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Review

Critical Analysis of the Past, Present, and Future of Dendrochemistry: A Systematic Literature Review

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Abstract: Dendrochemistry, the study of elements found within tree rings, has been used to understand environmental changes from both natural and anthropogenic sources. When used appropriately, dendrochemistry can provide a greater understanding of the elemental changes in the environment. However, environmental and species-specific processes have been shown to impact results, and research from the field has been scrutinized due to the need for a greater understanding that role-specific processes such as translocation play. This systematic literature review examines dendrochemistry's history, highlights how the field has changed, and hypothesizes where it might be headed. From this review, we recommend the following measures: (1) promoting the use of new experimental techniques and methods with faster data acquisition time to allow for a greater number of samples to be processed and included in studies to increase statistical significance; (2) that more studies focus on the two- and three-dimensional space that trees grow in and consider the complex physiological processes occurring in that space and over time and (3) more lab-based studies to reduce the variables that cannot be controlled when sampling in situ. Understanding the challenges and opportunities from the past, present, and future research of dendrochemistry is crucial to the advancement of the field.

Keywords: dendrochemistry; dendroanalysis; dendroisotopes; environmental chemistry monitoring; dendrochronology; tree-ring chemistry; pollution

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1. Introduction

Humans have been impacting and influencing the environment for thousands of years. Whether through prescribed burns to create grasslands, building dams to control water levels and generate power, or mining and smelting ore to recover valuable materials, human impact has altered the natural environment [1–3]. Natural processes also influence local and global environments, with the changes measured and studied through various proxies, including ice cores, lake sediments, coral, and tree rings [4]. Dendrochronology has aided in understanding historical processes, both natural, such as climate reconstructions and future climate-based growth predictions in dendroclimatology, and through anthropogenic activities such as dating wooden structures in dendroarchaeology [5–7]. In the last 50 years, a new subdiscipline of dendrochronology has emerged, using the elemental concentrations within tree rings to create a timeline of changes to environmental chemistry, the subdiscipline of dendrochemistry [8]. Dendrochemistry offers the opportunity to look back and assess changes in the chemistry of the environment in which trees are growing. This is especially beneficial when records of environmental contamination or other proxies for changes in environmental chemistry are not available [9–12]. Field sampling is often inexpensive and can require minimal training, tools, and time, making it an ideal introductory screening method to analyze potential contamination events [13]. Advances in technologies used in chemical analyses have allowed for lower detection limits and high-resolution mapping of elements within the tree structure [14–16]. Dendrochemistry has even been used in criminal and civil court cases as forensic evidence [17,18].

While promising, dendrochemistry is not without its difficulties and inconsistencies. Since the first applications of dendrochemistry, researchers in the field have continuously called for more research to fully understand the translocation and uptake of different elements by specific tree species [19–26]. The main reasons for the caution come from acknowledging that tree growth and elemental uptake depend on many factors [26]. In 1993, Cutter and Guyette published a formative paper on the many different influences on tree elemental uptake that they argued must be considered when conducting any dendrochemistry study. The review paper focused on many influences of tree elemental uptake, including habitat-based factors such as the type and depth of the soil or ecological amplitude of a species, xylem-based factors, and the factors impacting the elements present [26]. The recommendations made by Cutter and Guyette [26] provided a framework for future dendrochemical studies to outline the potential impacts of how experiments are designed and what elements and tree species are studied. Nearly every paper on dendrochemistry cites Cutter and Guyette [26], but not all studies follow their framework or fully explain the possible implications of the experimental design of their research and how results may be impacted.

The need for a more thorough description and considerations about study sites, tree species, elements, and experimental design continues from the now common debate and questioning of the reliability of dendrochemical research results. Because trees are not passive to the fluctuations and changes in the environments they grow in, and because of the many variables present from in situ experiments, multiple dendrochemistry studies will often find conflicting results [12,24,27–33]. While the confusion surrounding the appropriate methodologies for dendrochemistry has yet to reach a clear consensus, the number of publications has increased in recent years (Figure 1). Dendrochemistry has been applied to a variety of specific applications, including the quantification and timeline of heavy metals being released into the environment [11,34–37], acidification, nutrient retention and stability of forests [38–43], stable isotope research [44–50], and hydrocarbon contamination [51–56].

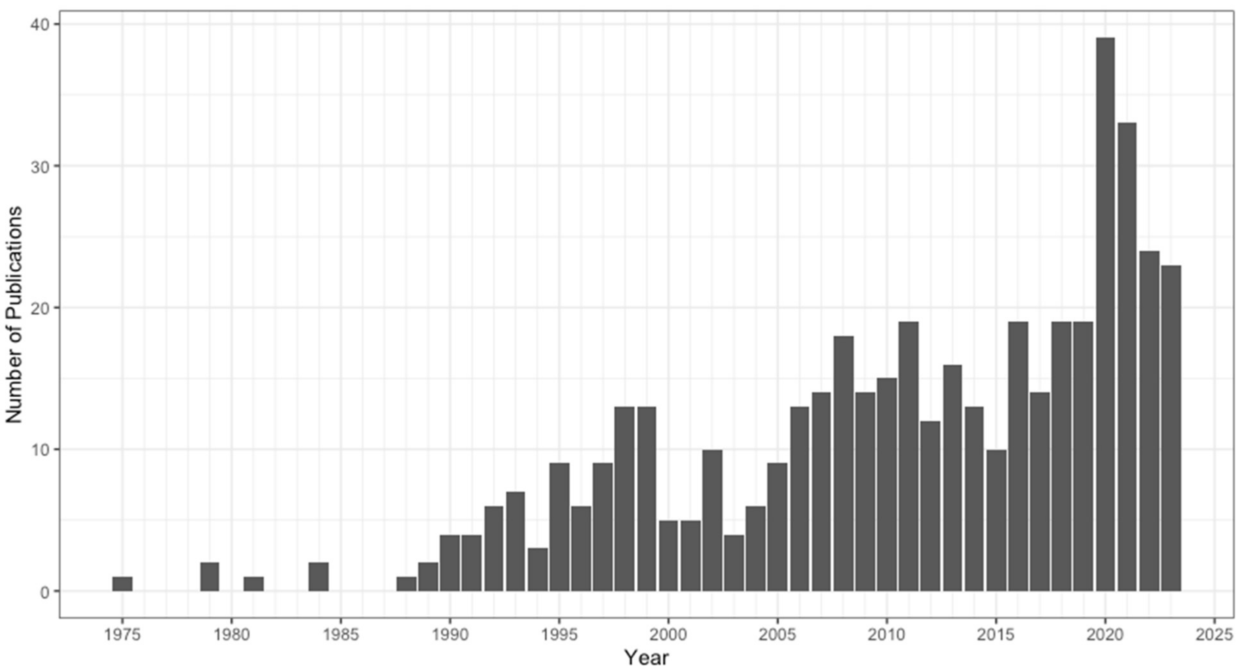

Figure 1. The increase in total dendrochemistry publications included in this study from 1975 to 2023.

