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Review

Human hair as a noninvasive matrix to assess exposure to micro-organic contaminants: State of the art review

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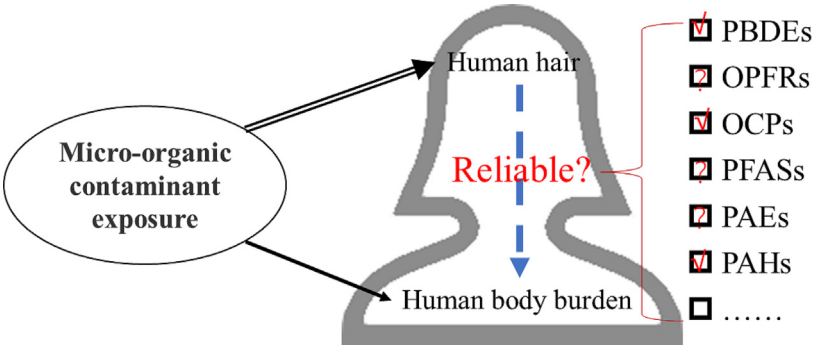
HIGHLIGHTS

- MOC incorporation pathways into hair are basically similar to drugs.
- Standardization of measuring procedure for human hair was proposed.
- Hair biomonitoring is reliable to assess internal exposure to POPs with high K_{ow}.
- Specific metabolite levels in hair can reliably reflect the internal dose of MOCs.
- Hair analysis is promising for health risk assessment of MOCs.

GRAPHICAL ABSTRACT



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ABSTRACT

Human biomonitoring has played an important role in assessing human exposure to micro-organic contaminants (MOCs), including chlorinated persistent organic pollutants, brominated flame retardants, organophosphorus flame retardants, non-persistent pesticides, *per*- and polyfluoroalkyl substances, phthalate esters, bisphenols, and polycyclic aromatic hydrocarbons. Specifically, human hair holds great promise as a noninvasive matrix for MOC biomonitoring. While human hair has been widely used to detect numerous MOCs over recent decades, its reliability of reflecting body burden is still disputable. As a premise for discussion, it is necessary to understand the mechanisms of MOC incorporation into hair from endogenous and exogenous exposures. Then, standardized protocols must be developed to ensure accurate and reliable results. This review article discusses these issues and provides evidence for the reliability of monitoring MOCs in hair by surveying past reports from various categories of MOCs. We find that most persistent organic pollutants - especially those with a higher octanol-water partition coefficient and lower volatility - can be reliably measured using hair analysis, while internal exposure can be accurately measured using MOC metabolites in hair. Finally, we explore the applications of hair analysis in large-scale surveys, retrospective cohort studies, and epidemiological investigations, highlighting the promise of hair analysis in studying the health risks of MOCs.

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Contents

1. Introduction

Human biomonitoring (measuring chemicals or their metabolites in blood, urine, or other matrices in the human body) is an essential tool to assess human exposure and associated risks (Boogaard et al., 2011; Choi et al., 2015; Esteban and Castano, 2009; Jones, 2020; Louro et al., 2019; Wittassek et al., 2011). However, blood or serum is an invasive matrix, which can discourage participation in research (Esteban and Castano, 2009; Rockett et al., 2004). Hair emerges from follicles surrounded by capillaries, and thus holds potential for monitoring chemicals in blood (Alves et al., 2014; Esteban and Castano, 2009). Human hair also holds several significant advantages, including noninvasive sampling, low cost, easy collection and transport, stable storage, and ethical acceptance (Alves et al., 2015; Schramm, 2008; Yusa et al., 2015; Zheng et al., 2021). Moreover, hair consists of 15–35 % water and l-9 % lipids, as well as 65–95 % protein (Harkey, 1993), which provides a suitable reservoir for both hydrophobic parent compounds and hydrophilic metabolites (Hardy et al., 2021; Schramm et al., 1992). Because hair grows 0.6–1.4 cm per month (Pragst and Balikova, 2006), hair samples can also offer larger monitoring windows (months/years) than urine or blood (hours/days) (Rotolo et al., 2021). In all, human hair is an essential tool for the development and application of human biomonitoring.

Human hair has already been proved a successful and reliable tool for assessing elements and drugs (Harkins and Susten, 2003; Mieczkowski, 2016; Mikulewicz et al., 2013; Pan and Li, 2015; Raposo et al., 2014; Skalny et al., 2015; Tagliaro et al., 1997). However, there is still limitations for hair as a reliable tool of assessing internal exposure to micro-organic contaminant (MOC). Two decades ago, a panel convened by the Agency for Toxic Substances and Disease Registry (ATSDR) reviewed and discussed the use of hair analysis in assessing exposures to contaminants. The panel noted several factors and data gaps that limit the reliability of hair in exposure assessment, up to now which are still significant for assessing reliability of hair analysis (Harkins and Susten, 2003). Firstly, many factors - including age, sex, hair treatments, and methods of sampling and purification - will affect the detection of MOCs in the hair matrix (Qiao et al., 2019) (Tang et al., 2023). Accordingly, standardized procedures for hair analysis of MOCs (including collection, preparation, etc.) are warranted, which will conduce to the establishment of reference ranges (Esteban and Castano, 2009). Secondly, another challenge involves distinguishing between endogenous and exogenous contamination of MOCs in hair: the former from incorporation through the blood and the latter from environmental contaminants in dust, air, or hair care products (Qiao et al., 2019; Zhang et al., 2007a; Zheng et al., 2014). Thirdly, the correlations between hair and blood/urine concentrations for each group of MOCs are limited, which are important in assessing systemic burden of contaminants (Harkins and Susten, 2003). Lastly, to fully determine the reliability of hair biomonitoring, we must clarify how and to what extent environmental contaminants are incorporated into hair (Harkins and Susten, 2003).

While numerous MOCs have been detected in human hair over recent decades, hair has not been widely used to assess the health risk of MOCs compared to blood/serum and urine. Thus, it is imperative to better understand the reliability of hair biomonitoring for assessing MOC exposure. In this review, we discuss the mechanisms of MOC incorporation into hair and propose standardized procedures for measuring MOCs in human hair. To further answer the question of whether human hair is a reliable tool for assessing MOC exposure, we have also reviewed past reports on techniques, experiments, and applications of analyzing MOCs in human hair.

2. Mechanisms for the incorporation of MOCs into hair

Several studies on the mechanism of incorporation of elements or drugs into hair have revealed that xenobiotics may enter hair from multiple sites, via multiple mechanisms, and at various times during the hair growth cycle (Bos et al., 1985; Henderson, 1993; Pragst and Balikova, 2006). Henderson

(1993) first proposed a multi-compartment model to explain how drugs get into hair. As generally accepted, not only drugs can enter the hair shaft through passive diffusion or active transport from the bloodstream feeding the hair follicles, but drugs can diffuse into the growing or mature hair fiber through sweat and other secretions, as well as vapors or powders in the air (Pragst and Balikova, 2006). Kamata et al. (2015) applying matrix-assisted laser desorption/ionization − time-of-flight tandem mass spectrometry (MALDI − TOF-MS/MS) imaging, demonstrated these two major drug incorporation sites. Kuwayama et al. (2022a) using micro-segmental hair analysis, revealed the differences in the distribution profiles of drugs in hairs with and without endogenous contamination. Based on some control studies about drugs, Pragst and Balikova (2006) concluded that three key factors (including melanin content, the lipophilicity and the basicity of substance itself) influenced drug incorporation. Different research approaches are conducting for further understanding of the mechanism of drug incorporation in hair (Kuwayama et al., 2022b).

Incorporation of MOCs into hair likely follows similar pathways with drugs. However, different groups of substances may present different mechanisms of incorporation, which is warranted to further verify. Some studies on segmented hair have reported accumulation of MOCs. For instance, measurement of hair segments found that the concentrations of most MOCs in distal hair segments were higher than those in proximal hair segments, suggesting that contaminants may accumulate along the hair shaft (Qiao et al., 2018; Qiao et al., 2016). This phenomenon was never reported in drug research, probably due to the low concentration of drugs in external environment of human. Furthermore, the rates of accumulation along the hair shaft for some MOCs were significantly correlated with the chemical's vapor pressure and the octanol-air partition coefficients (K_{OA}), indicating that the physicochemical properties of MOCs could influence their incorporation into hair (Qiao et al., 2016). A recent study on segmented hair from pigs and cattle found that structural damage of distal hair segments may facilitate the permeability and diffusion of exogenous contaminants (Otten et al., 2022). Disappointingly, there is no micro-segmental analysis or in situ analysis about MOC incorporation into hair, and more studies on mechanisms are necessary.

