Review

Tracers from Biomass Burning Emissions and Identification of Biomass Burning

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Abstract: The major organic compositions from biomass burning emissions are monosaccharide derivatives from the breakdown of cellulose, generally accompanied by small amounts of straight-chain, aliphatic, oxygenated compounds, and terpenoids from vegetation waxes, resins/gums, and other biopolymers. Levoglucosan from cellulose can be utilized as a specific or general indicator for biomass combustion emissions in aerosol samples. There are other important compounds, such as dehydroabietic acid, syringaldehyde, syringic acid, vanillic acid, vanillin, homovanillic acid, 4-hydroxybenzoic acid, and p-coumaric acid, which are additional key indicators of biomass burning. In this review, we will address these tracers from different types of biomass burning and the methods used to identify the sources in ambient aerosols. First, the methods of inferring biomass burning types by the ratio method are summarized, including levoglucosan/mannose, syringic acid/vanillic acid, levolgucosan/K^+, vanillic acid/4-hydroxybenzoic acid, levoglucosan/OC, and levoglucosan/EC to infer the sources of biomass burning, such as crop residual burning, wheat burning, leaf burning, peatland fire, and forest fire in Asia. Second, we present the source tracer ratio methods that determine the biomass combustion types and their contributions. Finally, we introduce the PCA (Principal component analysis) and PMF (Positive matrix factor) methods to identify the type of biomass burning and its contributions according to emission factors of different species in various plants such as softwood, hardwood, and grass.

Keywords: biomass burning emissions; PCA; PMF; tracers; sources; STR; ratio

Highlights

1. Key tracers from different biomass burning emissions are introduced in this review.
2. Methods applied to identify biomass burning types are introduced, and their advantages and disadvantages are also concluded.
3. The details of PMF to infer biomass burning types and source contributions are also included.

1. Introduction

Biomass burning such as wildfire, agricultural open-burning, residential biofuel combustion, forest fire, grass burning, and peatland fire can produce and release substantial amounts of pollutants into the atmosphere [1,2]. Aerosols released from biomass burning have a serious influence on climate change, the ecosystem, regional/global air quality, visibility, and human health, which had drawn global concern all over the world [3,4]. Biomass burning contributes much greater carbonaceous aerosol emissions in regions with

intense fires [5,6]. These emissions can be transported over long distances and affect areas far away from the source region [7–10]. Previous assessments of biomass burning emissions using combustion by-products as tracers have studied the following: (1) pollutants containing volatile, gaseous carbonaceous compounds such as CO, CO_2, CH_4, C_2H_4, HCN, HCO_2H, CH_3CO_2H, CH_3CN, and aerosols, especially organic carbon and black carbon; (2) macromolecular smoke particles in the form of soot or charcoal; (3) solvent-soluble organic compounds isolated from smoke particulate matter such as polynuclear aromatic hydrocarbons (PAH); and (4) the source of biomass burning in aerosols based on different methods such as the ratio of tracers [11–13]. Thus far, there is a lack of relative complete information about biomass burning emissions, especially using unique tracers to identify the source. In order to better understand biomass burning, comprehensive information about biomass tracers of different combustions and methods to infer burning types are illustrated in this review. For example, Figure 1 showed that the biomass burning sources, transport and deposition in environment and atmosphere. Furthermore, our future work is also displayed.
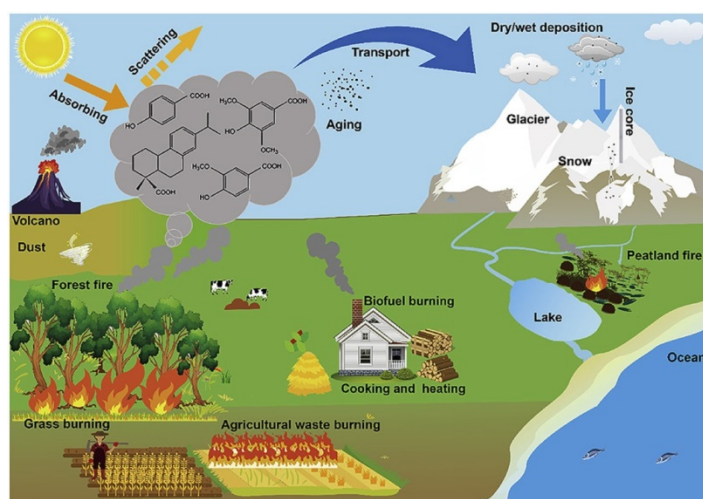

Figure 1. Schematic of the sources, transport, and deposition of biomass burning emissions [14].

1.1. Biomass Burning Process

The process of biomass burning is briefly described as follows. When biomass fuels are heated, with the increase of temperature, the compositions gradually begin to hydrolyze, oxidize, dehydrate, and pyrolyze, forming combustible volatile products, tarry substances, and highly active carbonaceous char [15]. At the ignition temperature of the tarry substances and volatiles, combustion reactions begin. As for complete combustion, carbon continues to be formed until no solid fuel remains in the ash and no combustion gas in the smoke. For incomplete combustion, there is still residual fuel in the smoke. The emissions produced by the combustion of fuels are directly dependent on the chemical composition of the fuel and the combustion conditions.

1.2. Organic Compounds from Biomass Burning

Due to the unique signatures of organic molecular tracers in the atmosphere, they play an important role in identifying biomass fuel combustion [16,17]. Cellulose, hemicellulose, lignin, sporopollenin, and suberin are important units of biomass fuels, and their pyrolysis products are widely used as unique tracers of biomass burning in the atmosphere [18,19]. Simoneit et al. identified a series of tracers in aerosols that are influenced by biomass burning, including alkane homologs, polycyclic aromatic hydrocarbons, pineic acids, sugars in cellulose, and methoxyphenols in lignin by Gas chromatography-mass spectrometry/mass spectrometry (GC-MS/MS) as shown in Figure 2 [14,16,17,19,20].
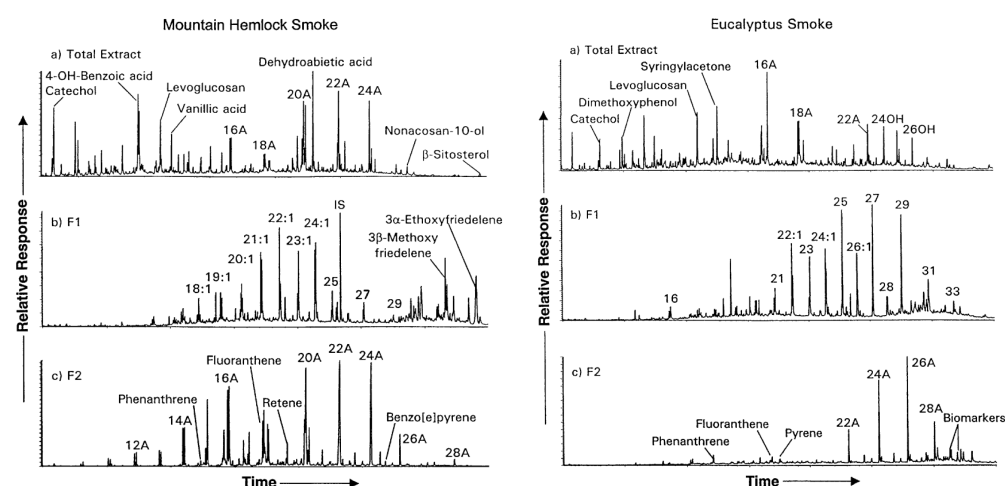
Figure 2. Chromatographic separation of biomass burning tracers using GC-MS/MS [17].

