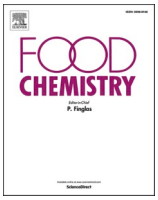
Food Chemistry 499 (2026) 147307

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem



From soil to fork: Are mulch films releasing additives to the soil and contaminating our food?

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A B S T R A C T

Climate change and rising food demand have increased plastic mulch use to boost yields; however, mulches release additives like phthalates, potentially contaminating soil and crops. This study examined additive transfer from conventional and biodegradable mulches to soil and strawberries over a two year-cycle. Substrates were collected at four times: before mulching (T0), first harvest (T1), six months after mulching (T2), second harvest (T3). Phthalate levels peaked at T2 (up to 746.18 ± 33.33 μg/kg) and declined at T3. Contrary to their eco-friendly image, PBAT released additives at levels comparable to polyethylene. Polypropylene and oxo-degradable polyethylene mulch consistently led to the highest contamination. Significant differences in di(2-ethylhexyl)-phthalate, dibutyl-phthalate, and diethyl-phthalate concentrations were found between treatments and controls. In strawberries, only dibutyl-phthalate (up to 164.32 ± 81.58 μg/kg) and acetyl-tributyl-citrate (up to 8.37 ± 2.68 μg/kg) were detected, with low estimated noncarcinogenic risk. The study urges assessment of mulch pollution and shows film types differ in additive release.

1. Introduction

Ensuring global food security and supply without compromising environmental protection is one of the greatest challenges of our time (Cui et al., 2024). While the effects of climate change are exacerbating water scarcity, restricting crop yields, and leading to substantial economic losses, enhancing agricultural productivity is essential to satisfying the food demands of a growing global population. Given these challenges, optimizing conventional farming techniques and integrating them with innovative approaches is crucial not only to fulfilling food demands but also to safeguarding natural resources and supporting environmental sustainability (Somanathan et al., 2022).

In the last decades, the agricultural sector has increasingly relied on plastic materials to boost crop yields and reduce production costs. Among these, plastic mulch films have become particularly widespread, with their annual usage reaching 6.5 million tons (Giacomo Scarascia-Mugnozza et al., 2012; Yates et al., 2021). Mulching is a widely adopted agricultural technique that enhances soil conditions, promotes crop growth and helps retain soil moisture, reducing irrigation needs (Liu et al., 2022; Wan et al., 2023; Yang et al., 2023; Zhang et al., 2022). Plastic mulches are often favoured over alternatives such as straw, grass, maize starch, paper, sawdust, or gravel, due to their affordability, flexibility, durability, and ease of application. Their performance and longevity are further enhanced by additives such as plasticizers, colour pigments, and ultraviolet stabilizers (Hahladakis et al., 2018).

The use of phthalates, also known as phthalic acid esters (PAEs), in plastic production has raised significant environmental and health concerns (Negev et al., 2022; Smith et al., 2022; Xie et al., 2016). These compounds, widely incorporated into plastic mulch films to enhance their flexibility and durability, pose a significant challenge due to their environmental persistence and potential toxicity (Hofmann et al., 2023; Zhou et al., 2023). Because these compounds are not chemically bound to the polymer structure, they can easily migrate from plastics into the environment (Amritha et al., 2022), posing a serious concern for agriculture, as PAEs present in mulching films can leach into the soil and be absorbed by crops, ultimately compromising food safety (Scopetani et al., 2023, 2024; Wang, Yu, et al., 2021). Indeed, PAEs are endocrine disruptors associated with various adverse health effects (Tuan Tran et al., 2022). Notably, di(2-ethylhexyl) phthalate (DEHP) has been identified as a major contributor to an increased risk of miscarriage,

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https://doi.org/10.1016/j.foodchem.2025.147307
Received 30 July 2025; Received in revised form 4 November 2025; Accepted 24 November 2025
Available online 25 November 2025

exhibiting a dose-response relationship (Ji et al., 2024). Additionally, research has found that higher maternal concentrations of monoethyl phthalate are linked to lower total gray matter volumes in 10-year-old children (Ghassabian et al., 2023). Exposure to PAEs has also been associated with infertility (Abdo et al., 2023), childhood neurodevelopmental disorders, obesity (Braun, 2017), and fatty liver disease (Foulds et al., 2017). Recognizing their risks, both the European Union and the United States Environmental Protection Agency have classified PAEs as priority pollutants, and their use in materials intended for prolonged contact with skin or mucous membranes is regulated by the European Commission Regulation 2018/2005.

In addition to the leaching of PAEs and other additives from intact mulch films, their end-of-life management presents challenges. Inadequate removal practices can lead to their accumulation in agricultural fields, while even properly discarded films often leave microplastic residues in the soil. Over time, these persistent fragments contribute to soil contamination and exacerbate the global plastic pollution crisis (El-Beltagi et al., 2022; Salama & Geyer, 2023). Once released, plastic debris can remain on the surface, where it undergoes further degradation by sunlight, or bury in the soil, causing long-term accumulation. In both cases, the slow degradation processes of the fragments will lead to the formation of microplastics and nanoplastics (Kasirajan & Ngouajio, 2012; Serrano-Ruiz et al., 2021).

With the purpose of limiting plastic pollution in terrestrial environments, biodegradable mulches have recently been introduced to the market. One of their primary commercial advantages is their supposed ability to biodegrade in the soil over time, potentially eliminating the need for removal, unlike conventional plastic mulches. This same commercial advantage raises environmental concerns as little is known about the substances these materials may release. Moreover, a few studies conducted on this topic indicate that biodegradable mulch films may release even greater amount of the same additives, including phthalates, than those found in conventional plastic mulches (Scopetani et al., 2025; Uzamurera et al., 2023). Because soil-biodegradable mulches can remain in agricultural fields, they may contribute to a greater release of additives into the soil which could then be absorbed by cultivated crops and pose a risk to human health. This risk is further amplified by their putative faster breakdown rate and shorter decomposition period compared to traditional mulches.