Although the details of contaminant incorporation into hair are still unknown, several methods have been developed to distinguish between exogenous and endogenous exposures. There are some reports that exogenous contaminants can be efficiently removed using appropriate washing solutions while more stable endogenous contaminants remain (Lin et al., 2019; Lu et al., 2014). However, Kucharska et al. (2015b) investigated different washing methods for hair samples externally exposed to brominated and phosphorus flame retardants and found that there was no washing medium able to entirely and exclusively remove exogenous contamination. Some researchers propose using an indirect approach to calculate the contribution of endogenous or exogenous contamination (He et al., 2017). Assuming that the ratios of different homologs or enantiomers are relatively constant when transferred to hair, a two-end member mixture model can be used to calculate the corresponding contribution rate of endogenous or exogenous contamination (Zheng et al., 2010). Using the ratios of dichloro-diphenyl-trichloroethane (DDT) analogues in hair and dust, He et al. (2017) estimated the exogenous source contributions of indoor dust as approximately 11 % and 20 % of the hair burden of DDTs in males and females respectively. Using enantiomer fractions of polychlorinated biphenyls-132 (PCB-132) in air, human serum, and segmented hair, Qiao et al. (2019) found that the contributions of PCB-132 in hair through external sources increased with the distance of the hair segment from the scalp. This approach is relatively complicated and needs to be verified in the lab. Considering some MOC metabolites are mainly incorporated via blood during hair growth and are not externally deposited, the measurement of MOC metabolites in hair provides a unique tool for assessing exposure (Schummer et al., 2009). This requires the identification of reliable markers for MOC exposure in hair as well as an assessment of the toxicokinetic characteristics of parent compounds and their metabolites. However, this method holds promise for establishing a more comprehensive risk assessment of systemic MOC exposure.

3. Standardized procedure for measuring MOCs in hair

Standardized protocols are necessary to produce accurate and reliable results. Analysis of metals in human hair has long been standardized, with the International Atomic Energy Agency and the World Health Organization recommending standardized protocols for collecting and treating hair samples in 1975 (Jenkins, 1979). However, due to the various classes and complicated structures of MOCs, it is difficult to create a single standardized protocol for MOC hair analysis. This section reviews what is known about MOC hair analysis technologies and provides recommendations on the standardization of measurement protocols.

3.1. Methods

Hair analysis methods were comprehensively reviewed by Schramm (2008), and thus this review will focus on developments after that period. Specifically, papers published between 2008 and 2023 were searched using the Web of Science Core Collection databases with Topic = hair analysis AND Year Published = (2008–2023) AND Abstract = hair AND (polybrominated OR polychlorinated OR dechlorane OR dichloro* OR hexachloro* OR perfluoro* OR polyfluo* OR phosphate OR phosphorus OR polycyclic OR phthalate OR bisphenol* OR pesticide OR endocrine disruptor OR organic pollutants). * represents a wildcard, which replaces any number of characters. After review articles were excluded, there were 724 results (up to May 15, 2023). Then articles whose MeSH headings are “Animals”, “Plants roots” “Minerals”, or “Arabidopsis” were excluded, the remaining 516 were listed in the Supplementary Information. Subsequently, 338 results were filtered out because they did not address MOCs in human scalp hair. For instance, it was excluded if the paper only reported on drugs or elements in human hair, or used only pubic hair or hair follicle, or reported only on the hair of rats or pets. In other instances, papers reported MOCs and metals in the human body but only used hair to detect mercury (Orenstein et al., 2014; Seabert et al., 2014; Valvi et al., 2017). Based on the remaining 178 papers, procedures including sample collection, sample preparation, and instrumental analysis are reviewed as follows:

3.2. Sample collection

>60 % of studies on MOCs in hair did not specifically state the site of hair collection (Fig. 1). In about 40 % of studies, hair samples were collected in the range from the nape to the vertex of the head (Fig. 1). Compared to other areas of the head, vertex hair has a more steady growth rate and hair amount (Boumba et al., 2006), while the nape tends to be the least exposed to external contamination (Jenkins, 1979). Thus, occipital hair is the preferred site of hair collection as recommended previously (Schramm, 2008), particularly in consecutive cohort studies.

Among studies which specifically stated the site of hair collection, about 40 % collected hair samples from the roots or close to the roots (Fig. 1). Some researchers have recommended that hair samples should be cut as close to the scalp as possible (Qiao et al., 2019; Schramm, 2008). For example, Qiao et al. performed correlation analysis and found that the polybrominated diphenyl ether (PBDE) congener profile in proximal segments was closer to that of serum while the PBDE congener profile in distal segments was closer to that of indoor dust (Qiao et al., 2018). Therefore, hair should be cut close to the roots for the purposes of assessing systemic MOC burden.

In most investigations, dyed or permed hair was excluded to avoid pitfalls in interpretation. Hair care and dyeing can lead to changes in hair construction and composition (Appenzeller and Tsatsakis, 2012; Schramm, 2008). For example, it has been reported that the oxygen content in melanin granules was increased by bleach treatment (Kojima et al., 2015). In addition, perms can reduce the levels of certain metals in hair such as mercury (Xue et al., 2014). However, studies that investigate differences in MOC measurements before and after hair treatments are scarce. Claessens et al. (2022) reported that hair treatments had no significant effect on the

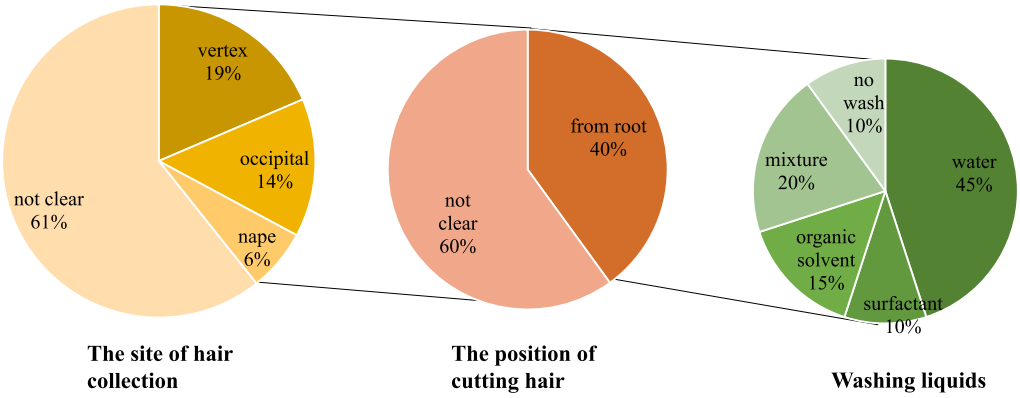

Fig. 1. Proportion of different measures about hair collection and purification for MOCs.

measurement of multiple endocrine disruptors. Thus, further studies on the influence of hair treatments on MOC measurement are required.

3.3. Sample preparation

In general, sample preparation for hair analysis includes decontamination, homogenization, extraction, and clean-up. Kucharska et al. (2015b) proposed that untreated hair could be used as a suitable indicator for integral (endogenous and exogenous) exposure and that any external contamination on the hair should be deposited from within the personal cloud of the sampled individual. However, in many situations, measurement of systemic MOC burden is desired and thus endogenous exposure must be distinguished from exogenous exposure. Therefore, a washing process is necessary to remove fat and sweat as well as contamination from the surface of hair. Optimal washing conditions involve warm solutions, multiple washes with small volumes, and ultrasonic baths (Zheng et al., 2013). Washing solutions vary, but often involve water, surfactant, and organic solvent. For drug testing in hair, organic solvent such as dichloromethane or acetone is generally accepted removing surface contamination (Cooper et al., 2012). As for MOCs, Toriba et al. (2003) indicated that hexane or dichloromethane could removal efficiently external contamination without extracting analytes from the hair matrix, but methanol wash resulted in over-extraction. Poon et al. (2015) found that water was the most effective at removing external contamination while maintaining analytes from the hair matrix. Comparison of different types of washing liquids using scanning electron microscopy (SEM) showed that hair shafts cleaned with warm Milli-Q water, hexane, acetone, or the hexane/dichloromethane mixture appeared smooth, while hair treated with dichloromethane exhibited probable structural damage (Zheng et al., 2013). Given decontamination efficiency and environmental impact, water washing is recommended as the preferred method to remove impurities from the surface of hair. As shown in Fig. 1, among studies in which hair samples were collected from nape, occipital or vertex and cut from hair root, 45 % papers used only water for decontamination.

After washed hair is dried and cut into small pieces (1–3 mm), approximately 100 mg of homogenized sample can be used to extract analytes due to the sensitivity of analytical methods (Li et al., 2013; Tang et al., 2021). Extraction methods depended on the nature of the analytes. For instance, polycyclic aromatic hydrocarbons (PAHs) and their metabolites should be extracted using dichloromethane following hydrolysis of hair specimens with NaOH, which could help generate homogeneous sample solutions and extract target analytes (Schummer et al., 2009). However, because OCPs (DDT and HCH) and higher-chlorinated PCBs can be destroyed by NaOH, hair samples should be digested with acids (Covaci and Schepens, 2001). For PBDEs, PAEs and OPFRs, acid digestion is also applied as a usual incubation method (Covaci and Schepens, 2001; He et al., 2018; Liang et al., 2016; Liu et al., 2016; Liu et al., 2015; Tadeo et al., 2009; Tang et al., 2021; Zheng et al., 2016). As for *per*- and polyfluoroalkyl substances (PFAS) and bisphenol A (BPA), organic solvent extraction from powdered samples is most efficient, since acid and alkaline digestions can lead to release of interfering compounds from the damaged biomineral structure of hair (Li et al., 2012; Martin et al., 2019). In particular, Di-(2-ethylhexyl)phthalate (DEHP) metabolites represented higher extraction efficiency when adding trifluoroacetic acid (TFA) (Chang et al., 2013; Hsu et al., 2022). Unlike hair test of drugs, extraction methods for analyzing MOCs from hair are mainly validated using authentic hair samples nor standard reference materials (SRM) (Welch et al., 2003). To increase the credibility of method validation, matrix effect assessment is warranted.

