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TOXICOLOGICAL ASPECTS OF FOOD PAPER PACKING

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ABSTRACT
Mostly used food packing are: paper, cartoon, plastics, glass, metals and multilayer materials. The plastics and paper/cartoon make about 70% of all food packing. These two have the biggest migration potential. The use of plastics is regulated by specific EU rules, but it is not yet the same about the use of paper packing. In order to ensure health assurance for the consumers, it is needed often to make an evaluation of the migration of the contaminants from paper/cartoon packing, especially in case of recycled papers, because of the fact that a recycled paper could contain many different contaminants, what is not the case regarding the originally produced paper. Migrating contaminants could originate from basic paper packing, but also from the components added in phase of forming, as well as from graphic finalisation trough the glue, paints, lacking etc. In this article is presented an overview of the typical contaminants from paper/cartoon packing, as well as their toxicological profile.

1. INTRODUCTION
Paper/cartoon and the plastics represent more than 70% of all food packing [1]. Paper/cartoon is so called primary packing. Any of them is in direct contact with a food, and can be used as the ordinary, graphically non-treated, polymer impregnated (like polyethylene, polypropylene etc), laminated by polymer or metallic films (for example by Al) and graphically treated by print paints [2]. Some examples of paper packing applications are the flour/sugar bags bakery products, pop-corn, packing for cheese, butter, chocolate, pots and caps for liquid food, thee, coffee, cartoon boxes for freeze food, fast food, cereals, cartoon brick-pack milk and juice packing, composite snack-spices jars [3]. Paper based materials can be in a contact with a food trough some other cases, like liquid food paper filtering, baking in a special baking paper [2].
In spite of a wide use of paper materials and contacts with a food, nowadays, there are big differences about precise info on chemical composition of different paper materials and the a.m. contacts, as well as a lack of the info on migration potential and toxicological effects of their components.

Paper is a thin material produced by pressing together moist fibres of cellulose pulp, sedimented from water suspension and drying them into flexible sheets. According to Castle, a paper is porous material, the plastics are transitive materials, but a metal and glass are non-transitive ones. Generally speaking, as a material is stronger, as more non-transitive it is, as lower diffusion trough it is, as lower migration is expected from contact surface/s. A migration goes relatively easy trough porous materials, because they have moist fibres characterized by air channels and inter-space of low molecular mass. Porous materials can have the migration from both, surface and inner contacts [4]. The papers and cartoons have the highest migration, until non-porous materials have generaly lower migration than porous ones [5]. The migrations of the contaminants from the papers/cartoons to a food could go by the mechanisms of diffusion, air-migration and by so called reflex or set-off migration [6]. So called secondary and terciary (transport-type) packings are not in a direct contact with a food, but some studies indicated air-migration to a food, by the processes of the evaporation and condensation [7].

EU legal framework defines two types of the a.m. migrations – global or total and specific migrations. Global migrations are total migrations of all (each and every) migrating materials form the packing to a food. Specific migration is the migration of every migrant separately [8]. Nowadays, there are no harmonized EU rules and regulations concerned with the papers (original and recycled) for direct contact with a food, printing paints, lacquerings and glues for a food packings. Anyhow these materials need to fulfill general requirements of the act 1935/2004, article 3, and accordingly its should not release the constituents from a food, up to a harmful level for the the human health, or up to unacceptable change in a chemical composition, neither organoleptic properties of a food [9]. Some EU states established their national standards, defining the rules and regulations about human health assurance, and also declaring potential contaminants and their limitations (from the papers and cartoons). These un-unified national standards cause a lot of problems to the producers and traders in the paper and cartoon industry in EU [10]. Mostly used a.m. national standard is the one issued by Federal German Directorate for Risk Assessment [2].