While there have been recent review publications on dendrochemistry, they focused on how and why dendrochemical studies had been conducted and the specific techniques used [19]. This review will not cover the many different methodologies and instruments used to determine concentrations of elements in dendrochemistry samples. For in-depth descriptions of techniques and methods used in dendrochemistry studies, please refer to Binda et al. [19]. This literature review aims to complement the review by Binda et al. [19] by looking through the lens of how the study of dendrochemistry has evolved over the last 50 years and hypothesizes where it may be heading in the future, along with providing recommendations for future research. This review focuses explicitly on quantifying past research from the field, including which countries and journals are publishing dendrochemistry studies, the types of trees being studied, and the elements of interest. This review will also cover limitations and novel techniques not always considered in most dendrochemistry research.

2. Methodology

From April to August 2023, dendrochemistry articles were searched in the Web of Science Core Collection (WoSCC) using the PRISMA 2020 checklist for systematic reviews [57]. Three keyword searches were conducted to try and account for as many studies and publications related to dendrochemistry as possible. The first search used 'All Fields' and the keyword 'Dendrochemistry.' The second search used the 'Topic' search, which only searches for article titles, abstracts, author keywords, and Keywords Plus in WoSCC using the keywords 'Tree-ring* AND Chemistry' (note * denotes a Boolean operator to search multiple endings of tree-ring). Finally, the third search used the 'All Fields' search and keywords 'tree ring*' AND 'Pollution' NOT 'Demndrochemi*'. The third search was broader to capture dendrochemistry studies not found by the previous keyword searches, but also aimed to limit the number of duplicate search results. Articles were independently screened and assessed to determine if they met specific criteria for this review by the authors. In this review, a dendrochemistry study is a study that uses tree rings (either cores or discs) and looks at the change over time present in tree rings in the form of either isotopes, heavy metals, nutrients, or hydrocarbons. Also included in this review are articles that add to either a specific technique or general methodology used in dendrochemistry that directly talks about how the findings can relate to dendrochemistry. To extract the most information from the list of articles curated from the three separate searches, the authors independently created a combined dataset that identified DOI, author(s), year of publication, affiliated country, journal of publication, tree species used, and elements studied. This dataset is available in the Supplementary Materials. Figures were created from this dataset in the program RStudio and the package ggplot2 to visually represent the data collected from the articles included in the dataset [58,59].

3. Results

As of 19 April 2023, the first search of 'Dendrochemistry' provided 169 results, of which 157 were scholarly articles, including a formal review paper, 20 proceedings papers and a handful of meeting abstracts, book chapters, and editorial materials. The total number of papers included in the first search was 146, ranging from publication dates of 1993 to 2023. As of 8 May 2023, the second search of 'Tree-ring* AND Chemistry' yielded 196 results, of which 176 were scholarly articles, 18 proceedings papers, and a total of 14 review articles, meeting abstracts, book chapters, and editorial materials. Sixty-seven duplicate results from the previous keyword searches were removed from the study, leaving 74 papers included from the second search with publication dates ranging from 1984 to 2023. The third keyword search was much broader in scope and therefore returned 669 results as of 10 August 2023. Many of the papers in the third search result were studies looking at the effects of pollution on tree ring widths (TRW) and comparing atmospheric pollution data to TRW [60–62]. These studies did not look at the elements within the tree rings and therefore are not considered dendrochemistry studies. After reviewing the 669 results, 206 additional articles were included in the literature review as dendrochemistry studies.

A PRISMA 2020 flow diagram describing the steps taken to create the database of articles included in this systematic literature review, the number of duplicates, and articles removed from the study, along with the reasoning, is included in the Supplementary Materials [57]. Articles excluded from this review included articles not written in English, conference citations included in WoSCC, articles not available through the institutional access provided by the University of Saskatchewan, and articles not considered by this review to be explicitly focused on dendrochemistry. Many of the articles not considered dendrochemistry papers in this review appeared in the search results due to either 'Tree-rings', 'Chemistry', or 'Pollution' being included in the Keywords Plus. A common example was the use of ice core or lake sediment chemistry that often mentioned other proxies, including tree rings, to infer past climate conditions [63–65]. These studies did not specifically use tree rings but still appeared in the search results and were excluded from this review. After combining the results of the three search terms, the total number of papers included in this literature review was 426.

The papers in this study do not account for *all* the dendrochemistry research conducted over the years. However, the articles included in this review provide a general snapshot of the significant research that has been ongoing over the last five decades. As previously described, the data collected from the individual papers were divided into specific areas, including affiliated country, species used, and elements of interest. Each area was analyzed to find general trends and how the field of dendrochemistry has changed or is changing.

3.1. Countries

From the 426 papers analyzed, 42 countries worldwide are considered primary countries that have published dendrochemistry studies (Figures 2–4). Primary publication countries were determined by where most co-authors were affiliated and where the research occurred. Most papers had co-author lists from the same country and often the same institutional affiliation [26,28,66]. However, if a paper had a roughly equal number of co-authors from two different counties, each country was considered a primary representative of that paper [67–69]. Recently, there have been several publications with co-authors affiliated with institutions from around the world; these studies were considered multinational (Figure 2a) [13,70,71].

