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Review

Smart films fabricated from natural pigments for measurement of total volatile basic nitrogen (TVB-N) content of meat for freshness evaluation: A systematic review

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ABSTRACT

Major databases were searched from January 2012 to August 2021 and 54 eligible studies were included in the *meta*-analysis to estimate the overall mean of total volatile basic nitrogen (TVB-N) in meat. The mean of TVB-N was 24.96 mg/100 g (95 % CI:23.10–26.82). The pooled estimate of naphthoquinone, curcumin, anthocyanins, alizarin and betalains were 25.98 mg/100 g (95 %CI:19.63–32.33), 30.03 mg/100 g (95 %CI: 24.15–35.91), 24.92 mg/100 g (95 %CI: 22.55–27.30), 23.37 mg/100 g (95 %CI:19.42–27.33) and 19.50 mg/100 g (95 % CI:17.87–21.12), respectively. Meanwhile, subgroups based on meat types showed that smart film was most used in aquatic products at 27.19 mg/100 g (95 %CI:24.97–29.42), followed by red meat at 19.69 mg/100 g (95 % CI:17.44–21.94). Furthermore, 4 °C was the most storage temperature used for testing the performance of smart films at 25.48 mg/100 g (95 %CI:23.05–27.90), followed by storage at 25 °C of 25.65 mg/100 g (95 % CI:22.17–29.13). Substantial heterogeneity was found across the eligible studies ($I^2 = 99$ %, $p = 0.00$). The results of the trim-and-fill method demonstrated publication bias was well controlled.

1. Introduction

Quality monitoring of high protein content foods such as fish, red meat, and chicken has become crucial because of their heavy consumption worldwide. Meat and meat products are rich in high-quality proteins; nevertheless, the disadvantage is the relatively short shelf-life of the products (Bekhit, Giteru, Holman, & Hopkins, 2021; Bekhit, Holman, Giteru, & Hopkins, 2021). Meat and meat products spoilage occurs due to metabolic processes or microbial action that alter their physicochemical and sensory characteristics. The level of rejection will vary amongst consumers; thus, depending on their culture, experience, and affordability (Font-i-Furnols & Guerrero, 2014; McCullough, Chervenak, Brent, & Hippen, 2012). Otherwise, regulatory agencies can set limits and be determined by safety and health-hazard studies.

Monitoring changes in the quality of foodstuffs is the main problem, particularly in the case of highly perishable food such as seafood, chicken, and meat-based products (Almasi, Forghani, & Moradi, 2022; Cheng et al., 2022; Oliveira Filho et al., 2021). Aroma is one of the critical attributes when assessing the freshness of food. Decomposition of protein and other nitrogen-containing ingredients resulting from microbial activities induces the accumulation of organic amines that are generally recognized as total volatile basic nitrogen (TVB-N) (Bekhit,

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Giteru, et al., 2021; Bekhit, Holman, et al., 2021). These compounds are toxic and risk factors for food-borne diarrhea that produces substantial color and odor changes (Bekhit, Holman, et al., 2021; Zhang et al., 2015) that influence the acceptability of meat products. The standard methods for monitoring meat freshness are time and resource-consuming. The urgent need for new, rapid, non-destructive, and time-effective sensing methodologies to measure microbial spoilage has increased in the last years (Carey et al., 2011). In previous studies, synthetic chemo-sensitive compounds such as bromocresol green, bromocresol purple, bromophenol blue, and cresol red were popular for the detection of volatile compounds in the headspace of food packaging (Aliabbasi, Fathi, & Emam-Djomeh, 2021; Elrasheid Tahir et al., 2021; Huang, Xin, & Zhao, 2011; Liu, Cui, Shang, & Zhong, 2021; Ma et al., 2021; Rodrigues, Souza, Coelhoso, & Fernando, 2021; Tahir, Xiaobo, Xiaowei, Jiyong, & Mariod, 2016; Xiao-wei, Xiao-bo, Ji-yong, Zhi-hua, & Jie-wen, 2018). Nevertheless, these chemo-sensitive compounds are not edible. Direct contact with foods can increase risks to the consumer because of their possible toxicity, which has restricted their utilization in assessing food quality and safety (Cheng et al., 2022; Dainelli, Gontard, Spyropoulos, Zondervan-van den Beuken, & Tobback, 2008; Oliveira Filho et al., 2021).

The common methods for smart films are mainly fabricated by casting, extrusion methods, and coating the filter paper or polymeric films in a specific dye solution. More advanced techniques such as layer-by-layer assembly, sol–gel, electrospinning, electrochemical writing, foam/pad development, and 3D printing have been applied for film fabrications (Almasi et al., 2022; Wu et al., 2018). Commonly, the smart film consists of solid support that is typically polymeric or biopolymeric base and natural or artificial dyes, which react with chemical compounds (i.e., TVB-N, organic acids, etc.) originating from microbial growth or chemical changes during food spoilage (Balbinot-Alfaro, Craveiro, Lima, Gouveia Costa, Lopes, & Prentice, 2019; Romero, Sharp, Dawson, Darby, & Cooksey, 2021; Singh, Nwabor, Syukri, & Voravuthikunchai, 2021). The resulting color change in the film might be useful for consumers to determine food's freshness.

Nowadays, developing a smart film based on biopolymers and natural pigments has gained significant attention. A brief overview of these examples is reported in Table 1S and Table 2S. Various biopolymers and pigments such as curcumin, anthocyanins, alizarin (1,2-dihydroxyanthraquinone), naphthoquinones, betacyanins, and betalains (Table 1) (Ardiyansyah, Apriliyanti, Wahyono, Fatoni, Poerwanto, & Suryaningsih, 2018; Huang et al., 2019; Jamróz, Kulawik, Guzik, & Duda, 2019; Oliveira Filho et al., 2021; Roy, Kim, & Rhim, 2021; Roy & Rhim, 2021b) were used to develop films with different mechanical properties (e.g., tensile strength, elongation break (%), water vapor, thickness) and pH responses range for monitoring meat and meat products freshness. Among the natural pigments used for colorimetric indicators, anthocyanins are the utmost frequently used for monitoring the freshness of meats and meat products (Table 1). Several studies have described the chemical composition, sources, and pH sensitivity of anthocyanins and underlined the perspective and challenges of using anthocyanins as a pH-responsive agent in intelligent food packaging implementation (Alizadeh-Sani, Mohammadian, Rhim, & Jafari, 2020; Priyadarshi et al., 2021; Roy & Rhim, 2021a; Yong & Liu, 2020).

Recently, many reviews have been published on the use of various biodegradable packaging materials from natural sources for monitoring food quality and safety (Ahari & Soufiani, 2021; Aliabbasi et al., 2021; Alizadeh-Sani et al., 2020; Cheng et al., 2022; Dodero, Escher, Bertucci, Castellano, & Lova, 2021; Josemar Gonçalves de Oliveira Filho et al., 2021; Pirsa, Sani, & Mirtalebi, 2022; Poyatos-Racionero, Ros-Lis, Vivancos, & Martínez-Máñez, 2018; Priyadarshi et al., 2021; Rodrigues et al., 2021; Subramanian et al., 2022; Yong & Liu, 2020).

To the best of our knowledge, no systematic reviews and *meta*-analyses have been performed regarding applying smart films prepared from natural pigments for tracking the freshness of meat and meat products. Thus, the present study systematically reviewed the potential use of smart film fabricated from natural pigments for detecting meat and meat product spoilage considering total volatile basic nitrogen (TVB-N) and implemented a *meta*-analysis of the collected data for the first time (Fig. 1S).

2. Methodology

2.1. Search strategy

An electronic, systematic database search was conducted in Web of Science, Scopus, and Google Scholar databases to find relevant articles reported in English, describing the potential use of smart film developed from natural pigments (e.g., anthocyanin, alizarin, curcumin, betalains, and naphthoquinone) for tracing the freshness of meat and meat products. The search was intended to obtain quality studies evaluated by the scientific community published from first January 2012 to first August 2021. The following keywords were utilized: (Anthocyanin extracts OR Curcumin extracts OR natural pigments) AND (smart film OR chromatic film OR pH-indicator OR intelligent food packaging OR color indicator films) AND (monitoring meats freshness OR monitoring meat products freshness OR total volatile basic nitrogen (TVB-N). Initially, two authors reviewed the titles and abstracts of the extracted papers independently. Subsequently, the obtained full texts were screened for eligibility. Furthermore, the list of references of the retrieved articles was reviewed to extract more relevant papers and enhance the precision of the search.

