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Review

Biomonitoring Atmospheric Pollution of Polycyclic Aromatic Hydrocarbons Using Mosses

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Abstract: Polycyclic aromatic hydrocarbons (PAHs), as the main components of petroleum hydrocarbons (PHCs), are carcinogenic organic pollutants that occur widely in the atmospheric environment with increasing concentration. Moreover, PAHs are widespread all over the world due to their high volatility and long-range transport potential. The monitoring of atmospheric PAHs is often limited by working conditions, especially around oil field operation areas and other industrial areas. Mosses (*Bryophyta*), the most sensitive atmosphere pollution indicators, can be easily collected and have been used to monitor atmospheric pollutants including PAHs. Thus, characteristics and influencing factors of mosses' absorption and accumulation of PAHs in the atmosphere were discussed, and the application of mosses in the biomonitoring of atmospheric PAHs were summarized. Biomonitoring mosses include *Dicranum scoparium*, *Hypnum cupressiforme*, *Thamnobryum alopecurum*, *Thuidium tamariscinum*, *Hylocomium splendens*, *Pleurozium scheberi*, etc. Currently, the main methods for monitoring PAHs by mosses are biomonitoring with the chemical analysis method, the index of atmospheric purity (IAP) method, the ecological survey method, and the Moss-bag technique. Biomonitoring of atmospheric PAHs using mosses has a relatively wide range of prospects.

Keywords: mosses; polycyclic aromatic hydrocarbons; atmospheric pollution; biomonitoring; pollution ecology

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1. Introduction

With the development of industry and the rapid growth of coal and petroleum fuels, the quality of the atmospheric environment is deteriorating daily. Atmosphere pollution has become one of the main harmful factors affecting the world environment and human health [1]. Petroleum hydrocarbons (PHCs) refer to a large group of compounds derived from crude oil. They are mainly composed of hydrogen and carbon, and they are important air pollutants [2]. Polycyclic aromatic hydrocarbons (PAHs), as the main components of petroleum hydrocarbons (PHCs), are carcinogenic organic pollutants that occur widely in the atmospheric environment with increasing concentration. Moreover, PAHs are widespread all over the world due to their high volatility and long-range transport potential [3]. Irrigation with sewage, discharge of oily wastewater, and volatilization of various petroleum products will also induce PAHs pollutions. PAHs pollution has attracted much attention because of its serious environmental harm, which poses a direct or potential threat to human health and the environment, and also changes the metabolic activities of microorganisms [4,5]. Atmospheric PAHs do great harm to the human body. Inhaling air is one of the most important ways for humans to come into contact with pollutants. Atmospheric pollutants will have different effects on human health. Mosses and lichens have been used to estimate human exposure to pollutants by inhalation [6,7]. Figueira [8] assessed the risk of arsenic exposure by using air deposition and water pollution in mosses. Augusto [9] reported that PCDD/F in lichens can be used as a spatial estimate of the potential risk of inhalation by the population living in the southern part of Portugal. Augusto [10] also assessed human exposure to (PAHs) through lichen data and calculated health risks.

Under the above circumstances, the high spatial resolution of biological monitoring data is essential for reliable environmental health research. Biomonitoring plays a vital role in assessing the risk of people exposed to the inhalation of air pollutants. However, most of the existing relevant literature research content focuses on monitoring and mechanism research, and there are few reports on the use of mosses to evaluate human exposure to PHCs in the atmosphere. This may become a research trend in the future.

The structure of PHCs is composed of approximately 57% aliphatic hydrocarbons, approximately 29% aromatic hydrocarbons, and approximately 14% asphaltenes with different polar compounds. In addition, aliphatic hydrocarbons can exist in linear or branched hydrocarbons with unsaturated or saturated forms [11]. They are listed as one of the key monitored chemical pollutants by the United Nations Environment Programme (UNEP) [12]. PHCs mainly refer to a compound with two or more benzene rings connected in fused or non-fused rings (Figure 1), divided into short-chain hydrocarbon, diesel oil, kerosene, and gasoline [13,14]. PAHs are one of the main toxic components of crude oil [15,16]. They have bioaccumulation effects in many biological chains in nature, and their content in nature is quite astounding; hence, they are considered the main organic pollutants that are harmful to human health [17]. Table 1 compares the concentrations of PAHs in different regions. Low molecular weight PAHs, i.e., NAP, ACE, ACY, FLU, and PHE, were significantly higher in the Delhi region than in Sao Paulo and Houston. ACY and ANT are highest in Delhi (16.1 ng/m^3), unlike Hong Kong, where PYR concentration is highest (6.35 ng/m^3). In previous studies, PAHs concentrations in the Houston area were the lowest, except for NAP.

Table 1. Comparison of polycyclic aromatic hydrocarbon concentrations in different regions (ng/m^3).