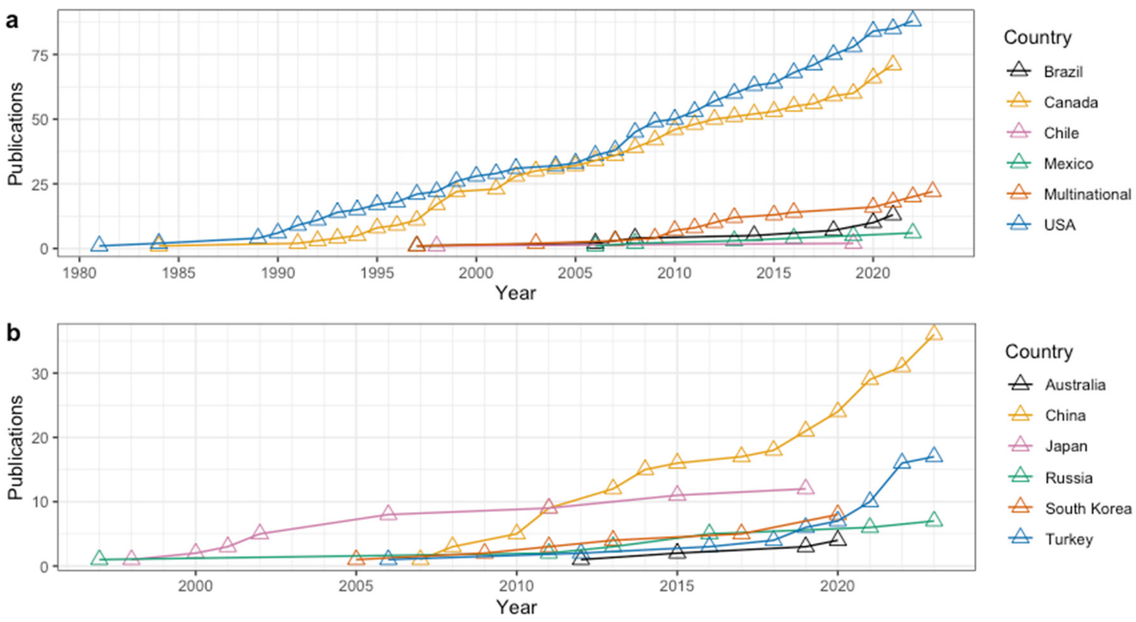

Figure 2. (**a**) The Americas and multinational countries' total dendrochemistry publications, (**b**) Asia and Pacific countries' total dendrochemistry publications over time.

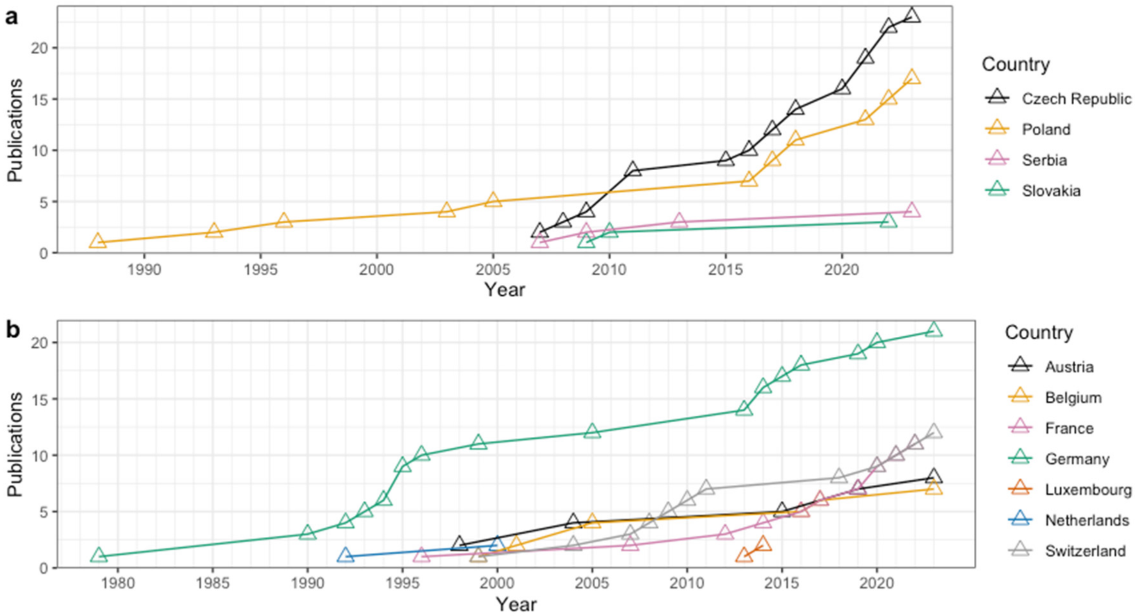

Figure 3. (**a**) Eastern European countries' total dendrochemistry publications, (**b**) Western European countries' total dendrochemistry publications over time.

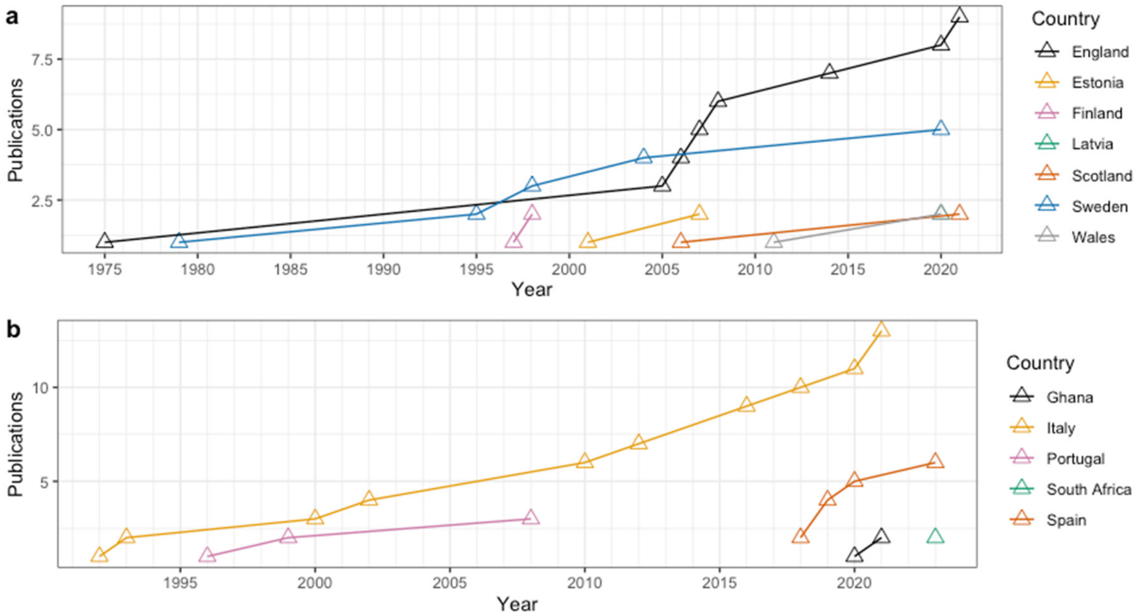

Figure 4. (**a**) Northern European countries' total dendrochemistry publications, (note, that as of 2020, Latvia and Wales both have two dendrochemistry publications) (**b**) Southern European and African countries' total dendrochemistry publications over time.