2.2. Inclusion and exclusion criteria

Studies that fulfilled the following criteria were included: (a) colorimetric labels developed from natural pigments and used to monitor the freshness of meat and meat products; (b) detailed the method of preparations; (c) study reported concentrations of total volatile basic nitrogen (mean and standard deviation); (d) study reported the sample size (mass) used while tracing the spoilage; (e) published in the period between first January 2012 and first August 2021; (f) published in the English language (to avoid mistakes during the translation), and (g) full-text article available.

The following articles or article subset types were excluded: (a) review articles and chapters in books; (b) studies that did not use natural dyes for fabrication of colorimetric sensors; (c) articles that did not focus on the current topic; (d) articles that lacked the concentration of total volatile basic nitrogen (mean and standard deviation); and (d) studies without precise sample size (weight of sample) were excluded. For subgroup analysis, if the study contains a mixture of pigments (e.g., curcumin/anthocyanins, anthocyanins/betacyanins), the studies involved the pigment group with a higher concentration. meta-analysis was conducted on TVB-N at the first-time point of spoilage (as defined by authors) in all included studies.

2.3. Data extraction

We collected the following data: first author's name, year of publication, country, sample size, type of pigments, and their concentrations, pigment sources and types of meat, visual color changes as spoilage indication, and TVB-N (initial level, at a first-time point of spoilage, and the end of storage) as well as the storage temperature and storage period (Table 1 and Table 2S). For articles that only reported the required information as figures, Origin Pro 2018 64-Bit (Origin Lab/. Corp., Northampton, MA) was applied to extract the numerical data about mean and standard deviation.

2.4. meta-analysis and meta-regression methods

In the present work, a *meta*-analysis regarding the application of smart films used for monitoring meat freshness was carried out with the aid of the mean and standard error (SE) and presented as a study effect

Table 1
Summary of the main characteristic of included studies.

Films matrix	Foods	SS (gram)	Storage (°C)	First time point of spoilage (days)	Initial TVBN (mg/100 g)	First spoilage TVBN (mg/100 g)	SD	SE	Final time point (days)	TVBN (mg/100 g)at the end of storage	SD	SE	Visual color change	Author
Bacterial cellulose membrane/ curcumin	Shrimp	100	4	7.00	17.85	26.02	0.00	0.00	34.84	10.00	0.84	0.08	Yellow → reddish-orange	(Kuswandi et al., 2012)
Starch/polyvinyl alcohol/ anthocyanins	Fish	20	4	6.25	6.61	21.80	1.10	0.25	14.00	28.53	1.10	0.25	Red → purple	(Zhai et al., 2017)
Cellulose acetate nanofibres/ alizarin	Fish	100	4	12.00	11.21	22.40	0.95	0.09	12.00	22.40	0.95	0.09	Yellow → violet	(Aghaei et al., 2018)
Ethylene-vinyl alcohol copolymer/ montmorillonite /Anthocyanin	Shrimp	15	25	1.00	0.67	20.39	0.89	0.23	1.25	24.89	1.43	0.37	Rose-red → light blue	(Kang et al., 2018)
K-carrageenan/ curcumin	Pork	50	25	3.00	4.91	31.11	0.05	0.01	3.00	31.11	0.05	0.01	Yellow → red	(Liu et al., 2018)
K-carrageenan/ curcumin	Shrimp	6	25	3.00	7.15	41.53	0.08	0.03	4.00	41.53	0.08	0.03	Yellow → red	(Liu et al., 2018)
Alginate/anthocyanins	Fish	30	25	1.50	3.32	20.66	0.49	0.09	2.67	15.86	0.70	0.13	Purple → yellow	(Majdinasab et al., 2018)
Alginate/anthocyanins	Fish	30	4	7.00	5.27	22.91	0.90	0.16	12.00	23.97	0.77	0.14	Purple → yellow	(Majdinasab et al., 2018)
Chitosan/agarose/anthocyanins hydrogel	Fish	200	25	0.83	5.66	21.89	0.76	0.05	0.83	21.89	0.76	0.05	Red → pink	(Wu et al., 2018)
Gelatin/ gellan gum/ anthocyanins	Fish	100	4	7.00	4.74	30.30	4.12	0.41	9.00	53.71	5.06	0.51	Orange-red → yellow-green	(Zhai et al., 2018)
Cellulose/chitosan/alizarin	Minced beef	30	4	4.00	11.66	20.47	0.73	0.13	6.00	23.53	1.60	0.29	Orange → reddish brown	(Ezati, Tajik, & Moradi, 2019)
Cellulose/starch/alizarin	Fish fillet	30	4	8.00	13.53	27.26	1.15	0.21	8.00	27.26	1.15	0.21	Orange → reddish brown	(Ezati, Tajik, & Moradi, 2019)
Agar /natural naphthoquinone	Fish	50	4	7.00	5.34	33.16	0.69	0.10	8.00	37.52	1.28	0.18	Pink → bluish violet	(Huang et al., 2019)
Agar /natural naphthoquinone	Fish	50	25	0.83	6.30	22.40	0.80	0.11	1.00	25.42	0.77	0.11	Pink color → bluish violet color	(Huang et al., 2019)
K-carrageenan/anthocyanins	Shrimp	30	25	3.00	6.94	39.53	0.08	0.01	3.00	39.53	0.08	0.01	Light gray → yellow	(Liu et al., 2019)
Gelatin /anthocyanins	Shrimp	25	25	1.00	4.80	22.77	0.37	0.07	2.00	83.01	1.47	0.29	Red-pink → greenish-blue	(Liu et al., 2019)
Bacterial cellulose/ anthocyanins	Rainbow trout	30	4	10.00	11.72	29.06	1.57	0.29	15.00	45.03	5.64	1.03	Deep carmine → khaki	(Moradi et al., 2019)
Bacterial cellulose/ anthocyanins	Common carp	30	4	10.00	13.21	29.60	1.96	0.36	15.00	43.13	0.78	0.14	Deep carmine → khaki	(Moradi et al., 2019)
Cassava starch/ anthocyanins	Pork	30	25	1.00	6.56	17.21	1.79	0.33	2.00	41.19	1.74	0.32	Pink/red/ purple → green/yellow	(Qin et al., 2019)
K-carrageenan/hydroxypropyl methylcellulose/ anthocyanins	Pork	30	25	1.50	6.48	19.26	0.84	0.15	2.00	26.57	0.72	0.13	Purple-red → blue	(G. Sun et al., 2019)
Chitosan/ anthocyanins	Pork	30	25	1.00	10.13	17.11	0.41	0.07	2.00	27.37	0.60	0.11	Red → blue	(Yong, Liu, Qin, Bai, Zhang, & Liu, 2019)
Gelatin/polyvinyl alcohol/ anthocyanin	Fish	10	25	1.00	6.30	30.80	2.29	0.72	1.00	30.80	2.29	0.72	Bright red → dark green	(Zeng et al., 2019)
Starch/polyvinyl alcohol/ anthocyanins	Pork	250	25	1.50	7.52	15.69	0.85	0.05	1.50	15.00	1.89	0.12	Red → green	(J. Zhang et al., 2019)
Starch/polyvinyl alcohol /curcumin / anthocyanins	Fish	100	4	8.00	7.44	25.83	2.04	0.20	10.00	34.83	2.88	0.29	Orange → green	(Chen et al., 2020)

(*continued on next page*)

Table 1 (*continued*)