1.2.1. Lignin Products

Lignin is an essential biopolymer of wood, which is derived primarily from three aromatic alcohols, which are named sinapyl, coniferyl, and p-coumaryl alcohols. The degradation products produced by lignin burning were mainly three kinds of compounds [14,21]. The proportions of these molecular markers vary greatly among different plant species. For example, lignins of hardwoods (angiosperms) are abundant products of sinapyl alcohol, whereas lignins of softwoods (gymnosperms) have a relatively high proportion of products from coniferyl alcohol, with a lesser composition from sinapyl alcohol; whereas grass products are dominantly from p-coumaryl alcohol [12,22]. The lignin combustion usually produces phenols, aldehydes, ketones, acids, and alcohols, which retain the components on the original benzene ring, such as hydroxyl and methoxy. During combustion, both softwood and hardwood can produce guaiacol derivatives such as 2-methodyphenol, but hardwood also can produce high levels of sryingol derivatives like 1,3-dimethoxyphenol [23,24]. Hawthorne et al. identified that guaiacol derivatives are potential tracers of wood-burning, while syringyl derivatives are indicators for hardwood burning [23,24]. Most woody plants are abundant in lignans, which are critical dimers of sinapyl, p-coumaryl, and coniferyl alcohols [25,26]. They provide supportive fillers, toxins, and other uses for plants [26].

Softwood Lignin

Methoxyphenol is the main compound produced by softwood lignin decomposition. Vanillic acid is derived from coniferyl alcohol, which is the main aromatic alcohol monomer unit in gymnosperm lignin and is a specific tracer of conifers [21]. For example, pine wood burning releases mainly vanillic acid and vanillin and lesser amounts of other pyrolysis compounds, syringic acid, syringaldehyde, and p-anisic acid. The diterpenoid dehydroabietic acid is determined in all samples and is the main product of resin acids, which are source-specific [21,27].

Hardwood Lignin

A series of methoxyphenols are also produced from the burning of hardwood lignin. These compounds are derived from syringyl alcohol, which is enriched in hardwood lignin and primarily includes syringyl acetone, acetosyringone, syringic acid, and the dimer species of disyringyl. Angiosperm lignin contains high levels of the coniferyl and sinapyl alcohol subunits, which are the precursors of oxidation and pyrolysis products of syringol and methoxyphenol [28]. For example, hardwoods, such as oak, have rich quantities of syringic acid and syringaldehyde in their smoke. There are different amounts of sinapine-type and other pyrolysis products, which are consistent with angiosperm lignin as the source, as well as a small amount of vanillin-type products. Hawthorne et al. and Simoneit et al. concluded that the syringyl molecular markers are indicators in smoke produced

by angiosperm fuels burning [21,29,30]. The molecular biomarkers, including ursana-2, 12-dien-18-oicacids, oleana-2, and 12-dien18-oic are the thermal degradation products from ursolic acids and oleanolic, respectively, and the major triterpenoids in angiosperm mucilages and gums [31,32].

Grass Lignin

In order to have a good knowledge of organic tracers released from grass burning, previous studies collected aerosol samples from the smoke of mixed ryegrasses consisting dominantly of n-alkanoic acids, levoglucosan, and phenolic compounds, including syringyl acetone, catechol, guaiacyl acetone, dimethoxyphenol, syringic acid, vanillyl acetic acid, and vanillic acid [17]. However, other studies have found that the burning of grass mainly releases p-anisic acid, p-anisaldehyde, and minor amounts of other p-coumaryl, vanillyl, and syringyl-type lignin pyrolysis products such as p-coumaric acid, 4-hydroxybenzoic acid, and syringic acid [12,17]. The grasses include all kinds of phenolic moieties of lignin precursor alcohols. Therefore, the thermal decomposition products are not representative tracers for grass burning, except for p-coumaric acid and 4-hydroxybenzoic acid, which are not prevalent in hardwood and softwood [12,33]. Therefore, the relative proportion of these compounds can also be used to identify grass burning.

1.2.2. Cellulose

The combustion process of cellulose and hemicellulose has been studied extensively in the past [15,34–36]. Cellulose decomposes via two alternative pathways when heated or exposed to an ignition source. At first, it is decomposed at 300 °C and then begins to bond, cleave, undergo fission and disproportionation, and produce tar anhydrous sugars and volatile products by transglycosylation. The second pathway produces molecular tracers from a specific source such as levoglucosan, the furanose isomer, and dianhydride. It is reported that L-glucan is present as fine particles from the burning of residential wood [37,38]; it is very stable in the atmosphere and has no chemical reaction after being exposed to sunlight and environmental conditions for 8 h [39–42]. Furthermore, L-glucan can have large-scale impacts on the chemical composition of atmospheric aerosols on a regional to global scale. Due to its water-solubility, levoglucosan contributes to water-soluble organic carbon in aerosols and significantly enhances the hygroscopic properties of atmospheric aerosols [43]. In addition, it is reported that levoglucosan exists in pyrolysis products of peat wood and lignocellulose burnings [44,45]. Mannosan and galactosan, isomers of levoglucosan, are produced by pyrolysis of cellulose and hemicellulose and can also be regarded as biomass burning tracers [17,46].

1.3. LC-MS/MS

Liquid chromatography-tandem mass spectrometry (LC-MS/MS) is utilized to quantify aromatic acids such as biomass burning tracers, is pretty effective, and has simplified the pretreatment of samples without needing derivatization of biomass burning products like vanillic acid, isovanillic acid, homovanillic acid, and syringic acid. For example, Hoffmann et al. developed and validated an HPLC-APCI-MS/MS (high-performance liquid chromatography-atmospheric pressure chemical ionization-tandem mass spectrometry) method for measuring biomass burning tracers [47,48]. The analysis time is short (only 15 min), the detection limit is low, and the recovery rate is high, which is suitable for the identification and quantification of aromatic acids in aerosol or other samples. More recently, in the research of Zangrando [8]., an effective and rapid HPLC-(-)-ESI-MS/MS (high-performance liquid chromatography-(-)-electrospray ionization-tandem mass spectrometry) analytical method was reported for quantitative determinations of aromatic acids, such as biomass burning tracers. An example of a total ion chromatographic analysis is shown in Figure 3 [12,49]. In addition, this approach shows considerably low limits of detection for vanillic acid and syringic acid. In our previous study, a fast and effective HPLC-MS/MS method was developed and validated, and 20 kinds of biomass burning