2. TIPICAL CONTAMINANTS FROM PAPER/CARTOON PACKING

Migrating constituents can get out from basic packing material, as weel as from the glues, printing paints or lackering-shaping, and that is why all possible migrants from the packings to a food and their limiting parameters (ADI, TDI, SML i QMA[1]) [11], should be taken into consideration. For a proper assesment of the a.m. tocsic migrants on human health, it is primary needed to define their genotocsic impact, which could be exposed on human tissue like mutogen, teratogen and cancerogen [12]. The contacts between the packing materials and a food are identified as the main source of potential chemical degradation [13].

Recycled paper/cartoon is more produced than the originally produced one, and it means a higher health risk for the consumers [14]. Some studies confirmed a migration (in the papers/cartoons) of NIAS constituents (non intentially added supstances) with mutogen or endocrine disruptor (ED) effects [15]. NIAS could be created as by products of chemical reactions in production or recycling process between the constituents and impurities, or in disintegration of the additives [16]. Some NIAS are foreseeable and well researched, but

[1] ADI-Acceptable Daily Intake; TDI- Tolerable Daily Intake ; SML -Specific migration limit; QMA - Maximum quantity allowed

some other ones are not, what causes a sort of higher risk [17]. The studies about a paper packing published in a last decade, identified a large spectrum of migrating constituents, as follows: perfluorated compounds like 3-chloro-1, 2-propandiol, then mineral oils like photoinniciators, phenols, phthalate, methyl-naphthalene, residuals etc [2].

2.1. Poly- and perfluorated alkyl substances

Poly- and perfluorated alcyl supstances (PFAS) are the cchemicals, which do not exist in the nature and they are consisted of at least three Fluor and/or fluorated C-group [18]. PFAS are firstly peoduced about the end of 1940-ies [19], and there is an estimation that there are at least 4.700 PFAS on global market. Small bags for the muffins, micro/wave pop-corn, packing for butter and chocolate are the examples containing PFAS [20]. Addition of PFSA (for example F-surfactants) to the raw materials or as a lacquering, makes a paper oil or fat resistant. Migration of F-covalent bonded chemicals in covalent bonded lacquering is significantly lower than in a pulp. Some of PFS (Poly-and perfluorated surfactants) as the perfluorated-octan acid and dialcalated polifluoroalcyl phosphate tenzids (diPAPS) are categorized as the toxic agents and there is a doubt that they act as the endocrine disruptors (ED). Alternative chemicals which occurred on the market, like the polifluoroalcyl phosphate tenzids (PAPS), also show the effects of ED [21].

The studies of the consumers protection groupes “Safer chemicals, healthy family and a future without toxic matters” from 2018, discovered that two of five paper packings from the american big food shops contain an increased level of Fluor. The very same situation is with 11 % of the backery and ice-cream products. It is indicated that PFAS causes reproductive, liver, kidney and immunology problems on lab-tested animals. That could be connected with a low natality and tyroide problems [22].

2.2. Chloral-compounds

A lot of polichloral dibenzo-p-dioxins and polichlor dibenzofurans, all of them namely dioxins, are used in paper packing production. Majority of the studies indicate that the dioxins are highly toxic, but the most toxic is the isomer 2,3,7,8-tetrachlorodibenzo-p-dioxin. Chlorophenol, which is highly concentrated used in the cartoon production (but in recycling mode), is actually precursor for the Chloroanisol formation. The fungi which exist in a fibre type cartoon participate in a methylation of Chlorophenol. At the other hand, the Chloroanisol from the packing migrates through a layer of polyethylene in a food (like candid fruits) and causes a fungizidation of a food [23]. There is the ban on use of many plastic one-use products according to EU Regulation 2019/904, starting from 2021, including plastic straws and the alternative was a paper straw production. But Swiss researchers reported from national testing-experience, that paper straws are contaminated by Chloropropanols, mineral oils and photoinniciators. Out of 12 tested paper straws, 7 of them (58 %) had an increased concentration of dichloropropanol and/or 3-monochloropropan-1,2-diola, two straws (17 %) were contaminated by mineral oils, two of them (17%) had the migration of the photoinnitiators, and six of them (50 %) had an impact on the taste of a drink. Chloropropanols can be cancerous and can be formed during the papers production [24].