Films matrix	Foods	SS (gram)	Storage (°C)	First time point of spoilage (days)	Initial TVBN (mg/100 g)	First spoilage TVBN (mg/100 g)	SD	SE	Final time point (days)	TVBN (mg/100 g)at the end of storage	SD	SE	Visual color change	Author
Gelatin/oxidized chitin nanocrystals /anthocyanins	Shrimp	15	25	1.00	0.00	41.48	1.62	0.42	1.00	41.48	1.62	0.42	Purple → gray-blue	(Ge, Li, Bai, Yuan, Wu, & Hu, 2020)
Pullulan-purple sweet potato extract/zein-glycerol-carvacrol	Pork	12	25	2.50	0.45	19.08	0.07	0.02	3.00	29.44	0.13	0.04	Dark pink → yellowish green	(Guo et al., 2020)
Carboxymethyl-cellulose/starch/ anthocyanins	Fish	10	25	1.50	6.56	21.55	1.08	0.34	2.00	36.84	0.70	0.22	Pink → blue	(Jiang et al., 2020)
Polyvinyl alcohol/okra mucilage polysaccharide / anthocyanins	Shrimp	15	25	1.00	5.02	21.00	1.61	0.42	1.33	25.01	2.61	0.67	Purple → yellow	(Kang et al., 2020)
Bacterial cellulose/anthocyanins	Shrimp	15	4	2.00	12.32	41.18	1.80	0.46	4.00	54.63	2.07	0.53	Fresh violet → yellow	(Mohammadalinejhad et al., 2020)
K-carrageenan/hydroxypropyl methylcellulose /anthocyanins	Pork	30	25	1.25	7.26	17.54	0.68	0.12	1.67	25.69	0.85	0.15	Dark-red → blue-black	(Sun, Chi, Xu, & Wang, 2020)
Nanocellulose/ anthocyanins	Minced beef	200	4	7.00	13.70	18.90	0.70	0.05	7.00	18.90	0.70	0.05	Bright red → blue	(Taherkhani et al., 2020)
Filter paper/Agarose/anthocyanin	Chicken patties	100	4	21.00	16.10	20.42	2.30	0.23	21.00	20.42	2.30	0.23	Bright red → blue	(Talukder et al., 2020)
Agar/ gellan gum /tio2/ anthocyanins/	Pork	200	4	10.00	7.55	18.24	1.00	0.07	14.00	23.79	1.59	0.11	Dark violet → yellow	(Zhai et al., 2020)
Agar/ gellan gum /tio2/ anthocyanins/	Silver carp-fish	200	4	10.00	8.23	23.71	1.27	0.09	15.00	33.25	2.44	0.17	Rose-red-→green	(Zhai et al., 2020)
Potato starch / anthocyanins / chondroitin sulfate	Seawater shrimp	23	4	1.50	9.57	39.20	0.68	0.14	1.50	39.20	0.68	0.14	Rose-red → green	(Bao et al., 2021)
Chitosan (CS)/oxidized-chitin nanocrystals /anthocyanins	Marine hairtail fish	25	25	2.08	2.42	35.09	1.64	0.33	2.00	35.09	1.64	0.33	Reddish purple → yellow	(Chen et al., 2021)
Anthocyanins / chitosan	Fish	10	23	2.00	7.10	38.80	0.50	0.16	2.00	38.80	0.50	0.16	Dark green → light green	(Li, Wu, Wang, & Li, 2021)
Polyvinyl alcohol/sodium carboxymethyl cellulose/ anthocyanin	Pork	20	23	1.00	6.36	18.78	1.30	0.29	1.00	18.78	1.30	0.29	Purple b → blue-black	(D Liu et al., 2021)
Corn starch/polyvinyl alcohol /curcumin	Fish	25	23	2.00	8.26	16.71	0.77	0.15	2.00	0.00	0.77	0.15	Bright yellow → reddish-brown	(Liu et al., 2021)
Cassava starch/polyvinyl alcohol/ anthocyanins-loaded nanocomplexes	Bass fillets	50	4	8.00	6.23	21.11	2.15	0.30	10.00	27.03	2.22	0.31	Bright purple → dark blue	(Qin et al., 2021)
Starch/ Polyvinyl alcohol/ anthocyanins	Tilapia fish	50	4	10.00	7.93	22.22	1.13	0.16	10.00	22.22	1.13	0.16	Pink → dark purple	(Shi et al., 2021)
Tara gum/ Polyvinyl alcohol/ anthocyanins	Tilapia fish	52	25	1.00	8.24	21.88	1.71	0.24	1.00	21.88	1.71	0.24	Pink → dark yellow	(Shi et al., 2021)
Degradable poly-l-lactic acid nanofibre / anthocyanin	Mutton	25	10	3.00	7.20	34.00	1.40	0.28	2.92	32.86	1.40	0.28	Pink → colorless	(Sun et al., 2021)
Mucilage of lallemantia iberica seed/ curcumin	Shrimp	6	25	3.00	6.66	36.33	3.87	1.58	5.00	43.89	3.15	1.29	Yellow → red	(Taghinia et al., 2021)
Soy protein isolate/ cellulose nanocrystals / polyvinylpyrrolidone nanocapsules/curcumin	Shrimp	6	4	5.00	5.22	25.41	0.52	0.21	5.00	25.41	0.52	0.21	Bright yellow → reddish brown	(Xiao et al., 2021)

(*continued on next page*)

Table 1 (*continued*)

Films matrix	Foods	SS (gram)	Storage (°C)	First time point of spoilage (days)	Initial TVBN (mg/100 g)	First spoilage TVBN (mg/100 g)	SD	SE	Final time point (days)	TVBN (mg/100 g)at the end of storage	SD	SE	Visual color change	Author
Zno nanoparticles/gelatin / gellan gum /anthocyanins	Fish	200	4	6.00	4.70	20.70	1.90	0.13	9.00	40.70	3.05	0.22	Pink → yellow-green	(Yang et al., 2021)
Starch/polyvinyl alcohol/ betacyanins.	Shrimp	80	20	1.00	4.90	21.88	0.61	0.07	1.00	69.30	0.81	0.09	Purple red → to pink	(Yao et al., 2021)
Chitosan /polyethylene oxide /curcumin	Chicken breas	10	4	6.00	7.01	36.85	3.35	1.06	6.00	36.85	3.35	1.06	Yellow → red	(Yildiz et al., 2021)
Polyvinyl alcohol/Sodium alginate/Polyvinylidene Fluoride/anthocyanins	Griskin meat	20	10	3.00	8.72	18.02	0.62	0.14	4.00	27.15	0.68	0.15	Pink → green	(Zhang et al., 2021)
Konjac glucomannan/camellia oil/ carrageenan/curcumin	Chicken meat	70	25	2.00	6.90	21.01	1.50	0.18	4.00	51.43	2.90	0.35	Dark red → reddish-brown	(Zhou et al., 2021)
Cassava starch/polyvinyl alcohol /grape skin/anthocyanin	Pork	100	4	1.50	3.52	18.17	0.22	0.02	2.50	48.73	1.34	0.13	Pink → yellow	(Zhu et al., 2021)
Cassava starch/polyvinyl alcohol /mangosteen rind /anthocyanin	Pork	100	4	1.50	3.52	18.17	0.22	0.02	2.50	48.73	1.34	0.13	Yellow → green	(Zhu et al., 2021)
Agar/Polyvinyl alcohol/curcumin	Freshwater shrimp	50	4	1.50	8.70	31.60	0.75	0.11	3.00	56.87	1.07	0.15	Yellow → orange-red	(Zhang et al., 2021)
Soft wood dissolving pulp/ Shikonin	Shrimp	10	4	3.00	7.43	29.43	0.48	0.15	5	33.00	0.77	0.24	Rose-red → bluish violet	(Dong et al., 2020)
Soft wood dissolving pulp/ shikonin	Pork	10	4	3.00	8.03	18.93	0.69	0.22	5	24.62	0.46	0.14	Rose-red → bluish violet	(Dong et al., 2020)
Tarch/polyvinyl alcohol/Betalains OR /betacyanins	Shrimp	30	20	1.00	7.07	19.38	0.08	0.01	2	35.17	0.22	0.04	Pink → pale yellow	(Qin, Liu, et al, 2020)
Quaternary ammonium chitosan/ fish gelatin/betalains	Freshwater shrimp	30	20	1.00	5.20	20.40	1.40	0.26	2	55.30	1.50	0.27	Pink → yellow	(Hu et al., 2020)
locust bean gum/polyvinyl alcohol / betacyanins	Shrimp	80	20	1.00	7.01	20.92	1.39	0.16	2	50.24	1.40	0.16	reddish-purple → brown/yellow	(Wu et al., 2021)
Pectin/watermelon peel/Betalains	Chilled beef	60	4	8.00	6.73	18.71	1.06	0.14	8	18.71	1.06	0.14	Pink → brownness	(Guo et al., 2021)
Bacterial cellulose/ pelargonidin	Fish -tilapia fillets	59	4	8.00	11.65	27.96	2.70	0.35	8	27.96	2.70	0.35	Red → gray-white	(Liu et al., 2021)
quaternary ammonium chitosan/ polyvinyl alcohol/ betalains	Shrimp	59	25	1.00	11.65	35.14	2.16	0.28	1	35.14	2.16	0.28	Purple → orange	(Yao et al., 2020)
Starch/polyvinyl alcohol/ anthocyanins + betacyanins (1:3)	Pork	10	20	1.00	4.70	20.10	0.30	0.09	2	54.30	1.60	0.51	Purple → yellow	(Qin, Xu, et al., 2020)

Sample size (SS); Standard deviation (SD); standard error (SE);

(ES) (Higgins, White, & Anzures-Cabrera, 2008; Quan & Zhang, 2003). The standard error of ES (i.e. TVB-N) of the tested meat was calculated using Equation (1)(Borenstein, Hedges, Higgins, & Rothstein, 2011).

$$SE = \frac{SD}{\sqrt{n}} \tag{1}$$

In this equation, SD is the standard deviation, and n represents the weight of meat samples used in the study.