The United States and Canada have published by far the majority of dendrochemistry studies (Figure 2a). Of all the articles analyzed, the U.S. and Canada were considered significant contributors to 159 articles, with the U.S. responsible for 88 articles and Canada responsible for the other 71. Some of the earliest dendrochemistry studies occurred in England, Germany, Canada, and the U.S. (Figures 2a, 3b and 4a). Seven of the forty-two countries only had one dendrochemistry publication in the search results. These countries were Hungary (2009), Macedonia (2010), Israel (2017), Lithuania (2017), Romania (2021), Croatia (2022) and New Zealand (2022).

3.2. Species

From the articles reviewed, there are 166 tree species, from 86 genera, across 35 families that have been used in dendrochemistry studies. Because many studies included multiple species of trees in their experiments, the total number of trees is greater than the number of articles reviewed. The majority of the trees that have been studied are within the Pinaceae family (271), followed by the Fagaceae family (104), and finally, the Sapindaceae family (40) (Figure 5). The diversity of tree species reflects the diversity of the researchers in dendrochemistry. Some articles did not report the species of trees used, and others grouped trees of the same genus that are similar ecologically to increase the number of samples in the study [13,56,72–76].

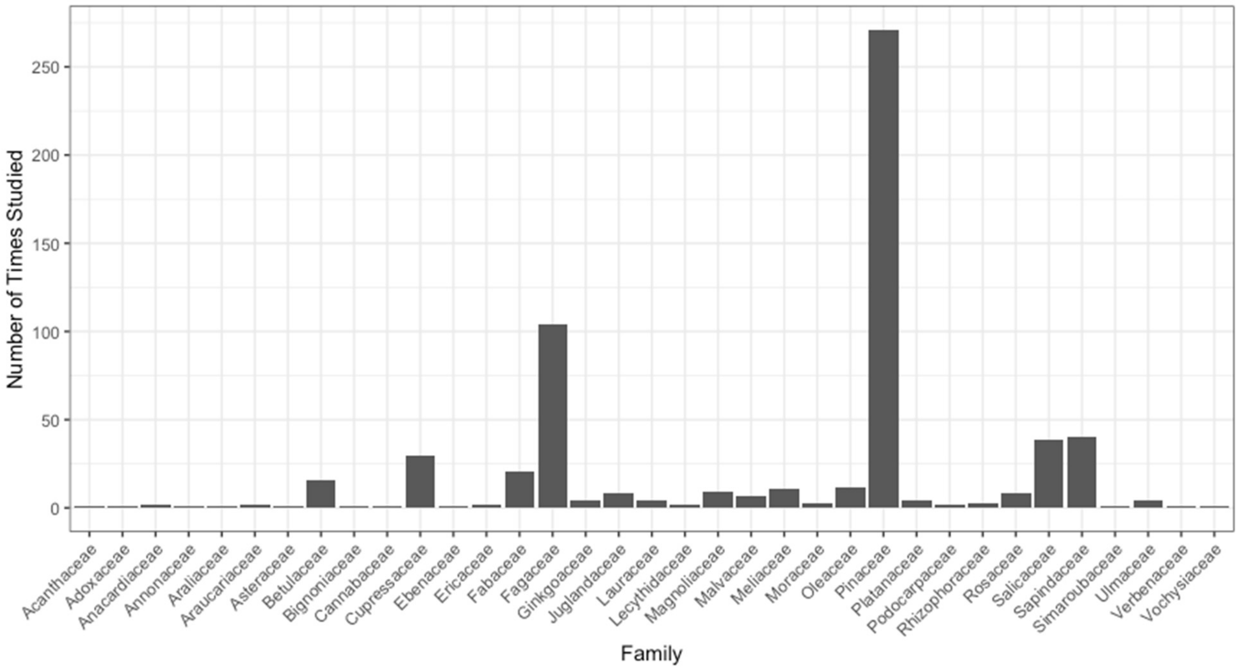

Figure 5. The 35 families of trees that have been studied in dendrochemistry from our dataset.

3.3. Elements

The studies reviewed looked at a wide range of elements and isotopes. Some of the studies focused on one specific element, like mercury (Hg) or lead (Pb) [29,35,77–82]. Other studies focused on specific isotopes and tracer elements [83–90]. Many studies focused on a handful of elements to identify environmental changes, either through acidification or the addition of heavy metals [71,91–98]. However, most studies looked at the general elemental profile of tree samples to identify elements of interest or of higher concentration than expected [22,68,99]. The most studied element in dendrochemistry from this review is lead (Pb), with 91 articles including it within the scope of the research (Figure 6). The second most studied is calcium (Ca), with 87 studies, followed by Zinc (Zn), with 76 studies and Manganese (Mn) at 74. The lighter elements on the periodic table, such as Ca, aluminum (Al), potassium (K), and magnesium (Mg), were among the most common elements studied, as well as the first full row of transition metals on the periodic table including copper (Cu), nickel (Ni), Zn, and iron (Fe) (Figure 6). The most common isotopes studies in dendrochemistry have been carbon (C)13 and nitrogen (N)15 (Figure 6). Many studies focused simply on the general elements present and did not focus their research until after determining elements of interest. From this review, a total of 55 different elements and isotopes have been studied in dendrochemistry.