Prior to instrumental analysis, impurities present in the aqueous extract or digestion solution should usually be removed by solid phase extraction (SPE) cartridges or clean-up columns. For example, extracts for analyzing non-polar to moderately polar compounds (e.g., PCBs, PBDEs, pesticides, phthalate esters (PAEs), bisphenols, alkylphenols and organophosphorus flame retardants (OPFRs)) can be purified using a Florisil cartridge or a HLB cartridges (Celik et al., 2021; Hsu et al., 2022; Lee et al., 2017; Li et al., 2021b; Liu et al., 2015; Tang et al., 2021; Yin et al., 2019), while strong acid substances, such as PFAS, are usually purified using weak anion exchange (WAX) cartridges (Kim et al., 2019; Liu et al., 2020; Wang et al., 2018b). In addition, Envi-Carb cartridge have been used to achieve quick cleanup of PFAS in hair (Feng et al., 2021; Ruan et al., 2019). Regarding pesticides in hair samples, a Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) method has been developed from pesticide analysis in food samples (Lehmann et al., 2018a; Park et al., 2021; Peng et al., 2020b). Obviously, developing a simple and efficient method for multi-class MOCs has been a trend. For example, a method by Iglesias-Gonzalez et al. (2020) used 50 mg of hair powder and only simple overnight incubation was carried out before performing solid-phase microextraction (SPME). Similarly, Ren et al. (2020) develop a method for simultaneous analysis of typical halogenated endocrine disrupting chemicals and metal(loid)s in human hair, and the supernatant phase of extraction by acetonitrile solvent was quantified directly by instrumental analysis. Such simple preparations may be due to smaller sample size.

3.4. Instrumental analysis

In general, liquid chromatography-tandem mass spectrometry (LC-MS/MS) is employed to analyze polar organic substances, while gas chromatography–mass spectrometry (GC–MS) or -tandem mass spectrometry (GC–MS/MS) is employed to analyze nonpolar organic substances. GC–MS analysis also performs well on derivatized polar analytes (Hardy et al., 2015; Kokkinaki et al., 2014; Soulard et al., 2022). Over the last decade, LC-MS/MS has been used more frequently, due to increasing interest in polar substances such as organophosphorus esters (OPEs) and PFAS. High resolution mass spectrometry has already proved useful for the measurement of chlorinated compounds (e.g. PCBs) (Helmfrid et al., 2015; Tzatzarakis et al., 2014), and time-of-flight mass spectrometry (TOF-MS) or Orbitrap mass spectrometry can aid in identifying novel compounds (Piva et al., 2021a; Shih et al., 2019). Finally, MALDI-MS has been employed to

image the incorporation of drugs into the hair matrix (Kamata et al., 2015), which may prove a novel analytical method for hair analysis for MOCs.

4. Detection of MOCs in human hair

While incorporation mechanisms and source differentiation have not been completely resolved, there is a growing body of data on MOC analysis in hair. Due to the chemical differences between MOCs, it is necessary to discuss the reliability of hair biomonitoring for different MOC groups separately.

4.1. Methods

In this review, Web of Science Core Collection databases were searched for articles that reported MOC contamination levels in human hair including chlorinated persistent organic pollutants (chlorinated POPs), brominated flame retardants (BFRs), organophosphorus flame retardants (OPFRs), non-persistent pesticides, PFAS, PAEs, bisphenols, alkylphenols, and PAHs. There is no limit on year published in this screening process. Keywords and search results for each type of MOC are listed in Table 1. Articles that did not discuss MOCs in human hair and review articles were excluded. For ease of reading, abbreviations for each of the mentioned chemicals are listed in Table 2.

4.2. Chlorinated POPs in human hair

PCDD/Fs and PCBs are likely produced unintentionally due to incomplete combustion or during the manufacture of pesticides and other chlorinated substances. To our knowledge, PCDD/Fs were the first reported MOCs in hair, in a study that compared washed hair and unwashed hair to reflect the systemic burden of airborne PCDD/F in single persons (Schramm et al., 1992). Many subsequent papers suggested that human hair can adequately indicate the atmospheric levels and total human exposure of dioxin-like compounds (Liu et al., 2019; Ma et al., 2011; Nakao et al., 2005). In contrast, PCBs are used widely in industry and consumer products as insulating fluids and additives. Many studies have reported the levels of PCBs in humans from different countries despite the fact that PCBs have been banned for a few decades (Barmpas et al., 2020; Malarvannan et al., 2013; Peng et al., 2020a; Wielgomas et al., 2012; Zheng et al., 2013). Peng et al. (2020a) found that French women had significantly higher detection frequencies (DFs) and concentrations of all PCBs than Chinese women using hair analysis. This result is in line with fact that France contributed to a larger proportion of historical global PCB consumption than China (Breivik et al., 2002; Peng et al., 2020a), suggesting that hair analysis could reflect long-term exposure (months/years).

Organochlorinated pesticides (OCPs: including DDTs, HCHs, HCB, Endosulfan, etc.) were widely used as pesticides in the past due to their efficiency, economy, and lack of acute toxicity in humans. However, their physical and chemical properties lead to long half-lives and widespread environmental distribution (Liu et al., 2016; Zheng et al., 2016). Numerous studies have identified OCP contaminants in human hair (Dahmardeh Behrooz et al., 2012; Lu et al., 2014; Peng et al., 2021; Yuan et al., 2017). He et al. (2017) reported DDT levels in the general population using large-scale hair sampling and found that while there was no significant difference in the hair concentrations of ΣDDTs between the rural and urban areas, hair concentrations of DDD and DDT in rural areas were significantly higher than those in urban areas. This result is not consistent with findings from children in rural and urban areas of Beijing by Zhang et al. (2007a). This may be because of age or geography on exposure.

DP is an additive chlorinated flame retardant using in many polymeric systems, like thermoplastics. Zheng et al. (2010) first determined *syn*- and *anti*-DP isomers in hair samples from an e-waste recycling area and two control areas in South China. And significantly positive correlation between DP concentrations in dust and hair indicated that dust may be one of the major routes for DP exposure. Segmental analysis found that the proximal segments of hair had low DFs of DPs (Qiao et al., 2018). In recent years, DPs in hair were less reported, possibly due to the ban on the substance (Tang et al., 2022).SCCPs are used as plasticizers and lubricants and were listed as POPs in the Stockholm Convention of 2017. However, research on human exposure to SCCPs is scarce (Jamdar and Osborne, 1982; Nevondo and Okonkwo, 2021). Han et al. (2021) first reported SCCP contamination patterns in hair in North China. There, concentrations of SCCP in hair samples were positively correlated with age, possibly due to different exposure times to pollutants or different metabolic/biotransformation abilities among different age groups (Han et al., 2021).

4.3. BFRs in human hair

BFRs have been widely used in consumer/industrial products (e.g., furniture, electronics, and building materials) to reduce the risk of fire (Rudel and Perovich, 2009). With the use and reuse of these products, BFRs are continuously exposed to residents and workers and can incorporate into human hair by endogenous and exogenous pathways. Hajeb et al. (2022) have reviewed analytical methods for the measurement of flame retardants in human hair until 2020, including PBDEs, HBCDs, and novel brominated flame retardants (NBFRs). PBDEs were detected in the hair of electronic waste (e-waste) recycling workers at up to 18-fold higher concentrations than that of general residents (Lin et al., 2019; Zhao et al., 2008; Zheng et al., 2011; Zheng et al., 2010). Malarvannan et al. (2013) found that primipara and multipara mothers living near a waste dumpsite in the Philippines had relatively high HBCDs in their hair. Since the production and use of PBDEs and HBCDs have been eliminated under the Stockholm Convention, NBFRs have started to draw widespread attention. In human hair, a variety of NBFRs (e. g. DBDPE, HBB, and BTBPE) were found in higher amounts in occupationally-exposed workers versus residents of an e-waste recycling area, which were in turn higher than residents

Table 1
Keywords and search results for each type of MOCs: papers identified in multiple categories were counted multiple times.

