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Saccharides in atmospheric particulate and sedimentary organic matter: Status overview and future perspectives



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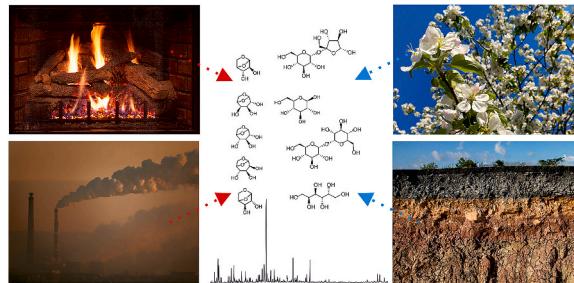
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HIGHLIGHTS

- Anhydrosaccharides are common biomass burning tracers, also formed from other sources, including coals.
- Mono- and disaccharides have multiple origins, but some are typical for specific sources.
- Fructose, sucrose, shikimic acid, D-pinitol, and quinic acid derive from pollen and plant debris.
- Xylose and arabinose are decomposition products from hemicellulose.
- Arabitol, mannitol, and trehalose are tracers from fungi.

GRAPHICAL ABSTRACT



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ABSTRACT

Saccharides are omnipresent compounds in terrestrial and marine ecosystems. Since the 2000s, their role in environmental and geochemical studies has significantly increased, but only anhydrosaccharides (mainly levoglucosan) have been reviewed. Here we present the wider knowledge about saccharides in organic matter of aerosols, bottom sediments, soils, dust, and sedimentary rocks. The main purpose here is to characterize the possible sources of saccharides, as well as sacharol formation, seasonal variability, and the possible applications in environmental and paleoenvironmental interpretations. Different saccharide sources were designated, including biomass burning, and particulate matter such as pollen, spores, lichen, and fungi, as well as polysaccharide decomposition as possible inputs of monosaccharides. The main focus was on the most common saccharides encountered in environmental samples and sedimentary rocks. These are the mono- and disaccharides glucose, fructose, sucrose, and trehalose, and sacharols arabitol and mannitol. The anhydrosaccharides levoglucosan, mannosan, and galactosan were evaluated as ancient wildfire indicators and industrialization tracers found in lacustrine sediments starting from Pleistocene to contemporary deposits. However, other anhydrosaccharides like xylosan and arabinosan were also found as products of fossil wood burning. These anhydrosaccharides have the potential to be further tracers of hemicellulose burning. Additional recommendations are proposed for future research, including environmental and paleoenvironmental topics that need to be addressed.

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1. Introduction

Saccharides (or carbohydrates, sugars) are ubiquitous constituents of all living organisms where they often constitute more than 50% of dry weight as significant components (Kamerling, 2007). Glucose is the dominant saccharide of metabolism with sucrose as the energy reserve in most biota. The identification of saccharides and their biological distributions have a long historical literature base. The assessments of the fate (diagenesis) and preservation of saccharides in the environment and geologic record were addressed early (Swain, 1969). Mono- di- and anhydro-saccharides and sacharols are important constituents of organic matter (OM) in ambient aerosols and soils (e.g., Simoneit et al., 2004a, 2004b, 2004c; Fu et al., 2012; Suciu and Masiello, 2019; Bhattacharai et al., 2019). Recently, their presence in sedimentary rocks has also been assessed in especially lignites and immature black shales (Marynowski et al., 2018, 2019). Although this broad group of compounds is commonly found in diverse living organisms, some are specific and can be used as organic tracers or biomarkers (Simoneit, 2002; Simoneit et al., 1999, 2004a, 2004b, 2004c, 2004a, 2004b, 2004c; Marynowski et al., 2019). Anhydrosaccharides, mainly levoglucosan, mannosan, and galactosan, are products of thermal decomposition of cellulose and hemicellulose and are frequently utilized as biomass burning tracers (e.g., Simoneit, 2002). Mannitol, arabitol, and trehalose are common in fungal spores (Bauer et al., 2008a; Marynowski et al., 2020a), while glucose, fructose, or sucrose have been interpreted as tracers for pollen (Yttri et al., 2007; Fu et al., 2012; Marynowski et al., 2020a). So far, there is no review summarizing the knowledge on the occurrence and sources of saccharides in aerosols, soils, and sedimentary rocks. Here we focus on the interpretation of anhydro-, mono- and disaccharide origins and demonstrate their possible utilization as organic tracers and biomarkers.

2. Saccharide determination for environmental studies

There are a plethora of sampling, extraction and analysis techniques for the characterization of individual or groups of compounds in the extractable OM of environmental samples (Jacobson et al., 2000; Suciu et al., 2019). Here we concentrate on the methods for saccharide determinations in especially atmospheric particulate matter (PM) and sediments. There is still a basic dichotomy in this research area. The quest for single compound identifications per sample (e.g., levoglucosan) versus multiple compounds (e.g., saccharides) has evolved into two approaches for extraction of analytes, namely water versus organic solvents (Suciu et al., 2019). There has been the additional misconception that saccharides and other polar compounds (e.g., oxalic or phosphoric acids) are insoluble in organic solvents, and therefore must be analyzed in aqueous extracts. This is not the case, organic solvent extraction has been in extensive use and dichloromethane/methanol (DCM/MeOH) was most effective for complete extraction of organic compounds including the polar molecules (e.g., saccharides; Abas et al., 2004a, 2004b; Simoneit et al., 2004a,b; Medeiros and Simoneit, 2007).

Furthermore, the detection methods and extensive workup procedures for water extracts have had low specificity and sensitivity, so new detection methods were developed (Suciu et al., 2019; Bhattacharai et al., 2019). However, specificity still must rely on molecular identification. Thus, DCM/MeOH (typically 1:1 or 2:1 v/v) extraction is recommended followed by derivatization with silylating and/or methylating reagents for gas chromatography-mass spectrometric analysis (Abas et al., 2004a, 2004b; Simoneit, 2006). However, some studies reported sequential extractions, first with DCM, and then with methanol, as more efficient than single extraction with the binary mixture (e.g., Alves et al., 2011). This allows the specific identifications of compounds comprising homologous lipid series, aromatic (i.e., PAH), polar (including saccharides), and synthetic molecules (e.g., Didyk et al., 2000; Simoneit, 2006; Simoneit et al., 2005). The detection sensitivity of specific trace components can also be enhanced by single ion monitoring (SIM) in

analytical series. There have been two interlaboratory intercomparisons of analytical methods for the determination of levoglucosan and saccharides (Yttri et al., 2015; Pietrogrande et al., 2017), and further such studies are needed to derive a standardized method.

The aversion by analysts to use silylation derivatives for GC-MS should also be allayed. Silylation methods of saccharides were established early for application in the biochemical, medical, food and agriculture sciences (e.g., Sweeley et al., 1963; Pierce, 1969), and are currently routine especially for microsamples (e.g., Simoneit et al., 2004c). Lastly, the complementary techniques like HP-LC-MS or HPAEC-PAD used for levoglucosan determinations were recently detailed (Wan and Yu, 2006; Yttri et al., 2015; Janoszka and Czaplicka, 2019; Bhattacharai et al., 2019; Suciu et al., 2019).

3. Saccharides in the atmospheric environment

Saccharides are common constituents of the OM in contemporary environmental compartments. For example here, in aerosols they are one of the most abundant groups of compounds in the soluble OM fraction. The following saccharide groups have been identified: anhydrosaccharides, mono- and disaccharides, and sacharols (e.g., Simoneit et al., 2004a,b; Yttri et al., 2007; Fu et al., 2008; Jia et al., 2010; Jia and Fraser, 2011; Wang et al., 2011). Trisaccharides, although they are also present in plants (Fischer and Shingleton, 2001), have only rarely been reported in aerosol samples (Wan and Yu, 2007), and this gap should be filled in the future with additional confirmations. Besides the soluble sugars in aerosols, the presence of complex saccharides such as cellulose, which is ubiquitous in the plant kingdom, has been reported (Kunit and Puxbaum, 1996; Puxbaum and Tenze-Kunit, 2003; Sánchez-Ochoa et al., 2007; for review see Alves, 2017). Polysaccharides as cellulose and probably also hemicellulose require further research to determine their specific sources and degradation rate in aerosols.

In the present review, we combined soluble saccharides found in snow with those reported from lake sediments because their common origin is associated with fires, and they were transported by air. The occurrence of saccharides in soils is described together with those in sedimentary rocks.

3.1. Anhydrosaccharides

Anhydrosaccharides (anhydromonosaccharides - AMs), including levoglucosan, mannosan and galactosan, as well as minor amounts of xylosan and arabinosan, are among the most common polar (water soluble) compounds present in aerosol particulate matter (PM) (Simoneit et al., 1999; Graham et al., 2002). In Fig. 1, we show the structures and mass spectra (as TMS) of the most common anhydrosaccharides detected in aerosols and sediments. They are readily identified by their GC retention time and MS key ions, in the elution order m/z 233 for arabinosan and xylosan, and m/z 217 for galactosan, mannosan, levoglucosan and 1,6-anhydroglucofuranose. Recently, levoglucosan has been the subject of review articles discussing its occurrence in the environment and its use as a biomass burning (BB) indicator (Vicente and Alves, 2018; Suciu et al., 2019; Bhattacharai et al., 2019). Here we focus on the origin, occurrence, and concentrations of AMs during various event episodes.