2.3. Printing paints

More than 5.000 different ingradients are used in printing paints production and only minority of them European Food Safety Authority, EFSA has researched and declared specified migration levels. Some important components of UV paints are: pigments, oligomers (epoxi acrylats, polyeter acrylats, uretan-acrylats); monomers (di-, tri-, tetrafunctional acrylats); photoinniciators (benzophenon derivates, alfa-hydroxy keton, amin-

synergists); polymer photoinniciators; additives (wax, silicon oils, stabiliyators). The dilutants on offset paints are: mineral oils, vegetable oils (lane, soya-bean, wood oils), the esters of fat acids [25].

2.3.1. Photoinniciators

Many photoinniciators are not, or are partly compaunded in printing paints, but because of low molecular mass they migrate in a food. Majority of the photoinniciators are not researched toxicologically [25,26]. The paints and lacquering for UV drying, usualy contain 5-10 % of the photoinniciators.

Most oftenly used photoinniciator is benzophenon (BP). It is used as well like an agent for a humidization of the pigments and for better viscosity of a paint. It is present in a recycled paper as well. BP migration can occur through an open structure of paper/cartoon to a food, and the studies about its toxicology indicate that it can act as a cancerogen [27, 23]. Nevertheless, the studies of BP's ED effect are not consistent. Some studies recognise ER, but at the other hand the other studies recognized low activity in ER research and a significant antiandogenic activity in some other testing [21].

IsopropylThioXanton (ITX) is a photoinniciator , which is used in UV paint and it's always mixed with cophotoinniciator (mostly EHDAB or 2-ethylhexyl-4- dimethylaminobenzoat). Italian authority for food safety discovered in 2005 a migration of ITX (i EHDAB) to UHT baby milk, packed in the cartoon laminated by plastic folia. ITX level was from 120-445 ppb in baby (over 12 months) milk – in one sample of the aromatized milk, ITX was found 600 ppb. Very similar situation was with chocolate cocoa, milky products from Austria and Germany, and in pomegranate/ananas juice, produced in Italy. NESTLE banned the distribution of these product and asked the provider of that packing not to use ITX any more, which accepted and declared that [28].

2.3.2. Heavy metals

Heavy metals exist in the printing paints. English Association of Printing Paints Producers declared that heavy metals as Titanium, Chromium, Molybdenum and Iron are used as the pigments in printing paints, until Cobalt and Manganese are used as the driers, Titanium oxide is used for so called pearl pigmenting [26]. The pigments based on Zinc and Cadmium give a sort of fluorescent property to a paper and at the same time increase the strength of of some lacquering on on a paper surface [29].

Yellow and green paints for the papers have the compounds like Led-chromate, Led-sulphate and Led-oxide. Cupper is the ingredient of blue and green paint. Led is usually used in white paints, but also in red, yellow and green ones [29].

Heavy metals are harmful regarding human health. Led is carcinogen, is harmful for the lungs, for immune system and especially for the children nervous system. Cadmium is carcinogen and is easy to be sediment in the lungs, kidneys, heart, as well as poisons the bones and lymphatic system. Six-valence Chromium is harmful to the skin, liver, kidneys and lungs, causing different diseases like lungs carcinoma, perforation in nose, dermatitis etc. Overdosing of Cupper can cause diaries, kidney problems, blood and nervous system malfunctioning [31].