Categories of target analytes	Keywords	Number of selected papers
PCDD/Fs	AB = (hair AND (PCDD/F OR dioxin) AND (concentration OR ng/g))	12
PCBs	AB = (hair AND (PCB OR polychlorinated biphenyl) AND (concentration OR ng/g))	40
OCPs	AB = (hair AND (DDT OR DDE OR HCH OR HCB OR OCPs OR organochlorine pesticides) AND (concentration OR ng/g))	46
DPs	AB = (hair AND (DPs OR Dechlorane Plus) AND (concentration OR ng/g))	4
SCCPs	AB = (hair AND (SCCPs OR chlorinated paraffins) AND (concentration OR ng/g))	2
BFRs	AB = (human hair AND (PBDE OR Deca-BDE OR HBCD OR Brominated flame retardant OR BFRs) AND (concentration OR ng/g))	20
OPFRs	AB = (hair AND (TPhP OR TEHP OR TCEP OR phosphate phosphorus flame retardants OR OPFR OR OPEs) AND (concentration OR level OR ng/g))	12
Non-persistent pesticides	AB = (hair AND (organophosphorus pesticide OR pyrethroid OR Terbuthylazine OR tebuconazole OR Glyphosate OR Folpet OR Mancozeb OR non-persistent pesticide) AND (concentration OR level OR ng/g))	22
PFAS	AB = (hair AND (PFOS OR PFOA OR PFAS OR PFCs) AND (concentration OR level OR ng/g))	19
PAEs and APs	AB = (hair AND (MEHP OR DEHP OR DPhP OR phthalate OR plasticizer) AND (concentration OR level OR ng/g))	19
Bisphenol analogs and alkylphenols	AB = (hair AND (BPA OR bisphenol OR alkylphenols) AND (concentration OR level OR ng/g))	15
PAHs	AB = (hair AND (PAH) AND (concentration OR level OR ng/g))	23

Table 2
The target analytes and their abbreviations mentioned in this review.

Target analytes	Abbreviations
polychlorinated dibenzo-p-dioxins	PCDD
polychlorinated dibenzofurans	PCDF
tetrachlorodibenzo-p-dioxin	TCDD
hexachlorodibenzo-p-dioxin	HxCDD
octachlorodibenzo-p-dioxin	OCDD
pentachlorodibenzofuran	PeCDF
polychlorinated biphenyls	PCBs
dichlorodiphenyltrichloroethane	DDT
dichlorodiphenyldichloroethane	DDD
hexachlorocyclohexanes	HCHs
hexachlorobenzene	HCB
dechlorane Plus	DPs
short-chain chlorinated paraffins	SCCPs
polybrominated diphenyl ethers	PBDEs
hexabromocylododecane	HBCDs
decabromodiphenylethane	DBDPE
hexabromobenzene	HBB
1,2-bis(2,4,6-tribromophenoxy)-ethane	BTBPE/TBE
pentabromobenzene	PBBz
di(2-ethylhexyl) tetrabromophthalate	BEH-TEBP/TBPH
pentabromotoluene	PBT
2-Ethylhexyl-2,3,4,5-tetrabromobenzoate	EH-TBB/TBB
pentabromoethylbenzene	PBEB
tetrabromobisphenolA	TBBPA
triphenyl phosphate	TPhP
tris(2-butoxyethyl) phosphate	TBOEP/TBEP
2-ethylhexyl diphenyl phosphate	EHDPhP
tri-n-butyl phosphate	TnBP
tris(1,3-dichloro-2-propyl) phosphate	TDCIPP/TDCPP
tri-cresyl phosphate/tris(methylphenyl) phosphate	TCP/TMPP
tris(chloroethyl) phosphate	TCEP
tris(2-ethylhexyl) phosphate	TEHP
tri-isobutyl phosphate	TiBP
Tris(2-chloroisopropy) phosphate	TCIPP/TCPP
triisopropyl phosphat	TiPP/TiPrP
tris(methylphenyl) phosphate	TMPP
tripropyl phosphate	TPRP
tripropyl phosphate	TPP/TPrP
triethyl phosphate	TEP
2,2-bis(chloromethyl)-propane-1,3- diyltetrakis(2-chloroethyl) bisphosphate	V6
isodecyl diphenyl phosphate	iDDPHP
bisphenol A bis (diphenyl phosphate)	BDP
resorcinol bis (diphenyl phosphate)	RDP
diphenyl phosphate	DPhP
bis(2-butoxyethyl) phosphate	BBOEP
pentachlorophenol	PCP
p- nitrophenol	PNP
3-methyl-4-nitrophenol	3Me4NP
2-isopropyl-4-methyl-6-hydroxypyrimidine	IMPy
perfluorobutanesulfonic acid	PFBS
perfluorohexanesulfonic acid	PFHxS
perfluorooctanesulfonic acid	PFOS
perfluorodecanesulfonic acid	PFDS
perfluorododecanesulfonic acid	PFDoS
perfluorobutanoic acid	PFBA
perfluorohexanoic acid	PFHxA
perfluoroheptanoic acid	PFHpA
perfluorooctanoic acid	PFOA
perfluorodecanoic acid	PFDA
perfluorododecanoic acid	PFDoA
C8 chlorinated polyfluoralkyl ether sulfonic acid	C8 Cl-PFESA
6:2 fluorotelomer unsaturated carboxylic acid	6:2 FTUCA
hexafluoropropylene oxide dimer acids	HFPO-DA
hexafluoropropylene oxide trimer acids	HFPO-TA
di-(2-ethylhexyl) phthalate	DEHP
mono-(2-ethylhexyl) phthalate	MEHP
mono-(2-ethyl-5-hydroxyhexyl) phthalate	MEHHP
mono-(2-ethyl-5-oxy-hexyl) phthalate	MEOHP
monoethylphthalate	MEP
bisphenol A	BPA
bisphenol S	BPS
4-tert-octylphenol	OP
4-nonylphenol	NP

of urban and rural areas (Qiao et al., 2016; Zheng et al., 2011). For the general population, HBB, PBBz, TBPH, PBT and TBB were also detected with relatively high DFs (> 50 %) and concentrations in hair samples (Li et al., 2018; Liu et al., 2016; Yuan et al., 2016). In addition, TBBPA, a reactive intermediate of some flame-retarded products and an additive flame retardant in antilock brake systems, has been mass produced and globally consumed (Hajeb et al., 2022). However, there is little information about TBBPA in human hair. Barghi et al. (2018) demonstrated the presence of TBBPA in human hair with nonspecific exposure and found significantly higher concentrations of TBBPA in hair samples from Korea than those from Iran.

4.4. OPFRs in human hair

OPFRs, as alternatives of BFRs, have been the subject of concern in recent years due to their global occurrence and endocrine disrupting toxicity. Kucharska et al. first developed methods to measure OPFRs in human hair and detected a variety of OPFRs (TPhP, TBOEP, EHDPhP, TnBP and TiBP) in all human hair samples from a Norwegian mother–child cohort (Kucharska et al., 2015a; Kucharska et al., 2014). He et al. (2018) found that the concentrations of OPFRs in urban hair were significantly higher than those in rural hair. In terms of occupational exposure, Qiao et al. (2019) reported levels of OPFRs in hair of female e-waste dismantling workers and found that the total concentration of OPFRs exceeded that of other flame retardants including PBDEs, DPs and NBFRs (HBCDs were not measured). Further, a study by Tang et al. (2022) on temporal changes in the levels of "legacy" and "emerging" flame retardants showed a significant increase in the ratios of TPhP and TCIPP ("emerging") to Deca-BDE ("legacy") from 2015 to 2019, implying a shift from legacy to emerging contaminants in recent years. Meanwhile, four emerging OPFRs including V6, iDDPHP, BDP, and RDP were detected in 100 % of the hair samples from e-waste dismantling workers, implying widespread use of these novel chemicals in the electronic products dismantled in this area (Tang et al., 2022). It is well known that OPFRs are readily metabolized in humans, and OPFR metabolites have been considered as biomarkers of internal exposure to OPFRs. Alves et al. (2017) developed an analytical method for OPFR metabolites in hair, but only DPhP and BBOEP were detected in hair samples from Norway (Xu et al., 2019).

4.5. Non-persistent pesticides in human hair

In addition to legacy pesticides, currently-used organophosphorus pesticide (OPPs), pyrethroid insecticides, and herbicides have long been a concern for environmental and occupational exposure, even though they are easily degraded and demonstrate low toxicity (Kokkinaki et al., 2014; Yusa et al., 2015). Hardy et al. (2015) first developed and validated multi-residue analytical methods for OCPs, OPPs, pyrethroids, carbamates, and other pesticides and their metabolites. Peng et al. (2021) evaluated multiple pesticide exposures in the general population using hair samples collected from the Observation of Risks and Cardiovascular Health in Luxembourg (ORISCAV-LUX) cohort. They observed 13 types of non-persistent pesticides (including OPPs, pyrethroids, carbamates, and phenylpyrazoles) in >50 % of the hair samples. Ostrea et al. compared several classes of pesticides in maternal (hair and blood) and infant (cord blood, infant hair, or meconium) samples and found that combined analysis of maternal hair and meconium further increased DFs and that maternal hair analysis had the advantage of detecting prenatal pesticide exposure (Ostrea Jr. et al., 2009; Ostrea Jr. et al., 2008). Iglesias-Gonzalez et al. (2020) accessed the cumulative exposure of French children to OPPs, pyrethroids, azoles, dinitroanilines, oxadiazines, phenylpyrazoles and other organic pollutants by hair analysis and observed PCP, PNP and 3Me4NP presented statistically significant higher concentration in younger children. Besides, many studies collected hair samples for biomonitoring of non-persistent pesticides and their metabolites, suggesting the significance of hair for the biomonitoring of exposure (Beranger et al., 2018; Lehmann et al., 2018b; Margariti and Tsatsakis, 2009; Polledri et al., 2021; Polledri et al., 2019; Schummer et al., 2012; Tsatsakis et al., 2008).