3.1.1. Anhydrosaccharide origin

Levoglucosan (1,6-anhydro- β -D-glucopyranose, LG) is a main product of the thermal decomposition (pyrolysis) of cellulose and hemicellulose, while mannosan (1,6-anhydro- β -D-mannopyranose, MA) and galactosan (1,6-anhydro- β -D-galactopyranose, GA) are pyrolytic degradation products of hemicelluloses (Simoneit et al., 1999; Nolte et al., 2001; Engling et al., 2006; Fabbri et al., 2008, 2009). All these compounds occur together in aerosol PM and are used as biomass burning tracers (Simoneit et al., 1999; Bhattacharai et al., 2019). However, relationships among the concentrations of LG, MA and GA differ

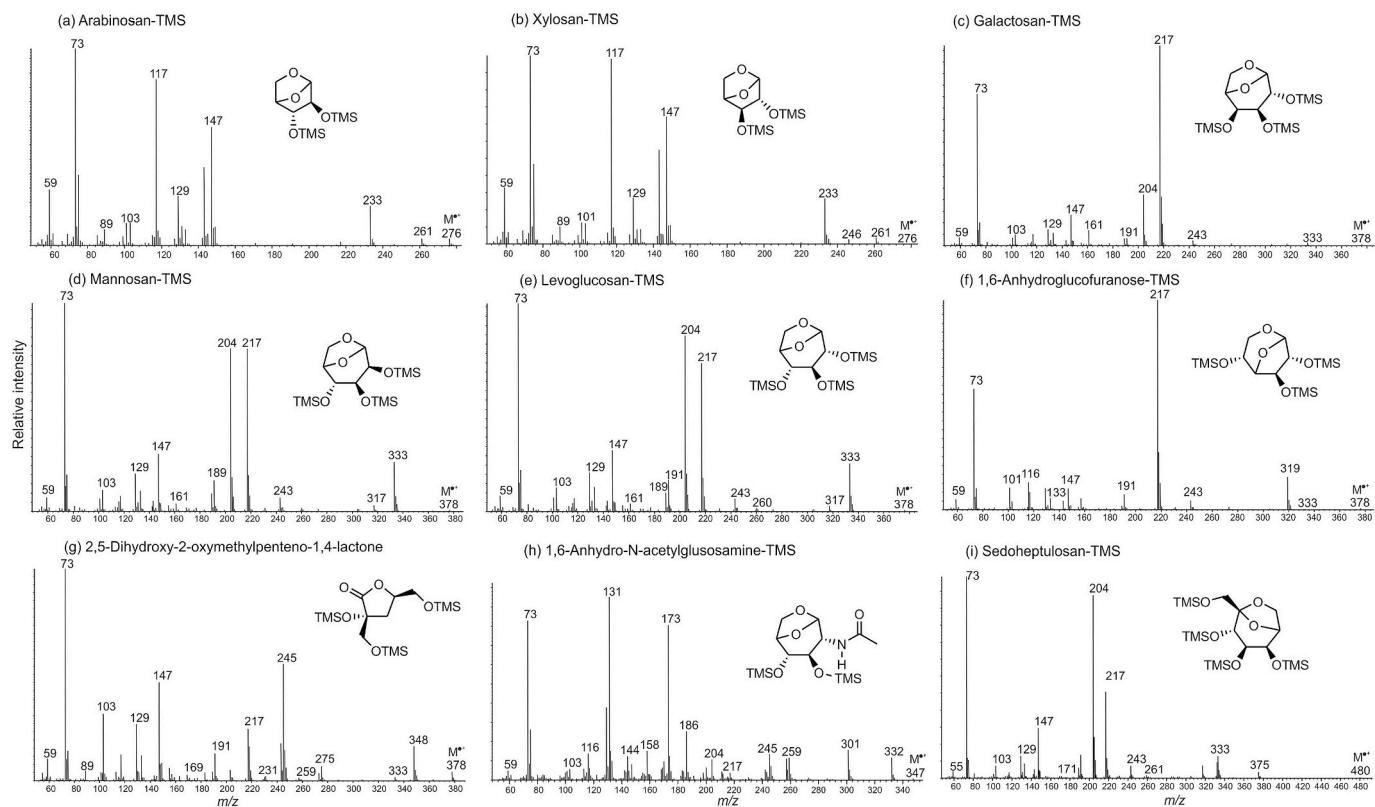


Fig. 1. Structures of the main anhydrosaccharides detected in aerosols, burn tests, and sediments, and their mass spectra (as TMS derivatives): (a) Arabinosan, (b) xylosan, (c) galactosan, (d) mannosan, (e) levoglucosan, (f) 1,6-anhydroglucofuranose, (g) 2,5-dihydroxy-2-oxymethylpenteno-1,4-lactone (a significant product from burning of sucrose), (h) 1,6-anhydro-N-acetylglucosamine (major product from burning of chitin), and (i) sedoheptulosen (used as internal standard).

depending on the type of biomass and possibly also other processes, including the burn temperature (Engling et al., 2006; Schmidl et al., 2008; Fabbri et al., 2009; Kuo et al., 2011a; Suciu et al., 2019) and subsequent degradation of AMs as a result of various, mainly free radical processes (Vicente and Alves, 2018; Bhattacharai et al., 2019 and references therein). It should be also mentioned that starch, the polysaccharide with β -glucosidic linkages to the C-4 hydroxyl of glucose, also produces high levels of LG under pyrolytic conditions (Fig. 2a).

The open burning and smoldering of plant biomass, organic waste or foods (grilling, baking, frying) are basically steam distillation/stripping processes, where the water content of the fuel is vaporized with dissolved and reformed organic products into the smoke. Char (i.e., black carbon) also forms and small particles, with adsorbed and occluded/trapped organic compounds, and inorganic ash particles are co-adveeted into smoke. There further adsorption/coagulation occurs upon cooling of the plume. DCM/MeOH extracts of PM dissolve all saccharides, including mainly LG in high yield (Medeiros and Simoneit, 2007), and upon extract concentration by blow down, which removes the DCM, the AMs (mainly LG) precipitate and crystallize. This indicates that AMs are steam stripped into smoke mainly as pure compounds and deplete during transport by washout due to their water solubility. The remnant AMs entrapped in the char are relatively unaltered as such, adsorbed to carbon or chemically bound (e.g., cellobiosan). It is this char/AM particulate matter that can be preserved in ice (from fallout) or sediments over significant time periods, whereas the free water-soluble AM compounds are degraded quickly (Bhattacharai et al., 2019 and references therein).

A discrimination diagram was presented based on the LG/MA and MA/GA ratios (Fig. 3; Xu et al., 2019). Such diagrams differentiated burn emissions from softwood/duff vs. grass/pine needles vs. hardwood based on the quantitative relations between cellulose and hemicellulose as well as the chemical character of hemicellulose in the particular

biomass. Differences in the LG/MA and MA/GA ratio values are usually seasonal in the ambient environment due to differences in the type of biomass combustion and variations connected with long-range aerosol transport (Fraser and Lakshmanan, 2000; Verma et al., 2015; Xu et al., 2019). However, these diagrams do not consider other less common and non-biomass sources of AMs (Fig. 3; see Wu et al., 2021).

For example, one such additional AM source is burning of lignite (and possibly other coals) (Fabbri et al., 2008, 2009; Yan et al., 2018; Rybicki et al., 2020a,b; He et al., 2021). The main AM compound formed during xylite combustion was LG, whereas MA and GA were low if present (Fabbri et al., 2009; Rybicki et al., 2020a). In the case of burning of detritic lignites, LG was usually a lesser component with no MA or GA (Rybicki et al., 2020a), but in samples rich in hemicellulose also xylosan and arabinosan were present besides MA and GA (Marynowski et al., 2021). The occurrence and concentration of AMs in burn emissions from different coal types are related to the holocellulose content, higher in xylites (up to 55%) and much lower in detritic coals (Marynowski et al., 2018). Recently, Rybicki et al. (2020b) showed differences between the stable carbon isotope ($\delta^{13}\text{C}$) values of LG generated from coal combustion and LG from burning of biomass (basket willow). This method can aid in the determination of LG sources.

There are numerous other potential point/fugitive sources of AMs, as for example in Asia from open burning of municipal and industrial waste (Bi et al., 2010; Christian et al., 2010; Giri et al., 2013), food cooking (He et al., 2004; Zhao et al., 2007, 2015, 2015; Vicente et al., 2021; Alves et al., 2021), fireworks burning (Cheng et al., 2014), and to a lesser extent smoldering incense (Tsai et al., 2010; Chuang et al., 2011). The general conclusion of Wu et al. (2021) was that although BB is still the main source of LG, non-BB sources can have a significant impact on regional LG budgets. An interesting AM emission source is burning/-roasting the biopolymer chitin, the exoskeleton of crustaceans, insects, and other fauna. The total extract of smoke PM from burning chitin had