Due to limited research on this topic and the widespread global use of both conventional and biodegradable mulches, there is an urgent need to gather data on the release of contaminants from these materials into the soil and their potential transfer to edible crops (Yates et al., 2021).

This study aims to fill this knowledge gap by evaluating the role of conventional and biodegradable mulch films in transferring chemicals to agricultural soil and crops, assess the fate of contaminants in the edible part of the plants, and determine whether these mulches pose a risk to terrestrial ecosystems. Additionally, it seeks to identify any significant differences between conventional and biodegradable mulches in terms of contaminant transport. To achieve this, a small-scale experiment was designed to replicate the strawberry cultivation cycle, where strawberry plants were grown for two years using four different mulch materials: polyethylene, oxo-degradable polyethylene, corn starch with polybutylene adipate terephthalate, and polypropylene. This study offers a deeper understanding of how these materials interact with their environment, addressing critical knowledge gaps and providing valuable insights to guide future agricultural practices.

2. Materials and methods

2.1. Chemicals

Phthalates (dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DBP), butyl benzyl phthalate (BBP), bis(2-ethylhexyl) phthalate (DEHP), di-n-octyl phthalate (DnOP)) and mono(ethylhexyl) phthalate (MEHP) (phthalate mix, O2Si Smart Solutions, North Charleston, SC, USA), acetyl tributyl citrate (ATBC) (Chiron, Trondheim, Norway), di-n-butyl sebacate (DBS) (Chiron, Trondheim, Norway), and tris(2-ethylhexyl) phosphate (TEHP) (Chiron, Trondheim, Norway) were used as standards for the calibration curve. All standards were certified reference materials (TraceCERT®) with a purity ≥99.0 %. DEHP-d4 (Chiron, Trondheim, Norway) and ATBC-d3 (Toronto Research Chemicals, Toronto, Canada) were used as internal standards. The internal standards had an initial concentration of 1000 μg/mL. For sample preparation, 100 μL of a working solution, diluted by a factor of 1000 from the original internal standard solution, were added to each sample. Hexane and acetone (Honeywell Research Chemicals, US, purity >99.8 %)were used for soil extraction. Soil sample clean-up was performed using columns of anhydrous sodium sulfate (Chem-Lab, Belgium). Agilent QuEChERS Extraction Kit, Original Method together with a dispersive SPE clean-up product (Thermo Scientific™ 60,105–205) was used to extract strawberry samples.

2.2. Experimental set-up

A small-scale experiment was conducted by growing strawberry plants (*Fragaria L.,* Camarosa cultivar) outdoors in clay pots, simulating field conditions and using shade nets. Each type of mulch film (50 cm^2) was assigned to five different pots (dimensions: 45 × 45 × 45 cm; volume: 91 L; supplied by Agraria di Vita s.r.l., Pescia, Italy). Each clay pot served as a replicate, with five replicates for each mulching condition: polyethylene (PE, thickness 0.05 μm), oxo-degradable polyethylene (Oxo-PE, thickness 15 μm), corn starch and polybutylene adipate terephthalate (PBAT, thickness 18 μm), and polypropylene (PP, thickness 450 μm). These materials were sourced from agricultural suppliers and chosen based on their widespread use, selecting the ones most utilized by farmers. The initial content of additives in the mulch films was previously characterized and is reported in our recent publication, based on analyses performed using two extraction approaches: release in water and extraction with organic solvents (Scopetani et al., 2025). The experimental design followed a randomized block statistical scheme. After filling the pots with the cultivation substrate(purchased from Agraria di Vita s.r.l., Pescia, Italy), a 1:1 *v*/v mixture of peat and lapillus (with lapillus particles sized 10/18 and pH = 6.2), the different mulching materials were applied to the surface to cover the peat. The cultivation substrate had the following physicochemical properties: Total Organic Carbon (TOC) = 4.83 %; Total Nitrogen (TN) = 0.08 (%); C/N ratio = 59.23; pH = 6.23; EC = 0.04 dS/m; CEC = 22.17 meq/100 g. Five pots were kept as controls without any mulch film.

The experiment was conducted between April 2023 and May 2024 in the experimental field of the Department of Agriculture, Food, Environment and Forestry (DAGRI), University of Florence, located in Sesto Fiorentino, Italy (55 m a.s.l.; 43°81′68″N, 11°19′99″E). The area is characterized by a typical Mediterranean climate, with hot and dry summers and mild, rainy winters. The selected strawberry plants were biennial, ensuring two fruiting seasons. Substrate samples (top 10 cm) were collected and freeze-dried at different time points: at the beginning of the trial (T0, mid-April 2023), during the first fruit harvest (T1, mid-June 2023), six months after mulch film application (T2, mid-October 2023), and during the second fruit harvest (T3, mid-May 2024). Strawberry samples were harvested and freeze-dried at full ripeness in June 2023 (T1) and May 2024 (T3).

During the experimental period, climatic conditions varied considerably. The warmest and driest phase occurred between mid-June and mid-October 2023 (T2), with mean maximum and minimum temperatures of 30.0 °C and 18.9 °C, respectively, and total precipitation of 53 mm. The coolest and wettest period extended from mid-October 2023 to mid-May 2024 (T3), showing mean maximum and minimum temperatures of 16.4 °C and 7.7 °C, and cumulative rainfall of 692.8 mm. The intermediate phase between mid-April and mid-June 2023 (T1) was characterized by moderate temperatures (mean maximum 23.1 °C; minimum 12.9 °C) and by a cumulative rainfall of 157.6 mm compared

to summer.