tracers in aerosol samples were successfully determined, with low limits of detection and high accuracy in 10 min analyses. The preparation of aerosol samples is a single-step derivation saving more time than before. The details of this method and aerosol sample preparation can be obtained in [50,51].
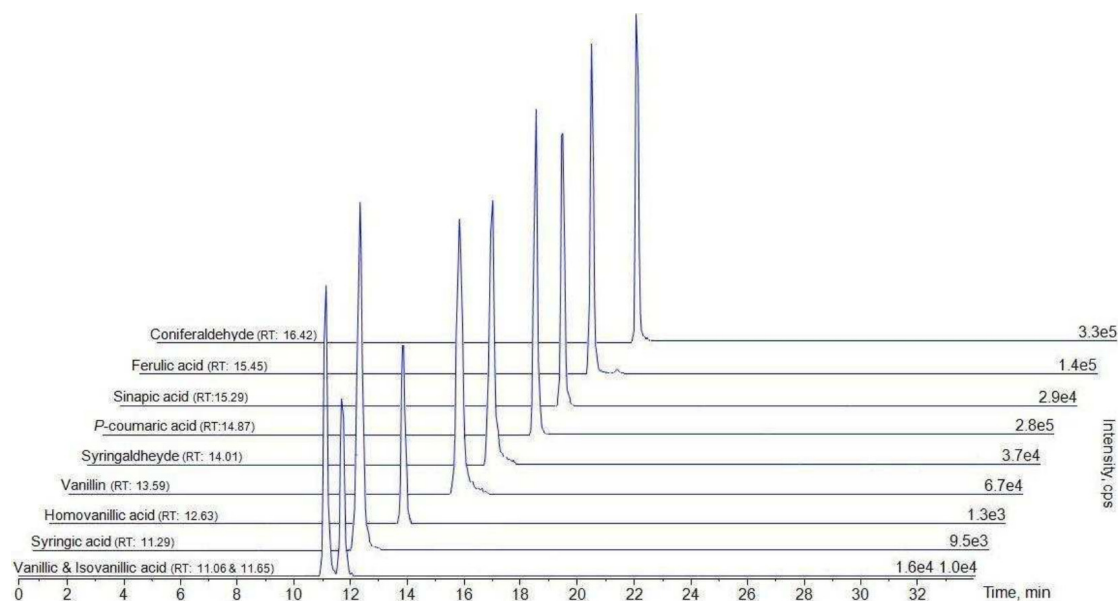

Figure 3. Chromatographic separation of biomass burning tracers by LC-MS/MS (1.6e4 means 1.6×10^4) [12].

2. Determination of Biomass Burning Types

Lignin pyrolysis products are an important component of the fine aerosol particles from wood and biomass burning [21,52]. However, the presence of individual syringyl, coumaryl, or vanillyl-type compounds in an aerosol or smoke sample cannot be a unique indicator for the original sources of biomass burning, but the relative proportions of such tracers can be used to distinguish the type of burned fuels. At present, there are many methods to determine biomass burning types, such as tracer ratios, principal component analysis, source tracer ratio, positive matrix factor analysis, etc. The following describes the advantages and disadvantages of these methods. Because of complex atmospheric processes like photochemical aging, phase-separation/partition and deposition, and varied chemical lifetimes, characteristic tracers and specific ratios have constraints in targeting transported pollution sources and classifying the contributions of the respective biofuels. Therefore, we select stable, long-lifetime compounds as biomass burning tracers to improve the results of identifying biomass burning types.

2.1. The Ratios of Tracers to Identify the Type of Biomass Burning

The most common and basic method for identifying the type of biomass burning in the atmosphere is the ratio method. In previous studies, the ratio method was divided into four types, including the ratios of levoglucosan to mannosan [53], levoglucosan to OC [54,55], levoglucosan to potassium, vanillic acid to syringic acid, and syringic acid to 4-hydroxybenzoic acid [56].

2.1.1. Levoglucosan/K$^+$

Previous studies used the levoglucosan/potassium (Levo/K$^+$) ratio to distinguish specific types of biomass burning in ambient aerosols [57]. For forest fire aerosols, the ratio of Levo/K$^+$ is typically above zero; for example, the ratio in Amazon forest fire smoke is 0.38–22.0, and the ratio in U.S. forest fire smoke is 4.35–58.8 [58–60]. Incineration of agricultural wastes is typically found to have a relatively low ratio of Levo/K$^+$ because potassium fertilizer is a large K$^+$ contributor. For example, Cheng et al. reported that

the burning of crop residuals such as wheat straw, rice straw, and corn straw had a Levo/K^+ ratio that was higher during the winter (0.51 ± 0.15) compared with the summer (0.21 ± 0.16), while the Levo/K^+ ratio averaged 0.11 ± 0.06 during the biomass burning period in Beijing aerosols [1,61]. The Mkoma study reported lower Levo/K^+ ratios with an average value of 0.37 ± 0.1 ranging from 0.82–0.57 in PM2.5 and 0.38 ± 0.2 ranging from 0.16–0.87 in PM2.5 during wet days, suggesting that the sampling site in Tanzania may be impacted by emissions of K^+ from the burning of plants and from the soils that contain a high abundance of K^+ [62]. In addition, the lower ratio of Levo/K^+ was 0.25 ± 0.7 ranging from 0.14–0.36 in PM2.5 and was 0.23 ± 0.06 ranging from 0.14–0.36 in PM_{10} dry burning days, which indicates that Tanzania burned agricultural crop residuals [62]. Several studies on crop burning residue in Asia have reported the Levo/K^+ ratio as ranging from 0.1 to 1.2 [61–64]. In the latest research conducted in the Chiang Mai Basin, due to the burning of crop residuals, the ratio of Levo/K^+ in PM_{10} ranged from 0.51–0.56 in the dry season [65]. In a study by Thepnuan et al., the ratio of Levo/K^+ was 0.92 ± 0.35, indicating that the ambient aerosols in Chiang Mai during the dry season in 2016 were influenced by burning forest and agricultural crop residuals, such as rice straw, maize, and sugar cane [56].

2.1.2. Levoglucosan/Mannosan

Galactosan and mannosan are isomers of levoglucosan and products of hemicellulose thermal decomposition, while levoglucosan is a product of cellulose thermal decomposition [17]. Hardwood contains a higher amount of cellulose than hemicellulose; therefore, the ratio of levoglucosan to mannosan (L/M) can also be used to distinguish the type of biomass burning [66] (i.e., softwood vs. hardwood) [17,67,68]. Previous studies have found that L/M ratios of burned crop residues are often > 40 [69], while the L/M ratios of burning hardwood are within the range of 15–25, and the L/M ratio of burning softwood is 3–10 [64,68,70]. For example, a high L/M ratio of biomass combustion aerosols produced by burning straw (40–42) was calculated [64]. In the North China Plain, aerosols are concentrated in the mountains, from severe levels of wheat straw burning (40–46) [70]. In a study by Fu et al. [71], the L/M ratios varied from 25 to 35, which were similar during sampling periods at the Hetian site, and the L/M ratios ranged from 33 to 60 and 23 to 59 in TSP and PM2.5 samples, respectively, at the Tazhong site. Therefore, high L/M ratios ranging from 23 to 60, found in their study, indicate that crop residues, hardwood, and herbaceous plants are the main fuel types for biomass burning in the Taklimakan region [71]. In the research of Thepnuan et al., the L/M ratios are in the range of 15.7–37.6, with an average of 20.4 ± 4.1, which was much higher than the values calculated from softwood burning (2.6–5.0) [64,67]. Similar to those calculated from hardwood burning (13–32) [60,64], Asian crop residue burning was 12–55, and corn straw burning was 19.5 ± 3.4 [1,56,61,64]. In Tanzania, East Africa, in 2011, during wet and dry seasons, it was found that the L/M ratios were fairly high, ranging from 10 to 13 in PM2.5 and from 9 to 13 in PM10. Such a high proportion indicates that the biomass burned in this land is dominantly hardwood and crop residues [62]. In Zhu's study, higher monthly average L/M ratios (38.9) in November–January suggest that the burning substrate is primarily hardwood, a mixture of softwood and hardwood, or a mixture of softwood and plant straws in MNA [66]. The low L/M ratios (2.1–4.8) in May–June, however, indicate that the aerosol in Okinawa is influenced by the burning of softwood [66]. Finally, the lower L/M ratios in Kang's study (average 4.2) suggested that the aerosols were mainly associated with the burning of softwood in the marine atmosphere over the East China Sea [72]. See Table 1 for more research on L/M identification of biomass combustion types.