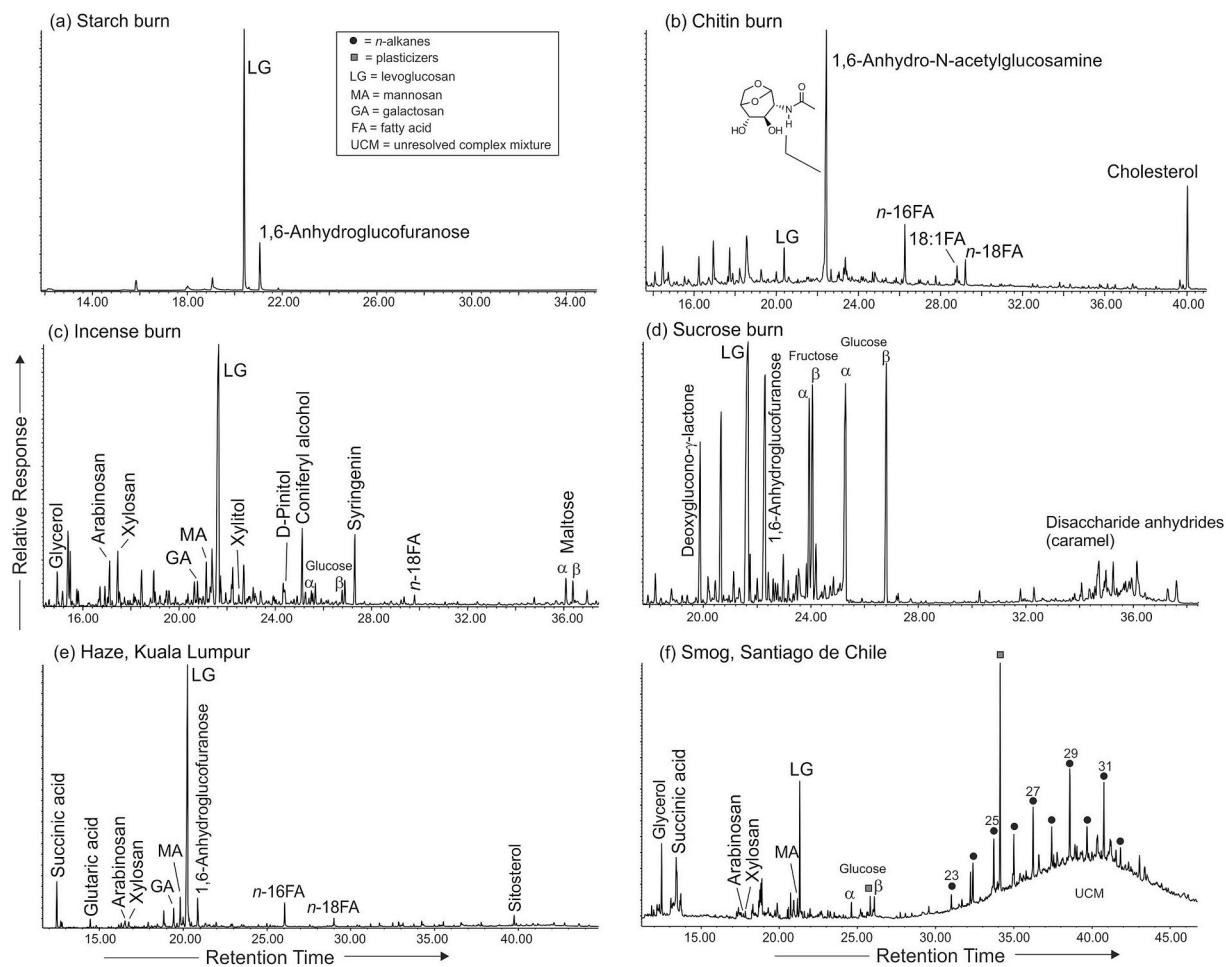


Fig. 2. Examples of total ion current (TIC) traces for GC-MS analyses of silylated total extracts of PM from burn tests and ambient air: (a) Potato starch, (b) shrimp exoskeleton (chitin, [Simoneit et al., 2000](#)), (c) Thai incense sticks, (d) sucrose, (e) haze, daytime Kuala Lumpur, Malaysia ([Abas et al., 2004b](#)), and (f) smog, Santiago, Chile ([Didyk et al., 2000](#)). The experimental procedures for these samples are described in the Supplemental Material section A.

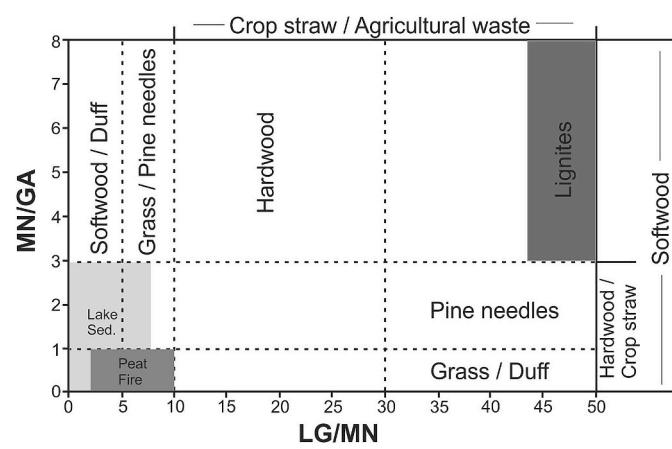


Fig. 3. The modified discrimination diagram, showing LG/MA and MA/GA ratios ([Xu et al., 2019](#)). Lignite and peat fire boxes were added. The marked boxes cover most of the data published, but exceptions are possible.

the dominant 1,6-anhydro-N-acetylglucosamine, with minor LG, fatty acids, cholesterol and 1,5-anhydro-3-acetamido-2-deoxyglucuronose ([Fig. 2b; Simoneit et al., 2000](#)). The 1,6-anhydro-N-acetylglucosamine can hydrolize to LG in the environment, thus adding to the total LG from this source. This emission should be considered as a LG input from urban restaurants/home cooking, mainly in Asia, but also in other regions.

Other emissions of AMs are wood pellet- and agro-fuels ([Alves et al., 2017](#)), whose importance in energy generation has recently increased. Although pellets generate lower concentrations of LG than other bio-fuels ([Alves et al., 2017](#)), it is difficult to estimate their significance in the global mass balance. We elaborate on the smoke PM composition from an example of incense from Thailand mentioned above. LG was the dominant compound in the total extract, with lesser coniferyl alcohol, syringenin, glycerol, mannosan, D-pinitol, glucose and maltose ([Fig. 2c; cf. Tsai et al., 2010](#)). Numerous minor peaks were phenolic derivatives from the lignin biopolymer burning/smoldering of the incense. Recently, [Alves et al. \(2020\)](#) reported that shredded tire chips and the wear particles forming from their interaction with pavement could release LG because cellulose is mixed with some rubber formulations during vulcanization. Such new fugitive and regional LG sources need estimation regionally and globally in future work.

3.1.2. Anhydrosaccharide occurrence, concentrations and applications

3.1.2.1. Anhydrosaccharides in ambient air. The occurrence and formation of LG in aerosols was recently reviewed ([Bhattarai et al., 2019; Suciu et al., 2019; Wu et al., 2021](#)). Here we focus on the extreme concentrations of LG and other AMs during smog and haze events and the seasonal AM concentrations determined for different parts of the globe.

Smog generally refers to urban fog of air contamination by emissions from traffic and industry, and haze is used to describe regional ultrafine

dust particles transported, for example, from deserts or wildfires. Whereas, smoke is the emission of fine carbonaceous particles close to the burn source. All these total suspended particles (TSP) as PM₁₀ or PM_{2.5} can cause respiratory problems and are distinguishable by their chemical compositions. Examples of the major compounds in TSP from haze in Malaysia and smog in Santiago, Chile are illustrated in Fig. 2 e and f. Haze from burning has dominant concentrations of AMs, and minor secondary homologous series (even/odd) of alkyl nitriles or amides derived from pyrolysis of OM rich in fatty acids and ammonia (Abas et al., 2004 a,b). Also, such haze PM contains the C₃₀ and C₃₁ moretane and hopane biomarkers typical of burning peat and duff biomass (Abas et al., 2004 b). The typical urban smog PM (e.g., Fig. 2f) has the predominant petroleum hydrocarbons (including alkanes, unresolved complex mixture of naphthalenes, PAHs and biomarkers), plasticizers, AMs, and other tracers from the multitude of emission sources (Didyk et al., 2000).

Table 1 lists the selected occurrences and concentrations of AMs in diverse global regions (data is from publications where mono- or disaccharides were also presented - see next sections). It is not surprising that during winter, the highest AM concentrations were detected in Asia and eastern Europe, which is consistent with the pollution of these regions (WHO Report, 2021). However, in some areas, AMs are also abundant in summer, which can reflect ambient conditions at the locale of the sampling points, long-range transport, or extensive BB in the region during the warmer seasons (Table 1).

3.1.2.2. Anhydrosaccharides from ancient wildfires. Anhydrosaccharides were identified in Neogene sedimentary deposits (mainly lake sediments) and ice cores, and were primarily interpreted as indicators of wildfires in past times. Elias et al. (2001) described the co-occurrence of LG and charcoal in a lake in southeastern Amazonia, Brazil, showing that major wildfires occurred in that region 0.7, 1.2, 5.0, and 7.0 kyr before present. The fire records from Lake Paru Co, Tibet, driven by climatic factors, were presented by Callegaro et al. (2018), and the LG concentration peaked during the early Holocene (10.7–7.5 kyr BP), with a second minor maximum during the middle Holocene (7.5–3.8 kyr). LG coupled with high-molecular-weight PAH concentrations reflected the wildfire intensity and character on the Tibetan plateau during the Holocene (Callegaro et al., 2018). Other studies reported wildfires and climate change during the middle to late Holocene using AM levels from different regions, including Guatemala (Schüpbach et al., 2015), East Africa (Battistel et al., 2017), New Zealand (Argiriadis et al., 2018), NW Africa (Schreuder et al., 2019a), Eastern Australia (Schreuder et al., 2019b), and Greece (Norström et al., 2021). The identification of AMs in young sediments (late Holocene) indicated the occurrence of both wildfires and human activity at different times and urban development in regions such as Puget Sound, WA (Kuo et al., 2011b), or the industrialization record in Lake Czechowskie, Poland (Dietze et al., 2019). In both cases, the main stages of modernization were defined over the past 250 years in the study areas.

Evidence of low-intensity BB was documented for much older sediments from Lake El'gygytgyn (far northeast of Russia), displaying 430 kyr of glacial-to-interglacial distribution changes of anhydrosaccharides (Dietze et al., 2020). AM influxes increased significantly during interglacials, when summer green boreal forests spread closer to the lake, and decreasing when tundra-steppe environments spread. Of special interest was the high concentrations of MA and GA compared to LG, with LG/MA ratios lower than 3 (but for most samples <1) and LG/(MA + GA) < 1 (most <0.5). These values are relatively low and matched with those reported from diverse BB (e.g. Nolte et al., 2001; Engling et al., 2006; Schmidl et al., 2008; Fabbri et al., 2009; Alves et al., 2010). AM ratios from Lake El'gygytgyn were also more than two times lower than those for other lake systems (Kirchgeorg et al., 2014; Schüpbach et al., 2015; Callegaro et al., 2018; Dietze et al., 2019), but data from Lake Allom, Fraser Island, Australia showed quite similar AM ratios

(Schreuder et al., 2019b) to that from Lake El'gygytgyn. Relatively low AM ratios were recently reported for the Agios Flores fen in Peloponnes, Greece (Norström et al., 2021). Two possibilities for high concentrations of MA and GA in relation to LG can be considered. The main possibility is the burning of specific types of biomass (Dietze et al., 2020). For example, burning of larch wood produces relatively high GA levels, as do moss-lichen mats common in boreal forests. In such cases only low-temperature surface wildfires can generate such AM proportions. Second, thermal influences can be excluded in such young sediments, but based on the complete aqueous solubility of the AM isomers, redox conditions and early-diagenetic oxidation/(bio)degradation should be considered.

Levoglucosan was also identified in Mesozoic and Cenozoic fossil wood, although at low concentrations. This compound was found in Bajocian-Bathonian mineralized wood fragments deposited in the Polish part of the Mid-European Epicontinental Basin (Marynowski et al., 2018), which is consistent with the detection of wildfires in the middle Jurassic clays from central Poland (Zakrzewski et al., 2020). Eocene fossil wood fragments from Eastern Poland also had traces of LG (Marynowski, unpublished data). No MA or GA were detectable in these samples.