PAH	Delhi, India [18]	Hong Kong, China [19]	Sau Paulo, Brazil [20]	Houston, US [21]	Lompoc, US [22]
NAP	13.1	ND	ND	ND	15.2
ACY	16.1	ND	0.09	0.04	ND
ACE	12.7	ND	0.35	0.01	2.56
FLU	17.1	0.25	ND	0.02	8.39
PHE	9.2	0.55	0.18	0.09	1.46
ANT	11.2	0.01	ND	0.02	2.05
FLT	6.0	5.47	0.68	0.07	4.51
PYR	4.1	6.35	0.52	0.08	5.51
BbF	3.4	6.64	1.23	0.05	11.7
BkF	1.2	0.19	0.76	0.14	5.8
BaP	1.9	2.06	0.52	0.04	9.01

ND—non-detectable; NAP—naphthalene; ACY—acenaphthylene; ACE—acenaphthene; FLU—fluorene; PHE—phenanthrene; ANT—anthracen; FLT—Fluoranthene; PYR—pyrene; BbF—benzo[b]fluoranthene; BkF—benzo[k]fluoranthene; BaP—benzo[a]pyrene.

Currently, many environmental monitoring units and agencies are conducting biomonitoring programs for hydrocarbons in the atmosphere. Petroleum hydrocarbon pollutants are diluted when released into the atmosphere, resulting in low concentrations and wide ranges that are difficult to monitor. Traditional atmospheric pollution monitoring methods are optical monitoring, filter-based sampling, and direct measurement. Monitoring also requires high-capacity or low-capacity samplers, power access, specific filters, a lot of manpower, and extremely high procurement costs. Biomonitoring can perform high-density sampling at a low cost on most required spatial and temporal scales, and it allows the measurement of a wide range of petroleum hydrocarbon pollutants [23–27]. Chemical monitoring is a random sampling of highly dynamic change systems, and only the instantaneous change of various environmental factors in the space is measured, which cannot indicate the actual situation of the life systems existing in this space for a long time [28]. The biological monitor can be used as a passive collector of PHCs in the atmosphere. It uses the changes in the life system to indicate environmental quality. All levels of the life system keep continuous records of the environmental quality, making the monitoring results reflect the historical status of the cumulative results of a particular area after pollution or ecological damage. It can be a long-term integrator of petroleum hydrocarbon pollutants [29–31]. This is also a major advantage of moss monitoring. Mosses are often used for passive biological monitoring, i.e., using native species in the study area. They are also sometimes used for active biological monitoring. When there is no native moss in the study area, grafts can be used for monitoring [32]. This is also a major advantage of moss monitoring.

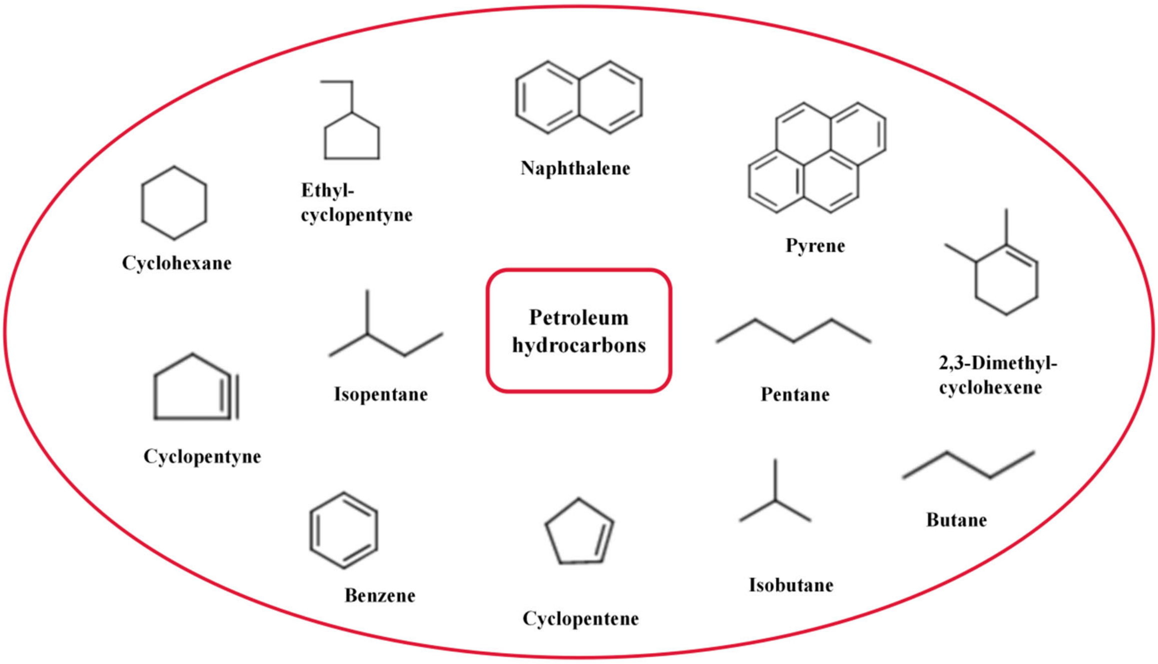

Figure 1. Main toxic PHCs in soil (created by author).

Mosses have a long growth cycle, low differentiation degree, and vigorous cell growth. The growth points at the tips of stems and branches usually stimulate the continuous or periodic division of cell groups in the lower part of mosses after dormancy or death; consequently, they are evergreen all year round to provide representative, applicable, and seasonal indications and monitoring [33–35]. Natural moss samples can integrate information on the long-term accumulation of pollutants due to their slow growth process. In addition, mosses' unique physiological adaptation mechanism enables them to survive in extreme environments where other terrestrial plants have difficulty surviving, such as high temperatures, high cold, drought, and dim light. They are widely distributed and diverse, and suitable for predicting atmospheric pollution in various geographical units. It can be recommended as an ideal material for global atmosphere pollution monitoring and verification [36]. In recent years, mosses have been increasingly used to monitor petroleum hydrocarbon pollutants. This review aims to critically discuss the application of mosses in monitoring PHCs pollutants.