Table 1. Ratios of anhydrosaccharides in source test emissions from lignite and biomass burning [46].

Sample Type		L/M	L/(M + G)	Data Source
Lignite	Jsp11	31	31	Fabbri et al. (2009) [46]
	S6s	92	92	Fabbri et al. (2009) [46]
	Lst6	40	40	Fabbri et al. (2009) [46]
	Average	54	54	
Hardwoods		22	17.6	Nolte et al. (2001) [73], Fine et al. (2002) [74]
		13–24		Fine et al. (2004) [59]
		14.5–14.6	8.5–9.9	Schmidl et al. (2008) [53]
		13.8–32.3	4.5–14	Engling et al. (2006) [75]
		3.3–8.4	1.5–2.1	Oros and Simoneit (2001b) [76]
Softwoods (conifer)		4	3.6	Nolte et al. (2001) [73], Fine et al. (2002) [74]
		3.9–6.7		Fine et al. (2004) [59]
		2.6–5.0	2.4–5.0	Engling et al. (2006) [75]
		3.6–3.9	1.8–2.8	Schmidl et al. (2008) [53]
		0.6–13.8	0.4–6.1	Oros and Simoneit (2001a) [77]
Charred pine wood		2.5	2.0	Otto et al. (2006) [78]
Charred pine cone		0.3	0.2	Otto et al. (2006) [78]
Forest fire smoke		4.8–5.6	3.2–3.9	Ward et al. (2006) [79]
Grasses		2.0–33.3	1.7–9.5	Oros et al. (2006) [80]
Green hardwood litter		4	2	Medeiros and Simoneit (2008) [81]
Green softwood litter		3.6	2.1	Medeiros and Simoneit (2008) [81]
Atmospheric aerosols		3.5–75	3.2–14	Pashynska et al. (2002) [82], Yttri et al. (2005) [82], Ward et al. (2006) [79], Simoneit et al. (2004b,c) [83,84], Medeiros et al. (2006) [85], Zdra'hal et al. (2002) [86], Sandradevi et al. (2008) [87]

There are many studies regarding the identification of biomass burning type by using levo/K+ and L/M ratios separately. However, as shown in [61], these two ratios are typically combined to infer potential sources of biomass burning. For example, in Thailand, the L/M ratios were approximate to those gained from leaf litter in dry dipterocarp forest and maize residues with an average of 19.0 ± 3.5 and 24.2 ± 12.3, respectively, while the Levo/K^+ ratio is about the available leaf litter in mixed deciduous forests, with an average of 0.90 ± 0.88. The results of these two ratios indicated an origin from agricultural waste burning and forest fires, which was presented in Figure 4 [56].

In addition, Jung et al. reported that hardwood combustion has a relatively high ratio of L/M and Levo/K^+, with average values of 26 and 28, respectively, and variation ranges of 2.2–195 and 11–146 [88]. The Levo/K^+ ratios from the burnings of softwood were similar to those from the burnings of hardwood, with an average of 46 and a range of 4.6–261. The L/M ratios from the burnings of softwood were, however, much lower than those from the burnings of hardwood, with an average of 4.3 and a range of 2.5–6.7. The L/M ratios from the burning of Asian crop residue were similar to those from burning hardwood, with an average of 29 and a range of 12–55, whereas the Levo/K^+ ratios from burning Asian crop residue were much lower than those from burning hardwood and softwood, with a value of 0.53 and a range of 0.1–1.2. In their study, Jung et al. reported aerosol samples collected from 5–8 November that showed a very low concentration of mannosan [88]. The Levo/K^+ ratio of this sample was similar to other data, while the L/M ratio was much higher. Therefore, the elevated L/M ratio in this aerosol sample could be explained by elevated emissions from burnings of crop residue, grass, or hardwood. The Levo/K^+ ratios calculated in this study were similar to those from burnings of grass and Asian crop residue, whereas the L/M ratios were similar to those obtained from the burnings of leaf and softwood. Calculated data in their study were located between the leaf/softwood cluster and the Asian crop residue/grass cluster; therefore, the tracers of biomass burning collected in their study might be a mixture of aerosols produced by burning grass, crop residue, leaf, and softwood, which was shown in Figure 5 [11,88].
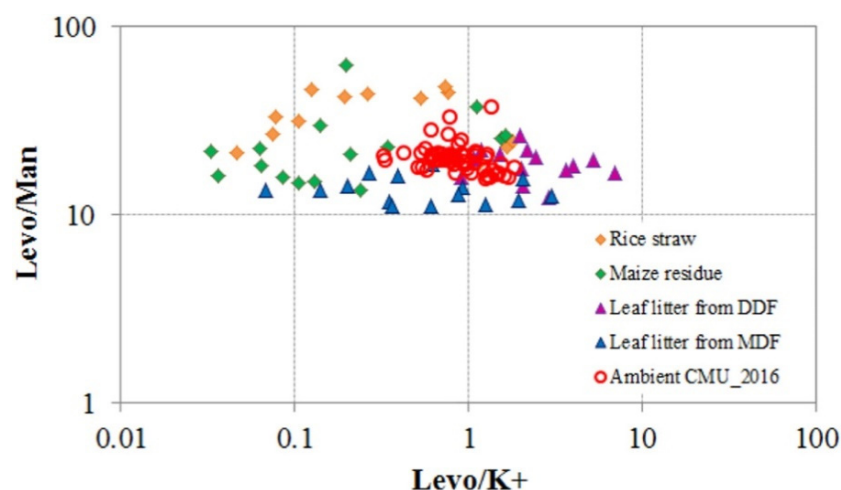
Figure 4. Scatter plot of Levoglucosan to K$^+$ ratios versus levoglucosan to mannosan ratios from different types of biomass burning [56].
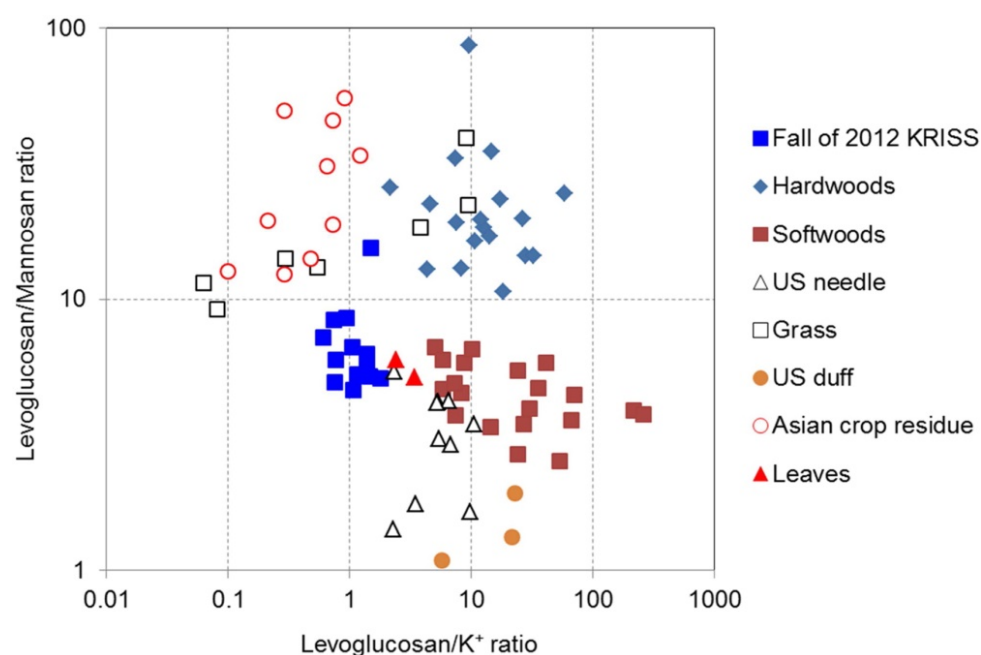

Figure 5. Scatter plot of levoglucosan to K$^+$ ratio versus levoglucosan to mannosan ratio of different types of biomass burning emissions [88].