LG has been reported in ice cores and snow, documenting historical wildfires and human-related BB, and was recently reviewed (You and Xu, 2018). Thus, we focus here only on some selected reports. The first results of AMs in ice were presented by Gambaro et al. (2008) for the Antarctic, where picogram quantities of LG were found and connected with long-range BB transport. The changes in LG concentrations over the past 300 years were reported for ice cores from the Kamchatka Peninsula, Northeast Asia (Kawamura et al., 2012). They showed a correlation between the occurrence of LG and the ambient temperatures at high northern latitudes, and interpreted the LG source from forest fires. Most studies have been conducted on the Tibetan Plateau (You and Xu, 2018, and references therein), where the highest concentrations of LG in ice and snow were found. This was associated with partial melting of snow thus concentrating LG in accumulation areas, as well as the relative proximities of BB-producing countries. In contrast, no ice melting was observed over the accumulation areas on Greenland and Antarctica (Zennaro et al., 2014) due to much lower annual temperatures. Hence, the detection of LG in Arctic or Antarctic ice cores documents long-range transport of aerosol PM and confirms forest fires in the past. The previous data support the utility of AMs as wildfire indicators in ancient environments. AMs, if present, seem to be more adequate wildfire tracers than for example polycyclic aromatic hydrocarbons (PAHs) because they directly confirm BB, while the origins of PAHs can be diverse (e.g., Simoneit and Fetzer, 1996; Grice et al., 2007). One exception is emissions from peat fires which also generate high concentrations of LG (Fujii et al., 2015; George et al., 2016; Popovicheva et al., 2019). However, linking AM concentrations to fire intensity can in some cases be misleading because the meteorological conditions and, above all, the prevailing atmospheric circulation, topography and transport range affect the compound concentrations (e.g., You and Xu, 2018).

3.2. Mono- and disaccharides

Saccharides are organic compounds of biological origins present in all eukaryotic and prokaryotic organisms (Pigman and Horton, 1980; Kamerling, 2007), and are common in the environment (e.g., Rogge et al., 2007; Jia et al., 2008). In the last decade, the number of aerosol studies in which saccharides were used as tracers increased significantly, hence the need for an overview summarizing the results of the research to date.

3.2.1. The origin of saccharides in aerosols

Although water-soluble organic compounds in atmospheric PM were studied extensively for many years (see review paper of Saxena and

Table 1

Average concentrations of selected saccharides and sphaerols in aerosol PM during the winter and summer seasons.

Location	Sample	Time period	Compounds [average concentrations in ng/m ³]							Reference
			Levoglucosan	Mannosan	Galactosan	Arabitol	Mannitol	Glucose	Trehalose	
Winter										
Chichi-Jima Island, Japan (WN Pacific, warm climate)	TSP	Dec.–Feb. 1990–1993	1.35	0.18	0.054	0.70	0.92	1.99	0.47	Chen et al. (2013)
Ghent, Belgium	PM ₁₀	2000–2001 Winter	420	61.0	25.0	26.0	26.0	73.0	–	Pashynska et al. (2002)
Jeju Island, S Korea	PM _{2.5}	Winter 2001	62	–	–	4.70	3.4	7.3	3.9	Fu et al. (2012)
Sapporo, Japan	TSP	Jan. 10–12, 2002	56.0	15.0	2.00	–	–	1.20	0.10	Simoneit et al. (2004a,b)
Elverum, Norway (suburban)	PM _{2.5}	Jan. 30–Mar. 15, 2002	–	–	–	4.30	2.80	20.0	9.80	Yttri et al. (2007)
Elverum, Norway (suburban)	PM ₁₀	Jan. 30–Mar. 15, 2002	605	167	4.0	5.30	4.20	22.0	13.0	Yttri et al. (2007)
Guangzhou, China	PM _{2.5}	Dec.–Feb. 2003	176	46.8	19.5	7.10 ^a	–	65.0	18.0	Ma et al. (2009)
Beijing, China (urban)	PM ₁₀	Winter 2003–2004	830	11.0	5.00	–	6.00 ^a	33.0	–	Zhou et al. (2009)
Beijing, China (suburban)	PM ₁₀	Winter 2003–2004	1014	18.0	10.0	–	–	40.0	7.00	Zhou et al. (2009)
Nanjing, China	PM _{2.5}	Jan. 2005	238	–	–	6.88	4.10	14.0	3.32	Wang and Kawamura (2005)
Hong Kong (suburban)	PM _{2.5}	Winter 2005	190	–	–	6.20 ^a	6.0	4.0	0.2	Wan and Yu (2007) ^c
Chichi-Jima Island, Japan (WN Pacific, warm climate)	TSP	Dec.–Feb. 2006–2009	1.25	0.21	0.055	2.11	2.52	2.03	1.09	Chen et al. (2013)
Seiffen, Saxony, Germany	PM ₁₀	Winter 2007	229	52.0	28.0	3.80	57.0	8.70	–	Iinuma et al. (2009) ^b
Chennai, tropical India	PM ₁₀	Jan. 23 – Feb. 6, 2007	112	13.4	8.07	1.05	0.54	3.86	1.22	Fu et al. (2010)
Baoji, central China	PM ₁₀	Winter 2008	901	54.0	96.0	17.0	5.00	55.0	7.90	Xie et al. (2010)
Bay of Bengal	PM _{2.5}	Dec. 27, 2008–Jan. 26, 2009	6.18	0.31	0.24	0.29	0.19	2.37	0.14	Bilkina et al. (2019)
Bologna, Italy (urban)	PM _{2.5}	Jan.–Feb. 2009	254	–	–	–	–	5.7	3	Pietrogrande and Bacco (2011)
Cork, Ireland	PM _{2.5}	Feb. 2 – Feb. 22, 2009	298	63.1	23.1	–	–	–	–	Kourtchev et al. (2011)
Rehovot, Israel	PM _{2.5}	Winter 2009	–	–	–	8.43	21.94	–	–	Burshtein et al. (2011)
Brno, Czech Rep.	PM _{2.5}	Winter 2010	578	99.2	37.8	2.70	1.66	5.34	2.47	Mikuška et al. (2017)
Ya'an, SE China	PM _{2.5}	Jun. 14–22, 2010	93.3	7.40	4.10	12.4	68.0	62.1	–	Li et al. (2013)
Ya'an, SE China	TSP	Jun. 14–22, 2010	131	9.40	5.80	34.6	188	227	–	Li et al. (2013)
Brno, Czech Rep.	PM _{2.5}	Winter 2011	806	113	44.4	3.15	1.60	6.54	2.27	Mikuška et al. (2017)
Beijing, China (urban)	PM _{2.5}	Dec. 1 – Jan. 30, 2012	590	60.0	–	–	–	–	–	Cheng et al. (2013) ^b
Shanghai, China	PM _{2.5}	Winter 2012	161	23	–	6.60	3.70	–	–	Li et al. (2016) ^b
Huaniao Island, China	PM _{2.5}	Winter 2012	43.0	8.20	–	1.50	1.00	–	–	Li et al. (2016) ^b
Ostrava, Czech Rep. (smog episode)	PM _{2.5}	Jan. 26 – Feb. 21, 2012	1471	248	80.6	1.44	0.87	3.95	0.57	Mikuška et al. (2015)
Ostrava, Czech Rep. (after smog episode)	PM _{2.5}	Jan. 26 – Feb. 21, 2012	613	111	28.9	0.91	0.63	1.98	0.30	Mikuška et al. (2015)
Dehli, India	PM _{2.5}	March 06, 2012	97.5	10.3	6.71	1.20	5.96	8.20	2.14	Kumar et al. (2017)
Bologna, Italy (urban)	PM _{2.5}	Winter 2013	259.4	63.6	31.6	–	–	5.6	–	Pietrogrande et al. (2014)
San Pietro near Bologna, Italy	PM _{2.5}	Winter 2013	252.9	61.5	32.0	–	–	5.4	–	Pietrogrande et al. (2014)
Kathmandu Valley, Nepal	TSP	Winter 2013	1391	116	85.6	26.1	18.1	39.9	20.1	Wan et al. (2019)
Shanghai, China	PM _{2.5}	Winter 2013–2014	392	127	–	7.30	36.8	12.7	–	Xiao et al. (2018)
Beijing, China	PM _{2.5}	Winter 2013–2014	189	39.2	25.6	3.34	4.38	16.0	7.45	Kang et al. (2018)
Barcelona, Spain (near cement plant)	PM _{2.5}	Nov. Dec. 2014	350	39.3	43.8	–	9.00	18.1	–	Sánchez-Sobrón et al. (2016)
Barcelona, Spain (near cement plant)	PM ₁₀	Nov. Dec. 2014	519	54.3	61.2	–	52.6	117	–	Sánchez-Sobrón et al. (2016)
Faisalabad, Pakistan	PM ₁₀	Winter 2015–2016	2760	100	70	90	80	270	120	Alvi et al. (2020) ^b
Chiang Mai, Thailand (smoke period)	PM _{2.5}	Winter – Spring 2016	1222	65.0	14.0	30.0	12.0	32.0	–	Thepnuan et al. (2019) ^b
Xi'an, N China	PM _{2.5}	Nov. 29 – Jan. 10, 2016	268.5	15.3	18.0	8.90	15.1	42.0	11.0	Wang et al. (2018)
Beijing, China	PM _{2.5}	Dec. 2017–Jan. 2018	559	72.9	46.3	14.1	21.2	86.4	61.1	Xu et al. (2020)
Sosnowiec, Poland	PM ₁₀	Winter 2017–2018 [4 m above ground]	1418	262	27.6	14.3	12.4	63.2	8.67	Marynowski et al. (2020a)

(continued on next page)

Table 1 (continued)