4.6. PFAS in human hair

PFAS are used in various products on account of their chemical stability, ability to lower surface tension, and hydrophobicity. Their widespread use, in addition to their environmental persistence and bioaccumulation, has led to their ubiquity in environmental, wildlife, and human samples (Meegoda et al., 2020). PFOA and PFOS were the most frequently detected PFAS in human hair, while PFBS, PFHxS, PFDS, PFDoS, PFBA, PFHxA, PFHpA, and PFDA were detected at relative high DFs in some regions (Alves et al., 2015; Feng et al., 2021; Kim and Oh, 2017; Li et al., 2013; Li et al., 2021a; Martin et al., 2016; Perez et al., 2012; Piva et al., 2021a; Rodriguez-Gomez et al., 2017; Ruan et al., 2019; Wang et al., 2018a; Wang et al., 2018b). Of note, a few alternatives to legacy PFAS, such as C8 Cl-PFESA, 6:2 FTUCA, HFPO-DA, and HFPO-TA, have also been found in human hair samples (Feng et al., 2021; Ruan et al., 2019; Wang et al., 2018a). In the general population, no significant difference in PFAS levels were observed between gender, age, hair treatments, and hair length (Claessens et al., 2022; Piva et al., 2021b).

4.7. Phthalate esters and alternative plasticizers in human hair

Phthalate esters (PAEs) are used in medical equipment, building materials, food packaging materials, and personal care products, and they are commonly found in indoor dust, air, and human bodily fluids such as blood, breast milk, and urine (Bornehag et al., 2005; Gaspar et al., 2014; Liu et al., 2022; Pei et al., 2013; Preuss et al., 2005). There is currently little information on exposure to PAEs in human hair. Chang et al. (2013) first assessed the exposure of DEHP in human hair, and identified three metabolites (MEHP, MEHHP, and MEOHP) with high levels and DFs. He et al. (2018) reported PAE parent compounds (including high molecular-weight PAEs and low molecular-weight PAEs) in human hair and found the PAE level in the hair of rural residents was higher than that of urban residents, while the opposite was true for OPFRs. As alternative plasticizers (APs) were introduced as replacements for PAEs, they have also been detected in the general population. Yin et al. (2019) performed a pilot biomonitoring study on AP metabolites in hair and identified four metabolites with a high DF. Based on previous results, hair is a better reservoir for more hydrophobic metabolites than blood and urine. A follow up on human volunteers with repeated urine and hair collection found that the variability of PAE concentration was much lower in hair, indicating hair providing more stable information over time than urine samples (Fays et al., 2021). In recent years, more studies on human biomonitoring have chosen hair as an alternative matrix to characterize chronic exposure of PAEs (Cai et al., 2023; Li et al., 2021b; Ruiz-Castell et al., 2023).

4.8. Bisphenols and alkylphenols in human hair

Bisphenols and alkylphenols are used in plastic products and personal care products (PCPs) and are also known to be classic endocrine disrupters (EDCs). BPA has been measured in different biological matrices, with Tzatzarakis et al. (2015) first reporting BPA in hair. Nehring et al. (2017) first measured BPA, OP, and NP in human hair simultaneously, and found that high EDCs in hair were significantly influenced by diet and lifestyle (e.g., preferring products of marine origin, heating food with plastic containers, using household chemicals without protective gloves, and wearing new clothes without pre-washing). Rodríguez-Gómez et al. (2017) develop a multi-class method for the determination of 21 EDCs including BPA and its chlorinated derivatives in human hair samples. Gonkowski et al. (2022) analyzed BPA and its analogue BPS in hair samples from Poland and found that BPS concentration levels were higher. Martin et al. (2019) found that the concentrations of BPA in adults' hair were higher than in children's but Karzi et al. (2018) observed the opposite result, perhaps because of different life habits in different regions. Compared with other matrix (including breast milk, serum, and urine), hair samples were found to have the highest levels of BPA and were least influenced by plastic products in the proceeding of collection and storage, revealing to be the best matrix for biomonitoring of BPA (Lee et al., 2017; Martin et al., 2019; Rodríguez-Gómez et al., 2017).

4.9. PAHs in human hair

PAHs are a large group of ubiquitous contaminants that are by-products of incomplete combustion of organic matter. People are exposed to PAHs mainly through dietary intake and inhalation of atmospheric and particulate matter (Urbancova et al., 2016). Toriba et al. (2003) first determined 14 PAHs in human hair samples and found the concentration of PAHs in hair tended to increase with decreasing of number of rings, which was consistent with profiles in cigarette smoke, as well as indoor and outdoor air. Not surprisingly, PAH levels were found to be higher in hair from smokers than those from non-smokers in multiple studies (Toriba et al., 2003; Wang et al., 2020). Schummer et al. (2009) was the first to detect hydroxylated PAH metabolites (OH-PAHs) in hair, providing a reliable tool for the assessment of chronic exposure to PAHs that avoids bias due to external contamination.

5. In vivo and in vitro experiments

With optimized protocols, hair is an acceptable and reliable measure for assessing the integral exposure of MOCs. When assessing internal (endogenous) exposure to MOCs, external contamination is a significant interference factor. Thus, In vivo experiments may be designed excluding external contamination to verify that hair samples reflect the internal dose of MOCs. Meanwhile several in vitro experiments have explored the influence of external contamination to MOCs on the hair matrix.

To clarify the origin of dioxins and dioxin-like compounds in human hair, Miyabara et al. (2005) determined the adsorption quantity of dioxins in human hair and the distribution of 2,3,7,8-TCDD in rats. Compared to hair samples unexposed to ambient air, hair samples exposed to air for one day showed a 51 % increase in their total Toxic Equivalent Quantity (TEQ), reflecting the capacity of human hair to adsorb dioxins from ambient air (Miyabara et al., 2005). In an animal experiment, the amount of TCDD in hair of TCDD-treated rats increased in a dose-dependent manner and showed a significant positive correlation to concentrations in adipose tissue (Miyabara et al., 2005). This experiment strongly suggests that hair can reflect the internal dose of dioxins and dioxin-like compounds.

PBDEs show similar persistence and lipid solubility to dioxins (Petreas et al., 2004), and thus would be expected to show similar results in animal experiments. In Poon et al. (2014), adult male Sprague − Dawley rats were exposed to increasing doses of a PBDE mixture. Except for BDE-28 and BDE-183, which displayed low DFs, significant correlations were observed for other PBDE congeners in rat hair, serum, liver, and fat across doses, with r values ranging from 0.803 to 0.988 (Poon et al., 2014). However, the correlation of BDE-209 between hair and other matrices was weaker than other congeners when exposed at lower levels ((r = 0.603–0.635) (Poon et al., 2014). This result suggests that different POPs may have slightly different distribution or transport mechanism in hair and other tissues and that correlations cannot be inferred from similar chemicals.

PBDEs and other BFRs have also been investigated in vitro. In a mimicking experiment, hair samples collected from one volunteer were exposed to OPFR and PBDE standards via evaporation of the solvent in plastic boxes. Levels of certain congeners (BDE-153, BDE-183) did not increase with exposure time (24 h, 48 h, 72 h and 10 days), while other PBDEs (BDE-28, −47, −100, −99, −154) did not reach steady-state during this same time frame, which suggests that the substances with high vapor pressure have relatively higher transfer efficiency (Kucharska et al., 2015b). In another investigation of HBCD and TBBPA, human hair samples were exposed via volatilization and deposition of standard solvents in a plastic box or via storage in an office with relatively high levels of indoor dust. HBCD and TBBPA concentrations in hair samples showed no increase in either the closed box or the open room after one week (Barghi et al., 2018). These results are consistent with a previous study which showed that lower-brominated PBDE congeners showed higher contributions from exogenous

exposure (Zheng et al., 2014). In other words, BFRs with higher octanol-water partition coefficients (K_{ow}) and lower volatility values are more likely to reflect internal exposure.

As for OPFRs, only TDCIPP has been studied in a controlled animal experiment. Mice administered 300 mg/kg/day TDCIPP for 35 days showed significantly higher loads of TDCIPP in hair than in other tissues. Four out of eleven types of TDCIPP metabolites were detected in hair samples, suggesting that hair could serve as a reliable biomarker for TDCIPP (Zhu et al., 2020). In an in vitro experiment measuring OPFRs and PBDEs, TDCIPP and TnBP reached steady-state in 24 h, while other OPFRs (TCP, TPhP, TBEP, TCEP, TCPP and TEHP) reached steady-state in 48 h, implying that air might be a potential source of exposure (Kucharska et al., 2015b). Thus, hair may not reflect systemic burden of OPFRs unless exogenous pollution is excluded.