2.1.3. Levoglucosan/OC or Levoglucosan/EC

The ratios of biomass burning tracers to OC and EC concentrations have also been used to identify various types of biomass burning [89]. Figure 6 shows a map of levoglucosan to OC ratios (Levo/OC ratios) and levoglucosan to EC ratios (Levo/EC ratios) from different types of biomass burning according to previous studies [53,55,60,63,68]. In the research of Jung et al. [88], the Levo/OC ratios and Levo/EC ratios suggested indicate the tendency of burning leaves and grass. Besides, the increasing trend was within the variability of crop residue burning in Asia [55,63]. Therefore, this study showed that grass, leaves, and crop residues were the main sources of biomass burning in the Daejeon air during the rice harvest season. Finally, this method of identifying the biomass burning types was similar to the previously reported method of burning agricultural residue during the fall rice harvest in Korea [90].
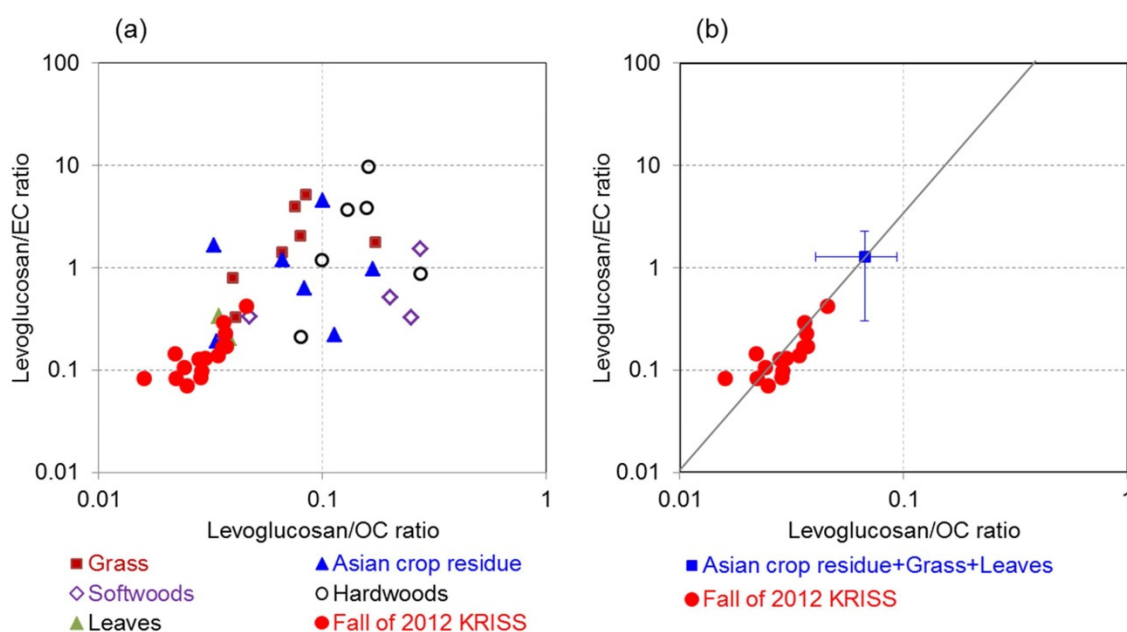
Figure 6. Scatter plots of levoglucosan to OC ratios versus levoglucosan to EC ratios of (**a**) different types of biomass-burning emissions and (**b**) average of the Asian crop residue, grass, and leaf burnings [88].

2.1.4. Vanillic Acid/Syringic Acid

The concentration of each tracer in an aerosol depends on the combustion conditions, amounts, and biomass burning type. For example, a low concentration of syringic acid may result from a lower amount of hardwood in burning emissions, while a higher concentration of p-hydroxybenzoic acid suggests a dominance of herbaceous plants [33]. Moreover, for the same quantity of different types of biofuels, the ratio of two biomass-burning tracers can be characteristics of the biomass, and it is more accurate than the single concentration. Therefore, concentration ratios of different aromatic acids are regarded as more important indicators to infer the type of biomass burning [21,33]. Recently, the ratio of vanillic acid/syringic acid (VA/SyA) has been used for identifying the biomass burning source in aerosols [1,17,33,59,91]. Figure 7 summarizes typical VA/SyA ratios of different potential sources (softwood, hardwood, herbaceous plants, grass, and other biomass burning sources) over the world. Briefly, the VA/SyA ratios ranged from 0.12 to 4.00 for hardwood burning and ranged from 8.57 to 11.9 for softwood burning, whereas the ratios of grass or herbaceous plants are between the hardwood burning and softwood burning, with values ranging from 0.40–5.03. The SA/VA ratios for burning woody angiosperms (hardwood) and non-woody angiosperms (herbaceous plants) were 1.90–2.44 and 0.10–2.00, respectively, while SA/VA ratios of burning gymnosperms (0.01) and non-woody gymnosperms (0.03–0.24) were much lower [22,33]. Therefore, if the proportions overlap, it is difficult to distinguish the type of biomass burning, especially when there are more than two emission sources. For example, Wan et al. reported VA/SyA ratios in aerosols collected in Lumbini, South Asia, ranging from 0.39 to 2.56 with a mean of 1.28 ± 0.50, which indicated herbaceous plants (crop residue) and hardwood as likely sources [92]. A previous study reported that the VA/SyA ratio was 1.70 ± 0.36 in Malaysia, while Indonesian peat-land fires period, which was approximately three times higher than the VA/SyA ratio (0.59 ± 0.27) on normal days, which provided a good indicator of the impact from the Indonesian peatland fires [93]. In aerosol samples from Beijing and Godavari, the SA/VA ratios were 2.21 (2.05–2.48) and 0.71 (0.62–0.96), respectively, suggesting that hardwood and grass (including agricultural residue) were more likely to be the sources for the biomass burning aerosols in these two regions [94]. In addition, the ratios of vanillin (VAN) and syringaldehyde (SyAH) were further applied to identify the sources of aerosol samples [49].
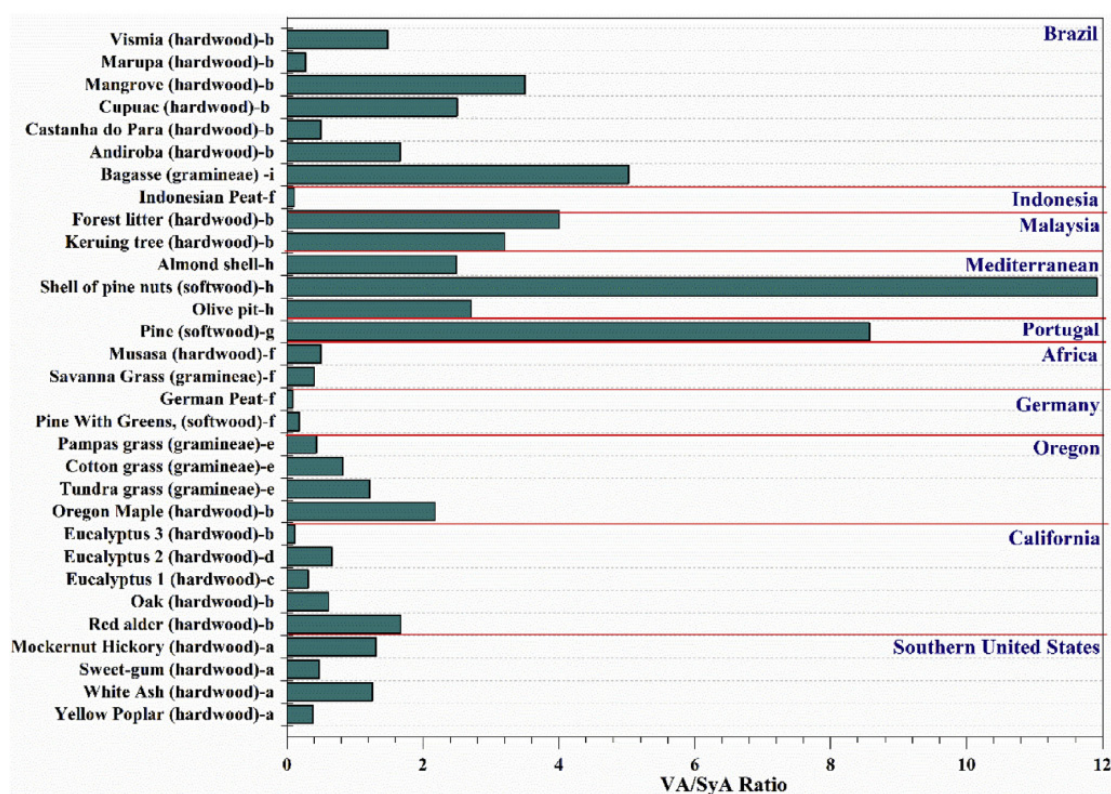
Figure 7. Comparison of VA/SyA mass ratios obtained from hardwood, softwood, grass, and other burning sources [1].