Location	Sample	Time period	Compounds [average concentrations in ng/m ³]							Reference
			Levoglucosan	Mannosan	Galactosan	Arabitol	Mannitol	Glucose	Trehalose	
Sosnowiec, Poland	PM ₁₀	Winter 2017–2018 [100 m above ground]	1009	174	19.4	10.0	7.73	46.7	6.17	Marynowski et al. (2020a)
Summer										
Chichi-Jima Island, Japan (WN Pacific, warm climate)	TSP	Jun.–Aug. 1990–1993	0.37	0.054	0.012	3.08	4.00	5.60	1.20	Chen et al. (2013)
Jeju Island, S Korea	PM _{2.5}	Summer 2001	8.0	–	–	9.6	11.0	9.6	3.3	Fu et al. (2012)
Ghent, Belgium	PM ₁₀	Summer 2001	19.1	3.00	1.02	105	97.0	270	–	Pashynska et al. (2002)
Sapporo, Japan	TSP	Jun. 28 – Jul. 5, 2001	6.40	0.20	–	18.0 ^a	–	33.5	7.70	Simoneit et al. (2004a,b)
Uryu Forest, Hokkaido, Japan	TSP	Aug. 13–15, 2001	7.30	1.00	0.30	16.0	13.0	16.0	3.70	Fu and Kawamura (2011)
Elverum, Norway (suburban)	PM _{2.5}	22 May – Jun. 28, 2002	–	–	–	2.00	2.00	5.20	4.10	Yttri et al. (2007)
Elverum, Norway (suburban)	PM ₁₀	22 May – Jun. 28, 2002	47.0	10.0	3.00	20.0	18.0	19.0	30.0	Yttri et al. (2007)
Howland Forest, Maine, USA	TSP	Jul. 9–23 2002	54.0	7.60	1.10	5.00	5.80	45.7	9.80	Medeiros et al. (2006)
Howland Forest, Maine, USA	TSP	Jul. 23 – Aug. 6, 2002	5.00	–	–	3.50	4.60	13.8	7.20	Medeiros et al. (2006)
K-puszta, Hungary	PM _{2.5}	Summer 2003	12.3	–	–	4.80	5.30	–	–	Ion et al. (2005)
Moitinhos, Portugal (intensive wildfires)	PM _{2.5}	Jul. 31 – Aug. 7, 2003	58	17.6	–	5.20	7.82	5.59	–	Pio et al. (2008)
Guangzhou, China	PM _{2.5}	Jun.–Aug. 2003	82.7	9.4	2.4	4.80 ^a	–	71.8	67.8	Ma et al. (2009)
Beijing, China (urban)	PM ₁₀	Summer 2003–2004	4980	442	306	–	630 ^a	2460	910	Zhou et al. (2009)
Beijing, China (suburban)	PM ₁₀	Summer 2003–2004	3088	164	123	–	380	3010	2191	Zhou et al. (2009)
Nanjing, China	PM _{2.5}	Jul. 2004	73.3	–	–	3.96	2.01	16.9	4.18	Wang and Kawamura (2005)
Hong Kong (suburban)	PM _{2.5}	Summer 2005	35	–	–	5.5 ^a	3.5	2.7	–	Wan and Yu (2007) ^c
Mt. Tai, Shandong, China	TSP	May 28–Jun. 28 2006	391	9.71	8.42	52.5	56.4	49.5	24.5	Fu et al. (2008)
Chichi-Jima Island, Japan (WN Pacific, warm climate)	TSP	Jun.–Aug. 2006–2009	0.22	0.03	0.009	11.4	19.4	8.47	5.93	Chen et al. (2013)
Chennai, tropical India	PM ₁₀	May 23–31, 2007	111	9.90	5.84	1.51	2.20	6.82	1.00	Fu et al. (2010)
Bologna, Italy (mixed area)	PM _{2.5}	Jun–Jul. 2008	1950	–	–	–	–	4.1	–	Pietrogrande and Bacco (2011)
Cork, Ireland	PM _{2.5}	Jul. 28 – Aug. 28, 2008	9.3	1.7	0.5	0.9	1.6	–	–	Kourtchev et al. (2011)
Arctic Ocean, MALINA cruise	TSP	Aug. 3–25, 2009	0.37	0.11	0.043	4.50	9.20	6.40	1.40	Fu et al. (2013)
Rehovot, Israel	PM _{2.5}	Summer 2009	–	–	–	10.59	15.07	–	–	Burshtein et al. (2011)
Hyytiälä, Finland (pine forest)	PM ₁₀	5 Aug.–2 Sept. 2009	4.6	0.81	0.18	24.0	30.0	–	13.0	Yttri et al. (2011)
Birkenes, Norway (boreal forest)	PM ₁₀	5 Aug.–2 Sept. 2009	2.1	0.39	0.06	16.0	16.0	–	5.80	Yttri et al. (2011)
Vavihill, Sweden (semi-rural)	PM ₁₀	5 Aug.–2 Sept. 2009	5.6	0.71	0.24	10.0	13.0	–	6.40	Yttri et al. (2011)
Lille Valby, Denmark (semi-rural)	PM ₁₀	5 Aug.–2 Sept. 2009	9.8	1.30	0.35	12.0	17.0	–	4.80	Yttri et al. (2011)
Ya'an, SE China	PM _{2.5}	Jun. 14–22, 2010	93.3	7.40	4.10	12.4	68.0	62.1	–	Li et al. (2013)
Ya'an, SE China	TSP	Jun. 14–22, 2010	131	9.40	5.80	34.6	188	227	–	Li et al. (2013)
Brno, Czech Rep.	PM _{2.5}	Summer 2011	79.3	13.0	3.58	7.71	7.35	11.4	3.92	Mikuška et al. (2017)
Beijing, China (urban)	PM _{2.5}	3 Jun. – Jul. 23, 2011	230	10	–	–	–	–	–	Cheng et al. (2013) ^b
Bologna, Italy (urban)	PM _{2.5}	Summer 2012	2.2	–	–	–	3.1	0.9	–	Pietrogrande et al. (2014)
San Pietro near Bologna	PM _{2.5}	Summer 2012	–	–	–	–	27.0	3.3	–	Pietrogrande et al. (2014)
Shanghai, China	PM _{2.5}	Summer 2012	28	12	–	6.90	21.0	–	–	Li et al. (2016) ^b
Huaniao Island, China	PM _{2.5}	Summer 2012	–	6.2	–	0.50	2.80	–	–	Li et al. (2016) ^b
Beijing, China	PM _{2.5}	Summer 2013–2014	12.4	1.41	0.72	0.46	1.04	2.84	1.09	Kang et al. (2018)
Shanghai, China	PM _{2.5}	Summer 2013–2014	46.5	9.70	–	14.9	172.7	14.9	–	Xiao et al. (2018)
Beijing (urban)	PM _{2.5}	Jun. 9–Jul. 8, 2014	55.5	3.54	2.51	–	13.0	21.0	–	Yan et al. (2019) ^b
Wangdu (rural)	PM _{2.5}	Jun. 9–Jul. 8, 2014	206	19.8	17.5	–	61.3	51.2	–	Yan et al. (2019) ^b
Xi'an, N China	PM _{2.5}	Jun. 2 – Aug. 20, 2015	100.4	5.6	6.3	10.6	15.5	60.5	21.7	Wang et al. (2018)

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Table 1 (continued)

Location	Sample	Time period	Compounds [average concentrations in ng/m ³]							Reference
			Levoglucosan	Mannosan	Galactosan	Arabitol	Mannitol	Glucose	Trehalose	
Mt. Emei, China	PM _{2.5}	Jun. 20 – Jul. 19, 2016	39.0	7.06	2.61	1.53	3.96	16.2	1.72	Zhao et al. (2020)
Faisalabad, Pakistan	PM ₁₀	Summer 2016	480	20	–	90	150	270	90	Alvi et al. (2020) ^b
Beijing, China	PM _{2.5}	June–July 2017	114	14.2	8.71	79.6	151	234	232	Xu et al. (2020)
Sosnowiec, Poland	PM ₁₀	Summer 2018 [4 m above ground]	189	24.5	1.84	106	97.0	132	55.4	Marynowski et al. (2020a)
Sosnowiec, Poland	PM ₁₀	Summer 2018 [100 m above ground]	123	17.9	2.39	89.0	106	98.6	39.1	Marynowski et al. (2020a)

^a Possibly arabitol was wrongly identified as xylitol and mannitol as sorbitol.

^b Saccharides were determined using high-performance anion-exchange chromatography with pulsed amperometric detection (HPAEC-PAD).

^c Saccharides were determined using liquid chromatography-mass spectrometry method (LC-MS). In all other cases the determination was by using GC-MS.

Hildemann, 1996), saccharides aroused the interest of scientists in the 2000s (Simoneit et al., 1999, 2004a,b; Graham et al., 2002). Mono- and disaccharides in aerosols were initially identified by Graham et al. (2002) in biomass burning aerosols of Amazonia (Rondônia, Brazil). Besides the most abundant LG, they identified 14 saccharides with a predominance of glucose, xylose, fructose, sucrose, and some sacharols. The AMs and some saccharides (e.g., xylose) were directly related to BB, while the sources of others (e.g., glucose, fructose, or trehalose) were proposed as diverse biogenic particles (Graham et al., 2002). Soil from that region of Amazonia had high levels of glucose, rhamnose, galactose, arabinose, sorbitol, xylitol, talose and trehalose, but low amounts of fructose and sucrose (Simoneit et al., 2004a). This supports the origin of the regional aerosol PM to be in part from biogenic matter (Graham et al., 2002). A detailed study of solvent extracts, including saccharides, from aerosols sampled in urban, rural, and marine locales, as well as natural and agricultural soils, was provided by Simoneit et al. (2004b). The mono- and disaccharides identified in the aerosol PM transported from Asia had a predominance of glucose and sucrose. Because the saccharides were present at relatively low concentrations during the early spring and sharply increased in the summer season, these authors linked their origin with soil, desert silt and unpaved road dust.

Later Yttri et al. (2007) and Jia et al. (2010) connected glucose, fructose and sucrose in aerosols with fragmented pollen grains and flower buds, and associated these compounds with the growing (vegetation) season. Although other sources (like spores, microbes or soil) of these particular saccharides were also reported (Graham et al., 2002; Simoneit et al., 2004a; Fu et al., 2012; Marynowski et al., 2020b), their highest concentrations in spring suggest vegetation as the important source in ambient air (Medeiros et al., 2006; Yttri et al., 2007; Jia et al., 2010; Zhu et al., 2015; Kang et al., 2018; Theodosi et al., 2018; Verma et al., 2018; Marynowski et al., 2020a; Oduber et al., 2021). The glucose and sucrose contents in the PM of the Sonoran Desert near Phoenix, USA, were much higher compared to the local soil, which correlated with input of primary plant and spore particles (Jia and Fraser, 2011). Furthermore, Marynowski et al. (2020a) showed statistically significant correlations between the annual concentration of glucose and fructose with meteorological elements such as the average air temperature, short-wave radiation balance, and duration of sunshine. The correlation between sucrose and the same meteorological factors was relatively low. That suggested glucose and fructose as the most robust candidates for tracers of pollen grains, but other sources like BB should still be considered for these saccharides (e.g., Pio et al., 2008; Sullivan et al., 2008; Scaramboni et al., 2015; Gonçalves et al., 2021).