2.3. Extraction of the substrate and strawberry samples

For substrate extraction, the procedure described by (Aparicio et al., 2007; Scopetani et al., 2023, 2024) was followed, with slight modifications. To analyse a representative portion of the substrate, the sample was collected using the quartering method. Approximately 2 g of substrate was then transferred into 50 mL glass bottles, to which 20 mL of acetone:hexane (1:1 *v*/v) mixture and 100 μL of the internal standards (DEHP-d4 and ATBC-d3) were added. The samples were vortexed for 1 min, sonicated for 30 min at room temperature, and subsequently centrifuged for 5 min at 300 rpm. The extraction procedure was performed in triplicate. Clean-up was carried out using an anhydrous sodium sulphate column, and concentration was achieved using a rotary evaporator. The samples were then concentrated to 1 mL under a gentle ultra -pure nitrogen stream, transferred into vials, and stored in the freezer until analysis.

After being homogenized, freeze-dried strawberry samples were extracted and purified using the QuEChERS method. Approximately 1 g of the sample was placed in a 50 mL tube, and DEHP-d4 and ATBC-d3 were added as internal standards, along with 10 mL of an acetone: hexane (1:1 v/v) mixture. The sample was vortexed for 1 min, after which the QuEChERS extraction salt mixture was added. The sample was vortexed again for 1 min and then centrifuged for 5 min at >3000 rpm. For the clean-up step, a dispersive SPE (Solid Phase Extraction) clean-up product was added, and the sample was vortexed for 1 min, followed by centrifugation for 5 min at >3000 rpm. The supernatant was collected, concentrated to 1 mL under a gentle nitrogen flow, and stored in the freezer until analysis.

2.4. GC–MS analysis

The selected plastic additives were analysed with an Agilent Technologies 6890 N (G1530N) gas chromatograph with an Agilent Technologies 5973 inert mass spectrometer and a DB-5MS 30 m, 0.250 mm, 0.25 μm internal diameter gas chromatograph column. The mass spectrometer was operated in selected ion monitoring (SIM) mode.

The ion source was maintained at 230 °C, while the quadrupole temperature was set to 150 °C. The temperature program started at 50 °C, increasing at a rate of 30 °C/min until reaching 280 °C, then at 15 °C/min up to 310 °C, with a total analysis time of 14.67 min. The injection was carried out in splitless mode using 1 μL of sample. The carrier gas (He) was kept at a constant flow of 1 mL/min.

Standard solution for calibration curves was stored at −20 °C. The calibration curves were prepared the same day of the analysis, and they ranged from 0.0001 to 1 ng/g for phthalates, ATBC, DBS, and TEHP. The limits of detection (LOD) and quantification (LOQ) were determined through replicated analyses ($n = 5$) of procedural blanks (Table S1). The LOD was calculated using a calibration slope of 3.8σ/slope (10σ/slope for LOQ), where σ represents the standard error of the regression, following the methodology proposed by the European Union (Thomas et al., 2016).

All the calibration curves had a data population of 10 points, with a correlation coefficient $R^2 \geq 0.991$ for each congener. The recovery range of the analytes was 91 %–112 %.

2.5. Quality assurance/quality control (QA/QC)

The field experiment was designed to minimize plastic exposure as much as possible. Strawberry plants were cultivated in clay pots, with five pots maintained as field controls. To avoid laboratory contamination from plastic additives, plastic tools were avoided, and all glassware and equipment underwent a thorough rinsing procedure: first with ultrapure water (18.2 MΩ), followed by acetone, and finally hexane. Throughout the analysis, procedural blanks were conducted at each stage to identify potential contamination sources, with necessary corrections applied as needed.

2.6. Statistical analysis

The collected data were analysed for normality and homogeneity using the Shapiro-Wilk and Levene tests with Origin 2024 (OriginLab) software. Since the data did not meet the normality criteria, non-parametric tests were used for comparisons. These included the Kruskal-Wallis test followed by Dunn's test. Differences between data sets were considered significant with a *p*-value lower than 0.05.

3. Results

3.1. Additives in substrate samples

In the substrate collected at T0 and T1, all the selected additives were detected across all treatments (controls included) except for BBP, DnOP, MEHP, and TEHP (Table 1). DMP was present in all treatments but absent in the controls and in T0. A similar pattern was observed in the substrate collected at T2 and T3, with the additional absence of DMP in all treatments (Table 1). (See Fig. 1.)

The mean concentration of ΣPAEs varied across substrates, ranging from 101.36 ± 4.48 to 781.34 ± 200.14 μg/kg in T1, 271.76 ± 38.09 to 746.18 ± 33.33 μg/kg in T2, and 114.40 ± 7.29 to 290.25 ± 39.57 μg/kg in T3. A general increase in PAE concentrations was observed from T1 to T2, with the Dunn test indicating this difference was significant for the control group ($p = 0.014$) and the PE-mulched substrate ($p = 0.014$). This was followed by a decrease from T1 to T3, which was statistically significant for the PBAT -mulched substrate ($p = 0.009$). In addition, the PP mulched substrate showed a significant increase in concentration from T0 to T1 and from T0 to T2 ($p = 0.021$ and $p = 0.020$, respectively). At all-time points, the highest concentrations were observed in substrates mulched with PP. In control samples, concentrations ranged from 101.36 ± 4.48 to 271.76 ± 38.09 μg/kg, generally the lowest among all treatments. Similarly, the T0 substrate had a mean concentration of 107.79 ± 6.23 μg/kg, closely aligning with the control samples, particularly at T1 (101.36 ± 4.48 μg/kg) and T3 (129.06 ± 14.11 μg/kg). Among individual PAEs, DMP was detected only in T1 and exclusively in the mulch treatments, while it was absent in control samples as well as in T0, T2, and T3. The most abundant congener across all time points was DEHP. ATBC concentrations were notably higher in T0 (86.74 ± 4.22 μg/kg) compared to all other treatments, where they ranged between 18.75 ± 2.78 and 0.65 ± 0.20 μg/kg. A steady decline in ATBC levels was observed from T0 to T1, and further from T1 to T2 and T3. The Kruskal–Wallis test followed by the Dunn test revealed significant differences from T0 to T2 and from T1 to T2 in the controls ($p = 0.040$ and $p = 0.024$, respectively), from T1 to T2 in the PE mulched substrate ($p = 0.014$), and from T1 to T3 in the PBAT, Oxo-PE-, and PP mulched substrates ($p = 0.009$, $p = 0.004$, and $p = 0.006$, respectively).