2.1.5. Vanillic/p-hydroxybenzoic Acid

The ratio of vanillic/p-hydroxybenzoic acid (VA/p-HBA) is also applied to identify types of biomass burning [1,95–97]. For example, the content of vanillic acid produced by burning conifers is higher than that of p-hydroxybenzoic acid, resulting in a higher VA/p-HBA ratio [88,98,99]. The VA/p-HBA ratio produced by pinewood in Europe is 8.75 [97], while the VA/p-HBA ratio after burning ponderosa pine was 19.75 [1,98]. In contrast, burning herbaceous plants contributes higher yields of p-HBA than VA, and Iinuma found that peat fires produced more p-HBA than VA [96]. In addition, P-HBA with almost no VA was released by grassland fires in the North American tundra [98,100]. In Gao's study, the ratio of VA to p-HBA was determined as 0.51 in PM_{10} samples from Beijing and 0.70 in PM2.5 samples from Godavari, South Asia. Such low values indicate that agricultural residue is substantially burned for cooking or heating for the residents of the North China Plain and South Asia [94]. The ratio of VA/p-HBA in aerosol samples collected in Washington, D.C. is 0.35, which indicates that the burning of herbaceous plants in this area is greater than that of conifers [94]. It should be noted that the relative abundance of aromatic acids in the environment is not only controlled by emission characteristics but also affected by possible transformations in the atmosphere (such as photochemical degradation). Therefore, the ratios of VA/p-HBA close to the source area of biomass combustion may be more reliable than that far away from the receptor area.

2.1.6. K$^+$/EC, char-EC/soot-EC, FLU/(FLU + PYR) and IP/(IP + BgP)

There are many other methods to identify biomass burning sources such as K$^+$/EC, char-EC/soot-EC, PAH, and others. Here we briefly introduce three ratio methods.

First, K$^+$/EC can be used to identify biomass burning. For example, in Cao et al.'s study, they examined aerosols during biomass burning periods. A strong positive correlation was observed among K$^+$, OC, EC, and WSOC during the biomass burning episodes, indicating an important contribution of crop residues burning to carbonaceous aerosols during this biomass burning period. The ratios of OC/EC, K$^+$/OC, K$^+$/EC, and WSOC/OC

for the agricultural-residue burning emissions were evaluated as 10.4 ± 0.5, 0.054 ± 0.006, 0.57 ± 0.07, and 0.48 ± 0.01, respectively [95]. The OC/EC ratio of 10.4, calculated in that study, was much higher than that for the wheat-residue burning (3.0) but similar to the previously reported ratio in paddy-residue burning (10.6) [96].

Second, the soot-EC/char-EC ratio depends on a mixture of different sources: motor vehicle emissions and possibly grass burning result in higher soot-EC/char-EC ratios, while wood combustion, particularly biomass burning by smoldering at low temperature, produces lower soot-EC/char-EC ratios [97,98]. In coal combustion, this ratio can be very low or high, depending on the type of coal [99]. In Lim's study, the EC2 + 3/EC1 ratio of 1.25 in summer was comparable to the ratio of 1.67 for motor vehicle exhaust [97,100].

Finally, a FLU/(FLU + PYR) > 0.5 indicates coal and biomass burning. While a ratio of IP/(IP + BgP) > 0.6 represents plant burning [101].

2.2. Principal Component Analysis

2.2.1. Introduction of PCA

The basic idea of principal component analysis is described in [102]. Briefly, a few principal components are derived from original variables, and the information of the original variables is retained as much as possible, though they are not related. It does not require information on the source component spectrum, but one principal component may contain multiple sources.

2.2.2. Application of PCA

In the field of aerosols, PCA is usually used to identify pollution sources. For example, Wang et al. collected aerosol samples with a large capacity sampler in the Yellow Sea. Then, the ions and metals were quantified by ICS-3000 and ICP-MS, and the combustion source of light oil combustion, sea salt, SOA (secondary organic aerosol), and soil/metal melting was determined by PCA [103]. Xu et al. collected PM10 aerosol samples in Fuzhou with a high volume sampler. Ions were quantified by ICS-3000, and a mixture of secondary inorganic aerosols, marine aerosols, and traffic sources were identified by PCA [104]. Thepnuan et al. collected PM2.5 aerosol samples in Chiang Mai Province, Thailand, using a mini volume air sampler. ICS-5000 and ICS-3000 were used to quantify the ions and sugar alcohols and sugars in samples. Then PCA was applied to identify the sources of pollutants in this field. Five factors, such as biomass burning of agricultural wastes, traffic-related emission, photochemical formation of SO_2 generated by traffic/industrial activity, biomass burning, dust, and sea salt [56]. However, there is no research on using PCA to identify the types of biomass burning, such as softwood, hardwood, grass, peat wood, and seed oil combustion. If sufficient data are available, PCA can be applied to identify these different types of burn products. The advantage of PCA is that before PMF analysis, PCA can be selected to preliminarily determine the sources of pollution in the studied area. The disadvantages are that there must be enough samples and that one sample may contain more sources of pollution.