2. Physiological Structure and Biological Characteristics of Mosses

Mosses are an autotrophic higher plant group that transits from aquatic to terrestrial. The primary sources of nutrients include rainwater, dew, and sediments produced by impact with atmospheric dust [37,38]. Except for a few genera and species, the leaves are primarily single-layer cells with no waxy cuticle coating on the body surface. They are straightforward when reacting to pollution factors in the atmospheric environment and reflect the type and approximate concentration of pollutants through specific diseases [39]. Mosses can have biological effects on environmental pollution factors to achieve the purpose of atmosphere pollution monitoring. Mosses grow in polluted environments for a long time, and their physiological and plant functional characteristics show obvious damage symptoms. For example, the chlorophyll in mosses can be destroyed, leading to the rupture of leaf cells and sometimes even plants' severe decline or disappearance [40]. In addition, the change in air pollutant components results in a series of changes in biological characteristics or index parameters, such as mortality, leaf cell size, number, etc. (Table 2) [33,41,42]. Moss monitoring of atmospheric pollution is based on biological performance, monitoring various interference effects through different mosses' injury symptoms, as well as pollutant content in plants and moss species, and then making a corresponding judgment on atmospheric pollution.

Figure 2 shows the morphology and life cycle of mosses. Mosses usually have low individuals and simple anatomical structures. They are composed of only a single layer or a few layers of cells. No real roots (only supporting pseudo roots) and vascular bundles exist. Because it has extraordinary physiological phenomena, such as the proximal end of the plant rots (that is, the part of the plant close to the ground rots), and the tissue does not contact the ground at all, it does not absorb nutrients from the soil or matrix. However, it can directly exchange air, water, and nutrients on the body's surface. Mosses take up nutrients exclusively via atmospheric deposition (essentially precipitation and sedimentation). With correspondingly low vegetation cover of the soil, the soil matrix with its nutrients can also reach mosses through splash erosion during precipitation. If mosses grow under trees and bushes, nutrients washed out of them can be taken up by mosses. Especially, those species that are epiphytic on the trunk are almost entirely free from the interference of their substrate. The exposed surface area of mosses in the atmosphere is about 1.6 (m^2/g) [43]. A waxy cuticle does not cover the plant surface, and its dorsal and ventral surfaces can directly accept pollutants in the atmosphere. Therefore, they are susceptible to atmosphere pollution and can quickly reflect the pollutants concentrated in rainwater and dew through their specific symptoms. Moss monitoring of atmospheric pollution is based on biology. Different reactions, such as mosses' injury symptoms and injury types, can help make corresponding judgments on the status of atmospheric pollution. In terrestrial ecosystems, the sedimentation flux of PAHs is bidirectional, and there is a continuous exchange between the soil, air, and organisms (Figure 3). The absorption and release of PAHs by terrestrial ecosystems vary greatly in space and time, are greatly affected by climate and environmental conditions, and also depend on the metabolic activities of the biological surface and plants involved [44]. Mosses have a large proportion of leaf surface, many cation exchange sites, strong resistance and adsorption to environmental pollutants, and their sensitivity characteristics are far superior to most higher plants. It is easy to obtain materials, and the investigation and detection methods are simple, which can objectively reveal the migration law and Spatio-temporal change characteristics of air pollutants.

Table 2. Performance of different mosses in atmospheric pollutants' monitoring.

Mosses Species	Pollutant Type	Symptom
Bryum argenteum, *Ceretodon Purpud*, *Glyphomitrium humillimu*, *Grimmia pulvinata*, *Hypnum cupressiforme*, *H. yokohamae var. kusatsuense*, *Tortula muralis*	Sulfur compounds	The cytoplasmic wall was separated and the tissue cells were necrotic. Chlorophyll decomposes downward from the leaf tip until it disappears [33].
Aulacomium androgynum, *Pohlianutans*, *Orthotrichaceae*	Oxides of carbon, hydrocarbon, carbohydroxides, fluorides, and ozone	Cell wall separation occurs, respiration rate decreases, carotene increases, chloroplasts are slightly damaged, leaf tips turn brown or dark brown, and plants show green deficiency spots [39].
Dicranum scopariunm, *Entodon compressus*, *Hyloconium splendens*, *Isothecium stoloniferum*, *Philonotis fontana*, *Pleurozium shreberi*, *Polytrichum commume*, *P. juniperinum*, *P. acutum*, *Sphagnum rigensohnii*, *Taxiphyllum taxirameum*	Particulate matter	Abnormal nuclear division occurs, spores cannot usually germinate or grow into short or even deformed protonema, chloroplasts in cells decrease, and plants grow abnormally [41].