The weight of the study is correlated with SE; when increasing the SE decreases, the weight of the study (*Wi* and it was calculated using equation (2). The relative weight was calculated using equation (3) (Hedges, Gurevitch, & Curtis, 1999).

$$Wi = \frac{1}{SE^2} \tag{2}$$

$$RW = \frac{Wi}{\sum W} \times 100 \tag{3}$$

Where $\sum$W is the sum *Wi*.

The Chi-square (I^2) analysis was carried out to examine the heterogeneity of studies. If the I^2 value is higher than 75 %, heterogeneity is considerable, and when the I^2 index is less than 50 %, heterogeneity is low (Higgins & Thompson, 2002). In this study, I^2 was higher than 75 %. Therefore, we applied the random effect model (REM) for *meta*-analysis.

Meta-regression, random-effects *meta*-analysis, and the Hunter-Schmidt model were also used to find the possible sources of heterogeneity between included studies. Furthermore, it can aid in determining whether collinearity might give an alternative justification for some of the significant results (S. Li & Kallas, 2021). This *meta*-regression used seven covariates (samples size, geographical origin of samples, storage temperature, storage period, food types, pigment types, and pigments concentrations). Before the *meta*-regression analysis, the concentrations of pigments were recalculated for the standardization of units of measurement (mg/g). Due to few studies that reported pH values, total viable count, and color change (ΔE), we could not perform *meta*-regression analyses on these attributes that might be additional sources of high heterogeneity that we observed in the research outcomes. The possibility of publication bias was evaluated using Egger's (Egger, Smith, Schneider, & Minder, 1997), Begg's (Begg & Mazumdar, 1994) weighted regression tests, trim-and-fill correction, and funnel plot. All analyses were conducted using Stata 16.0 software (Stata Corp., College Station, TX, USA).

3. Results and discussion

3.1. *Systematic review process*

In the initial study selection, 2173 articles were retrieved. After removing duplicate reports, 1991 articles were screened based on title and abstract. After the screening, 110 full-text articles were evaluated for eligibility based on the inclusion and exclusion criteria (56 full texts were excluded, see Supplementary Material). Finally, 54 articles met the

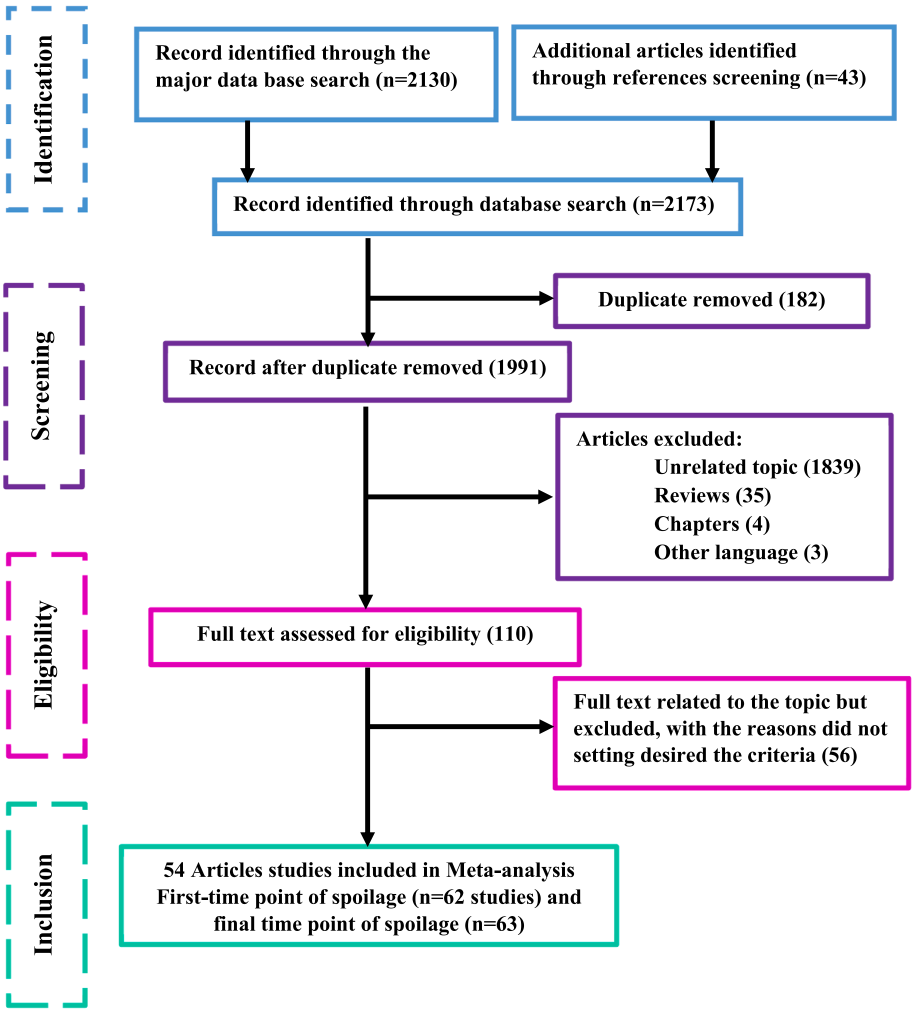


Fig. 1. Summarized search strategy of systematic review and meta-Analysis.

criteria and were included in the *meta*-analysis analysis. Fig. 1 indicates the study selection route. If the article was reported multi-studies, all studies that fulfilled the criteria were included.

3.2. Study characteristics and findings

All 54 articles included in the analysis described the potential use of smart film for assessing the freshness of meat during the storage period. The 54 included articles were distributed as follows; China (44), Iran (7), India (1), Indonesia (1), and Turkey (1). TVB-N values as quality index were measured in all included studies. The sample weight of individual literature was also varied. The maximum sample weight was 250 g, which was used to trace pork spoilage (Zhang, Zou, Zhai, Huang, Jiang, & Holmes, 2019), whereas the minimum sample size was used to trace the freshness of shrimp (Liu et al., 2018; Taghinia, Abdolshahi, Sedaghati, & Shokrollahi, 2021; Xiao, Liu, Kang, Cui, & Xu, 2021). Among the included 62 studies (Table 1), aquatic products were 66.67 %, followed by meat products (pork, beef, and mutton) (28.57 %) whereas a few studies were reported on chicken (4.76 %). According to the findings, anthocyanin from flowers, fruits, and vegetables were the most pigments used (constituting 40 studies) to prepare colorimetric labels due to their high sensitivity to pH change (Table 1). Curcumin is another natural pigment for developing food packaging systems (Aliabbasi et al., 2021; Tambawala, Batra, Shirapure, & More, 2022). Moreover, curcumin-loaded packaging systems were successfully used to detect meat spoilage (Chen, Zhang, Bhandari, & Yang, 2020; Huang et al., 2019; Kuswandi, Jayus, Larasati, Abdullah, & Heng, 2012; Liu, Dang, Zhang, Munsop, & Li, 2021; Liu et al., 2018; Oliveira Filho et al., 2021; Taghinia et al., 2021; Xiao et al., 2021; Yildiz, Sumnu, & Kahyaoglu, 2021; Zhang et al., 2021; Zhou et al., 2021). Based on the literature, few studies successfully applied a pH indicator with alizarin and naphthoquinones to monitor meat and meat products' freshness (Aghaei, Emadzadeh, Ghorani, & Kadkhodaee, 2018; Alizadeh-Sani et al., 2020; Dong et al., 2020; P Ezati, Tajik, Moradi, & Molaei, 2019). Moreover, numerous anthocyanin sources have been used in recent years; red cabbage, berries, and rose/roselle were the most commonly used sources (Table 2S). Betalains are water-soluble nitrogen-containing natural colors consisting of red–purple betacyanins and are widely used as food additives in many foods such as meat and poultry (Priyadarshi et al., 2021). Films containing betalain and betacyanins were successfully correlated with shrimp and pork quality parameters during storage (Table 1).

Concerning the origins of curcumin, in most of the studies, the pigment was purchased (analytical grade). The detailed information on natural pigment sources is presented in Table 2S. Based on the database searches, only one article was included in 2012, while no studies were included from 2013 to 2016. In 2017 only one article satisfied the inclusion criteria. The increase in publications was observed between 2018 and 2021, which might indicate the growing interest of the consumers, food quality and safety authorities, and researchers using a rapid method for tracing or indicating meat spoilage in these years. Table 1 and Table 2S summarize the characteristics of included reports in this study.