In contrast to the mono- and disaccharides above, xylose showed a good correlation with typical BB tracers, such as AMs (Chen et al., 2013; Mikuška et al., 2015; Kang et al., 2018; Theodosi et al., 2018; Gonçalves et al., 2021). It was interpreted as a thermal degradation product of hemicellulose, where it is one of the primary monomers. Also, arabinose is a frequent component of hemicellulose and its origin in organic

aerosols was also associated with the combustion of biomass (Medeiros et al., 2006). Xylosan and arabinosan, the anhydrides of these monosaccharides, were reported as BB tracers with the other AMs in Amazon aerosol PM (Graham et al., 2002, 2003). The disaccharide maltose, occurring in the same aerosol PM, showed the best correlation with BB tracers (Medeiros et al., 2006; Fu et al., 2010).

Another disaccharide that has commonly been found in aerosols is trehalose (a.k.a. Mycose). Graham et al. (2002) identified trehalose in Amazon aerosols, and Simoneit et al. (2004b,c) found this disaccharide in Santiago de Chile, Sapporo, Japan, and Datong, China aerosol PM. Jia and Fraser (2011) reported enriched concentrations of trehalose in fungal spores in the Phoenix desert area. Later, Zhu et al. (2016) observed elevated concentrations of trehalose (together with mannitol and arabitol, see below) during night time in aerosol PM of a temperate coniferous forest in Wakayama, Japan. That concentration rise coincided with the nocturnal sporulation of fungi (see also Graham et al., 2002). Trehalose concentrations had a good correlation with temperature (Fig. 4) in a temperate climatic region and were statistically significant with the presence of discrete Alternaria, Cladosporium, Epicoccum, Ganoderma, and Periconia fungal spores (Marynowski et al., 2020a). Although trehalose is known to be present in some higher plants and bacteria (Müller et al., 1995; Argüelles, 2000), fungi and their spores appear to be its main source (Thevelein, 1984), especially because it exhibits the seasonally elevated concentrations associated with sporulation of fungi (Graham et al., 2002; Zhu et al., 2016; Marynowski et al., 2020a). Moreover, the fungal (and lichen; see Montiel, 2000) origin of trehalose explains its elevated concentrations in soil and road dust (Simoneit et al., 2004a; Rogge et al., 2007, 2012).

It is worth noting that saccharides can also be emitted from seawater to the marine boundary layer through bubble bursting processes from primary and heterotrophic bacterial productivity (Sempéré et al., 2008; Kawamura et al., 2017). Three major saccharide classes were detected in the Southeast Pacific, the aldohexoses (glucose, galactose, and mannose) as most abundant, deoxysaccharides (fucose and rhamnose), and aldopentoses (arabinose, xylose, and ribose) (Sempéré et al., 2008). Other studies carried out in the Equatorial Pacific and central Arctic Oceans indicated glucose (47–79%), fructose (15–16%), and arabinose (5–15%) as dominant free dissolved saccharides (Rich et al., 1996, 1997). On the contrary, Jayaratne et al. (2016) found the highest concentrations of mannose, besides glucose, fructose, and arabinose in sea spray aerosol and elevated concentrations of mannose, xylose, arabinose, and fructose in the sea surface microlayer.

There are other potentially significant points or fugitive sources of saccharides. Various compounds, including AMs, have been reported in aerosol PM (mainly indoor) from cooking/restaurant activities (He et al., 2004; Zhao et al., 2007; Alves et al., 2021), which needs further clarification. The mono- and especially disaccharides are not volatile organic compounds (VOC), and do not vaporize into smoke during burning. This is illustrated with the open burning test of sucrose

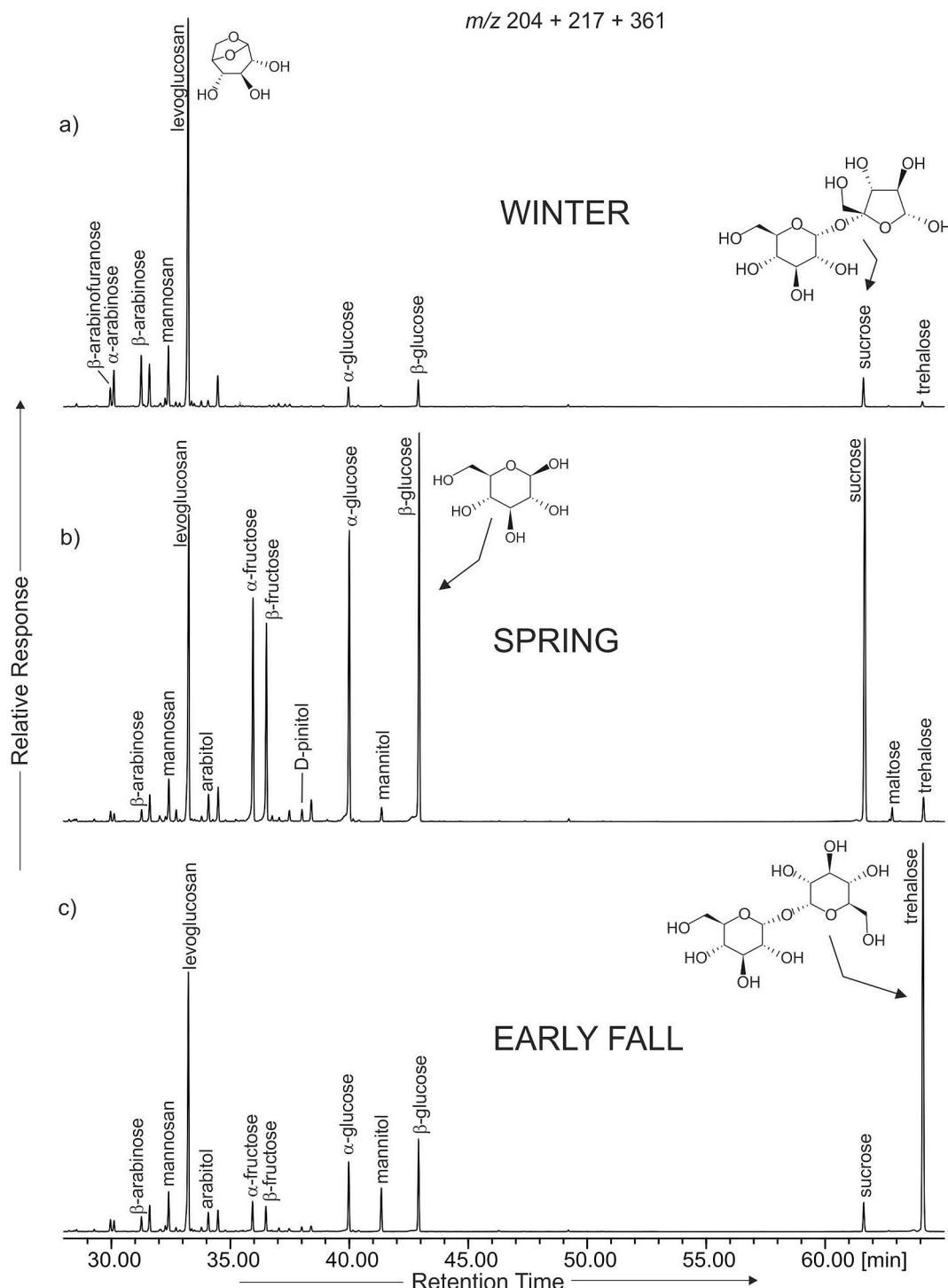


Fig. 4. Summed mass chromatogram ($m/z 204 + 217 + 361$) showing seasonal variation in saccharide distribution in a temperate climate (Sosnowiec city, Poland). Note the high concentration of LG in winter, fructose, glucose, and sucrose in spring, and trehalose, mannitol, and arabitol in early fall. Data are from Marynowski et al. (2020a).

(Fig. 2d), where sucrose is not detectable in the smoke PM and the dominant compounds are LG, glucose and fructose, with minor monosaccharide lactones and disaccharide dimers (caramel, Table SM-1). Thus, the monosaccharides present in the PM appear to be dissolved or entrained in steam microdrops advected into smoke, and with regard to cooking operations this steam stripping carries low levels of tracer compounds, including monosaccharides, into the plume.

3.3. Sacharols as tracers of aerosol PM

The most commonly identified sacharols (sugar alcohols, polyols) in ambient air PM are arabitol and mannitol (Bauer et al., 2002, 2008a; Graham et al., 2002; Simoneit et al., 2004a; Medeiros et al., 2007; Burshtein et al., 2011), but many others, like glycerol, threitol, erythritol, ribitol, xylitol, sorbitol, and inositol (including D-pinitol, myo- and scyllo-inositol) have also been reported for some regions (e.g., Medeiros and Simoneit, 2007, 2008; Kundu et al., 2010; Zangrando