In addition, mosses also have some unique biological characteristics, making them suitable for monitoring and researching environmental pollution. First of all, due to its low degree of differentiation, the growth potential of the plant cells is relatively strong. Therefore, when the growth point at the tip of the stem and branch is dormant or dead, the meristem development at the lower part of the stem and leaf is stimulated to promote the rapid growth of new branches and keep the plant evergreen all year round [45]. Then, there is no waxy cuticle on the surface of the mosses, and their dorsal and ventral surfaces can directly accept pollutants in the air. Therefore, they are particularly sensitive to atmospheric pollution and can quickly reflect the pollutants concentrated in rainwater and dew through their specific diseases, thereby providing a representative and applicable year-round indication and monitoring. In addition, many mosses are perennial plants that can be used as long-term cumulative species of atmosphere pollution [46]. Long-term biomonitoring for a particular area or a specific pollution source increases the stability and reliability of the determination results.

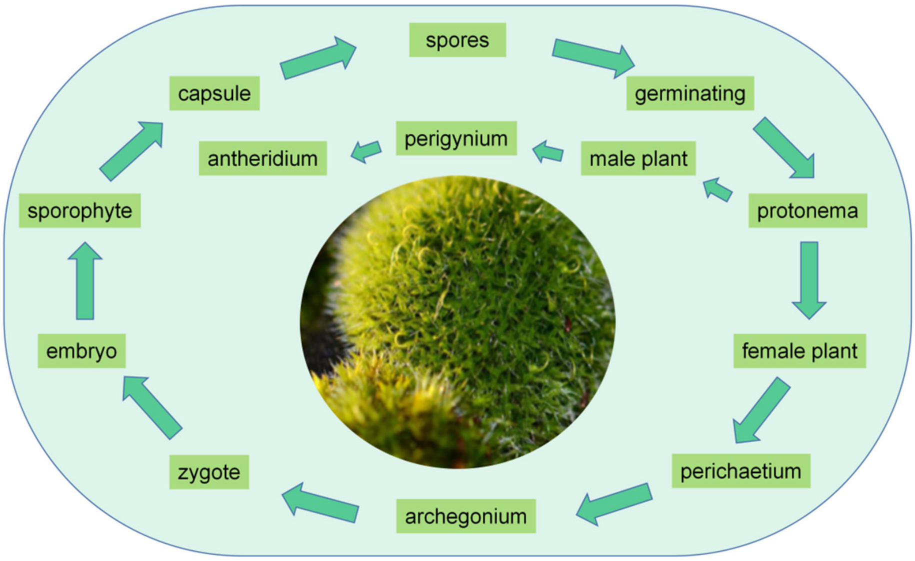

Figure 2. Moss morphology and life cycle (created by author).

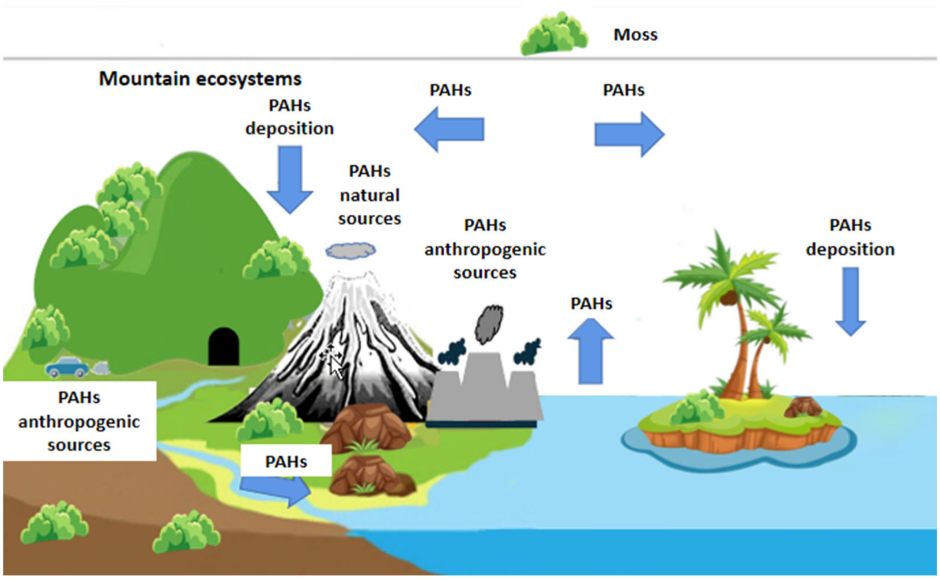

Figure 3. Moss biomonitoring system (created by author).

3. Influencing Factors of PHCs Biomonitoring by Mosses

Atmospheric PHCs pollutions result from the long-term accumulation and synergy of various pollutants, which existed in persistence and complexity. Because mosses are sensitive to environmental changes, their monitoring function for atmospheric PHCs may be restricted by many factors. In the process of monitoring, it is not only necessary to understand the influence of pollutants on the content of relevant matter in mosses, but also to understand the surface structural characteristics of mosses, the condition of the rhizoid attachment matrix, and even the length of stem tip and whether there are appendages on the plant surface, which will affect the ability of mosses to capture pollutants in the atmosphere. In this paper, referring to the existing cases, the main influencing factors are summarized in three aspects: moss species, growth substrate, and habitat status.

