alone or with small ESBO and PA	India, Pakistan
1	**Mesamoll**	India
1	14 % ESBO, 5.6 % **DEHP** + 10 % DEHA + Ehol	Thailand
1	32 % **DIDP** + **DINP** + 5 % ESBO	China
1	17 % **DEHP**, 17 DEHT, 6 % ES	India
1	25 % PA + 13 % ESBO * 10 % **TXIB**	Thailand
1	38 % ESBO + **Ehol**	Spain
1	42 % ESBO, 1 % ATBC + **Ehol**	Denmark

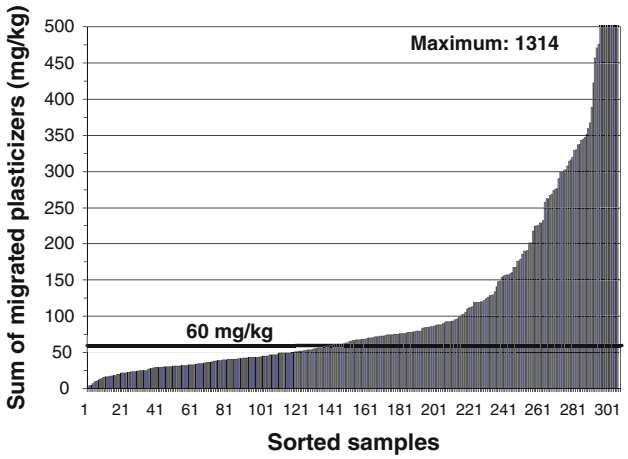

Fig. 2 The 308 products with free oil in contact with the lid ("relevant" in Table 1) sorted by increasing concentration (mg/kg) for the sum of the migrated plasticizers included in the SML(T)32 of EU Regulation 10/2011

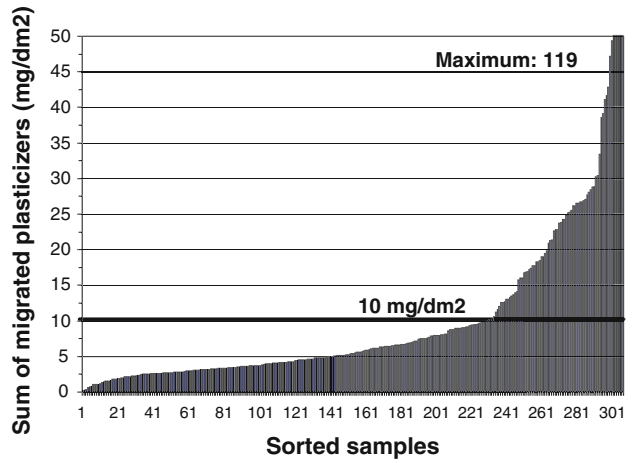

Fig. 3 Products with free oil in contact with the lid sorted by increasing migration calculated in terms of mg/dm^2 for the sum of the plasticizers included in the SML(T)32 of EU Regulation 10/2011

The SML(T)32, the migration limit for the sum of the plasticizers, was exceeded by 36 products. In 27 of these also at least one plasticizer exceeded its SML, that is, merely for 9 products the exceeded SML(T)32 was the only cause of non-compliance.

In the 27 samples listed as "uncertain" in Table 1, the migration was at the legal limit within the uncertainty of the measurement. For 20 of these, the critical parameter was the SML(T)32, for 4 the migration of ESBO and for 2 each that of PA and ATBC, respectively. For these products, the results neither showed compliance nor non-compliance. For all except of two the end of the shelf life was more than a year ahead, that is, it is assumed that they would end up as non-compliant.

Specific migration of plasticizers

Migration can be related to the content of the plasticizer in the gasket, for example by calculating the migration in terms of mg/dm^2 for a concentration in the gasket of, for example, 10 %. This classifies plasticizers by their tendency to be transferred into food (specific migration). The data obtained scatter, however, since they are from products differing in composition, age as well as jar and lid size, which is why Table 6 only includes data for plasticizers for which at least 30 data points were available. As the migration of a plasticizer also depends on the presence of other plasticizers [15, 26], only lids containing the substance at fairly high concentration were taken into consideration, the lower cut-off depending on the data available (specified in the third column in Table 6); for instance, DBS was only encountered at low concentrations. The second column lists the number of lids used for this evaluation.

The lowest migration was observed for PA, with a mean of 0.65 mg/dm^2 for a PA concentration of 10 % in the gasket and a median of 0.5 mg/dm^2/10 %. Migration also depended on the type of polyadipate: the migration of the PA from the lids exclusively containing PA, presumably all from the same manufacturer, was approximately half of that of the others—not because it was present as only plasticizer, but because it was a PA of particularly low migration.

Migration of ESBO was 4–5 times higher than that of average PAs and roughly 10 times above that of the best PA. It was similar to that of ARMG. The specific migration of ATBC resulted surprisingly low when considering its modest molecular mass: It was lower than that of ESBO. The highest specific migrations were determined for DBS and DINCH.