2.3.3. Mineral oils

Carbon-hydrates from mineral oils (MOH) are complex mixture of Carbon-hydrates from raw mineral oils [32]. MOH are divided in two groups: saturated Carbon-hydrates from MOSH

and aromatic Carbon-hydrates from MOAH. MOSH are alkaline and alkyl-subsidized cikloalkans, until MOAH are poly-aromatic alkyl-subsidized Carbon-hydrates. MOAH can be cancerogen and mutagen and they enter in contact with a food through the additives, paints, processing equipment or by using recycled paper [33]. Two studies from 2010, carried out by Swiss researchers confirmed that primary sources of MOH are the paints and recycled paper/cartoon, due to low quality of recycling process. In such a way MOH enter a food; it takes a few days trough cartoon, or a few weeks-months trough polyethylene (PE). It is accepted that MOAH have cancerogen effect because of its structure, which is similar to polycyclic aromatic Carbon-hydrates. That is why MOAH is to be avoided. High fat content as well as high temperatures during where-housing or distribution, can enhance a migration of MOH in a food [32]. After MOH identification in New Year chocolates in 2012, German Federal Institute for Risk Assessment (BFR) warned that some aromatic Carbon-hydrates could be cancerogen. In the a.m. case a chocolate was in touch with a cartoon, without any barrier in between, and MOH easily migrated through a cartoon [34]. EFSA has an estimation that the humans get daily from 0,03 – 0,3 mg MOSH per kg of body mass, and the children even more. At the other hand, the elderly get 0.005 – 0,06 mg MOAH per kg of body mass. EFSA considers that as a critical level, because of their mutogen and cancerogen potential. In spite of it, still there no in EU rules any official limitation in MOSH and MOAH migration, or the limitations in content of the a.m. compounds in the paints [33].

2.3.4. Phthalates

Phthalates are most often group of compounds in printing paints, glues and lacquering for food packing. They are always present in recycled paper/cartoon. They can be used as well as the paper softeners [35]. Di- butyl-phthalate (DBP) i di (2-ethylhexsil) phthalate (DEHP) are mostly used phthalates in the paper/cartoon packing. Phthalates migrate through the paper/cartoon packing and contaminate a food. DBP and DEHP show some ED effects on the mousses, causing more problems to the females than to males. Metabolitates DEHP and the other phthalates are found in the urine both, elderly and children [36]. There are also some proofs that the phthalates decrease the activity of lipids metabolism [37,21].

2.4. Phenols

The use of phenols in the paper/cartoon industry vary from bisphenol A (BPA), which is used in a termic paper, up to the pentachlorophenol, as a biocide in the paper production. Oktilphenol, 4-nonilphenol and 4-terc-oktilphenol are used in polymer resin, which are used in a preparation of the paints, and nonilphenol is a part of some paints based on epoxy resins, glues etc. BPA is also found in recycled paper [38,39]. BPA is thoroughly researched because of a number of the toxicological effects ED [21].

2.5. Glues

The glues in paper-carton packing are often of complex composition comprising glue components and modification components, like glue, solidifier, inhibitor, dilutor, plastificator, flexibiliator, antioxidant, surface layer and humidization agent [40].

The researcher Aznar with its team analyzed 45 paper/cartoon glue market samples (29 different formulation), and their lamination with plastic films, detecting four components in these glues, being on top of toxic list according to Kramer [41].

2.6. Dispersators

During the recycling, majority of the papers/cartoons undergo deinking of the paints, glues and the other contaminants. It is highly probably that some chemicals (like Na-hydroxide,

Na- silicate or H-peroxide in a combination with a dispersion matter like stearin acid) for a deinking, will remain in the fibres of recycled papers and will come to a food [42].

2.7. Dimensioning matters

Dimensioning matters for a paper, like abietic acid (AA) i dehydro-abietic acid (DHAA) are added to enhance the absorption and/or fragility of paper fibres and to increase printing properties. Derivates AA are also used in the lacquering and glues for the papers/cartoons. Some studies identified the resin acids AA and DHAA in the packing of recycled paper/cartoons as genotoxic [41,21].