3.3. Meta-analysis results

Concerning the natural pigments used for smart films, the data of anthocyanin from 40 studies, curcumin from 8 studies, alizarin from 3 studies, naphthoquinones from 4 studies, and betalains /betacyanins from 7 studies. Based on this *meta*-analysis (Fig. 2), the overall results proved the effectiveness of smart films developed from natural pigments for tracing meat freshness (pooled estimate (TVB-N) 24.96 mg/100 g, 95 % confidence intervals (CI), 23.10 to 26.82, $p = 0.00$). The overall I^2 value was 100 %, demonstrating substantial heterogeneity within studies in this research.

3.3.1. Subgroup results

Meta-analysis showed that the subgroup of curcumin obtained the highest TVB-N estimate of 30.03 mg/100 g (95 %CI: 24.15–35.91), followed by naphthoquinones 25.98 mg/100 g (95 %CI: 19.63–32.33) whereas betalains showed the lowest value 19.50 mg/100 g (95 %CI: 17.87–21.12). The TVB-N estimates of the anthocyanin were 24.92 mg/100 g (95 %CI: 22.55–27.30). The CIs around curcumin and naphthoquinone is very wide, demonstrating that, more extensive studies on using these pigments may be warranted. Subgroup analysis by pigments types demonstrated great variability in TVB-N values ($p = 0.00$) between curcumin, anthocyanin, alizarin, naphthoquinones, and betalains with significant heterogeneity of TVB-N between the various pigments (I^2 over 99 %) (Table 2).

Regarding the results of a subgroup of aquatic products, it was observed that some studies reported very high TVB-N values (Table 2). The results of the subgroup analysis showed that aquatic products presented the highest TVB-N value with an average of 27.19 mg/100 g (95 %CI: 24.97–29.42). Regarding the findings of the subgroup for chicken products, it indicated that these products had a high value of 26.05 mg/100 g (95 %CI: 15.55–36.55). Based on the results, the red meat subgroup showed the lowest value of TVB-N, 19.69 mg/100 g (95 %CI: 17.44–21.94). The chicken subgroup was recorded in only three studies with an average value of 26.05 mg/100 g (95 %CI: 15.54–36.55); thus, the outcomes should be interpreted with caution. Overall, the test of group differences demonstrated significant differences in within-group and between-group effect size heterogeneity (Table 2). Subgroup analysis by food types approached showed significant differences in TVB-N levels between studies conducted on aquatic products, red meat, and chicken. The I^2 statistics of all three subgroups were above 99 %, which confirmed the presence of substantial heterogeneity.

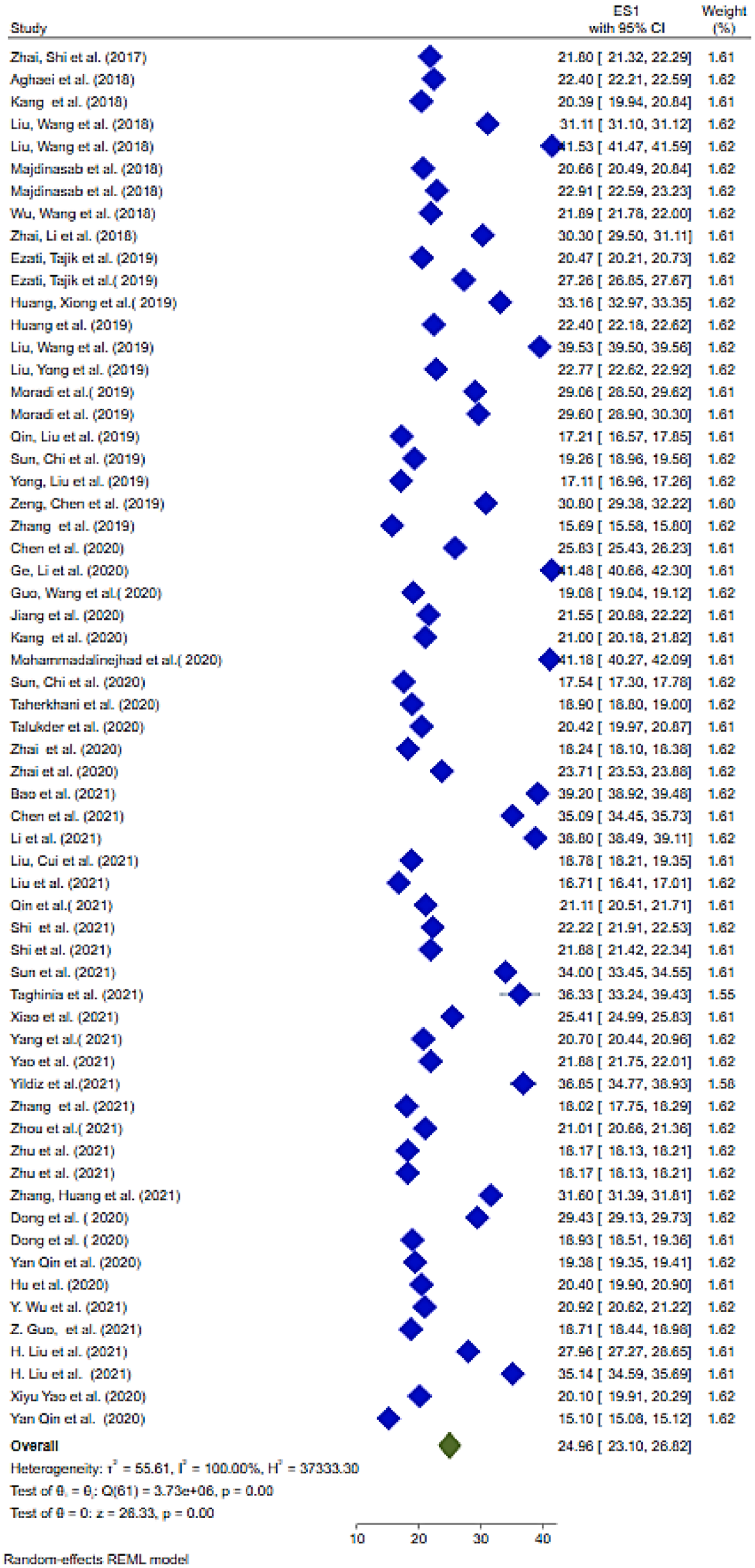
As for subgroup results for storage temperature, studies mainly were conducted at 4 °C (46.03 %) and 25 °C (36.51 %). Few studies were conducted at 10 °C (3.17 %), 23 °C (4.76 %), and 20 °C (9.52), with inclusive evidence. Since these three subgroups of storage temperatures had few studies, the outcomes should be interpreted with caution. The results of the storage temperature subgroups (Table 2) showed that the TVB-N estimate of 4 °C 25.48 mg/100 g (95 %CI:23.05–27.90) and 25 °C 25.65 mg/100 g (95 %CI:22.17–29.13) were both higher than 10 °C 26.01 mg/100 g (95 %CI:10.35–41.67), 20 °C 19.63 mg/100 g (95 %CI:17.73–21.53) and 23 °C 24.76 mg/100 g (95 %CI:10.96–38.57). Test of group differences showed significant differences in within-group and between-group effect size heterogeneity (Table 2).

3.4. Meta-regression results

Meta-analysis was performed to capture the source heterogeneity. The results were reported in Table 3 and Fig. 2S. The findings presented the coefficient (Coef.), standard error (Std. Err.), p-value, and 95 % confidence interval. The overall p-value was $p = 0.001$ less than 0.05, demonstrating significant differences and evidence of a relationship with at least one attribute (Table 3). The I^2 statistic was 99.95 %, and it indicated residual variation as a result of heterogeneity, whereas 80.38 % was the percentage of between-study variance described. Tau^2 value was 9.66, showing the regression model was appropriate. The results of the *meta*-regression model suggested the association among TVB-N values, storage sample size (Coef.-0.03, $p = 0.00$, storage period (Coef -0.21, $p = 0.16$), storage temperature (Coef -0.08, $p = 0.10$), sample geographical origin Coef 3.20, $p = 0.00$), type of foods (Coef -5.24. $p = 0.00$), type of pigments (Coef -2.09, $p = 0.00$), and pigment concentrations (Coef. -0.01, $p = 0.28$) (Table 3 and Fig. 1S).