3.1. Moss Species

Different mosses have different tolerance, absorption capacity, and sensitivity to pollutants. The sensitivity increases gradually from mat-like, layered, intertwined growth phylloid moss to epiphytic species. For example, *Orthotrichaceae*, whose trunk is epiphytic, is usually more sensitive to atmospheric pollution than *Polytrichaceae* and *Hylocomiaceae* [47], which are influenced by the mosses' morphology, leaf area, leaf number, and stomatal openness; the larger the surface area of mosses, the stronger the ability to capture particulate pollutants. Capozzi [48] collected PAHs content of mosses in spring (213 ng/g), which was significantly higher than that of mosses collected in autumn (177 ng/g); thus, collected PAHs content may depend on different seasons. The higher PAHs content in mosses collected in spring may reflect PAHs accumulated in the previous winter, while the lower PAHs content in mosses collected in autumn may be affected by PAHs degradation and loss in the previous summer. Most PAHs in the air enter vascular plants through pores or cuticle wax. Due to the large surface area, the leaf interface is considered to be the main channel for the accumulation of organic chemicals [49,50]. Bryophyte has strong drought tolerance, and its sensitivity to environmental factors is ten times that of seed plants [51]. In addition, the plant leaves with fluff have high surface roughness and a large contact area with the atmosphere, which can intercept pollutants. The dense fluff can also effectively prolong the retention time of particles on the leaf surface [52].

3.2. The Growth Substrate and Habitat of Mosses

The growth substrate fixed by mosses' rhizoids will affect the physiological characteristics of mosses and their tolerance and absorption capacity for pollutants. The sensitivity of mosses in different growth states to atmospheric PHCs pollutions are different, and the performance trends from weak to strong are epiphytic mosses in stone, epiphytic mosses in soil, and epiphytic mosses in trees [53]. Thus, the same mosses growing on different substrates may also have different tolerance and absorption capacity for atmospheric PAHs pollutants. Therefore, to reduce and avoid the difficulties of monitoring data analysis caused by the interference of environmental factors, epiphytic mosses are most suitable for evaluating the status of atmospheric environmental quality.

The resistance and absorption capacity of mosses to petroleum hydrocarbon pollutants are affected by their conditions and habitat conditions, such as canopy coverage, meteorological conditions, vegetation types, pollutant concentration, and contact time. [54,55]. Most mosses are shade loving, and the saturation light intensity required for photosynthesis is far lower than that of higher plants. Therefore, the light intensity and light quality changes in the atmosphere will have a certain impact on the growth of mosses. Too strong light will lead to the weakening of the absorption capacity of atmospheric pollutants. In addition, the absorption capacity of mosses for PAHs is influenced by exposure time. In Bertiz nature reserve, Foan [56] counted the seasonal variation of monthly atmospheric deposition of PAHs and its total content in mosses, and found that there was a similar trend ($r = 0.464$, $p < 0.01$). The mosses in warm and humid areas have lower pollution tolerance than those in cold and dry areas. The results show that the absorption and accumulation capacity of mosses for pollutants is directly proportional to the concentration and exposure time of atmospheric pollutants within their tolerance range [57].

3.3. Limitations of Moss Biomonitoring

Environmental monitoring with biota can be used to evaluate environmental change in time and space or to track point sources after some specific event. According to Bignert [58], continuous and long-term annual pollutant monitoring studies based on biota samples can provide a very useful aid in describing environmental processes, as long as interannual variability is taken into account. Mosses or lichens can be used to monitor PAHs in the air [59–63]. Although it may be difficult to calculate absolute air concentrations based on levels in vegetation [64], mosses or lichens can be used to map relative differences in air concentrations and deposition between different areas, although a better understanding of the uptake mechanisms is needed to interpret the data [64–66]. Although it is not easy to properly perform such environmental monitoring with biota, the available references on biota monitoring in local space and time make it possible. Previous studies showed that mosses are somewhat limiting if the intention is to monitor environmental changes over a wide range of time and space. However, that mosses (a single species) can be used for local monitoring of individual events cannot be questioned—if all samples are taken at the same time and under roughly similar temperature and humidity [67].

4. Application of Mosses in Biomonitoring Atmospheric PAHs

4.1. Biomonitoring with Chemical Analysis Methods

Much of current research is based on biomonitoring, supplemented by chemical detection, which is a widely used integrated monitoring method. Sweden first used moss materials on the ground for chemical analysis and environmental monitoring in 1969 [68]. The first European mosses survey was conducted in 1990 and has been repeated every five years since. The samples were collected according to the standardization protocol, and the atmospheric pollutants in mosses were analyzed by chemical methods [69]. Colantuono [70] used gas chromatography to analyze PHCs concentrations in moss samples collected one month after a fire near the Antarctic station in Brazil, and PAHs concentrations in the moss ranged from 131–1235 (ng g^{-1} dw). Cabrerizo [71] conducted two Antarctic expeditions (2005 and 2009) and analyzed hydrocarbon levels in remote areas and near current and abandoned Antarctic Research settlements to assess potential sources of pollutants. The PAHs in the samples were dominated by low molecular weight (LMW) compounds. The most abundant compounds were PHE, FLU, PYR, and CRY. ΣPAHs were found in the moss concentration range of 4.4–34 (ng/g dw). Concha-Graña [72] determined 19 PHCs in mosses using a matrix solid-phase dispersion method to monitor atmospheric pollution. Aboal [73], Carballeira [74], Jonathan [75], Gerdol [76], and Grodzinska [77] also used this method to conduct much research. The main technical links include sampling, sample processing, and chemical analysis. Table 3 shows the different methods used for the analysis of PHCs in mosses. Soxhlet extraction is commonly used to extract PHCs from mosses, and gas chromatography coupled with mass spectrometry is the most common technique used to determine PHCs.