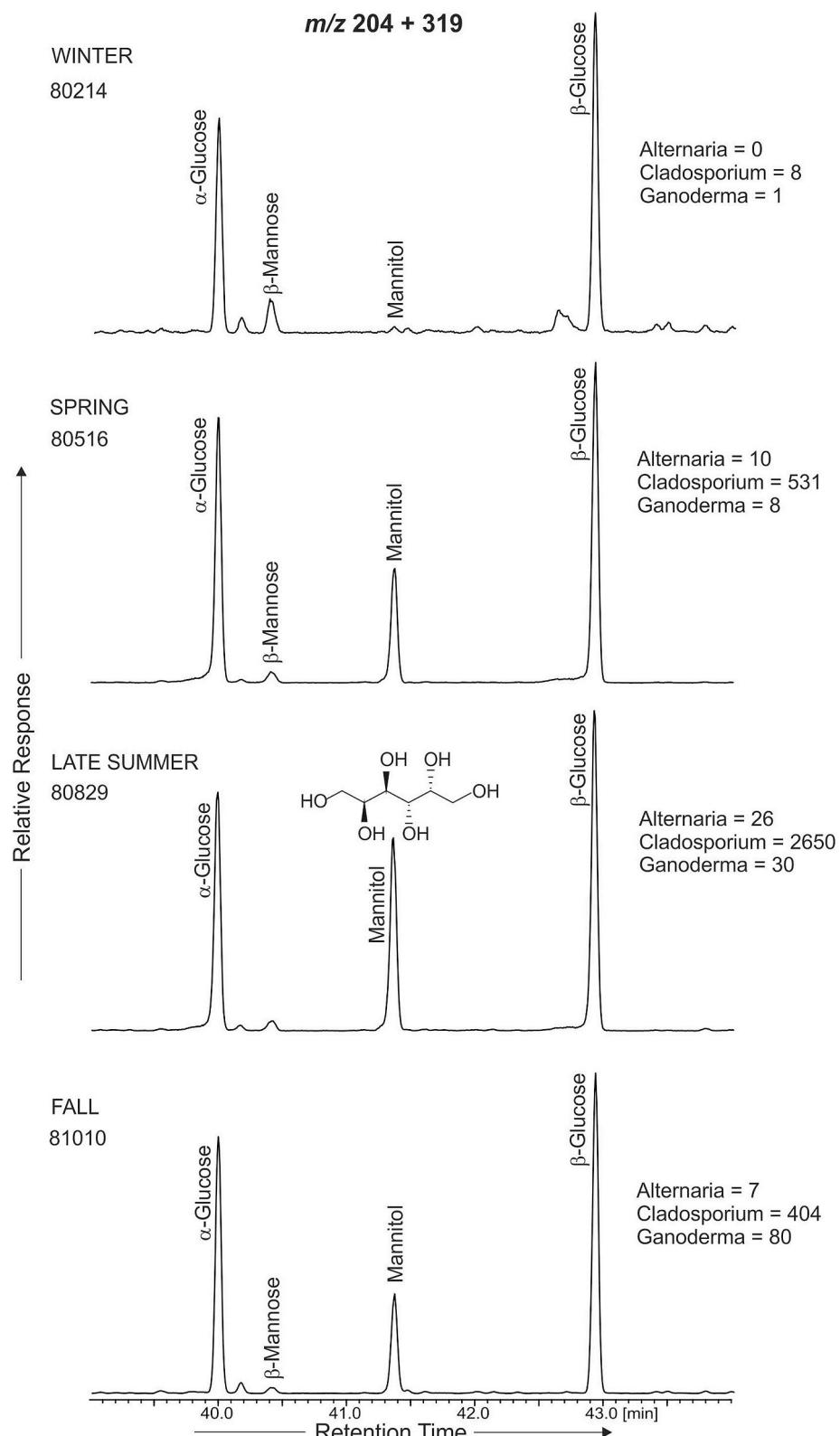


Fig. 5. Summed mass chromatogram (m/z 204 + 319) showing the distribution of glucose and mannitol in relation to seasonal variation and the total number of most common fungal spores. The abundance of mannitol is proportional to fungal spore counts. Data are from Marynowski et al. (2020a).

et al., 2016; Zheng et al., 2018). The origin of arabitol and mannitol from fungal spores showed a good correlation between spores and both sacharols ($R^2 > 0.7$; Bauer et al., 2008a). Later other studies also reported excellent correlations for both sacharols and spores suggesting their same source (Zhu et al., 2016; Zangrande et al., 2016; Verma et al.,

2018; Emygdio et al., 2018; Samaké et al., 2019a; Li et al., 2020; Xu et al., 2020; Marynowski et al., 2020a). A weaker correlation was explained by the varied composition of sacharols among different species of fungi (Fu et al., 2016 and references therein). Moreover, the peak concentration of these compounds occurs during summer and early fall

(Fu et al., 2010, 2012; Verma et al., 2018; Marynowski et al., 2020a). For example, a high concentration of arabitol and mannitol in aerosol PM coincided with fungal spores during late summer in a Nordic rural background environment (see Fig. 5; Yttri et al., 2011). The same was observed in various European cities (Pashynska et al., 2002; Zangrando et al., 2016; Samaké et al., 2019a; Marynowski et al., 2020a); Chichi-jima Island in the North Pacific (Verma et al., 2018); North Carolina, USA (Mullaugh et al., 2014); Texas, USA (Jia et al., 2010); Jeju Island, South Korea (Fu et al., 2012); Baoji City, China (Xie et al., 2010); or Beijing, China (Liang et al., 2016).

The highest average concentration of sacharols in Antarctica was measured during the summer (Deng et al., 2021). These authors interpreted their results as biological PM of terrestrial spores of fungi in ice-free soil ecosystem areas, but not excluding marine sources from the sea surface. All the data above support increased fungal activity, common during summer and fall. Moreover, both sacharols prevailed in coarse-grained PM fractions rich in fungal spores, in contrast to the BB tracers, indicating that humid conditions following rainfall events are favorable for increased concentrations of fungal spores, and hence, elevated arabitol and mannitol concentrations in ambient PM (Barbaro et al., 2015; Samaké et al., 2019a,b; Fetracco et al., 2020; Xu et al., 2020; Oduber et al., 2021). Also, the importance of air temperature and humidity has been emphasized in the variation of the sacharol concentrations (Samaké et al., 2019b). Although there are some exceptions, for example, Alternaria fungal spores bloom at elevated temperatures (23–27 °C) and low relative humidity (Oduber et al., 2021). Furthermore, sacharols correlate well ($p < 0.05$) with the abundances of the bacterial genera *Massilia* and *Pseudomonas* (Samaké et al., 2021).

In order to avoid future errors, the misassignment of sorbitol as a compound related to fungi (Li et al., 2019) should be mentioned. These authors based their assumption on the work of Shamrikova et al. (2014) and Bauer et al. (2008a,b), but neither of those papers mentioned a correlative link between sorbitol and fungi.

Although many reports confirm that arabitol and mannitol are dominant in fungi, these compounds are also found in many plants and microorganisms (Burshtein et al., 2011; Kordowska-Wiater, 2015; Patel and Williamson, 2016 and reference therein), and therefore their source may be diverse in specific cases (Fig. 6). These sacharols have been connected with wildfire and BB emissions, where for example both were detected during intense wildfires in Amazonia (Graham et al., 2002; Clays et al., 2010; Kundu et al., 2010) and in source tests (Medeiros and Simoneit, 2008). Yang et al. (2012) found elevated concentrations of arabitol and mannitol at Chengdu, China, in spring 2009, which correlated well with BB indicators like LG and K^+ . A good correlation between sacharols and LG was also reported for atmospheric PM from Portugal during the intense summer forest fire period of 2003 (Pio et al., 2008). Furthermore, here we show, based on multiple data from Table 1, a good correlation between the levoglucosan to arabitol and mannitol (LG/A + M) vs. levoglucosan to glucose (LG/GLU) ratios during winter, suggesting a biomass burning source of arabitol and mannitol (Fig. 6). On the contrary, no correlation was found during summer which indicates diverse sources of these saccharols and glucose in the summer season. Also, Li et al. (2016) suggested that BB is a crucial source of sacharols in fine aerosol PM over Shanghai, China. Large concentrations of arabitol and mannitol were generated during burn experiments of maize straw and wood branches, with lower levels from wheat straw (Sun et al.,

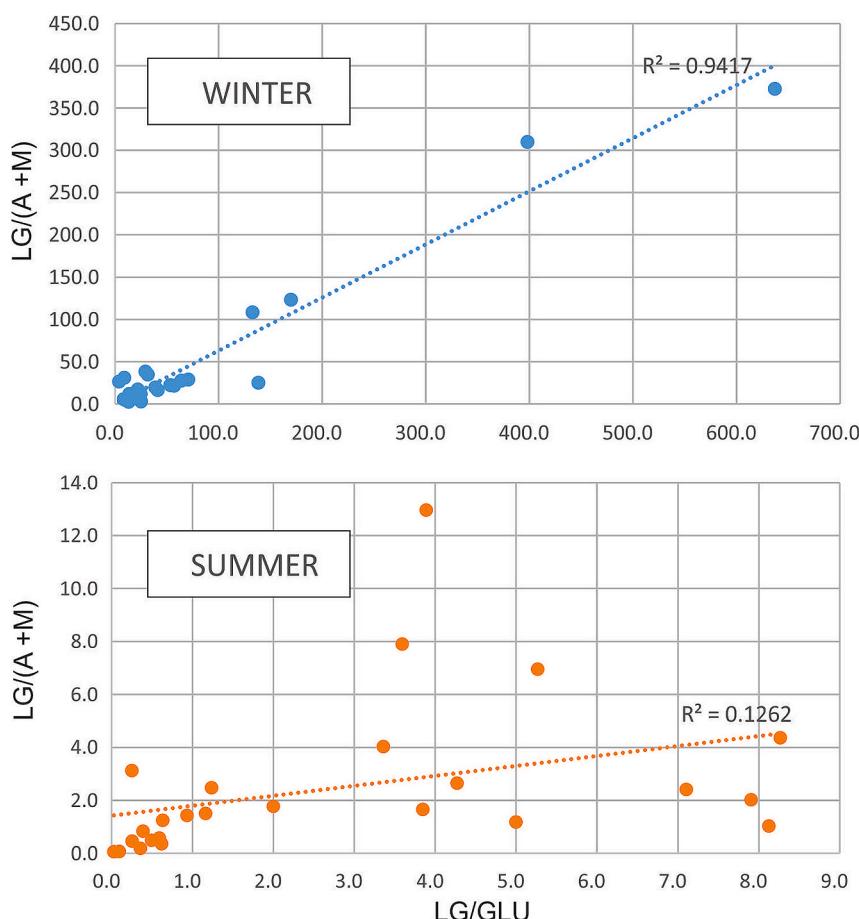


Fig. 6. Cross plots of levoglucosan to arabitol and mannitol (LG/A + M) vs. levoglucosan to glucose (LG/GLU) ratio, showing differences in correlation during winter and summer (based on data from Table 1). The good correlation during winter suggests the main sources of arabitol and mannitol are from biomass burning. Lack of correlation during summer suggests a diverse source of these compounds.

2019). However, these concentrations were ten to a hundred times lower than the LG concentrations. Besides, xylitol was found to be well correlated with anhydrosaccharides and interpreted to be a sacharol of a BB origin (Gonçalves et al., 2021).

Aerosol PM from long-lasting smoldering burns of boreal peatlands produced arabitol, mannitol, threitol and methyl-threitol, but at much lower concentrations than the AMs (Engling et al., 2014; Popovicheva et al., 2019), which may be related to the burning of fungal hyphae, typical in peats (Thormann, 2006). In addition, D-pinitol, *myo*-inositol, glycerol and xylitol have also been found in smoke from incense burning (e.g., Fig. 2c; Tsai et al., 2010). Little is known about the origin of these sacharols in aerosols and further data is needed. D-pinitol and *scylo*-*myo*-inositol are typical plant constituents, while *myo*-inositol is also common in yeast and fungi (Kersting et al., 2003; Vallurua and Van den Ende, 2011; Poongothai and Sripathi, 2013), and glycerol is the ubiquitous component of lipid fatty acid esters in all biota (e.g., Nolte et al., 1999).