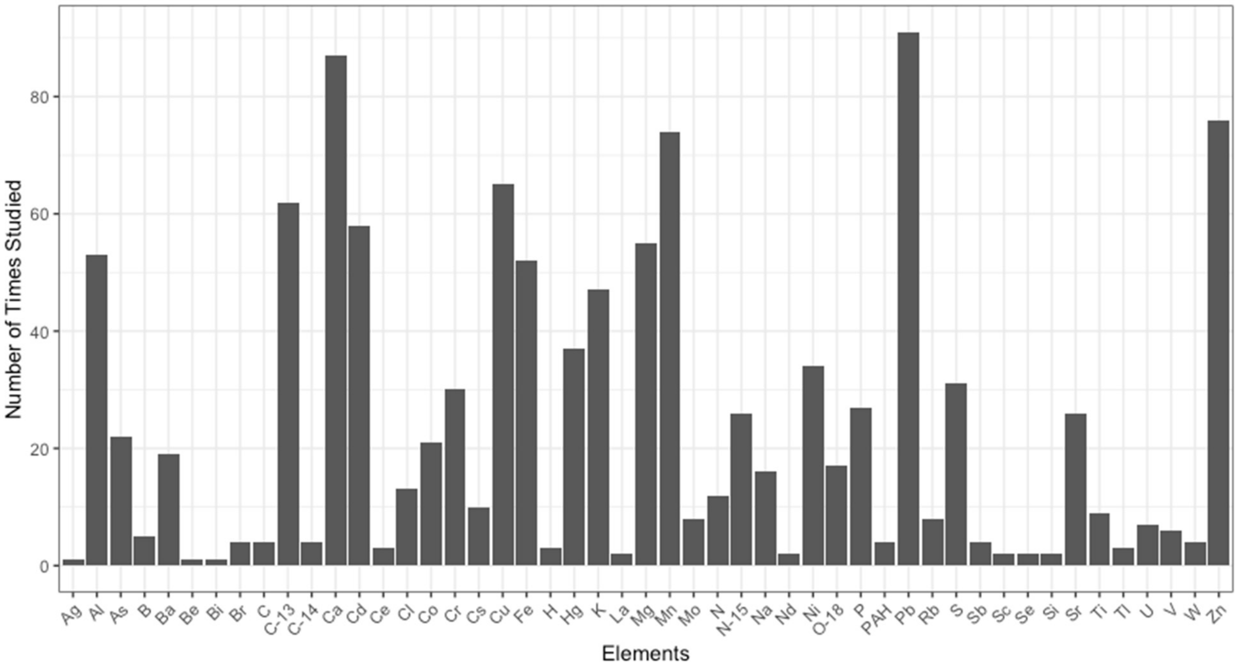

Figure 6. The top 50 most studied elements in this dendrochemistry dataset.

4. Discussion

Over the last five decades, dendrochemistry has changed and evolved considerably. Everything from the techniques used, to the research questions being asked, or specific types of studies being conducted has changed or been reassessed at some point. We are curious to understand how the field got to where it is today and where it may be heading in the next five decades. To accomplish this, we use the findings from the literature review to evaluate how the early research shaped what is being achieved today, how the field is changing, and what main themes may be the focus of dendrochemistry in the future. We also take the opportunity to highlight areas where future research could be better directed that is currently under studied.

4.1. The Past

The earliest days of dendrochemistry included in this review started in 1975. Early papers are proof-of-concept focused, where studies are designed to determine if there is a change in the tree chemistry related to the change in the soil chemistry of a region. Many of the early dendrochemistry papers focused on available nutrients and the effects of soil acidification on tree ring chemistry [38,100–106]. Acidification of soils and aquatic environments was a primary environmental concern starting in the 1970s due to the atmospheric anthropogenic additions of sulphur dioxide (SO_2) and various sources of atmospheric N, including nitric oxide (NO), nitrogen dioxide (NO_2) (NO_x emissions for both) and ammonia (NH_3) [107]. The sources of these emissions came from many systems, including the volatilization of fertilizers, industrial processes, and the burning of fossil fuels [108]. Researchers from industrialized regions of Eastern North America and Europe saw an increase in soil acidification and questioned how this change to the natural balance of the S and N cycles would impact forests [109,110].

Early dendrochemistry research in this area was focused on the changes in concentrations of nutrients like Ca, Mn, and Mg and metals like Al. The research focused on determining if changes in the soil were reflected in the chemistry of the xylem. The focus on the impacts of soil acidification on forests was mainly driven by the concern that the decrease in soil pH and loss of base cations would lower the overall soil fertility in forests, and there would be a potential for large-scale dieback. Joslin et al. [104] reviewed the potential link between soil acidification in the Eastern United States and the noticeable decline in red spruce (*Picea rubens* Sarg.). It is known that forest soils will have lower soil pH over time due to the uptake of base cations by trees and other plants as a forest stand matures, and if harvested, those cations are then removed from the ecosystem entirely and not replaced [111]. Joslin et al. [104] found many studies, including long-term soil pH studies in forest stands and dendrochemistry studies, showing a decreasing pH trend as a forest stand matured in both the soil and tree rings. However, the soil acidification rate could not be accounted for solely by the maturing of forests in both the U.S. and Europe [103,105,109]. Joslin et al. [104] conclude that many factors likely contribute to the acidification of forest stands in Eastern North America; however, they estimate that roughly half of the losses of base cations could be traced back to acidic deposition. The authors could not say that the previously observed declines in red spruce in the region were due to the loss of nutrients from the soil [104].

Fears for the impacts of atmospheric pollution and acidification of forests were not only constrained to North America. In the late 1970s up until about the 1990s, central regions of Europe were also experiencing substantial dieback of forests [110,112,113]. Many European researchers hypothesized the dieback was linked in some way to acidification. Wallner [110] looked at trees from two sites, one heavily polluted with SO_2 and another less impacted site. Trees were felled and analyzed using neutron activation analysis and inductively coupled plasma atomic emission spectroscopy (ICP-AES) for Mg measurements. The results from this study agreed that Mn is an important health metric for tree health and noted that the Mn values at the more heavily polluted site were much greater compared to the less polluted site and this was associated with a decrease in the ring width of the trees at the polluted site [110]. Researchers around the world had observed the increase in die offs of large sections of forests and dendrochemistry became a go-to tool and methodology to determine the extent that acidification had on tree and environmental health.