2.3. Source Tracer Ratio

2.3.1. Introduction of Source Tracer Ratio

The source tracer ratio method assumes that there is a fixed emission ratio between the compound emitted from a certain source and the tracer of the source and that the ratio will not change during the transmission process. Through the emission ratio and the concentration of the tracer, the contribution ratio of the compound from this primary source can be calculated as follows [105]:

$$[\text{Compound}] = [\text{Compound}]_{\text{pri}} + [\text{Compound}]_{\text{other}} + [\text{Compound}]_{\text{bg}} \quad (1)$$

$$\begin{aligned}
[\text{Compound}]_{\text{other}} &= [\text{Compound}] - [\text{Compound}]_{\text{pri}} - [\text{Compound}]_{\text{bg}} \\
&= [\text{Compound}] - \text{ER} \times [\text{tracer}] - [\text{Compound}]_{\text{bg}}
\end{aligned} \quad (2)$$

In the formula, [Compound], [Compound]$_{\text{pri}}$, [Compound]$_{\text{bg}}$ represent the measured concentration of the compound, the concentration from the primary source, and the concentration from other sources, respectively. ER is the ratio of this compound to the tracer, that is, the emission ratio.

The emission ratio was determined through observation data. The specific calculation method assumes that ER is a known value, and then [Compound]$_{\text{other}}$ can be obtained according to Equation (2) by subtracting the primary emission concentration and the background concentration from the measured concentration. Because [Compound]$_{\text{other}}$ has nothing to do with the selected discharge, there should be no correlation between [Compound]$_{\text{other}}$ and [Compound]$_{\text{pri}}$. By testing different ER values within a certain range, the square correlation coefficient r^2 of [Compound]$_{\text{other}}$ and [Compound]$_{\text{pri}}$ corresponding to different ERs can be obtained. The ER corresponding to the lowest value in this series of r^2 is considered to be the actual emission ratio.

2.3.2. Application of Source Tracer Ratio

When applying the source tracer ratio method to determine the type of biomass burning, each term in the formula can be defined by itself. For example, in our previous study [106], SO$_2$ was selected as the tracer for coal burning, and then the concentration of levoglucosan was divided into two parts (Equation (3)): the concentration of levoglucosan from coal burning ([Levoglucosan]$_{\text{coal}}$) and the concentration of levoglucosan from other types of biomass burning ([Levoglucosan]$_{\text{other}}$). The enhancement ratio (ERs) can be determined when the r^2 between the concentration of levoglucosan from other biomass burning and the concentration of SO$_2$ is the minimum. Finally, the contribution of coal burning to levoglucosan is calculated to be 15%, indicating that the types of biomass burning include coal, such as peat and lignite burning.

$$
\begin{aligned}
[\text{Levoglucosan}] &= [\text{Levoglucosan}]_{\text{coal}} + [\text{Levoglucosan}]_{\text{other}} \\
&= \text{ER} \times \left([\text{SO}_2] - [\text{SO}_2]_{\text{bg}} \right) + [\text{Levoglucosan}]_{\text{other}}
\end{aligned}
\tag{3}
$$

The application of the source tracer ratio method is not prevalent. According to its principle, appropriate tracers can be selected to identify the type of biomass burning, such as peat, lignite, tobacco, etc. At the same time, the contribution of corresponding biomass burning to the total biomass burning can be determined. The advantage of using the ratio of source tracer is that it is simple and requires less data, but the disadvantage is that it relies too much on the choice of tracers.

2.4. PMF

2.4.1. Introduction of PMF

Positive matrix factor analysis is an effective technique for source analysis, which estimates the composition of pollution sources and their contribution to the environmental concentration based on measured data at the receptor site. There is no need to require source component spectrum information. Therefore, it is more suitable for areas where the pollution sources are not very clear. The data matrix X with m rows and n columns (m and n are the number of samples and the number of species, respectively) is decomposed into the product of two matrices G (m multiplied by p) and F (p multiplied by n), and adding a residual matrix E:

$$
x_{ij} = \sum_{k=1}^{p} g_{ik} \, f_{kj} + e_{ij}
\tag{4}
$$

in which X_{ij} is the concentration of species j in sample i, p is the number of sources, g is the contribution of the source, f is the mass ratio of the species to the source, and e is the residual.

The PMF's G and F are derived by minimizing the objective function Q as follows:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{e_{ij}}{u_{ij}} \right]^2 \tag{5}$$

The US EPA PMF 5.0 software is used for analysis. In the input data, The uncertainty matrix u_{ij} of the sample is calculated according to the recommended method in the operation manual: ① If the concentration of the substance in the environmental sample is lower than or equal to the detection limit of the analytical method, the uncertainty $U_{nc} = (5/6) \times MDL$, where MDL is the detection limit of the analytical method; ② If the concentration of the substance in the environmental sample is higher than the detection limit of the analytical method, the calculation formula of the uncertainty is:

$$\sqrt{(\text{ErrorFraction} \times \text{Concentration})^2 + (0.5 \times \text{MDL})^2} \tag{6}$$

In order to obtain reliable simulation results, the species are screened according to the data quality. In the calculation, PMF models can decrease uncertainty by operating a bootstrap from −1 to 1, and the conditions are continuously optimized until obtaining excellent results.

2.4.2. The Application of PMF

In previous studies, PMF was also used to identify the sources of pollutants in the atmosphere [107–110]. For example, Sowlat et al. collected the quantity and size of atmospheric particles in the range of 14–760 nm (mobility diameter) by using a scanning mobility particle sizer (SMPSTM, TSI Model 3081) and particles in the range of 0.3–10 μm (optical diameter) were measured using an optical particle sizer in the University of Southern California's (USC) park campus [111]. Then, PMF was applied to analyze the sources of organic compounds. Six sources, including nucleation, traffic 1, traffic 2, urban background aerosol, secondary aerosol, and soil/road dust, were determined during the sampling period.

However, only two studies have used PMF to determine the type of biomass burning. Bari et al. applied the PMF method to identify the sources of pollution from November 2005 to March 2006, and six source types were determined, including hardwood burning, softwood burning, light oil burning, traffic, road dust, and other, as shown in Table 2 [89]. The contrition of each source was 43.1%, 5.8%, 13.4%, 7.2%, 8.2%, and 21.5%, respectively. PMF was also used to study the type of biomass burning in Nanjing aerosols in the year 2016. In this study, the contribution of hardwood burning, softwood burning, and grass burning to the total biomass burning in four seasons was calculated, and factor profiles, time-series variation of contribution, and contribution of each type of biomass burning in four seasons are given in Figures 8 and 9 [51]. PMF can not only identify the type of biomass burning but also calculate the contribution of each burning source in a time series. The disadvantage is that the quantity of data required is large; at least 90 samples should be analyzed, and the more data, the more accurate it is.

Table 2. Organic compound mass fractions (%) of the six identified source factor mass concentrations and contribution (%) of each source to the mass concentration of the determined organic compounds in residential areas [89].