Several studies on pesticides indicate that hair analysis is a reliable tool to investigate exposure-associated adverse health effects. Tutudaki and Tsatsakis (2005) found that Sprague-Dawley rats systemically exposed to two doses of the pesticide diazinon showed dose-dependent diazinon concentrations in hair, presenting the possibility of using hair analysis for low-level exposure monitoring of diazinon. Chata et al. (2016) investigated the correlation between blood and hair concentrations of 23 pesticides/metabolites from different chemical classes in rats submitted to chronic controlled exposure, and all the investigated compounds demonstrated significant association between hair and plasma concentrations without the influence of the physicochemical parameters. Appenzeller et al. (2017) also observed a linear relationship between exposure intensity and the concentration of pesticides in rat hair, and a comparison with urine and plasma samples demonstrated the accuracy of hair analysis. Thus, hair can reliably reflect the internal exposure of pesticides.

Unlike other POPs, PFAS have been found to bind to some proteins (e.g., serum albumin, β-lipoproteins, and fatty acid-binding proteins) (Sznajder-Katarzyńska et al., 2019). Considering hair is composed mainly of protein, it is necessary to systematically investigate the correlation between hair and serum concentrations of PFAS in animal models. Gao et al. (2015) exposed adult male and female rats to PFOA, PFNA, and PFOS for 90 days, and these PFAS were detected in rat hair in a dose-dependent manner. Significant positive correlations and similar PFAS profiles were observed between hair, serum, and other tissues, suggesting that hair could be used as an exposure bioindicator (Gao et al., 2015). However, as PFAS exposure increased, hair-to-serum ratio was decreased (Gao et al., 2015). This phenomenon implies that high level PFAS incorporation into hair may not only depend on passive fusion, and further mechanisms will need to be investigated.

Exposure experiments on PAEs are scarce. Hsu et al. evaluated long-term exposure to DINP using rat hair. DINP metabolites showed a positive correlation with increasing administered dose, and significant positive correlations for MINP, MOINP and MHINP were found between hair and urine (Hsu et al., 2015). Given similar metabolic mechanisms of different PAEs, it can be inferred that hair is a reliable tool in the assessment of long-term exposure to PAEs. In fact, DINP metabolites in urine showed earlier saturation than those in hair, demonstrating the advantages of hair analysis for PAEs (Hsu et al., 2015).

Like PAEs, PAHs are easily metabolized. Some animal experiments have been conducted on PAHs and their metabolites. In a 28-day exposure experiment, rats were injected intraperitoneally with 12 PAHs at 0.01, 0.1, and 1 mg/kg, and consequently 20 out of 50 metabolites were detected in hair samples that corresponded to nine parent PAHs, demonstrating the incorporation of PAH metabolites in hair and sufficient sensitivity of hair analysis for most PAHs (Grova et al., 2013). In another 90-day exposure experiment, a stronger linear relationship was observed between exposure level and concentration of OH-PAH in rat hair than in rat urine for 28 out of 54 OH-PAH (Grova et al., 2018). In addition, urine showed the highest concentration levels for OH-PAHs with less than five aromatic rings, while hair showed the highest concentration levels for OH-PAHs with five aromatic rings (Grova et al., 2018). Thus, hair is not only a reliable alternative matrix but also an irreplaceable tool for assessing exposure to PAHs.

In conclusion, exposure experiments indicate that the internal exposure of most POPs, especially pollutants with higher K_{ow} and lower volatility, can be reliably assessed using hair analysis of parent compounds, while internal exposure of other chemicals can be assessed by hair analysis of their metabolites. However, some classes of MOCs, such as PCBs, SCCPs, and BPA, have not been verified in in vivo experiments, so their reliability remains unknown.

6. Correlation studies on human biomonitoring for MOCs

In addition to controlled in vivo and in vitro experiments, the correlation between MOC levels in hair and other tissues from exposed populations can demonstrate the reliability of hair biomonitoring to some extent. Although the reflection periods vary across biospecimens, stable working environment of occupational populations is likely to result in consistency between short-term exposure and long-term exposure. If hair specimen can reliably reflect internal exposure to MOCs, good correlation of MOCs concentrations between hair and blood/urine sample is expected to observe.

For chlorinated POPs, many correlation studies indicate that hair is a useful matrix for monitoring human exposure. Li et al. (2020) determined 17 congeners of PCDD/F in paired hair and serum of municipal solid waste incineration plant workers and observed strong positive correlations for two PCDDs (1,2,3,6,7,8-HxCDD and OCDD). In previous studies, the concentrations of 2,3,4,7,8-PeCDF between paired serum and hair samples from six non-occupationally exposed volunteers exhibited significant positive correlations, and the TEQ values in human hair from 10 pregnant woman exhibited significant positive correlations with those in placenta and human milk (Chan et al., 2007; Nakao et al., 2002).

For PCBs, correlations between hair and serum samples have been investigated with different results. Nakao et al. (2002) reported that coplanar polychlorinated biphenyls (Co-PCBs) showed no correlation between hair and blood from 6 Japanese volunteers, although the sample size was small. Altshul et al. (2004) found moderate correlations for some of PCB congeners from 10 volunteers but other PCB congeners showed no correlation or weak correlation. Zheng et al. (2016) analyzed 34 matched hair and serum samples from e-waste recycling workers, and observed that almost all PCB congeners showed significant correlations, although correlations for lower chlorinated congeners were stronger than those of highly chlorinated congeners. Barmpas et al. (2020) determined a statistically significant correlation for PCB101 based on 120 paired hair-serum samples from pregnant women in Greece, but PCB congeners in human matrices had relatively low DFs. Amir et al. (2021) focused on infertile men in Pakistan and found that hair samples had no significant association with serum/urine concentrations of PCBs. However, it is a possibility that PCBs in hair samples were lost due to washing with n-hexane/dichloromethane in this study. In general, measuring procedures and detected sample amounts may influence correlations between hair and serum for PCBs and affect the reliability of hair biomonitoring.

There are many studies on PBDE occupational or non-occupational exposure using both hair and serum (Liang et al., 2016; Liu et al., 2016; Poon et al., 2014; Qiao et al., 2018; Zhao et al., 2020; Zheng et al., 2014). Zhao et al. (2021) performed a meta-analysis by pooling the correlation coefficients of the five main congeners of PBDEs from 7 studies and established human hair as a reliable indicator of exposure to PBDE congeners. As for DPs and other BFRs, studies measuring both hair and blood are relatively rare. The concentrations of DPs in paired blood and hair samples from e-waste recycling workers or DP workers showed positive correlations, indicating that DP in human hair could be used as a reliable biomonitoring tool (Chen et al., 2015; Zhang et al., 2013; Zhao et al., 2020). Qiao et al. (2018) measured DBDPE in paired human serum and hair but found no significant correlation. Thus, more studies are necessary to fully assess the reliability of human hair analysis for NBFRs.

Correlations between OPFR levels in hair and blood were not reported, mainly due to rare detection in blood. Some studies discussed correlations between OPFR/metabolites levels in hair and urine. DPhP is often detected

in urine as the common metabolite of TPhP and other OPFR (e.g. EHDPhP, RDP, iDDPHP, BDP), and it is not surprising to observe no significant association between DPhP in urine and DPhP/TPhP in hair (Kucharska et al., 2015a; Xu et al., 2019). BDCIPP is a primary metabolite of TDCIPP. (Kucharska et al., 2015a) found a correlation between TDCIPP in hair and BDCIPP in urine ($r = 0.352$, $p = 0.02$ for morning spot urine samples, $r = 0.422$, $p = 0.003$ for median urine samples), but in case of mothers only one positive correlation was found for log-transformed TDCIPP in hair and log-transformed BDCIPP in morning spot urine samples ($r = 0.395$, $p = 0.01$). It implies that TDCIPP levels in hair may not reflect levels of corresponding metabolites in urine. Indeed, hair segment sample (1–5 cm) reflects generally a long period of exposure (one month or more), while urine correspond to short-term exposure (may in the past hours). Therefore, to what extent OPFR/metabolites levels in hair are related with levels of OPFR long-time exposure is more noteworthy. According to the available research, it is hard to conclude the reliability of assessing OPFR long-time exposure using human hair.

Although animal experiments have proved the reliability of hair biomonitoring for pesticides, the correlations between human hair and blood samples are not always consistent. In Çelik's and Amir's studies, no correlation between pesticides detected in human hair and blood or urine samples was observed (Amir et al., 2021; Celik et al., 2021). We noticed that these two studies adopted dichloromethane as washing reagent, which may bias their results. As for pesticide metabolites, Kokkinaki et al. (2014) analyzed non-specific metabolites of OPPs and found that the sum of metabolite concentrations in hair and urine was significantly correlated. By contrast, Hardy et al. (2021) found no correlation between hair and urine concentrations for most pesticides and their metabolites, while Fays et al. (2021) found that out of four detected pesticides, only IMPy exhibited a correlation between the hair and urine. These differences may be to the different lengths of pesticide exposure. Considering that hair can reflect longer-term exposure than urine or blood, we still recommend hair as a reliable tool for assessing chronic exposure to pesticides in agricultural areas.