In contrast, DBS concentrations showed a significant decrease only from T1 to T2 in the controls and PP mulched substrate ($p = 0.021$ and $p = 0.019$, respectively).

Statistical analyses revealed significant differences at different time points. At sampling time T1, the Kruskal-Wallis test indicated overall differences across treatments, with the Dunn test confirming significant differences in ΣPAEs concentrations between the controls and the substrates mulched with Oxo-PE ($p = 0.033$) and PP ($p = 0.002$). In this text, where the Dunn test indicates a significant difference between two treatments, the second treatment mentioned has the higher concentration compared to the first. Significant differences were also detected in DEP concentrations between the controls and the PBAT mulched substrate ($p = 0.030$), in DBP concentrations between the controls and the Oxo-PE mulched substrate ($p = 0.022$) and the PP mulched substrate ($p = 0.002$), and in DEHP concentrations between the controls and the PP mulched substrate ($p = 0.004$). However, no significant differences were

Table 1
Concentration of additives extracted from substrate samples (μg/kg) - Mean ± Standard Error ($n = 5$). LOD stands for limit of detection. * Indicates a significant difference compared to the controls.

		Σ PAEs	DMP	DEP	DBP	DEHP	DBS	ATBC
T0 substrate		1.08E+02 ± 6.23E+00	<LOD	8.98E+00 ± 1.96E+00	1.80E+01 ± 2.30E+00	8.00E+01 ± 1.01E+01	2.03E+02 ± 1.23E+01	8.67E+01 ± 4.22E+00
	C	1.01E+02 ± 4.48E+00	<LOD	5.81E+00 ± 8.46E-01	1.03E+01 ± 9.57E-02	8.53E+01 ± 4.75E+00	1.47E+02 ± 2.80E+01	1.21E+01 ± 6.05E-01
	PE	1.25E+02 ± 1.09E+01	5.46E-01 ± 2.96E-01	7.36E+00 ± 1.04E+00	1.48E+01 ± 2.44E+00	1.02E+02 ± 8.15E+00	1.77E+02 ± 1.55E+01	1.35E+01 ± 1.05E+00
T1 substrate	**PBAT**	1.51E+02 ± 2.73E+01	6.39E-01 ± 3.32E-01	1.16E+01 ± 8.76E-01*	1.87E+01 ± 4.04E+00	1.20E+02 ± 2.27E+01	1.17E+02 ± 2.29E+01	1.39E+01 ± 1.25E+00
	Oxo-PE	3.51E+02 ± 1.03E+02*	1.43E+00 ± 4.28E-01	8.58E+00 ± 9.20E-01	5.31E+01 ± 8.62E+00*	2.88E+02 ± 9.48E+01	1.18E+02 ± 3.08E+01	1.67E+01 ± 2.25E+00
	PP	7.81E+02 ± 2.00E+02*	1.08E+00 ± 5.23E-01	1.17E+01 ± 2.47E+00	9.26E+01 ± 2.12E+01*	6.76E+02 ± 1.78E+02*	9.14E+01 ± 1.13E+01	1.88E+01 ± 2.78E+00
	C	2.72E+02 ± 3.81E+01	<LOD	8.09E+01 ± 1.35E+01	5.30E+01 ± 1.66E+01	1.38E+02 ± 2.25E+01	3.76E+02 ± 1.60E+01	6.52E-01 ± 2.00E-01
	PE	7.38E+02 ± 1.14E+02	<LOD	1.16E+02 ± 1.54E+01	1.46E+02 ± 5.29E+01	4.76E+02 ± 8.34E+01	3.54E+02 ± 4.88E+01	1.42E+00 ± 5.89E-01
T2 substrate	**PBAT**	6.29E+02 ± 6.81E+01	<LOD	9.70E+01 ± 1.29E+01	9.51E+01 ± 2.75E+01	4.37E+02 ± 5.26E+01	3.09E+02 ± 3.70E+01	1.97E+00 ± 3.64E-01
	Oxo-PE	6.02E+02 ± 2.06E+01	<LOD	1.09E+02 ± 2.06E+01	7.52E+01 ± 2.15E+01	4.17E+02 ± 8.56E+01	3.51E+02 ± 3.24E+01	5.18E+00 ± 1.70E+00*
	PP	7.46E+02 ± 3.33E+01	<LOD	1.72E+02 ± 2.91E+01	9.90E+01 ± 2.47E+01	4.76E+02 ± 2.23E+01	3.90E+02 ± 4.32E+01	2.87E+00 ± 4.23E-01
	C	1.29E+02 ± 1.41E+01	<LOD	2.22E+01 ± 6.13E+00	1.30E+01 ± 1.46E+00	9.38E+01 ± 1.73E+01	2.34E+02 ± 1.37E+01	9.47E-01 ± 1.07E-01
	PE	1.14E+02 ± 7.29E+00	<LOD	2.63E+01 ± 4.79E+00	2.05E+01 ± 1.81E+00	8.68E+01 ± 1.47E+01	2.77E+02 ± 3.83E+01	1.93E+00 ± 5.60E-01
T3 substrate	**PBAT**	1.26E+02 ± 1.62E+01	<LOD	2.71E+01 ± 9.08E+00	1.50E+01 ± 2.60E+00	8.37E+01 ± 1.29E+01	1.62E+02 ± 2.30E+01	8.39E-01 ± 2.72E-01
	Oxo-PE	1.49E+02 ± 3.49E+01	<LOD	1.47E+01 ± 3.43E+00	1.65E+01 ± 1.51E+00	1.18E+02 ± 3.37E+01	2.52E+02 ± 3.02E+01	9.31E-01 ± 2.57E-01
	PP	2.90E+02 ± 3.96E+01	<LOD	1.94E+01 ± 6.49E-01	1.86E+01 ± 2.82E+00	2.52E+02 ± 4.20E+01	2.68E+02 ± 1.57E+01	2.25E+00 ± 2.47E-01

found for ATBC and DBS concentrations.