In order to understand the historical scale of the impacts of acidification of forests on tree species, researchers looked to dendrochemistry to create a timeline of the changes in soil chemistry reflected by the changes in xylem chemistry. However, few early studies explored the possible limitations of dendrochemistry, primarily the possibility of translocation of elements across ring boundaries, the multiple pathways for elemental uptake (atmospheric deposition and through the root system), and the physiology of different tree species [106]. Dewalle et al. [106] looked at the five most recent years of growth in the sapwood of six different Eastern North American species, including red oak (*Quercus ruba* L.), chestnut oak (*Quercus prinus* L.), black cherry (*Prunus serotina* Ehrh.), eastern white pine (*Pinus strobus* L.) eastern hemlock (*Tsuga canadensis* [L.] Carr.), and pignut hickory (*Carya glabra* [Mill.] Sweet) across five different sites with a range of soil pHs. Elements in the wood samples were analyzed using inductively coupled plasma mass spectrometry (ICP-MS). They found that most of the species tested showed changes in the elemental concentrations of Sr, Ca, Na, Mn, Mg, Al, Be and Ba. Only chestnut oak and pignut hickory showed similar sapwood chemistry when growing in different soil pH [106]. This illustrated that even trees in the same genus, such as the chestnut oak and red oak, may have different responses and sensitivities to the environment they grow in and how they are impacted. It is also interesting to note that DeWalle et al. [106] assessed the sapwood of six species but only looked at the most recent five years of growth. The number of rings within sapwood varies between species, and it is possible that comparing five-year increments between each species is not the most representative comparison.

After initial studies in Eastern North America looking at the potential impacts of soil acidification on forests, fertilization studies attempted to reproduce the impacts of acid deposition through experiments. The purpose of these studies was to determine with greater certainty the specific impacts of additional NO_x and SO_x in the environment on species such as sugar maple (*Acer saccharum* Marsh.) in Eastern Canada [114]. Similar to other Eastern North American species, in the 1980s, foresters saw considerable dieback of sugar maple, with causes being attributed to a variety of factors, including drought, insect and disease outbreaks as well as an increase in the availability of Al from acid deposition [39,114,115]. Over three years, Hutchinson et al. [114] conducted a fertilizer experiment where selected plots were treated with ammonium sulphate ($(NH_4)_2\ SO_4$). By the end of the study, there was no visible decline in the tree's health, but there were changes to the soil chemistry, including a reduction in pH and mycorrhizal infection and association, as well as an increase in concentrations of Cr, As Co, Ni, Zn and Mn in the growth rings during the years of fertilization [114]. A notable result in this study was the overall difference between the two sites where the experiments were conducted. Both sites had treatment and control plots and were classified as Sombric Ferrohumic Podzols with similar parent material, with one site being located within 100 km of a known source of heavy metals [114]. The results illustrated that one of the sites had limited statistical differences between the control plots and the fertilizer treatments, while the other site saw significant differences in foliar concentrations of nutrients like N, Mg Mn and Cd and an increase in heavy metals in the tree rings.

The substantial differences between the results at the two sites in Hutchinson et al.'s [114] study highlight the complexity of the many interconnected processes occurring in situ and the need for better controls for experiments in dendrochemistry. While acidification is not the only type of dendrochemistry study from the discipline's early days, it is often overlooked and not as common in ongoing dendrochemistry research. Given the disproportionate number of researchers from the U.S. and Canada who published early dendrochemistry studies, it makes sense that they would focus on environmental issues close to home (Figure 2a). However, the results from these early acidification studies also helped to inform the possible shortcomings and advantages of the discipline for future studies not focused on the impacts of acidification.

4.2. The Present

The majority of current studies in dendrochemistry are expanding on previous research areas to focus on new elements of interest and new species of interest. In the opinion of the authors, the main areas of current research include heavy metal and dendroisotope research. While the concepts of tracing heavy metals and isotopes over time using tree rings are not new, the ability for researchers to use new methodologies with lower detection limits or faster acquisition times to allow for a greater number of samples to be processed adds new elements to the research and greater research possibilities.

4.2.1. Heavy Metals in Tree Rings

One of the most significant environmental concerns worldwide is the increase in heavy metals entering the environment from anthropogenic sources such as resource extraction and contamination events [116]. Since the early days of dendrochemistry, researchers have attempted to show the connection between the increase in heavy metals in a region and the uptake of those metals documented within the rings of trees growing in the same area [20,23,28,37,117–121]. However, while there have been multiple studies that have shown the ability of trees to keep a minimal record of heavy metal contamination events, many similar studies have also shown high rates of translocation and call into question the reliability of the results locked into a timeline [11,12,24,28–30,121,122]. Many studies that find a correlation between known contamination dates and the general elemental concentrations in tree rings present their results with caution and highlight the range of concentrations between samples [11,122].

Recently, more research has emphasized specific elements with more promising results from elements like mercury (Hg). Several studies have been published on the ability of trees to be used as recorders of Hg emissions in both regional and national settings [9,32,77,78,123–125]. However, comparing any of these studies can be difficult due to site- and region-specific influences and individual studies using different species of trees. For example, Abreu et al. [31] assessed the ability of only black poplar (*Populus nigra*) to monitor Hg levels in aquatic environments. The authors used three black poplar trees growing close to a point source of Hg, cores from the trees were cut into 1cm segments that represented around two years of growth, and the total Hg per segment was determined through atomic absorption spectroscopy (AAS) [31]. Abreu et al. (2008) found that the location of the trees played a prominent role in the amount of Hg present in the core, with the tree being closest to the point source of Hg emissions having the highest concentration and the tree the furthest from the source having the lowest concentrations. The authors did conclude that black poplar reliably recorded the Hg emissions in the area and generally followed the trends of the industrial activities in the area [31].