3.5. Publication bias results

A total of 54 articles were included in *meta*-analyses (Fig. 2) that have met the criteria specified in this study. Funnel plot, trim-and-fill, Begg's and Egger's tests have been applied to recognize any



Study	ES1 with 95% CI	Weight (%)
Zhai, Shi et al. (2017)	21.80 [21.32, 22.29]	1.61
Aghaei et al. (2018)	22.40 [22.21, 22.59]	1.62
Kang et al. (2018)	20.39 [19.94, 20.84]	1.61
Liu, Wang et al. (2018)	31.11 [31.10, 31.12]	1.62
Liu, Wang et al. (2018)	41.53 [41.47, 41.59]	1.62
Majdinasab et al. (2018)	20.66 [20.49, 20.84]	1.62
Majdinasab et al. (2018)	22.91 [22.59, 23.23]	1.62
Wu, Wang et al. (2018)	21.89 [21.78, 22.00]	1.62
Zhai, Li et al. (2018)	30.30 [29.50, 31.11]	1.61
Ezati, Tajik et al. (2019)	20.47 [20.21, 20.73]	1.62
Ezati, Tajik et al.(2019)	27.26 [26.85, 27.67]	1.61
Huang, Xiong et al.(2019)	33.16 [32.97, 33.35]	1.62
Huang et al. (2019)	22.40 [22.18, 22.62]	1.62
Liu, Wang et al. (2019)	39.53 [39.50, 39.56]	1.62
Liu, Yong et al. (2019)	22.77 [22.62, 22.92]	1.62
Moradi et al.(2019)	29.06 [28.50, 29.62]	1.61
Moradi et al. (2019)	29.60 [28.90, 30.30]	1.61
Qin, Liu et al. (2019)	17.21 [16.57, 17.85]	1.61
Sun, Chi et al. (2019)	19.26 [18.96, 19.56]	1.62
Yong, Liu et al. (2019)	17.11 [16.96, 17.26]	1.62
Zeng, Chen et al. (2019)	30.80 [29.38, 32.22]	1.60
Zhang et al. (2019)	15.69 [15.58, 15.80]	1.62
Chen et al. (2020)	25.83 [25.43, 26.23]	1.61
Ge, Li et al. (2020)	41.48 [40.66, 42.30]	1.61
Guo, Wang et al.(2020)	19.08 [19.04, 19.12]	1.62
Jiang et al. (2020)	21.55 [20.88, 22.22]	1.61
Kang et al. (2020)	21.00 [20.18, 21.82]	1.61
Mohammadalinejhad et al.(2020)	41.18 [40.27, 42.09]	1.61
Sun, Chi et al. (2020)	17.54 [17.30, 17.78]	1.62
Taherkhani et al. (2020)	18.90 [18.80, 19.00]	1.62
Talukder et al. (2020)	20.42 [19.97, 20.87]	1.61
Zhai et al. (2020)	18.24 [18.10, 18.38]	1.62
Zhai et al. (2020)	23.71 [23.53, 23.88]	1.62
Bao et al. (2021)	39.20 [38.92, 39.48]	1.62
Chen et al. (2021)	35.09 [34.45, 35.73]	1.61
Li et al. (2021)	38.80 [38.49, 39.11]	1.62
Liu, Cui et al. (2021)	18.78 [18.21, 19.35]	1.61
Liu et al. (2021)	16.71 [16.41, 17.01]	1.62
Qin et al.(2021)	21.11 [20.51, 21.71]	1.61
Shi et al. (2021)	22.22 [21.91, 22.53]	1.62
Shi et al. (2021)	21.88 [21.42, 22.34]	1.61
Sun et al. (2021)	34.00 [33.45, 34.55]	1.61
Taghinia et al. (2021)	36.33 [33.24, 39.43]	1.55
Xiao et al. (2021)	25.41 [24.99, 25.83]	1.61
Yang et al.(2021)	20.70 [20.44, 20.96]	1.62
Yao et al. (2021)	21.88 [21.75, 22.01]	1.62
Yildiz et al.(2021)	36.85 [34.77, 38.93]	1.58
Zhang et al. (2021)	18.02 [17.75, 18.29]	1.62
Zhou et al.(2021)	21.01 [20.66, 21.36]	1.62
Zhu et al. (2021)	18.17 [18.13, 18.21]	1.62
Zhu et al. (2021)	18.17 [18.13, 18.21]	1.62
Zhang, Huang et al. (2021)	31.60 [31.39, 31.81]	1.62
Dong et al. (2020)	29.43 [29.13, 29.73]	1.62
Dong et al. (2020)	18.93 [18.51, 19.36]	1.61
Yan Qin et al. (2020)	19.38 [19.35, 19.41]	1.62
Hu et al. (2020)	20.40 [19.90, 20.90]	1.61
Y. Wu et al. (2021)	20.92 [20.62, 21.22]	1.62
Z. Guo, et al. (2021)	18.71 [18.44, 18.98]	1.62
H. Liu et al. (2021)	27.96 [27.27, 28.65]	1.61
H. Liu et al. (2021)	35.14 [34.59, 35.69]	1.61
Xiyu Yao et al. (2020)	20.10 [19.91, 20.29]	1.62
Yan Qin et al. (2020)	15.10 [15.08, 15.12]	1.62
Overall	24.96 [23.10, 26.82]	

Heterogeneity: $\tau^2 = 55.61$, $I^2 = 100.00\%$, $H^2 = 37333.30$

Test of $\theta_i = \theta_j$: $Q(61) = 3.73e+06$, $p = 0.00$

Test of $\theta = 0$: $z = 26.33$, $p = 0.00$

Fig. 2. Forest plot showing the total basic volatile nitrogen (TVB-N) in various meats measured by smart films fabricated from natural pigments.

Table 2
Summary of the results from subgroup analysis based on types of pigments, types of pigments, and storage temperature.

Subgroups	Number of studies	TVB-N estimates mg/100 g	Lower 95 % CI	Upper 95 % CI	Weight %	DF	Heterogeneity p value	τ^2	I^2 (%)	H^2
Types of pigments										
Curcumin	8	30.03	24.15	35.91	12.84	7	0.00	71.59	100.00	21959.19
Anthocyanins	40	24.92	22.55	27.30	64.59	39	0.00	58.84	99.99	16731.86
Alizarin	3	23.37	19.42	27.33	4.85	2	0.00	12.21	99.85	648.01
Naphthoquinones	4	25.98	19.63	32.33	6.47	3	0.00	41.94	99.96	2348.26
Betalains /betacyanins	7	19.50	17.87	21.12	11.33	6	0.00	4.82	99.98	5959.67
Overall	62	24.96	23.10	26.82	100.08	61	0.00	55.62	100.00	37333.30
Test of group differences: Q_b = chi2(3) = 24.18 Prob > Q_b = 0.000										
Type of foods										
Aquatic products	41	27.19	24.97	29.42	66.15	40	0.00	52.87	99.99	11540.48
Red Meat products	18	19.69	17.44	21.94	29.12	17	0.00	23.67	100.00	36301.35
Chicken products	3	26.05	15.55	36.55	4.81	2	0.00	85.73	99.91	1069.10
Overall	**62**	**24.96**	**23.10**	**26.82**	**100.0**	**61**	**0.00**	**55.62**	**100.00**	**37333.30**
Test of group differences: Q_b = chi2(2) = 21.80 Prob > Q_b = 0.000										
Storage temperature										
4 °C	28	25.48	23.05	27.90	45.20	27	0.00	42.90	99.98	6562.99
10 °C	2	26.01	10.35	41.67	3.23	1	0.00	127.63	99.96	2618.84
20 °C	6	19.63	17.73	21.53	9.71	5	0.0	5.63	99.99	8273.01
23 °C	3	24.76	10.96	38.57	4.85	2	0.00	148.82	99.98	4210.86
25 °C	23	25.65	22.17	29.13	37.09	22	000	72.34	100.00	49430.00
Overall	**62**	**24.96**	**23.10**	**26.82**	**100.00**	**61**	**0.00**	**55.62**	**100.00**	**37333.30**
Test of group differences: Q_b = chi2(4) = 17.98 Prob > Q_b = 0.001										

total volatile basic nitrogen (TVB-N mg/100 g), I^2 means the variation in TVB-N attributable to heterogeneity and all values are more than 99.0 %, showing the presence of high heterogeneity; CI = confidence interval; degree of freedom (df), tau2 (τ_j^2) is the between-study variance, a measure of between-study heterogeneity.

Table 3
Results of the meta-Regression (based on TVBN values at the first spoilage time).