Table 3. The different methods used for the analysis of PHCs in mosses. NM: not mentioned. ASE: accelerated solvent extraction. MAE: microwave-assisted extraction.

Compounds	Sample	Extraction	Determination	Sensitivity
13 PAHs [78]	*Dicranum scopariumHypnum cupressiformeThamnobryum alopecurumThuidium tamariscinum*	Soxtec (moss + sodium sulphate + Florisil)	HPLC-FLD	LOQ 3–52 pg instrumental
16 PAHs [79]	*Hylocomium splendensPleurozium scheberi*	ASE, DCM	GC–MS	LOQ 1–5 ng g^{-1}

Table 3. *Cont.*

Compounds	Sample	Extraction	Determination	Sensitivity
19 PAHs [80]	*Pseudoscleropodium purum*	MAE, 20 mL H: A (90:10)	GC–MS/MS	MQL: 0.1–1.7 ng g^{-1}
16 PAHs [81]	*Hypnum cupressiforme*	Sonication 5 g + 100 mL H:A (1:1), twice	HPLC	NM
11 PAHs [82]	*Fontinalis antipyretica*	Soxhlet 200 mL DCM 16 h	HPLC-FLD	NM
15 PAHs [32]	*Pleurozium scheberi*	Soxhlet 200 mL DCM 16 h	GC–MS	NM
18 PAHs [83]	*Tortula muralis*	Sonication 5 g 30 min, 100 mL H	GC–MS	NM
16 PAHs [84]	*Hypnum plumaeformae*	ASE 5 g, 1500 psi, 100 °C, 2 cycles, 5 min DCM:A (1:1)	GC–MS	MDL:3.3–7.8 ng g^{-1}
16 PAHs [85]	*Hypnum cupressiforme*	Soxhlet, 8 h DCM. Sulphuric clean up	GC–MS	0.3–1 ng g^{-1}
16 PAHs [86]	*Leptodon smithii*	Sonication, 3 g, 3 × 100 mL DCM:A (1:1)	GC–MS	LOD 1–3 ng mL^{-1}

Currently, research mainly focuses on the PAHs in PHCs and less on other components. Ares [86] proposed a simple and economical method to identify small-scale pollution sources of PAHs. This method is used to test whether there is a decreasing gradient of PAHs concentration in mosses related to the distance between different emission sources. The concentration of PAHs was measured at 35 atmospheres of sampling points. Each atmosphere of sampling points was 1 km away. Ötvös [81] explored the source of PAHs in Hungarian moss samples and compared the PAHs concentration measured in moss samples with other forms of vegetation in the world in order to determine the accumulation mechanism of PAHs on mosses according to the octanol atmosphere partition coefficient of various PAHs. In the initial stage, mosses began to be used to monitor the deposition of atmospheric pollutants. Most of the research objects mainly focused on organic pollutants, such as organochlorine pesticides, pesticides, and polychlorinated biphenyls, and then gradually involved studying PAHs in PHCs. Currently, some studies have been carried out around the world using mosses as passive sampling materials of natural plants to monitor the pollution level of PAHs in the region and indicate the spatio-temporal variation of PAHs. Gerdol [83] collected 19 samples of mosses from industrial areas, residential areas, urban centers, and suburbs of Italy to analyze the content of 16 kinds of PAHs, indicating that the content of PAHs in urban areas was significantly higher than that in rural areas. In addition, the concentration of phenanthrene, fluoranthene, and pyrene was the most abundant and the highest in industrial areas, while the concentration of low molecular weight PAHs was the highest in rural areas. Galuszka [79] analyzed 16 PAHs and 33 inorganic elements in *Hylocomium splendens* and *Pleurozium schreberi* at 10 sampling sites of 2 forest types (dry pine forest and continental coniferous forest) in the mountains of southern Poland. The analysis showed that PAHs with different ring numbers were distributed differently in mosses. Four-ring hydrocarbons accounted for the largest proportion of all aromatic groups in the moss species examined. Three- and five-ring hydrocarbons show similar second-highest concentrations and were significantly superior to the six-ring equivalents.

4.2. Index of Atmospheric Purity (IAP)

As a classical ecological method, the atmosphere cleanliness index method has positive significance in monitoring atmosphere pollution and has been widely used worldwide [87–89].

Based on the sensitivity of mosses to atmosphere pollution, a method for calculating atmosphere purity is designed. The formula is:

$$\mathrm{IAP} = \sum_{i=1}^{n} (Q \times f)/10$$

i: Natural number from 1 to N, indicating the ith mosses in a particular area;
n: The number of species of all epiphytic mosses in a specific area;
Q: Ecological index of epiphytic mosses in the test area; that is, the average number of all other epiphytic moss species coexisting with epiphytic mosses in all sample areas.
F: Subjective estimates of the coverage and frequency of each epiphytic mosses in the same area. Generally, five levels are adopted, and the classification method is shown in Table 4.