Studies on PFAS exposure also come to different conclusions. Li et al. (2013) evaluated the application of nail, hair, and urine for human biomonitoring of PFOS and PFOA, and found that PFOS in hair had a statistically significant relationship with that in serum samples, but that the relationship between PFOA in hair and serum was weaker than that in nails and serum. In contrast, Kim et al. (2019) did not observe relationships between serum and hair levels for most PFAS. This may be due to the interference of exogenous exposures like indoor dust. Feng et al. (2021) have found strong correlations between the concentrations of short-chain perfluoroalkyl carboxylic acids (PFCAs) in hair and those in indoor dust samples, suggesting indoor dust was an important source of short-chain PFCAs in human hair. Finally, it is possible that specific PFAS transfer mechanisms from blood to hair leads to weak correlations between hair and serum, given PFAS-protein binding effects (Chen and Guo, 2009). Although in vivo experiments suggest that PFAS in hair can reliably indicate PFAS exposure, before transfer mechanisms into human hair are defined, we do not yet know whether human biomonitoring for PFAS in hair will accurately assess systemic burden.

Correlation studies for plasticizer exposure are still lacking. Fays et al. (2021) collected hair and urine samples from a 6-month follow-up on human volunteers to test phthalates and bisphenols, and observed no significant correlation between the two matrices for most compounds (except MEP and 5-oxo-MEHP), probably due to the lack of representative of urinary biomarkers. Nevertheless, Li et al. (2021a) detected a significant correlation between BPF concentrations in hair and urine samples from 17 kindergarteners in Hong Kong. Thus, analyzing PAEs and other plasticizers using hair shows promise. More studies including human biomonitoring and animal experiments need to be conducted to improve hair analysis for PAEs and other plasticizers.

Since OH-PAHs in human hair come from endogenous exposure rather than external deposition, OH-PAHs in hair can reliably indicate internal exposure of PAHs (Lin et al., 2019). Lin et al. (2020) measured both PAHs and OH-PAHs in paired hair and urine samples from e-waste dismantling workers, verifying the reliability of OH-PAHs in hair as a biomarker for PAHs exposure. Although most of the individual OH-PAHs were not significantly correlated in hair and urine, the two matrices could be considered valid and complementary bioindicators due to differences in biotransformation and enrichment behaviors (Lin et al., 2020).

7. Application of human hair MOC analysis

According to in vivo/in vitro experiments, the reliability of human hair for biomonitoring has been verified for most MOCs. Combined with the unique advantages of hair samples, human hair should be widely used for MOC measurements. Here, we summarize several applications of human hair MOC analysis, which highlights the usefulness of human hair for MOC exposure assessment.

7.1. Large-scale surveys on MOCs

Hair analysis has the advantages of being non-invasive, low-cost, and easy to transport, and thus is uniquely suitable to monitor and evaluate human exposure to MOCs on a large scale. Based on large-scale surveys, we hope that reference (or background) ranges of MOCs in human hair can be established, so that researchers can interpret whether detected levels in hair are elevated due to environmental releases (Harkins and Susten, 2003). In fact, hair Hg already has set reference values and a reported reference range (Lu-lu et al., 2009; McDowell et al., 2004; Wu et al., 2021). While there are many reports on MOC levels in human hair from different geographical locations, there are few large-scale surveys on hair MOCs.

Although the Canadian Health Measures Survey (CHMS), a department providing general population human biomonitoring data in Canada, planned to analyze metals and trace elements in hair as part of cycles 5 and 6, analysis of organic compounds in human hair was not planned (Haines et al., 2017). Ruan et al. (2019) performed the first nationwide survey of hair to evaluate PFAS exposure in India, but only 39 samples were collected. Beranger et al. (2018) collected 311 hair samples from pregnant women enrolled in the ELFE birth cohort and analyzed about 180 pesticides and/or metabolites. Peng et al. (2021) assessed the exposure of 497 adults from Luxembourg to 67 organic pollutants (e. g. PCBs, PBDEs, OCPs) via hair analysis. Recently, a nationwide survey in China determined hair samples of adult residents in 10 and 17 provincial capital cities, fulfilling evaluating human exposure to MOCs in urban residents at a large scale (Li et al., 2023). In another recent research letter, an database and data visualization of POPs in China was released online (https://pops.hhra.net/) (Dong et al., 2021). Although the area is limited to China and human hair is not listed separately, it greatly improves exposure assessment of MOCs all over China. In the future, more nationwide data of MOC exposure in the general population is needed to establish reference ranges and background levels of various MOCs.

7.2. Retrospective cohort studies on MOCs

Hair samples are well known for their stability and easy storage, as well as the fact that they can be sampled multiple times. In addition, segmented hair analysis can provide valuable retrospective information on the history of MOC exposure. Therefore, hair biomonitoring provides an advantage for retrospective cohort studies and has been employed in a wide variety of contexts, including pre-natal drug exposure, anti-doping control, and so on (Barbosa et al., 2013). However, studies on retrospective exposure assessment of MOCs are scarce.

In one example, a retrospective cohort study that measured persistent organochlorine contaminants in hair samples collected in 1968, 1989, and 2009 observed a significant and continuous decrease of PCBs over the studied period, while p, p'-DDT were still present at high concentrations (Wielgomas et al., 2012). Interestingly, the samples in 1968 and 1989 were originally collected for estimating exposure to heavy metals and hydrogen fluoride in workers (Wielgomas et al., 2012). In a recent study, Tang et al. (2022) reported hair levels of PCBs, PBDEs, and OPFRs over ten years

Table 3
Evidence and conclusions on the reliability of hair analysis for different groups of MOCs.

Chemicals	Detection in human hair	In vivo experiments	In vitro experiments	Correlation studies	Applications	Conclusions of reliability
Dioxins	PCDD/Fs (Liu et al., 2019; Ma et al., 2011; Nakao et al., 2005; Schramm et al., 1992)	2,3,7,8-TCDD in rats (Miyabara et al., 2005)	No reports	2,3,4,7,8-PeCDF, 1,2,3,6,7,8-HxCDD and OCDD in human hair and serum (Li et al., 2020; Nakao et al., 2002); TEQ values in human hair and placenta or human milk (Chan et al., 2007)	No reports	Reliable to assess internal exposure
PCBs	PCBs (Barmpas et al., 2020; Malarvannan et al., 2013; Peng et al., 2020a; Wielgomas et al., 2012; Zheng et al., 2013)	No reports	No reports	PCBs in human hair and serum (Altshul et al., 2004)[a]	Large-scale surveys and retrospective cohort studies on PCBs (Peng et al., 2021; Tang et al., 2022; Wielgomas et al., 2012).	Reliable but probably influenced by measuring methods when assessing internal exposure
SCCPs	SCCPs (Han et al., 2021)	No reports	No reports	No reports	No reports	No evidence for the reliability of assessing internal exposure
BFRs	PBDEs, HBCDs, DBDPE, HBB, BTBPE PBBz, TBPH, PBT, TBB, and TBBPA (Barghi et al., 2018; Hajeb et al., 2022; Li et al., 2018; Liu et al., 2016; Qiao et al., 2016; Yuan et al., 2016; Zheng et al., 2011)	PBDEs in rats (Poon et al., 2014)	PBDEs, HBCD and TBBPA in in vitro hair (Barghi et al., 2018; Kucharska et al., 2015b)	PBDEs in human hair and serum (Liang et al., 2016; Liu et al., 2016; Poon et al., 2014; Qiao et al., 2018; Zhao et al., 2020; Zheng et al., 2014)[a]	Large-scale surveys, retrospective cohort studies and epidemiological investigations on PBDEs (Goodyer et al., 2017; Li et al., 2023; Peng et al., 2021; Tang et al., 2022).	Reliable for chemicals with higher K_{ow} and lower volatility values to assess internal exposure
OPFRs	TPhP, TBOEP, EHDPhP, TnBP, TiBP, V6, iDDPHP, BDP, RDP, DPhP, and BBOEP(He et al., 2018; Kucharska et al., 2015a; Kucharska et al., 2014; Qiao et al., 2019; Tang et al., 2022; Xu et al., 2019)	TDCIPP in mice (Zhu et al., 2020)	TDCIPP, TnBP, TCP, TPhP, TBEP, TCEP, TCPP and TEHP in in vitro hair (Kucharska et al., 2015b)	TDCIPP in human hair and BDCIPP in urine (Kucharska et al., 2015a)[a]	Large-scale surveys and Retrospective cohort studies on EHDPhP, TPhP, TBOEP, TCP, TEHP, TCEP, TCIPP, and TDCIPP (Tang et al., 2022) (Li et al., 2023)	Lack evidence for the reliability of assessing internal exposure
Pesticide	OCPs, OPPs, pyrethroids, carbamates, phenylpyrazoles and so on (Dahmardeh Behrooz et al., 2012; He et al., 2017; Lu et al., 2014; Ostrea Jr. et al., 2009; Ostrea Jr. et al., 2008; Peng et al., 2021; Yuan et al., 2017; Zhang et al., 2007b)	OCPs, OPPs, pyrethroids, carbamates, phenylpyrazoles and so on in rats (Appenzeller et al., 2017; Chata et al., 2016; Tutudaki and Tsatsakis, 2005)	No reports	ΣOPP metabolites, diazinon metabolites, and pentachlorophenol in human hair and urine (Fays et al., 2021; Hardy et al., 2021; Kokkinaki et al., 2014)[a]	Large-scale surveys and epidemiological investigations on pesticides (Anguiano-Vega et al., 2020; Beranger et al., 2020; Beranger et al., 2018; Kanavouras et al., 2011; Li et al., 2023; Michalakis et al., 2014; Peng et al., 2021)	Reliable to assess internal exposure and chronic exposure
PFAS	PFBS, PFHxS, PFDS, PFDoS, PFBA, PFHxA, PFHpA, PFDA, C8 Cl-PFESA, 6:2 FTUCA, HFPO-DA and HFPO-TA (Alves et al., 2015; Claessens et al., 2022; Feng et al., 2021; Kim and Oh, 2017; Li et al., 2013; Li et al., 2021a; Martin et al., 2016; Perez et al., 2012; Piva et al., 2021a; Piva et al., 2021b; Rodriguez-Gomez et al., 2017; Ruan et al., 2019; Wang et al., 2018a; Wang et al., 2018b)	PFOA, PFNA, and PFOS in rats (Gao et al., 2015)	No reports	PFOS, and PFOA in human hair and serum (Li et al., 2013)[a]	No reports	No consistent conclusion on the reliability of assessing internal exposure
PAEs	DEHP, MEHP, MEHHP, MEOHP, and AP metabolites (Chang et al., 2013; He et al., 2018; Yin et al., 2019)	DINP in rats (Hsu et al., 2015)	No reports	DEHP metabolites and DEP metabolites in human hair and urine (Fays et al., 2021)	No reports	Lack evidence for the reliability of assessing internal exposure
Bisphenol analogs	BPA(Martin et al., 2019; Nehring et al., 2017; Tzatzarakis et al., 2015)	No reports	No reports	BPF in human hair and urine (Li et al., 2021a)	No reports	Lack evidence for the reliability of assessing internal exposure
Alkylphenols	OP and NP (Nehring et al., 2017)	No reports	No reports	No reports	No reports	No evidence for the reliability of assessing internal exposure
PAHs	PAHs and OH-PAHs (Schummer et al., 2009; Toriba et al., 2003; Wang et al., 2020)	Chrysene, pyrene, benz [*a*]anthracene, naphthalene, and so on in rats (Grova et al., 2018; Grova et al., 2013)	No reports	Naphthalene metabolite in human hair and urine (Lin et al., 2020)[a]	Epidemiological investigations on acenaphthylene (Pang et al., 2020; Wang et al., 2016)	Reliable to assess internal exposure via metabolites