At sampling time T2, the Kruskal-Wallis test suggested no significant differences in ΣPAEs ($p = 0.045$) and DEHP concentrations ($p = 0.046$). Conversely, the Dunn test indicated a significant variation in ATBC concentrations between the controls and the Oxo-PE-mulched substrate ($p = 0.028$).

At T3, the Kruskal-Wallis test identified no significant differences.

DBS concentrations remained relatively stable, with no significant variations across treatments.

3.2. Additives in strawberry samples

Most of the selected additives were not detected in the strawberries. In T1, DBP and ATBC were the only compounds detected, with DBP concentrations ranging from 44.69 ± 10.12 to 69.52 ± 16.38 μg/kg, while ATBC concentrations ranged from 1.14 ± 0.50 to 8.37 ± 2.68 μg/kg (Table 2). ATBC was not detected in the control strawberries. The Kruskal-Wallis test identified significant differences in ATBC concentrations ($p = 0.011$), though the Dunn test did not confirm any differences between mulching treatments.

In T2 strawberries, only DBP was detected, with concentrations ranging from 58.87 ± 14.79 to 164.32 ± 81.58 μg/kg, but no significant differences were highlighted by statistical tests (See Fig. 2).

DEHP was not detected in the strawberry samples. This absence may be due to concentrations below the limit of quantification (LOQ) or to the possible transformation of DEHP into metabolites different from MEHP, which were not identified in this study.

3.3. Estimated daily intake and risk assessment

To estimate the non-carcinogenic risks associated with exposure to DBP and ATBC through the consumption of strawberries cultivated in this study, the estimated daily intake (EDI) and the hazard quotient (HQ) were evaluated following the methodology described by (Martellini et al., 2016; Shao et al., 2021a). The HQ is defined as the ratio between the estimated exposure and a reference dose that is without appreciable risk to human health.

The formulas (1 and 2) used to calculate these risk parameters are as follows:

$$EDI = C \times RC/BW \quad (1)$$

$$HQ = EDI/TDI \quad (2)$$

where C is (μg/kg) is the average concentration of the contaminant in the strawberry samples, RC is the recommended daily consumption (kg dry wight), set to 150 g of fresh strawberries (equivalent to 15 g dry weight) based on recommendations from SINU (Italian Society for the Human Nutrition) and previous studies (Charoenwoodhipong et al., 2024; Ellis et al., 2011). BW is the average body weight, assumed to be 70 kg according to (Staskal et al., 2008). TDI (or ADI) represents the tolerable (or acceptable) daily intake. The TDI values for DBP and ATBC were 0.01 mg/kg/day and 1 mg/kg/day, respectively, based on data from (Ennis et al., 2018; Kim et al., 2018) and regulatory sources such as the European Medicines Agency (EMA), and the European Food Safety Authority (EFSA). EDI and HQ values are reported in Table 3. No THQ values exceeded 1 through strawberry consumption.

4. Discussion

Although detected at lower concentrations than in the treatments with applied mulches, PAEs, ATBC, and DBS were present in the control samples at all time points. These findings indicate that mulches are not the sole source of plastic additives. Plastic materials are widely used throughout agriculture, from substrate storage bags and fertilizer packaging to irrigation systems and greenhouse coverings. Collectively, these various sources contribute to the broader issue of plastic pollution