_ Covariates	Coef.	Std. Err.	z	P > z	[95 %CI]	
Sample Size (gram)	−0.03	0.01	−4.55	0.00	−0.05	−0.02
Storage periods (first spoilage time point)	−0.21	0.15	−1.39	0.16	−0.49	0.08
Storage temperature (°C)	−0.08	0.05	−1.62	0.10	−0.18	0.02
Sample origin	3.20	0.85	3.77	0.00	1.54	4.87
Type of foods	−5.24	0.69	−7.58	0.00	−6.59	−3.88
Type of pigments	−2.09	0.33	−6.26	0.00	−2.74	−1.44
_cons	36.98	1.95	18.98	0.00	33.16	40.80
Random-effects *meta*-regression						
Method: Hunter-Schmidt						
Residual heterogeneity:						
$tau^2 = 8.46$						
I^2 (%) = 99.96						
H2 = 2768.70						
R-squared (%) = 88.10						
Wald chi2(6) = 138.77						
Prob > chi2 = 0.00						

Note: * Significance level: 0.05; metaregression was used to formally assess differences between subgroups; t2 is the between-study variance, a measure of between-study heterogeneity; CI = confidence interval.

publication bias, which demonstrated publication bias between included studies. The results indicated significant bias between the studies ($p = 0.0000$ for Begg's and $p = 0.0009$ for Egger's). Thus, we applied the random effects trim and fill analysis. No studies were added to the funnel plot and there was no change in the mean of TVB-N (Fig. 3), indicating the absence of publication bias.

4. Key findings

In this *meta*-analysis, we attempted to clarify the potential use of smart film to predict the freshness of meat and meat products. The varying reports of TVB-N values are reflected in the substantial heterogeneity across the included studies (I^2 over 99 %). However, the efficacy of smart film for tracing freshness was reported in all included studies.

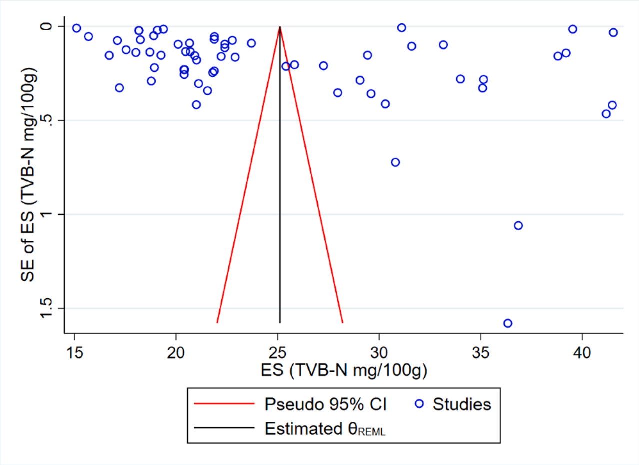


Fig. 3. Forest plot to assess the publication bias.

According to the pigment type's subgroup, it has been noted that the number of studies conducted to assess the potential use of smart films for tracing the freshness of meat varies between pigments. Among the pigments that have been used for smart packaging for tracing the freshness of meat, anthocyanins and curcumin have attracted the attention of food scientists' due to their biological activities, such as antioxidant and antimicrobial potential (Aliabbasi et al., 2021; Oliveira Filho et al., 2021). This is also due to the high sensitivity to the change in pH and gas (Hashim et al., 2021; Sun, Chi, Zhang, Xu, Li, & Wang, 2019; Yang et al., 2021; Zhai et al., 2018; Zhang et al., 2021; Zhou et al., 2021). Furthermore, many studies found that the types and concentrations of natural pigments incorporated into smart labels affect (positively or negatively) physicomechanical characteristics of the film (Huang et al., 2019; Koosha & Hamedi, 2019; Priyadarshi et al., 2021; Taherkhani, Moradi, Tajik, Molaei, & Ezati, 2020). Therefore, these factors should be considered in developing this type of film. Another essential factor that should be considered when producing smart films is the compatibility of pigments with the polymeric base. For example, bacterial nanocellulose, has distinctive properties such as a large number of functional groups (HO–), unique porosity, and water absorption ability besides being economically practicable for monitoring seafood freshness (Mohammadalinejhad, Almasi, & Moradi, 2020; Moradi, Tajik, Almasi, Forough, & Ezati, 2019; Taherkhani et al., 2020). Furthermore, polymeric bases in the films can protect the pigments from oxidation by tapping or encapsulating them, therefore, improving the quality of the films (Qin, Yun, Xu, Chen, Kan, & Liu, 2021; Wu et al., 2018). The pH of the film-forming solutions also significantly affects the sensitivity of the final films. In a study, Liu, Shi, Sun, Zhang, & Ji, (2021) investigated the potential use of bacterial cellulose and pelargonidin (at pH 3.0–10.0) to monitor the freshness of fish. The results revealed that the smart film with pH 3.0 is more suitable for storage at 4 °C, while the film with pH 4.0 is more appropriate for storage at 25 °C.

Based on the systematic review, the method of preparation of smart films (e.g., casting, extrusion, dipping, electrochemical writing, multi-layers, etc.) could contribute to the high between-study heterogeneity (τ^2) in the pigment type's subgroup (Table 2), since it can affect the sensitivity of the films. The narrow confidence intervals of the betalains /betacyanins subgroup (17.87–21.12) and anthocyanins subgroup (22.55–27.30) indicated a high quality of evidence and absent sources of bias.

The extensive studies on anthocyanin have been attributed to the availability and researchers' access since there are various sources of these pigments. Additionally, several extraction methods were used to obtain the anthocyanins to incorporate their extracts onto the films (Oliveira Filho et al., 2021; X Zhai et al., 2017; Zhang et al., 2021; Zhang et al., 2019; Zhu, Lu, Qin, Cheng, Yuan, & Li, 2021). Another possible reason is that films containing anthocyanins may serve as active and smart, as evidenced by many studies (Liu, Yong, Liu, Qin, Kan, & Liu, 2019; Qin, Liu, Yong, Liu, Zhang, & Liu, 2019; Wu et al., 2021; Yao, Qin, Zhang, Zhang, Qian, & Liu, 2021).

In many studies, the potential applications of the film incorporated with curcumin in tracing meat freshness food were reported (Aliabbasi et al., 2021; Chen et al., 2020; Kuswandi et al., 2012; Liu et al., 2021; Liu et al., 2018; Taghinia et al., 2021; Xiao et al., 2021; Yildiz et al., 2021; Zhang et al., 2021; Zhou et al., 2021). A study reported the potential use of the mixture of pigments (curcumin/anthocyanin 1:4) on films to detect bighead carp quality changes (H.-z. Chen et al., 2020). The film successfully presented three different colors corresponding to freshness, medium freshness, and spoilage.

Alizarin is natural orange-brown color, usually employed as a colorant agent in textile fabrics (Trovato, Mezzi, Brucale, Rosace, & Rosaria Plutino, 2022). Compared with other natural pigments, few studies successfully reported the potential application of alizarin in food packaging (Table 1) (Almasi et al., 2022; Priyadarshi et al., 2021). Therefore, comprehensive studies are needed before adapting alizarin as sensing materials in food packaging.

Naphthoquinone and naphthoquinone derivatives (e.g., shikonin and alkannin) are commonly used in textile dyeing agents and food colorants (Almasi et al., 2022; Ordoudi, Tsermentseli, Nenadis, Assimopoulou, Tsimidou, & Papageorgiou, 2011). Recently, naphthoquinones are promising new natural colorants for developing colorimetric films (Dong et al., 2020; Parya Ezati, Bang, & Rhim, 2021). Films containing naphthoquinone or naphthoquinone derivatives demonstrated high sensitivity and rapid color change when used for monitoring food freshness (Table 1). The color changes of naphthoquinones could be attributed to the transformations of the chromophore molecular structure, which is unstable and degrades at alkaline pH conditions (Dong et al., 2020). In some smart films, the addition of naphthoquinones increases the hydrophobicity and moister resistance of the developed films, demonstrating their potential application in food industries (Almasi et al., 2022; Dong et al., 2020; Huang et al., 2019).

Because of their high pH-sensitive features, betalains are greatly promising natural pigments that can be used for monitoring meat and seafood freshness (Guo, Ge, Li, Yang, Han, & Yu, 2021; Hu, Yao, Qin, Yong, & Liu, 2020; Qin, Liu, Zhang, & Liu, 2020; Wu, Tang, Quan, Zhang, Wang, & Liu, 2021; Yao, Hu, Qin, & Liu, 2020). Recently, Qin, Xu, Yuan, Hu, Yao, & Liu, (2020) investigated the effectiveness of smart films containing anthocyanins and betacyanins. The results showed that film containing the mixture of anthocyanins/ betacyanins in the ratio of 1:3 was appropriate to monitor pork spoilage. These results could be attributed to betacyanins' thermal and acidic stability compared to anthocyanins, suggesting they are appropriate sensing materials for low acid foods (Ravichandran et al., 2013).