[a] There were correlation studies in which there were no significant correlations between hair and serum/urine.

(2009–2019) from a former e-waste area, suggesting that banned or restricted MOCs decreased significantly as a result of regulations. As the reliability of hair biomonitoring for MOCs becomes more accepted, more retrospective MOC studies should be completed using hair samples.

7.3. Epidemiological investigations on MOCs

Since hair, as part of the human body, can reflect systemic burdens of MOCs, hair samples can be used as valid biomarkers for epidemiological investigation. Meanwhile, hair sampling does not exclude special populations such as infants and other vulnerable individuals as blood sampling does. Thus, hair is an ideal tool to trace MOC exposure or screen major factors contributing to health risks over time and in special populations (Alves et al., 2014; Panneel et al., 2021; Zhao et al., 2021). In fact, analysis of trace elements in hair is often used in epidemiological investigations (Sazakli and Leotsinidis, 2017). So far, there are some epidemiological investigations on MOCs using human hair.

Kanavouras et al. (2011) report the case of a patient developing progressive motor neuron disease, with significant levels of DDTs and OPPs in hair as well as levels of organic solvents (n-hexane and toluene) in blood. In another case, Michalakis et al. (2014) measured several pesticides and their metabolites in hair (and blood) collected from children with hypospadias and their parents, discovering that hypospadiac boys and their parents were exposed to pesticides. Other cases employed hair as a unique matrix for epidemiological investigations. Goodyer et al. (2017) examined whether there was a relation between maternal hair PBDE concentrations and the risk of cryptorchidism in male infants, and found that every 10-fold increase in the concentration of maternal hair BDE-99 or BDE-100 more than doubled the risk of cryptorchidism. Wang et al. (2016) recruited 405 women for a cross-sectional study (including 170 with hypertension) and found that acenaphthylene in hair was associated with an increased risk of hypertension after adjustment for multiple testing and potential confounders. Beranger et al. (2020) explored the relationship between maternal hair concentrations of 64 pesticides and their metabolites and newborn birth measurements using the ELFE French nationwide birth cohort, and found statistically significant associations between maternal hair concentrations of seven pesticides/metabolites and birth measurements. A study determining OCP concentrations in hair samples and total nuclear abnormalities in buccal cells from children at two elementary schools found that the risk of >0.2 % of micronuclei increased 7.97 times once OCP concentrations exceeded 0.447 μg/g, suggesting that hair may be a useful biomarker of genotoxic damage in children exposed to highly-toxic compounds (Anguiano-Vega et al., 2020). A case study explored the associations of PAHs and metal(loid)s with influenza-like illness risk among housewives in Shanxi, China, and of the nine PAHs detected, only acenaphthylene concentration was significantly associated with illness risk (Pang et al., 2020). In this case, hair was employed as a passive sampler integrating multiple exposures including fuel combustion, passive smoking, vehicle exhaust, and other pollution sources, highlighting the advantage of hair sampling in epidemiological investigation. As the technology of multi-class analysis improves and concerns over the health effects of MOCs grow, more extensive epidemiological investigations on MOCs in hair should be conducted.

8. Conclusions and future perspectives

This review summarizes numerous reports on MOCs in hair and indicates that hair is a reliable tool for MOC exposure assessment in most cases when standardized protocols are followed. Conclusions on the reliability of human hair biomonitoring for different MOC groups are summarized in Table 3. For dioxins, PCBs, OCPs, OPPs, and PAHs, some in vivo/in vitro experiments or correlation studies indicate that hair analysis is reliable to assess internal exposure to them; For BFRs, it depends on their higher Kow and lower volatility values; As for PFAS, there is no consistent conclusion on the reliability of assessing internal exposure; For others, there is no sufficient evidence for the reliability of assessing internal exposure. In sum, for POPs with higher K_{ow} and lower volatility values, hair biomonitoring can reliably assess internal exposure; for MOCs that are easy to metabolize, hair biomonitoring can reliably assess internal exposure using specific metabolites; and for MOCs that are capable of binding to protein, hair analysis should be used with caution. Nevertheless, it must be admitted that MOCs, especially emerging MOCs, include too many varieties to take an exhaustive list. Also, different chemicals in the same group are different from each other, and therefore their assessment of hair biomonitoring may also be different. Extensive and intensive research for more MOCs not mentioned in this article is warranted.

In addition, we attempted to describe the incorporation of MOCs into hair from endogenous and exogenous exposures by referencing studies on metals and drugs. Considering their discrepancy in exposure situations, however, MOCs must be independently investigated and compared to our fundamental of drug/metal exposure. While human hair is being employed in more epidemiological investigations, it is mainly considered a biomarker of metallic elements or an auxiliary matrix. As thus, its advantages have not yet been fully exploited. To do so, fundamental work must first be conducted, such as the verification of correlations between blood/urine and hair, the standardization of measurement methods, and the establishment of reference (or background) ranges.

In the future, some technologies may help solve these related problems. For instance, secondary ion mass spectrometry (SIMS) and MALDI-MS have the capacity to perform in situ micro-analysis, and have been applied to investigate the adsorption, penetration, and distribution of metal ions and drugs in hair (Audinot et al., 2004; Beasley et al., 2016; Cuypers et al., 2016; Erne et al., 2019; Poetzsch et al., 2015; Smart et al., 2009). Thus, in situ analysis using SIMS and MALDI-MS holds promise to explore mechanisms of MOC incorporation into the hair matrix. Secondly, with respect to PFAS, some protein-binding studies have employed X-ray crystallography, equilibrium dialysis, site-specific fluorescence, and other techniques to show that some PFAS homologs tend to bind serum albumin (Chen and Guo, 2009). However, the interactions between PFAS and keratin in hair have not yet been investigated. Finally, since the list of MOCs is long and constantly increasing, nontargeted identification of MOCs in human hair is necessary to provide complete information on human exposure to MOCs and may prove important in the further regulation of MOC production. In conclusion, more cutting-edge technology should be applied to MOC hair analysis to fully exploit the advantages of this crucial analytical matrix.

CRediT authorship contribution statement

Shiyi Zhang: Conceptualization, Methodology, Writing – original draft, Writing – review & editing. Xiao Yan: Supervision, Validation, Writing – review & editing. Bin Tang: Validation, Writing – review & editing. Weikeng Luo: Visualization, Writing – review & editing. Shejun Chen: Writing – review & editing, Grammar and logical check. Xiaojun Luo: Writing – review & editing, Structuring. Jing Zheng: Funding acquisition, Resources, Supervision, Writing – review & editing. Bixian Mai: Supervision, Writing – review & editing. Yunjiang Yu: Resources, Supervision, Writing – review & editing.

Data availability

Data will be made available on request.

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.

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Appendix A. Supplementary data

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