2.8. Nitrozamins

Nitrozamins are genotoxic cancerogen matters which could be found in a food. They come to a food from the papers and waxed cartoons. N-nitrosomorpholine and morpholine migrate in a longer period of time, from the a.m. matters to a food [23].

2.9. Methyl-naftalen

Kellogg Company withdrew in 2010 about 28 millions cereal boxes because of an increased level of methyl-naftalen, which got out from the paper boxes to a food. The exact negative effects of the consumption in the a.m. case are not defined, but many consumers were complaining about it and had some health problems [43].

3. RISK FROM RECYLED PAPER/CARTOON

In the year 2017 more than 71% of the paper and cartoon were recycled in Europe. Cellulose fibres could be recycled up to seven times, before they become too short for that use [17]. Paper and cartoon which are produced partly or totally from recycled fibres, are already in use in Europe. That is very characteristic in fast food industry of many European countries. Recycled paper is mostly used in direct contact with dry food, like flour, cereals, rice, spaghetti, eggs, fruits and vegetables [10]. The list of ponential migrants from the paper/cartoon containing recycled fibres is much longer, than in the case of originaly produced ones. Besides of all migrants from the originaly produced papers, the additives also come on the a.m. list , and they are used to be added in a process of the recycling, but especially NIAS matters. Recycling process can bring even several unknown NIAS compounds in the packing. For example in a mixture of the papers for recycling. It could be paper packing used for a food packing, and after an use a consumer put some uneatible items in it and some migrants from these items enter in a paper. During the recycling process the a.m. migrants will enter newly produced food packing [44]. Recycled papers are a source of known NIAS, among them are often present phthalates, paint components, photoinniciators and mineral oils [35,26,45]. NIAS components in the paints could emerge from the raw materials for the paints, from drying and solidifying process, from the interaction with the other components of a packing or from mutual contamination in a production or in use of a paint [17].

Multilayered recycled papers/cartoons have above of recycled fibres layer, another layer of original primary fibres which is in touch with a food. Such a packing have the advantage that the original (clean) primary fibres layer is in touch with a food, but still it does not fully prevent possible contaminants migration from the recycled paper fibres layer. In that case it is advisable to use an Al-folia or a polymer film as a safety barrier. It is still in process of research if a polyethylene film is an adequate barrier for evaporating components from the recycled papers/cartoons, since it is known that polyethylene is transparent for some evaporating compounds [21]. Although a recycled cartoon is used for secondary or terciary

food packing, which is not in direct contact with a food, still it is possible a migration of evaporating compounds or an air migration through a transparent primary packing [33].

4. MIGRATION INFLUENCING FACTORS

There are several migration influencing factors like: type of contact (direct or indirect), type of food, packing material properties, concentration and characteristic of the migrants, time and temperature of a contact. Direct contact, bigger contact area, longer time and higher temperature make a bigger potential of a migration. The migrations are more intensive in case of liquid or semi-liquid food, as well as with fat-food. Higher concentration of the migrants will increase a migration level. Migrant constituents of high molar mass have a lower migration level, than the ones of lower molar mass. The migrants with a complex type of configuration (spherical oriented molecules, side-chained molecules) have a lower migration level. Migration from a paper is much faster than a migration from a plastic. There is a higher level of a migration from thin packing, than from a thick one. Porous structure of a paper, more additives in production/recycling and NIAS components (all of them) increase migration potential [5,6,23].

5. CONCLUSIONS

The papers and cartoons have an important application role in a food industry and it will be increased further. Paper packing which will be in direct contact with a food, need to be produced in accordance with a high hygienic standard and should not chemically react with a food, neither release any chemical substances and influence a food properties. Migration potential from these packing is high. That is why at least EU should prescribe a sort of harmonized legal framework for the papers/cartoons regarding the limits in a specific contaminants migration, from these packing, as well as for the paints and glues used in that packing production industry. There is a trend of substitution of one-use plastic packing by paper ones and that is additional reason for the a.m. EU action, as well as an increased level of a paper recycling.

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