TVB-N estimate from our *meta*-analysis suggested that all pigments had a great potential to trace the freshness of meat and products within a specific period. There were significant differences in subgroup between the pigments type subgroups ($p = 0.00$, Table 2) when the *meta*-analysis was applied to values of TVBN at first spoilage.

As indicated in Table 1, when various meat and meat product storage times increased, the level of TVB-N produced by microbial action increased; then, the response pigments increased and varied significantly. A similar trend was observed when various synthetic chemo-sensitive dyes were used for monitoring fish freshness (Morsy et al., 2016).

Based on the available data in the studies, there is evidence of variations in the speed of the visual detection of meat spoilage smart films prepared with natural pigments (Table 1). Curcumin smart films can detect the meat deterioration with storage periods ranging from 1.5 to 6 days, anthocyanins from 0.83 to 21 days, alizarin from 4 to 12 days, naphthoquinone from 0.83 to 7 days, and betalains /betacyanins from 1 to 8 days. The variations in the speed of visual changes of films could be attributed to many factors such as the concentration of TVB-N during food spoilage, the concentration of pigments, structure of the pigments, types of polymeric base, methods of film production, pH film-forming solutions and many others.

Many studies indicated that the sensitivity of intelligent labels is dependent on dye concentrations (Huang et al., 2019; Moradi et al., 2019; Taherkhani et al., 2020). Therefore, attempts have been made to correlate the pigment concentrations and TVB-N. Unfortunately, it was impossible to conduct a *meta*-regression analysis using all included studies due to the inconsistency of the methods of calculating the concentrations and additions; therefore, some articles (Chen, Yan, Huang, Zhou, & Hu, 2021; Dong et al., 2020; Kang et al., 2018; Kuswandi et al., 2012; Liu et al., 2021; Liu et al., 2021; Liu et al., 2021; Mohammadalinejhad et al., 2020; Moradi et al., 2019; Qin, Xu, et al., 2020; Taherkhani et al., 2020; Talukder et al., 2020; Yao et al., 2020; Yao et al., 2021) were excluded from *meta*-regression test. Generally, low concentrations of pigments were preferred in many studies (Fig. 1S and Table 2S). However, Zhai and coauthors observed weak color of extremely low concentration compared with the very strong color of high concentration (X Zhai et al., 2017). Using low concentrations in anthocyanin and curcumin could be due to the rapid color change of low concentration compared with the high concentration. Based on the *meta*-regression result, there was no significant relationship between TVB-N values and pigments concentrations (Fig. 2S).

A higher concentration of TVB-N found in aquatic foods could be attributed to the accumulation of trimethylamine (TMA), which is the main contributor to TVB-N. Many studies have linked the relationship between TVB-N limits and the freshness of seafood and other meat products (Table 1). From these studies, we can see the widespread utilization of TVB-N to evaluate the freshness of aquatic foods. However, there are contrary positions that limit it as an indicator of seafood freshness. Different TVB-N limits were used to assess meat freshness in the included studies. For example, studies from China and India have used the limit of Chinese standards GB2733-2015 and GB 2733. According to Chinese standards, the limit of TVB-N level for shrimp and fish is 20 mg/100 g and 30 mg/100 g for marine fish and marine shrimp

(Bao et al., 2021; Chen et al., 2020; Talukder et al., 2020; Zhang et al., 2021). While the limits used in studies published from Iran were varied, some authors identified TVB-N levels of 25.5 mg/100 g and suggested this as a critical limit. In contrast, others compared the concentrations of TVB-N with the other authors (Ezati, Tajik, & Moradi, 2019; Majdinasab, Hosseini, Sepidname, Negahdarifar, & Li, 2018; Mohammadalinejhad et al., 2020; Taghinia et al., 2021). Our research data showed that smart films could be used successfully for tracing the freshness of aquatic food with an average limit of 27.19 mg/100 g and a confidence interval of 24.97–29.42. The subgroup analysis indicates that the chicken products have a higher TVB-N value, while the red meat products have the lowest value. These results agree with the standard limit for chicken and red meat (Ezati et al., 2019; Taherkhani et al., 2020; Talukder et al., 2020). According to the acceptable limit of TVB-N, the storage temperature subgroup shows that smart films have been successfully used to distinguish the spoilage status of various meat products. As can be seen in Table 2, the smart films can discriminate the spoilage meats stored at 4 °C with storage time ranging from 1.5 to 21 days, at 25 °C with a limit of storage ranging from 0.83 to 3 days, at 23 °C with storage time from 1 to 2 days, and at 20 °C with storage time from 1 to 1.83 days. Few studies have used 10 °C for tracing the freshness of mutton and griskin meat (stored for three days) (Sun et al., 2021; Zhang et al., 2021) (Yao et al., 2021).

According to the results, smart films' effectiveness was mainly tested at 4 °C and 25 °C. The TVB-N estimate of 4 °C was slightly higher than that of 25 °C. This finding could be attributed to TVB- increases with the storage time of meat to rapid bacterial growths, producing various amine compounds that directly change the film's color when TVB-N exceeds acceptable limits.

Many authors have reported the higher values of TVB-N of samples stored at low temperature (4 °C) compared with the ambient temperature (Huang et al., 2019; Majdinasab et al., 2018; Shi, Zhang, Jia, Yang, & Zhou, 2021). However, some studies showed that smart films did not exhibit good color change despite the progressed fish spoilage stored at 2 °C (Jamróz et al., 2019) and 5 °C and −5°C(Amiri, Piri, Akbari, & Moradi, 2020). Based on *meta*-regression analysis, sample size sample, geographical origin of samples, type of foods, and type of pigments were the main covariates that contributed to the variations of TVB-N values at the first spoilage point. In contrast, at the end of the storage, all covariates affect the TVB-N values except sample size (Table 3S).

Overall, the findings of this study can be used to guide scientists, food processors, and food authorities when looking for rapid techniques for tracing the freshness of meat and meat products.

5. Limitations and strengths

There are some limitations of the present study. The first is related to the natural pigments (i.e., stability). Various concentrations of pigments (single or in combination) were used in film development. Second, some factors affected the quality of the results not tested in some studies (e.g., microbial analysis, pH of the tested sample, and the color change (ΔE) of the film during the monitoring process). This could be due to a lack of guidelines for the preparation and application of this type of film. Third, the results showed a high degree of heterogeneity ($I^2 > 99$ %) and might be other factors affecting heterogeneity that have been measured. Fourth, only an article published in English was included, which may have caused us to overlook relevant articles written in other languages. Fifth, no safety and possible side effects evaluation of the smart film fabricated from natural extracted pigments were reported. Sixth, due to the limited number of studies on the chicken's subgroup, some pigments subgroups (naphthoquinone and alizarin) and some storage temperature subgroups (10 °C, and 23 °C) have been reported; the results of these subgroups should be interpreted with caution. Seventh, this research mainly focuses on China and Iran because most of the included studies are from these countries. The main strength of this *meta*-analysis is that it is the first systematic review and *meta*-analysis that evaluated the potential use of smart film fabricated from natural pigments.

6. Conclusion

In brief, the current *meta*-analysis suggested that the smart film fabricated from natural pigments including curcumin, anthocyanins, alizarin, naphthoquinone, and betalains may be used for tracking the freshness of meat and meat products. To provide better guidelines for the designation of smart film to be used in the rapid tracing of meat and meat products' freshness, additional studies should evaluate the optimum concentration of pigments. The appropriate matrix can preserve the pigments without affecting their release during the actual application of the film and attempt to identify better sources of the pigments with high stability and higher sensitivity to changes in pH. Furthermore, micro and nanoencapsulation should be included to enhance the stability of the natural dyes in films. Moreover, pH, microbial analysis, ΔE, the detailed information of tested samples (e.g., whole fish, fillets, slaughter method, etc.), and sensory evaluation of packaged meat and meat products should be included in all future studies to improve the quality of the results. The results showed that smart films can discriminate at 4 °C with storage time ranging from 1.5 to 21 days, at 25 °C with a limit of storage ranging from 0.83 to 3 days, at 23 °C with storage time from 1 to 2 days, and at 20 °C with storage time from 1 to 1.83 days. Overall, our research data showed that smart films developed from various natural pigments could be used successfully to trace the level of TVB-N with an average limit of 27.69 mg/100 g and a confidence interval of 25.18–30.21 mg/100 g. The results of *meta*-regression analysis show that sample size, sample geographical origin, food typesand pigment types covariates were the primary sources of TVB-N variations. This information would be guidelines for academia, food safety, and food quality assurance authorities to establish the detection limit of smart films to monitor meat freshness. Finally, this review showed that these films are only the beginning of these works, and more investigations should be conducted to develop other robust *meta*-analysis studies.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2022.133674.

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