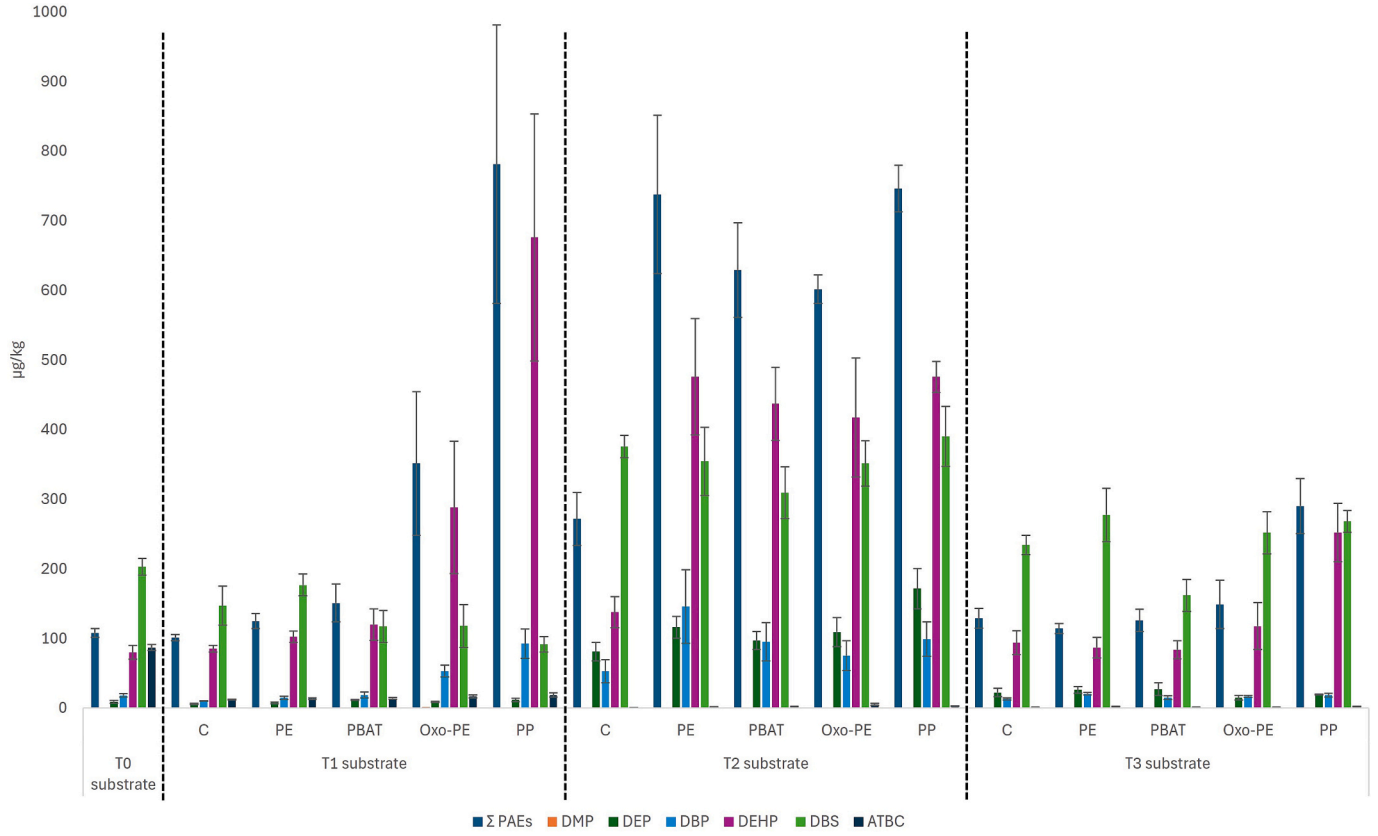

Fig. 1. Additives extracted from the substrate at different time points. Mean ± Standard Error ($n = 5$).

Table 2
Concentration of additives extracted from strawberry samples (µg/kg dry). Mean ± Standard Error ($n = 5$). LOD stands for limit of detection.

		DBP	ATBC
T1 strawberries	C	4.47E+01 ± 1.01E+01	<LOD
	PE	4.60E+01 ± 8.62E+00	8.37E+00 ± 2.68E+00
	PBAT	5.99E+01 ± 8.81E+00	8.14E+00 ± 1.73E+00
	Oxo-PE	6.79E+01 ± 6.37E+00	1.76E+00 ± 9.51E-01
	PP	6.95E+01 ± 1.64E+01	1.14E+00 ± 5.01E-01
T3 strawberries	C	5.89E+01 ± 1.48E+01	<LOD
	PE	6.80E+01 ± 3.41E+01	<LOD
	PBAT	9.61E+01 ± 2.05E+01	<LOD
	Oxo-PE	1.64E+02 ± 8.16E+01	<LOD
	PP	1.58E+02 ± 7.53E+01	<LOD

in agricultural environments.

Significant differences in ΣPAEs concentrations among treatments were most evident three months after the start of the experiment (T1), indicating that the initial period following mulch application is critical for additive release. These differences appeared to lessen over time, although some variation was still detectable at T2 (six months after the start). This trend may reflect the establishment of an equilibrium between additive release from the mulch films into the soil and into the atmosphere (J. Cao et al., 2024) and their degradation and downward migration through the substrate, potentially facilitated by rainfall. Indeed, (J. Cao et al., 2024) found out the up to 56.71 % of PAEs released from mulch films can go into the atmosphere, and the rest end up into the soil.

Although marketed as eco-friendly, biodegradable PBAT mulches emitted additives at rates comparable to PE, while PP and Oxo-PE consistently showed the greatest release. Indeed, PP consistently resulted in the highest concentrations of phthalates in the substrate, followed by Oxo-PE. These findings align with those of (Scopetani et al., 2025), who reported significantly higher phthalate release from PP and Oxo-PE mulches compared to PBAT and PE in aqueous environments. Together, these results suggest that certain mulch types, particularly PP and Oxo-PE, may pose greater environmental risks due to their higher additive release profiles.

The overall rise in PAEs from T1 to T2 corresponds with the summer months (June to August), when increased temperatures and solar radiation may have accelerated the degradation and release of additives. Conversely, the decline from T2 to T3 may be attributed to autumn and winter rainfall, which could have promoted leaching and transport of additives through drainage holes in the pots, reducing their concentration in the upper substrate layers. (Wang, Xi, et al., 2021) reported similar findings in their study on PAE concentrations in the soil-crop system. They observed higher PAE levels in mulched soils (450–810 µg/kg) compared to non-mulched soils (370–730 µg /kg), with peak concentrations occurring during the warmest months (July) and decreasing by autumn (October).

The presence of DMP exclusively at T1 and only in mulched treatments (absent in controls) suggests that this phthalate was released by the mulch films within the first three months of the experiment. Its absence in T0, T2, and T3 substrates, as well as in the control samples at T1, supports this conclusion. The lack of DMP detection in strawberry fruits further implies that it may have either degraded into metabolites or been leached from the substrate during autumn rainfall. No significant differences in DMP concentrations were observed between mulch types, indicating that all tested films, PE, Oxo-PE, PBAT, and PP, released this compound at comparable levels.