4. Saccharides in soils and sedimentary rocks

4.1. Soil saccharides and their origin

Saccharides, including neutral saccharides, are frequent constituents of soils and dust (e.g., Amelung et al., 1996; Nierop et al., 2001; Kögel-Knabner, 2002; Simoneit et al., 2004a; Otto and Simpson, 2005; Medeiros et al., 2006; Rushdi et al., 2006, 2016; Rogge et al., 2007, 2012; Gunina and Kuzyakov, 2015). In soils, carbohydrate carbon can reach 20–30% of total carbon, but a significant part of that total is constituted of polysaccharides (Oades, 1993). Nevertheless, saccharides are an important component of the water-soluble organic compounds in soils and dust (Rogge et al., 2007, 2012, 2012; Marynowski et al., 2020b). Many neutral saccharides are fungal, plant and microbial metabolites that serve as a carbon and energy source, and cell protectants against abiotic stress (Solomon et al., 2007; Hybelbauerová et al., 2008;

Rushdi et al., 2016).

4.1.1. Mono-, disaccharides and sacharols

The most common saccharides in soil and dust are glucose, sucrose and trehalose (Table 2). Glucose and sucrose are usually associated with plant debris, while trehalose with fungal and microbial activity (Simoneit et al., 2004a; Medeiros et al., 2006; Otto and Simpson, 2007; Rushdi et al., 2016). However, in the first stage of soil formation (so-called arenosols), the major saccharide is sucrose, genetically associated with algal-cyanobacterial mats. Trehalose, together with arabitol and mannitol, appear in substantial amounts at the later stages of initial soil formation and are associated with the activity of fungi and lichen (Marynowski et al., 2020b). Other common compounds in soils and dust are inositol (especially D-pinitol), fructose, and to a lesser extent glycerol, mannose and galactose (Table 2). Fugitive dust can also be a component introducing saccharides to aerosol PM during dry and windy seasons (Simoneit et al., 2004b; Medeiros and Simoneit, 2007). Furthermore, saccharides were identified in lacustrine sediments with a dominance of glucose, rhamnose and fucose (Al-Mutlaq et al., 2008).

4.1.2. Anhydrosaccharides

Levoglucosan, although plentiful in aerosol PM and detectable in lake sediments, has rarely been found in soils and dust, and if so, LG always occurs at low or trace amounts. LG was barely detectable in roadside dust particles in Kuala Lumpur, while in air PM, its concentrations reached ca. 60 ng/m³ (Omar et al., 2007). Soils from different crop fields in the San Joaquin Valley, USA, had LG in safflower fields at 66.2 ng/g mean concentration, which was low compared to the mono- and disaccharides (trehalose was 5903 ng/g), and it was presumably from field burn residues (Rogge et al., 2007). Later, Rogge et al. (2012) studied paved and unpaved road dust from that area, and reported a minor LG concentration in urban paved road dust (11.3 ng/g) with sucrose or trehalose mean concentrations >600 ng/g. Low LG levels were

Table 2
Concentrations of saccharides in soil and dust samples^a.

Sample ^a	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Anhydrosaccharides														
Levoglucosan	24	2	—	—	—	—	—	—	—	66.2	—	11.3	—	—
Mannosan	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Galactosan	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Sugaralcohols														
Glycerol	3379	41	25	—	—	—	—	—	—	175	3405	73.4	—	—
Threitol	32	1	0	—	—	—	—	—	—	—	—	—	—	—
Erythritol	174	6	0	—	—	—	—	—	—	—	—	—	—	—
Arabitol	495	134	28	—	—	—	—	—	—	—	—	—	—	—
Adonitol	113	323	35	—	—	—	—	—	—	—	—	—	—	—
D-pinitol	443	18	235	—	—	—	—	1276 ^b	2465 ^b	858 ^b	323 ^b	—	—	—
Xylitol	—	—	—	—	—	—	—	—	—	—	—	—	43.2	—
Mannitol	2288	513	222	—	—	—	—	—	572	—	—	—	—	—
Sorbitol	763	136	120	—	—	—	—	—	—	—	—	3361	—	—
<i>scylo</i> -Inositol	30	0	0	—	—	—	—	—	—	—	—	—	—	—
<i>myo</i> -Inositol	2069	36	204	—	—	—	—	—	—	—	—	—	—	—
Monosaccharides														
α- + β-Arabinose	163	24	21	—	—	—	—	—	—	—	—	—	—	—
α- + β-Fructose	160	89	141	—	—	—	14	6307	11,435	3806	—	—	45.8	—
Mannose + Galactose	273	82	72	40	—	—	—	897	1970	277	—	—	—	—
Talose	—	—	—	—	—	—	—	1880	1134	808	—	—	1300	—
α- + β-Glucose	2559	96	270	30	54	58	44	5901	15,419	4565	998	2256	293	1700
Disaccharides														
Sucrose	15,558	9410	44,908	220	398	1388	170	34,196	60,058	60,129	1311	1848	762	4500
Trehalose (Mycose)	5549	661	1095	4300	760	1807	321	226	1695	3520	5903	30,206	618	—

^a 1, 2, 3 = Błędowska Desert arenosols at different development stages (Marynowski et al., 2020b). 4 = Dark black grassland soil from western Alberta (Feng and Simpson, 2007). Concentrations of 1–4 in µg/g TOC. 5 = Soil and sand sample from highway outside Kuwait city (Rushdi et al., 2006). 6 = Soil around market place in a rural area, Kuwait city (Rushdi et al., 2006). 7 = Sand outside Kuwait city (Rushdi et al., 2006). 8 = Soil and sand in the center of Riyadh city (Al-Mutlaq et al., 2007). 9 and 10 = Soil around market place in Riyadh (Al-Mutlaq et al., 2007). 11 = Surface soils of safflower fields and 12 = surface soils of almond fields. Both from the San Joaquin Valley, California (Rogge et al., 2007). 13 = Urban paved road dust from the San Joaquin Valley (Rogge et al., 2012). 14 = Prairie grassland soils, orthic brown, Canada (Otto et al., 2005). Concentrations of 5–14 in ng/g sample.

^b Identified as inositol.

detected in the initial soil of the Błędowska desert, Poland (<24 µg/g TOC), and comparably the most abundant sucrose was 15,558 µg/g TOC in the same sample (Marynowski et al., 2020b). To date, no mannosan, galactosan, xylosan or arabinosan have been reported in soil samples (Table 2). Current data suggest that input of AMs by fallout seems low and rapidly degrades in oxic conditions of soils and dust by bacteria and/or fungi. Studies on microbial degradation of LG and other AMs in different redox conditions are needed as previously postulated (Bhat-tarai et al., 2019).

4.2. Lignites, fossil wood and black shales

The first records about the occurrence of saccharides in sedimentary OM are known from the 1960s, but their extraction was by aqueous 0.5 M H₂SO₄ leaching and subsequent characterization by comparison with standards using paper and later gas chromatography (Vallentyne, 1963; Swain, 1969, and references therein). Other reports describing the unmistakable presence of saccharides in Mesozoic and Cenozoic rocks were published in the 1990s, along with the development of gas chromatographic and mass spectrometric techniques (Moers et al., 1989, 1990a,b; 1994). However, these authors still used acid hydrolysis as the main method of saccharide extraction, thus analyzing monosaccharides with their hydrolysates from polysaccharides. Later the presence of mono-saccharides was reported in peats using the same hydrolysis method (e.g., Comont et al., 2006; Jia et al., 2008; Disnar et al., 2008). The neutral anhydro-, mono- and disaccharides were recently identified in solvent extracts (DCM/MeOH) from Mesozoic (Middle Jurassic and Cretaceous) xylites (fossil wood), lignites and shales, and from Miocene lignites (Marynowski et al., 2018, 2019). Those saccharides could be designated according to source into four groups:

- (i) Levoglucosan – BB tracer from ancient wildfires;
- (ii) fructose, glucose, sucrose, shikimic acid, D-pinitol and quinic acid – compounds representative of pollen grains and plant debris present in lignites and shales. Glucose can also be from decomposition of hemicellulose and cellulose;
- (iii) xylose and arabinose – decomposition products from hemicellulose. If they co-occur with LG then can also derive from wildfires; and
- (iv) arabitol, mannitol, trehalose and possibly erythritol – fungal biomarkers. If present in xylites, they may have originated from wood degrading fungi (Marynowski et al., 2018, 2019, 2019; Jaroszewicz et al., 2018).

Microbial decomposition of lignites in laboratory culturing experiments formed numerous monosaccharides which had not been reported previously in the raw lignite samples (Detman et al., 2018). This indicated that some saccharides are bound to the kerogen structure, which was supported by the high contents of holocellulose in xylites and a moderate level in detritic lignites (Marynowski et al., 2018). Certainly, saccharides should only be expected in thermally immature rocks, although early diagenetic sulfurization processes can “vulcanize” them via sulfur cross-linkages (van Kaam-Peters et al., 1998; Sinnighe Damsté et al., 1998). Definitely, more work is required to fully understand the sources and utility of saccharides in sedimentary rocks.

5. Summary and prognosis

This review shows multiple uses of saccharides as organic tracers and biomarkers in organic aerosols, soils, and sedimentary rocks. Although saccharides have only been widely used since the 2000s, their current importance is high, and the potential still seems not to be fully utilized. It is recommended that saccharide tracers be applied in conjunction with other biomarkers, e.g., PAHs, lignin derivatives, lipids, fossil terpenoids, etc., in the samples under consideration to assess the origin and fate of their OM. The genesis of some of the saccharides found in brown coal

extracts and the generation of anhydrosaccharides from coals (especially of higher rank) during combustion still require clarification. More research into biomass and coal burning source tests is needed, especially that conclusions derived from aerosol analyses during wildfire episodes may be misleading because extreme fire temperatures can increase advection and vaporization of biomass fuel components. Moreover, the preservation potential of anhydro-, mono- and disaccharides requires clarification for the context of archeo- and paleo-event reconstructions. The diagenetic potential of saccharides needs to be elucidated to understand the conditions required for their preservation in sediments, and which microorganisms preferentially utilize saccharides in soils and sediments. Despite most (if not all) of the saccharides having multiple origins, these compounds are useful source indicators and increase our understanding of organic aerosols, soils and sedimentary rocks. Future research should focus on the statistical dependencies of individual saccharides that could potentially define the origin based on the co-occurrence of individual compounds.

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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