Table 4. Subjective estimation of epiphytic coverage and occurrence frequency.

	Coverage	Frequency of Occurrence
1	Rare species with low coverage	0~20%
2	Uncommon or low coverage species	21~40%
3	Uncommon or moderately covered species on some trees	41~60%
4	Species with high coverage on some trees	61~80%
5	Common species with high coverage on all trees	81~100%

IAP values of various areas are calculated according to the survey results. According to the IAP value, the degree of atmosphere pollution can be analyzed [90]. According to this, the pollution is classified into serious pollution area, pollution area, and pure area, and the distribution map of atmosphere pollution is drawn [91]. However, the atmosphere cleanliness index cannot directly indicate the types of pollution sources and pollutant concentrations.

4.3. Ecological Survey Method

The ecological survey method is the most common and commonly used method to determine a large area's atmospheric pollution [92]. Ecological investigation of epiphytic mosses includes species, coverage, frequency, growth status, growth status of attached trees, and the location of observation points. Based on the data obtained, each epiphytic moss was given a coverage series, and the average percentage was calculated. By integrating the data from all sample areas and analyzing the levels of pollutants in the mosses, the air pollution status of an area can be derived [93]. Since 1990, according to a standardized program, moss samples have been collected and monitored every five years in Europe. Pollutants analyzed include metals (since 1990), nitrogen (since 2005), persistent organic pollutants (since 2010), and micro plastics (since 2020). It is believed that mosses provide an effective and inexpensive method for high-resolution monitoring of pollution trends in Europe [59]. Regarding sampling sites, the goal of each European country is to collect at least 1.5 moss samples per 1000 km^2. The samples shall be uniformly and objectively distributed as much as possible. The sampling point shall be in the open grassland or peat land, and sampling under the shrub canopy or large leaf herbaceous plants shall be avoided, as well as the area with flowing water on the hillside, so as to avoid being affected by the dripping water from the canopy [94]. Foan [95] set 61 sites in France, Spain, and Switzerland as sampling sites. The content of PAHs in mosses was determined by pressurized liquid extraction (PLE) and solid phase extraction (SPE) purification combined with high performance liquid chromatography fluorescence detection (HPLC-FLD) analysis. Moss monitoring maps were drawn using ArcMap 10.7.1 (ESRI, West Redlands, CA, USA), a part of the integrated geographic information system (GIS), based on a 50×50 km^2 grid, showing the average pollutant concentration of each unit [96]. Bekteshi selected sampling sites and collected moss samples nationwide and determined different elements by chemical methods. The degree of pollution was assessed by calculating enrichment factors while determining the most likely local anthropogenic emission sources and evaluating the relative contribution of anthropogenic and natural element deposition sources from the atmosphere to the terrestrial system [97].

4.4. Moss-Bag Technique

The moss-bag method uses mosses as materials to monitor and analyze the pollution status by using their high absorption and accumulation instinct of no selection and rejection of metal pollutants in the atmosphere. The moss-bag method is convenient and practical for monitoring when no moss is distributed in a polluted area or densely populated cities or factories. Aničić Urošević used moss-bags to confirm the hypothesis that the botanical garden can be a background area of urban atmosphere pollution [98]. Capozzi studied the possibility of using moss-bags to detect pollution input. Twenty kinds of PHCs were analyzed in some agricultural sites; 4- and 5-ring PAHs were the most abundant, especially pyrene, fluoranthene, and pyrene. At the same time, moss-bag has proved to be a very sensitive tool, which can distinguish different land use scenarios and the proximity to roads in mixed urban and rural landscapes. A large number of air pollutants (metals and PHCs) were analyzed in exposed mosses from 40 sites in 5 cities. Total PAHs concentrations in the mosses before exposure were 59 ng g^{-1} d.w., and total PAHs concentrations after exposure ranged from 72 to 95 ng g^{-1}. PAHs were grouped according to ring number, with 2-, 3-, and 4-ring PAHs (Figure 4) accounting for 24, 32, and 40% of the total content, respectively, before exposure. After exposure, the mean values of PAHs accumulated at all sites (i.e., after exposure minus pre-exposure values) were still generally dominated by 4-ring PAHs (51% of total PAHs), followed by 5-ring PAHs (24% of total PAHs). Four- and five-ringed PAHs were the most abundant PAHs in the atmosphere, including chrysene, fluoranthene, and pyrene. The results showed that the pollutants investigated had similar spatial distribution patterns throughout the study area, with the highest levels of 4-ring PAHs mainly associated with road traffic, while agricultural practices were the main sources of diffuse pollution [99]. The traditional moss-bag method was improved by multi-mosses species detection technology, and a variety of mosses were used alternately for large-scale PHCs pollutions monitoring [32]. Wegener [100] collected Sphagnum to make spherical moss-bags, which were placed in the open atmosphere 1 km from an aluminum electrode manufacturing plant in Rotterdam, the Netherlands. The concentrations of six PAHs in moss-bags were analyzed and compared with those in moss-bags in relatively clean areas. The results show that mosses can be used not only to monitor heavy metals, but also to monitor PAHs in the atmosphere. The "moss-bag technique" is a common type of moss biomonitoring in the literature. The content of PAHs pollutants in the atmosphere is very low. If only moss-bag technology is used, the results may be inaccurate. Therefore, monitoring using this technique may require higher sensitivity. In the future, moss monitoring and physicochemical monitoring methods can be combined to improve the sensitivity and accuracy of the results.