At T1, significantly higher concentrations of DEHP and DBP were found in the substrate mulched with PP compared to the controls ($p = 0.004$ and $p = 0.002$, respectively). Similarly, substrates mulched with Oxo-PE showed elevated DBP levels compared to controls ($p = 0.022$). Notably, DBP was the only phthalate detected in strawberry samples, with the highest concentrations observed in fruits grown in PP- and Oxo-

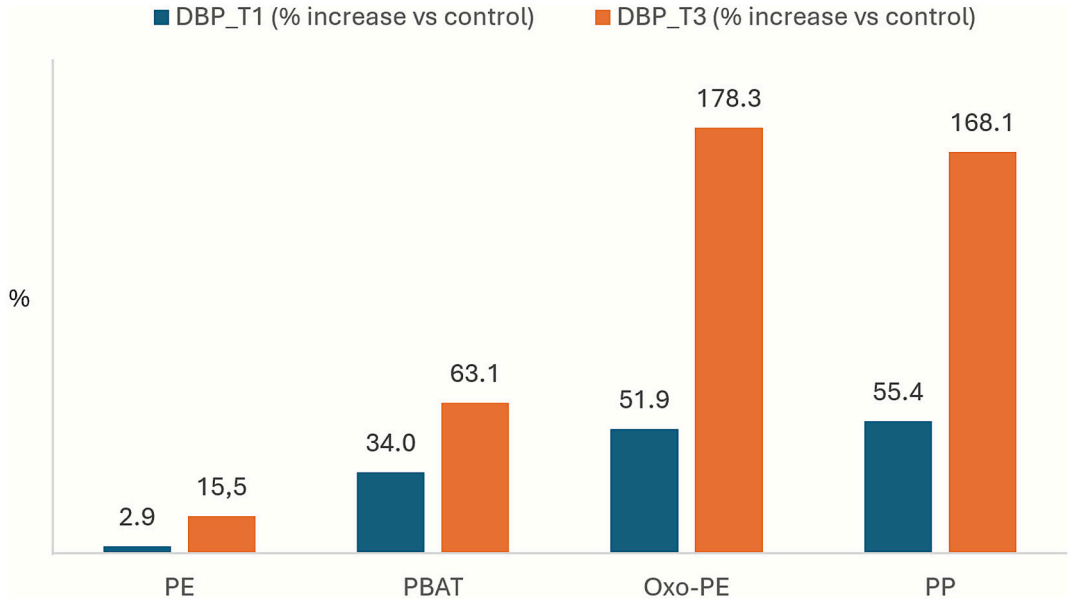

Fig. 2. Increase (%) of DBP compared to control (T1 vs T3) in strawberries.

Table 3
Estimated dietary intake (EDI in ng/day-kg) and hazard quotient (HQ).

		DBP			ATBC		
		C (µg/kg)	EDI	HQ	C (µg/kg)	EDI	HQ
	C	4.47E+01	9.58E-03	9.58E-04	–	–	–
	PE	4.60E+01	9.86E-03	9.86E-04	8.37E+00	1.79E-03	1.79357E-06
	PBAT	5.99E+01	1.28E-02	1.28E-03	8.14E+00	1.74E-03	1.74429E-06
	Oxo-PE	6.79E+01	1.45E-02	1.45E-03	1.76E+00	3.77E-04	3.77143E-07
T1	PP	6.95E+01	1.49E-02	1.49E-03	1.14E+00	2.44E-04	2.44286E-07
	C	5.89E+01	1.26E-02	1.26E-03	–	–	–
	PE	6.80E+01	1.46E-02	1.46E-03	–	–	–
	PBAT	9.61E+01	2.06E-02	2.06E-03	–	–	–
	Oxo-PE	1.64E+02	3.52E-02	3.52E-03	–	–	–
T3	PP	1.58E+02	3.39E-02	3.39E-03	–	–	–

PE-mulched substrates (157.99 ± 75.34 µg/kg and 164.32 ± 81.58 µg/kg, respectively). These are the same mulch types that released the highest DBP levels into the soil, suggesting a potential direct transfer from mulch to soil and ultimately into the edible part of the plant and raising concerns for both soil health and food safety.

At T1, the substrate mulched with PBAT showed significantly higher concentrations of DEP compared to the controls ($p = 0.030$). These findings are consistent with results from a parallel study conducted in our laboratories, where additives were extracted from PE, Oxo-PE, PP, and PBAT mulches using both organic and aqueous solvents (Scopetani et al., 2025). Considering DEP's classification as an endocrine disruptor (Weaver et al., 2020), this suggests that even biodegradable mulches like PBAT may release chemicals with potential environmental and health risks, challenging the assumption that these materials are inherently safer.

(J. Cao et al., 2024) conducted pot experiments with maize to investigate the release patterns of PAEs throughout the lifecycle of mulching plastic films, including biodegradable plastics. Like our findings, their study reported that phthalate esters, particularly DBP, are released from mulch films into the soil and can be taken up by plants, though to a limited extent. Both studies indicate that PAEs predominantly accumulate in the soil, with only minor translocation to edible plant parts, highlighting the soil as the primary environmental sink for these contaminants. Additionally, (J. Cao et al., 2024) found that biodegradable films composed of PBAT and polylactic acid (PLA) released comparable or even higher amounts of additives than conventional plastics, further supporting our conclusions.

Additional evidence supporting this hypothesis comes from (Reay et al., 2025), who evaluated the leaching of low molecular weight organic additives, metals, and metalloids from various mulch films—including low-density polyethylene (LDPE), LDPE oxo-biodegradable, and biodegradable blends of PBAT and PLA, as well as PBAT alone. Their results showed that biodegradable plastics exhibited higher overall leaching of both inorganic and organic additives.

Phthalates are known to contaminate a wide range of foods, from baby food to meat, dairy, grains, and vegetables (X. L. Cao, 2010; Ma et al., 2021; D. Wang et al., 2021). In line with these findings, (Shao et al., 2021b) reported mean DBP concentrations of 283 µg/kg and DEHP concentrations of 250 µg/kg in fresh strawberries from suburban Shanghai. While DEHP and MEHP, one of DEHP's metabolites, were not detected in the strawberries in the present study, DBP concentrations were within the same range as those reported in the previously cited study. The absence of DEHP and MEHP in the strawberries in our study may indicate either that DEHP did not migrate into the fruits, or that it underwent further degradation and transformed into other metabolites. Although DEHP was not detected in the strawberries, it emerged as the most abundant phthalate congener in the substrate samples across all time points. These findings are consistent with those reported by (Cui et al., 2024), who reviewed the presence and release of PAEs from mulch films and identified DEHP, along with DBP, as the predominant congeners.