Wright et al. [74] looked at the ability of trees to record regional and national changes in atmospheric Hg emissions along a gradient from the coast to the interior of the United States. Based on the previous findings of Cutter and Guyette [17], pines were the selected species due to their presence across a wide geographical area, the minimal number of rings in the sapwood, and the low moisture content of the heartwood to minimize the possibility of translocation. Due to the known variability of elemental concentrations within a single ring and between multiple trees [117], Wright et al. [74] sampled three trees from each site and collected multiple cores per tree. Wright et al. [74] found that proximity to potential sources of Hg, such as lakes and oceans, the elevation of the sampled tree, and the various forms of Hg can all impact the result of Hg concentrations. They also include the possibility that the trees' age and species play a role in influencing the results, however, only to a minimal extent. The authors concluded that dendrochemistry is a potential technique for recording the general trends of atmospheric Hg concentrations [74]. Both studies found that the trees used worked and provided reliable results, but there were considerable differences between experimental designs, such as the number of sites and samples between the studies.

Based on the promising results from Wright et al. [74], most studies focused on various pine species and looked at the impacts of both point and nonpoint sources of Hg in the environment. Many of these studies showed translocation within the rings [9,123,125]. A study by Arnold et al. [125] used potted saplings from a common genetic source to determine their ability to take up and deposit Hg within their leaves, bark, and tree rings. Four-year-old trembling aspen (*Populus tremuloides*) and Austrian pine (*Pinus nigra*) saplings were given four different treatments of atmospheric Hg conditions: ambient greenhouse air, greenhouse increased Hg, greenhouse Hg emitting mine tailings (only at night), and outside adjacent to a busy interstate highway [125]. The greenhouse trials were also spiked with a $HgBr_2$ solution in the soil to test the pathway from the roots to the tree rings and through to the foliage. The results showed that Hg uptake was primarily determined by the total gaseous mercury concentrations in the air and not what was added to the soil. It also illustrated that the total uptake was not dependent on the type of mercury compound present. This was, however, contradictory to other results from previous studies, and it was stated that further investigation was needed to confirm these results [125].

Arnold et al.'s study [125] is one of the few greenhouse sapling studies conducted in dendrochemistry, but it had many experimental issues. An aphid infestation impacted the trembling aspen saplings, and the application of pesticide caused the deterioration of the foliage for both species in all greenhouses and only two months of data are reported due to the reduced health of the trees from the pesticide and not the additional Hg [125]. A main finding from this study was the evidence of radial translocation of Hg across ring boundaries where the ring grown in the increased atmospheric Hg trial had significantly more Hg present than the control tree's same growth ring [125]. Another result from Arnold et al. [125] was that radial growth had a limited impact on the overall concentrations of Hg, as shown by the trial next to the highway, which had the greatest growth, but the lowest Hg/mm ratio compared to the trail with increased atmospheric Hg. This result shows that trees likely reflect atmospheric Hg concentrations independent of their radial growth rate [125].

Based on the findings from Arnold et al. [125], a subsequent study by Peckham et al. [77] aimed to answer some of the main questions in dendrochemistry, which are as follows: (1) is there spatial and temporal consistency between tree rings? (2) How many samples are required to have a robust experimental design in dendrochemistry? Moreover, (3) do the Hg concentrations remain consistent in trees over time? Peckham et al. [77] sampled a new site in the eastern Sierra Range at two locations, high- and low-elevation sites oriented west and east to each other. Half of the tree cores from the higher elevation site were cored randomly on the trees, and the other half were cored, all facing the south side of the trees. At the lower elevation site, all cores were extracted from the north side of the trees [77]. Sites that were previously sampled by Wright et al. [74] were revisited and resampled by Peckham et al. [77] in two of the locations, the same trees were sampled as in Wright et al. [74]. From the first site, it was found that Hg concentrations were highly variable between trees at the same site. However, the correlation improved when all the cores were sampled from the same side of the tree [77]. Although the concentrations varied, the general trends seen over time remained consistent between trees. Peckham et al. [77] also noted that the two sampling locations with different elevations at the same site had considerable differences in Hg concentrations. However, the lack of agreement between trees at different elevations could be because the trees growing at the higher elevation were older, and the spikes that were identified likely occurred when the lower-elevation trees were saplings [77]. This suggests that the physiological condition of the trees, including the age and possibly time of sampling, plays a more significant role than previously thought. The samples from the same sites as Wright et al. [74] were analyzed, but a different instrument calibration was used, resulting in a difference in concentration levels between the two studies. The sites where the same trees were sampled in both studies had similar trends of Hg concentration, but due to not sampling the exact same trees in both studies, the known variability was high between trees. The overall correlation between the majority of samples from Wright et al. [74] and Peckham et al. [77] was low. However, Peckham et al. [77] showed that the higher number of samples included at specific sites increased the overall correlation between the two studies. The findings from Peckham et al. [77] led to the following recommendations for dendrochemistry: (1) researchers should attempt to minimize the effects of resource competition between trees as a way to account for the physiological differences between them; (2) researchers should consider the impacts of stand composition including age, height and growth rate of the trees being sampled; and, (3) to account for the variability in the radial growth around a tree, multiple samples from around the trunk should be collected and analyzed. Peckham et al. [77] also called for the need to use non-destructive and higher sensitivity detection methods in the future.

4.2.2. Tree Ring Isotopes

Just as researchers have looked at the heavy metals present in tree rings to determine ongoing chemical changes in the environment, isotopes of δ^2H, δ^{13}C, δ^{18}O, $\delta^{206/207}$Pb and δ^{15}N have been used to reconstruct and understand changes in past climates, air quality or pollution records and even recently to understand physiological growth after a fire disturbance [33,48,83–85,126–133]. The study of these isotopes can show the changes in the different uptake mechanisms used by trees to grow and how changes to their growing environment are being impacted. For example, δC isotopes are controlled by the assimilation of atmospheric CO_2 and can record changes in atmospheric chemistry, whereas δH, δO, and δN are impacted primarily by the chemical composition of the soil water trees use, as well as the minimal effects of foliar assimilation for δN [48]. A promising area of research is the ability of dendroisotopes to show possible impacts on tree growth from pollution stress, specifically in areas where pollution records are short, or non-existent and especially when paired with other proxies for environmental change [48]. Another research area of interest is paleoclimate reconstructions, focusing specifically on the availability of moisture and the climactic changes over time [127].