Non-compliance by producing countries

Table 7 lists the samples by the labeled country of production. The first column refers to the number of samples considered pertinent for this campaign (sorting criterion). It shows that Italy was the by far predominant supplier: Italian origin was declared for 118 products (38 % of the samples considered relevant). The second column shows, for instance, that 9 Italian products were non-compliant (nC), which is 8 % of those considered relevant (third column); totally 14 were non-compliant or uncertain (u; 12 %; fourth column). Among the countries of origin of a substantial number of products, lower proportions of non-compliances were noted only for Germany (no non-compliance among 17 products) and for Austria (none among 9).

Among the Asian countries having delivered several products, high proportions of non-compliance were noted

Table 5 Non-compliances by causes and extent by which high migration exceeded the legal limit

	Nr. non-compliant	Legal limit (mg/dm^2)	Maximum (mg/dm^2)	Factor above limit	Mean of non-compliants (mg/dm^2)
PA (SML(T)31)	0	5	5.2		
acPG	0	10	3.8		
DBS	0	10	7.6		
DINP/DIDP	1	(1.5)	26	17	
TXIB	1	(0.83)	12	14	
Mesamoll	1	(0.0083)	11		
Ehol	2	?	9.5		
DEHA	4	3	22	7	
ATBC	4	10	28	3	19
DEHP	4	(0.25)	8.6	34	7
DEHT	4	10	73	7	58
ARMG	5	10	29	3	22
DINCH	6	10	55	6	32
SML(T)32	36	10	120	12	30
ESBO	40	10	119	12	24

Table 6 Classification of plasticizers by specific migration: The migration measured (mg/dm^2) for a 10 % concentration of the plasticizer in the gasket

	Number of lids	Range content in gasket (%)	Specific migration (mg/dm^2 per 10 % in gasket)	
			Mean	Median
PA all	150	10–44	0.65	0.5
PA exclusively	14	40–44	0.36	0.38
PA not excl.	136	10–40	0.7	0.68
ESBO	138	10–44	4.7	2.6
ATBC	122	6–37	3.1	1.7
ARMG	26	5–40	4.8	3
DBS	46	1.5–4.3	8.9	5.6
DINCH	30	6–41	8	6

for Thailand (8 from 16 products), India (4 from 9), China (5 from 6) and Pakistan (3 from 5). In fact, of the 42 Asian products, 23 were non-compliant (55 %).

Nonetheless, the majority of the non-compliant products (48) were from European producers; 19 % of the products with declared European origin were non-compliant. The highest proportions were noted for products from Portugal (5 from 8, 63 %), Slovenia (3 from 5, 60 %), France (2 from 5, 40 %), Spain (12 from 31, 39 %) and Greece (5 from 13, 38 %).

Conclusions

The campaign had two targets. Firstly, it was an experiment to explore whether such a joint enforcement procedure would work and be as effective as anticipated. It was encouraging to note that 21 countries participated. Most authorities took action to enforce compliance with legislation, including notification in the European Rapid Alert System for Food and Feed (RASFF). However, also obstacles in national regulation hindering such joint campaigns became apparent.

Secondly, the campaign should determine whether the migration from the gaskets of lids in contact with oily foods is finally under control. Indeed, the situation strongly improved compared to 2004: Lids have become available, which seem to systematically comply with the migration limits for plasticizers at least for a limited range of lid sizes per jar content. However, the overall result is still not acceptable for enforcement: 74 non-compliant products out of 308 samples (24 %), and it should be kept in mind that most of the products still had 1–3 years of shelf life ahead. Many products exceeded the limits massively (by more than a factor of 10), and lid producers should have known that these lids will not respect the legal limits.

Table 7 Number of products by labeled country of production, number of samples considered relevant for this survey, number of non-compliant (nC) or non-compliant + uncertain (nC + u) products as well as percentages of these related to the number of relevant products from the given country

	Samples	nC	% nC	nC + u	% nC + u		Samples	nC	% nC	nC + u	% nC + u
Italy	114	9	8	14	12	Malta	3	1	33	1	33
Spain	31	12	39	15	48	Serbia	2	1	50	2	100
Germany	17	0	0	2	12	Switzerland	2	0	0	0	0
Thailand	16	8	50	9	56	Australia	1	0	0	0	0
Greece	13	5	38	7	54	Columbia	1	1	100	1	100
United Kingdom	10	2	20	2	20	Croatia	1	1	100	1	100
Austria	9	0	0	0	0	Denmark	1	0	0	0	0
India	9	4	44	4	44	Hong Kong	1	0	0	0	0
The Netherland	9	0	0	0	0	Indonesia	1	0	0	0	0
Poland	8	2	25	4	50	Lebanon	1	0	0	1	100
Portugal	8	5	63	6	75	Lietuva	1	0	0	0	0
Turkey	8	2	25	3	38	Mauritius	1	0	0	0	0
China	6	5	83	5	83	Moldavia	1	0	0	1	100
France	5	2	40	4	80	Taiwan	1	0	0	0	0
Pakistan	5	3	60	5	100	Tunisia	1	0	0	0	0
Slovenia	5	3	60	3	60	Estonia	0	0	0	0	0
Belgium	4	1	25	1	25	Finland	0	0	0	0	0
Morocco	4	2	50	3	75	Luxemburg	0	0	0	0	0
Sweden	4	2	50	3	75	Peru	0	0	0	0	0
Malaysia	3	1	33	1	33	Not declared	2	2	100	2	100

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