Compounds	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6
Anthracene (Ant)	0.03	1.02	0.16	0.52	0.34	0.00
Retene (Ret)	1.27	2.02	0.00	0.85	0.32	0.30
Fluoranthene (Flut)	0.80	0.18	10.61	4.80	0.61	0.00
Pyrene (Py)	0.75	3.03	8.89	1.51	3.94	0.23
Chrysene (Chr)	0.37	1.30	2.90	1.77	3.35	0.95
Benzo[a]anthracene (BaA)	0.66	0.31	3.42	2.86	4.17	1.39
Benzo[k + b]fluoranthene (B[k + b]F)	0.54	1.82	2.11	1.33	3.88	1.60
Benzo[a]pyrene (BaP)	0.21	1.63	2.77	0.88	3.64	1.29
Benzo[e]pyrene (BeP)	0.42	1.21	1.94	1.44	3.07	0.98

Table 2. *Cont.*

| Compounds	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6
Perylene (Prl)	0.92	1.29	0.23	1.56	0.00	0.00
Dibenzo[*a,h*]anthracene (DBahA)	0.05	0.80	0.00	1.17	0.61	0.01
Indeno[1,2,3-*cd*]pyrene (IP)	0.32	1.53	0.62	1.05	2.40	1.36
Benzo[*ghi*]perylene (BghiP)	0.19	1.78	1.11	2.80	2.84	1.83
Syringaldehyde	18.84	1.44	0.00	0.00	0.00	6.53
Acetosyringone	52.54	1.46	0.00	0.00	0.00	29.92
Propionylsyringol	9.87	1.19	0.00	0.57	0.00	6.73
Sinapylaldehyde	21.98	8.34	0.00	0.00	0.00	1.35
Vanillin	0.19	1.57	0.00	0.00	0.00	1.28
Acetovanillone	1.13	1.72	0.00	0.00	0.00	0.35
Guaiacylacetone	5.27	5.73	0.00	0.00	0.00	1.66
Coniferylaldehyde	1.15	2.65	0.00	0.00	0.05	0.66
Levoglucosan	34.80	51.41	0.00	0.00	0.00	3.76
Dehydroabietic acid	6.29	29.65	0.00	0.00	0.00	0.93
Biomass burning types	Hardwood	Softwood	Light oil	Traffic	Road dust	Others
Contribution (%)	43.1	5.8	13.4	7.2	8.2	21.5
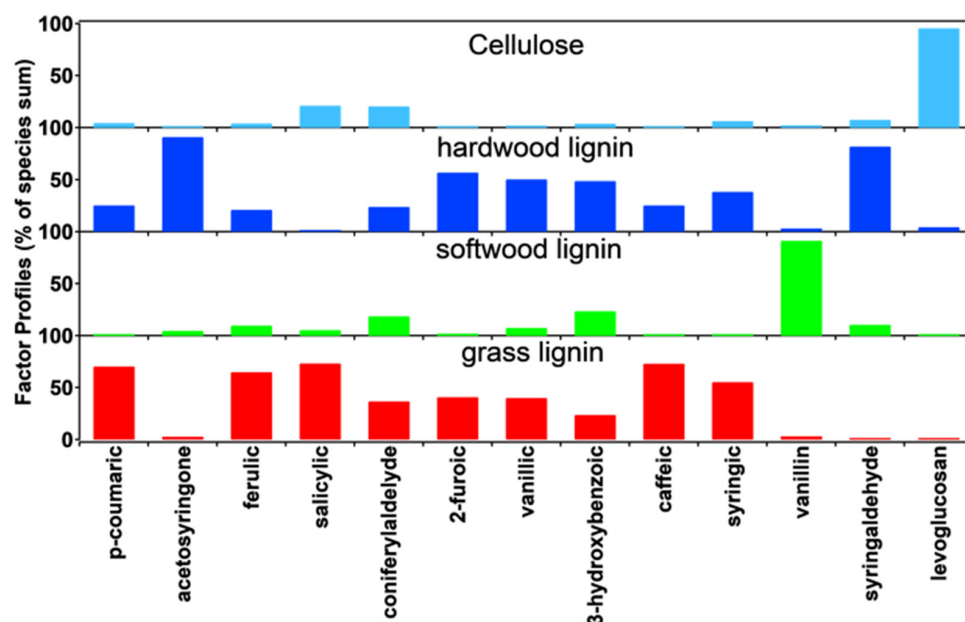
Figure 8. Profiles of type of biomass burning in Nanjing aerosols in 2016 [51].
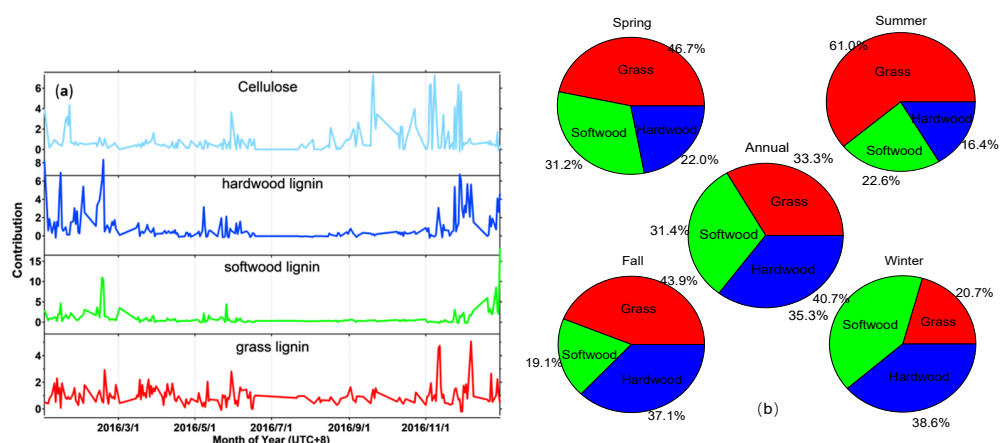

Figure 9. Changes of contribution of different types of biomass burning on days (**a**) and in four seasons (**b**) [51].

3. Summary and Perspectives

In this review, we introduced various types of biomass burning, including Asian crop residuals, wildfire, peatland, leaf, forest, grass, herbaceous plants, hardwood, softwood, wheat, oil, and lignite, as well as the main components released. First, different tracers of biomass burning were introduced. Levoglucosan is a common tracer of biomass burning in the atmosphere. Methoxyphenols such as p-hydroxybenzoic acid, vanillic acid, isovanillic acid, syringic acid, p-coumaric acid, ferulic acid, and homovanillic acid are unique trace molecular markers of biomass burning in atmospheric aerosols. 1,6-anhydro-2-acetamido-2-deoxyglucose is regarded as a specific marker for chitin in biomass burning emissions; wax esters are regarded as unique tracers for this source. Nicotine is a tracer of tobacco burning in aerosol samples.

Second, comprehensive methods to infer sources of biomass burning were illustrated. Single tracers cannot readily identify the source of biomass burning because hardwood and softwood can release the same tracers. However, the relative proportion of tracers is different in various plant species. To distinguish the types of biomass burning, five proportion methods have been adopted in this field, levoglucosan to K^+, levoglucosan to mannosan, levoglucosan to OC, levoglucosan to EC, vanillic acid to syringic acid, and vanillic acid to 4-hydroxybenzoic acid. Other simple methods introduced include K^+/EC, char-EC/soot-EC, and characteristic ratios of PAHs.

Finally, this paper summarizes three other methods, including source tracer ratio, principal component analysis, and the positive matrix fraction method. Both advantages and disadvantages are shown in this review, as well as the limitations of the ratio method to identify sources. Therefore, in the future, PCA, STR, and PMF, combined with the ratio method, will play a dominant role in the identification of biomass burning sources.

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