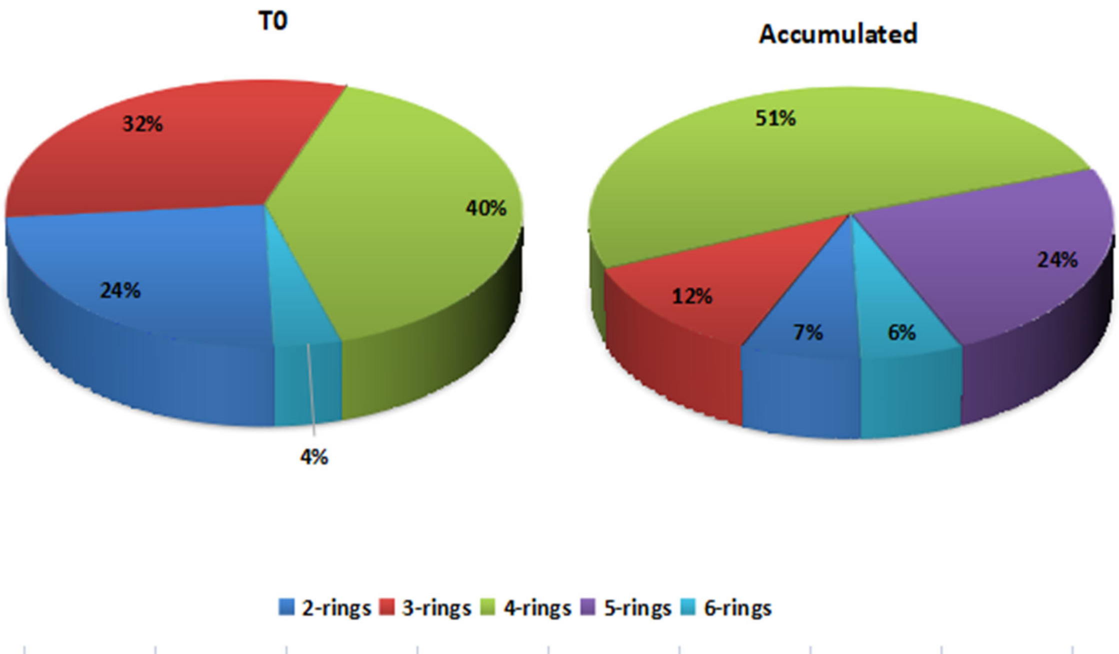

Figure 4. Grouped by ring number, as a percentage of total PAHs, in pre-exposure moss (T0) and accumulated in exposed moss pockets (created by author).

5. Conclusions and Perspectives

Monitoring atmospheric PHCs pollutions by mosses has broad research prospects. During oil exploitation, processing, and transportation, use of PHCs will inevitably release them into the environment and cause PHCs pollutions. PHCs pollution has attracted much attention because of its serious environmental harm, which poses a direct or potential threat to human health and the environment, and changes the metabolic activities of microorganisms. Traditional atmospheric pollution monitoring methods are optical monitoring, filter-based sampling, and direct measurement. They also require high-capacity or low-capacity samplers, power access, specific filters, a lot of manpower, and extremely high procurement costs. Biomonitoring can perform high-density sampling at a low cost on required spatial and temporal scales, and it allows the of a wide range of PHCs pollutants. Currently, existing research mainly focuses on quantitative analysis of pollutant levels in moss samples to indicate the local and regional environmental quality or to reflect the spatial and temporal distribution of pollutants. However, studies on the enrichment mechanism and physiological characteristics of PAHs (as the main components of PHCs) in mosses are not deep enough. Therefore, the biomonitoring effect of mosses on atmospheric PAHs should be further studied comprehensively through the growth characteristics of mosses and various physiological indicators in the polluted environment. However, mosses have excellent research potential in phytoremediation, with wide distribution and prominent leaf surface areas that can adsorb a large number of atmospheric pollutants. Therefore, it will be a research measurement focus in the future to apply mosses to the field of phytoremediation to exert their ecological effects. In addition, a combination of biochemistry, plant physiology, and physics is beneficial for mosses to improve their biomonitoring ability and sensitivity. In some cases, the concentration of PAHs pollutants in moss is directly related to the measured atmospheric concentration, and meteorological parameters must be considered. Mosses are more or less limited if the intention is to monitor environmental change over a wide range of time and space. However, if all samples are collected at the same time and at roughly similar temperatures and humidity, there is no doubt that mosses (a single species) can be used for local monitoring of individual events. According to the current progress in the field, we suggested that the following three aspects need to be improved and strengthened: (i) the relationship between the accumulation of PAHs in different mosses and the concentration, (ii) mosses should be combined with physical and chemical methods to improve their sensitivity, and (iii) how to better reflect human health problems by mosses in biomonitoring of atmospheric PAHs.

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