At T2, the Kruskal-Wallis test indicated significant differences in

both total PAEs ($p = 0.045$) and DEHP concentrations ($p = 0.046$). However, these differences were not confirmed by the Dunn post hoc test, implying that although variation exists among treatment groups, no single pairwise comparison reached statistical significance after correction for multiple testing. This suggests a broader pattern of variation in contaminant levels without strong differentiation between specific mulch types at this stage.

The highest mean concentration of ATBC was observed in the substrate at T0, suggesting possible contamination during substrate production, storage, or transport. From T0 to T3, ATBC concentrations declined steadily, most notably between T0 and T1, likely due to microbial degradation, transformation into metabolites, leaching from rain or migration to the strawberry plants as it will be discussed later. Despite this decline, ATBC remained detectable in substrates at all time points. At T2, ATBC concentrations in control samples were significantly lower than in Oxo-PE-mulched substrates ($p = 0.028$) and noticeably lower in comparison to PP, though the latter difference was not statistically significant. This indicates that both Oxo-PE and PP mulches may contribute to the persistence of ATBC in soil by adding to the initial contamination.

Notably, ATBC was detected in strawberries from all mulched treatments at T1, but not in the controls. This suggests that ATBC may be taken up by the plant and translocated to the fruit. Although the European Chemicals Agency (ECHA) considers ATBC a safer alternative to traditional plasticizers like phthalates, emerging evidence suggests potential endocrine disrupting activity. For instance, (Horie et al., 2022) reported that ATBC at 194.5 μg/L could disrupt hormonal activity and impair fish embryo development.

A similar pattern was observed for DBS, which was already present at T0 in concentrations comparable to those at later time points. Unlike ATBC, however, DBS did not show a continuous decrease. Instead, its concentration increased from T1 to T2, likely due to summer heat accelerating its release, before declining at T3, possibly due to leaching from autumn and winter rainfall. No significant differences in DBS concentrations were found between treatments and controls. Its presence across all time points, including T0, suggests that mulch films are a marginal source of DBS, and there are other sources used in strawberry cultivation, such as substrate components or potential contamination during production or handling.

Regarding the risk assessment of DBP and ATBC detected in the strawberries, none of the HQ values exceeded 1, indicating that dietary exposure to these substances does not pose a significant non-carcinogenic risk.

The results of this research show that mulch films can introduce certain plastic additives into the soil and fruits of strawberry plants, although only to a limited extent. What makes these findings particularly alarming is that, in our study, the mulch films remained intact and did not undergo significant fragmentation. We suggest that if the films were to fragment, the release of additives could be even more substantial. Supporting this concern, (Ouyang et al., 2023) demonstrated that DEHP release from mulch films was negligible when the films were intact, but increased significantly during mechanical abrasion, a process that also led to microplastic formation. The release of DEHP was strongly correlated with the extent of fragmentation, highlighting the environmental risk posed by deteriorating mulch films.

The detection of additives even in the control samples shows that plastic derived contaminants are present in agricultural systems regardless of mulching film application. These results highlight that the problem of plastic and its released additives exists even in the absence of mulching films; however, the use of mulching films contributes to this contamination.

5. Conclusions

In this study, the release of plastic additives from different mulch films (PBAT, PP, Oxo-PE, and PE) into the substrate and the transfer of those to strawberry fruits was evaluated.

Contrary to their eco-friendly image, biodegradable PBAT mulches released additives at levels comparable to PE. Critically, PP and Oxo-PE mulches were consistently identified as the highest-emitting materials. Indeed, among the different material tested PP and Oxo-PE were associated with the highest levels of contaminants in soil and fruit, suggesting greater environmental and food safety concerns compared to PE and biodegradable PBAT.

The assumption that biodegradable films are more environmentally sustainable is challenged by the fact that they do not always release fewer additives into the substrate compared to conventional plastics like PE. Indeed, in some cases, the release of additives is comparable to, or even exceeds, that of conventional plastics.

ATBC was likely already present in the substrate due to contamination during its production, storage, or transport, as indicated by the highest concentrations observed in the substrate at the beginning of the experiment. As for phthalates, the fact that ATBC concentrations were higher in substrates mulched with Oxo-PE and PP at the first harvesting time suggests that these materials may contribute additional ATBC to the soil. Moreover, its presence in strawberries from all mulched treatments (but not in controls) supports the idea that ATBC can be absorbed by plants and transferred to edible parts. These observations raise concerns about its environmental persistence and potential health implications, especially given growing evidence of its endocrine-disrupting effects.

To assess the sustainability of mulch films, it is essential to look beyond their degradability and consider the timing and extent of additive release under different environmental conditions. Further research into the long-term effects of both conventional and biodegradable mulches, including their breakdown products, is crucial for guiding more sustainable agricultural practices. Furthermore, in this study, substrate samples were collected from the top 10 cm. Future studies should examine the distribution of additives in substrate samples at different depths to better understand their vertical mobility and potential accumulation patterns.

CRediT authorship contribution statement

Costanza Scopetani: Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Saul Santini:** Formal analysis. **Giulia Selvolini:** Writing – review & editing, Funding acquisition, Conceptualization. **Agnese Bellabarba:** Writing – review & editing, Funding acquisition, Conceptualization. **Tania Martellini:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Carlo Viti:** Writing – review & editing, Conceptualization. **Alessandra Cincinelli:** Writing – review & editing, Supervision, Methodology, Conceptualization.

Funding

This research is supported by the European Union-Next Generation EU, UNIFI Young Independent Researchers Call – MuSC (Analysis and sensing of contaminants in agriculture: from Mulches to Soil and Crops) work was funded by the European Union – Next Generation EU, within the MuSC project (Analysis and sensing of contaminants in agriculture: from Mulches to Soil and Crops).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

We would like to acknowledge the support of Francesca Ricciardi

and Ludovica Galli for their help during the running of the experimental tests.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2025.147307.

Data availability

Data will be made available on request.

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