The ability to reconstruct and infer about past climates is critical to understanding how ecosystems are able to adapt to changing weather patterns and their overall growth or performance trajectories. Stable isotope ratios of carbon 12 and 13 can provide a climate specific reconstruction of tree growth better than more general tree-ring width measurements. The isotope ratios are controlled by the partial pressure of CO_2 in leaves and needles [127]. Depending on the rates of carbon assimilation and stomatal conductance, the ratios represent a climactic growth response in line with their environment. Bale et al. [127] used this technique to reconstruct precipitation trends over 920 years from an annually resolved bristlecone pine (*Pinus longaeva*) from the White Mountain range in California $\delta^{13}C$ isotope chronology. Negative values of $\delta^{13}C$ ratios indicate stomatal closure or narrowing due to moisture stress in drought prone environments [127,134,135]. Bale et al. [127] found their reconstruction to be statistically significant and sensitive to severe and extreme El Nino events from 1500 BCE to 2005 CE and hypothesized at least 13 other events prior to 1500 BCE. Their results also showed significant similarities between reconstructions of Colorado River flows to an extent [127]. Other isotope climate reconstructions include research from the boreal forest in Eastern North America where Alvarez et al. [84] looked at the impacts of where isotope ($\delta^{13}C$ and $\delta^{18}O$) sampling occurs within the ring for black spruce trees (*Picea mariana*). Previous research has shown that depending on the species, latewood and whole ring values of $\delta^{13}C$ can show different results [84,136,137]. The results from Alvarez et al. [84] clearly showed similar trends and relationships of precipitation and climate reconstruction when using either latewood or whole ring isotope ratios for $\delta^{13}C$ and $\delta^{18}O$. Whether reconstructing paleoclimates or more recent climate trends, it is important to understand how divergences from the known climate conditions can occur, and how to best prevent those divergences from occurring. Savard and Daux [50] describe five of the main reasons researchers may encounter isotopic divergence from known climate records and provide methods to limit or identify the divergences. Among them are the impacts of stand dynamics such as the juvenile effect, changing climates that alter the previously known primary causes of changes in isotopic ratios, and the impacts of atmospheric pollution, which has the ability to impact the primary uptake mechanisms in leaves and roots [50].

The ability to monitor and detect the impacts of pollution through the use of isotopes in tree rings is a promising field of study. Determining when ecological stress due to high levels of pollution had occurred at an annual resolution allows for the ability to both determine likely causes of pollution and understand the short- and long-term impacts on growth. In a recent study, Savard et al. [138] aimed to better understand the link between the water use efficiency of boreal forest trees and the rise in atmospheric CO_2 concentrations along with increased atmospheric pollution to determine how those processes combined are impacting isotopes and tree growth and water use. Their results showed that trees in sites exposed to high acidifying atmospheric emissions had pronounced increases in their water use efficiency, but no increase in the growth increments of stems. The results also showed a decoupling of isotopic ratios from climate responses due to the increased pollution, showing that areas with known atmospheric pollution should be avoided when sampling for climate reconstructions using tree-ring isotopes.

Dendroisotopes have the potential to be used as active monitors of pollution, but also as a historical reference for large pollution events depending on the mobility and source of the isotopes of interest. Francova et al. [131] looked at the three environmental samples including tree rings to determine if they were suitable record holders for environmental pollution, and if the isotope ratios present in the samples would determine the source of the pollution. Their results showed that the isotopic ratios of $\delta^{206/207}Pb$ and $\delta^{208/206}Pb$ reflected the changes in sources of known atmospheric Pb pollution in the study region in line with past studies [33,131–133]. The field of dendrochemistry around the world is growing quickly (Figures 1–4). Good dendrochemistry research has been conducted for years and will continue. However, researchers should utilize advancements in technologies for increased efficiency as well as to strive to come to agreements on recommended procedures for the field.

4.3. Challenges and Recommendations for the Future of Dendrochemistry

The third keyword search (Tree ring* AND Pollution NOT Dendrochemi*) nearly doubled the number of articles included in this study due to the broad nature of the search term 'pollution', which is common in many environmental science papers. Using the search term pollution also included many dendrochemistry studies that did not use the term 'dendrochemistry' but instead used terms such as dendroanalysis [139–141] or dendrogeochemistry [36,142,143]. Many papers used the term dendrochemistry in introductions or discussions but did not include it in the keywords or abstract [144–148]. The multiple names being used to define the same study subject makes it challenging to find all of the relevant articles in the field. One recommendation is for future articles to either include multiple terms in keywords to make it easier for other researchers to finds their tree-ring chemical pollution research.

Much of the focus in dendrochemistry has been on the historical dating of contamination events or environmental change, usually due to anthropogenic influences. Many of the studies included in this review have taken place in North America and focus on the species and elements of concern in that area (Figures 2a, 5 and 6). However, more diverse research from outside North America is increasing (Figures 2–4). With more researchers working on dendrochemistry studies, the challenge for the field is to address the long-standing issues surrounding the impacts of processes such as translocation, the uptake of different elements, and how the tree species impact results. Returning to seemingly basic or general questions and understanding fundamental processes will make the field more applicable and reliable. We recommend (1) the promotion of the use of new experimental techniques and methods with faster data acquisition time to allow for a greater number of samples to be processed and included in studies to increase statistical significance; (2) that more studies focus on the two and three-dimensional space that trees grow in and consider the complex physiological processes occurring in that space and over time; and, (3) more lab-based studies to reduce the variables that cannot be controlled when sampling in situ.

4.3.1. New Techniques to Address Old and New Questions

Many of the questions raised in this review are complex, and many of the answers will likely be location-, species-, and element-specific. These answers will likely require advancements or alterations in traditional dendrochemical experimental design, sampling, and techniques. Dendrochemistry is useful because researchers can take samples from multiple trees in the same area that have all been growing under the same conditions. However, every result of the elemental make-up of the samples will be unique to a tree and sometimes even susceptible to the specific core in a two- and three-dimensional space within the core sample (See Section 4.3.2). In traditional dendrochronology, the individual growth rates of trees growing in response to environmental or site-specific conditions (i.e., precipitation variation from year-to-year and trees growing on the side of a slope) result in the rings around the entire stem not being perfectly uniform. Researchers are then required to create a master chronology for a specific area, meaning multiple samples, typically consisting of over 30 trees and cores, taken 90° from each other to capture the overall variation to make a master chronology [8]. However, due to the cost and time to prepare and process samples, the high sample depth required in dendrochronology is often not feasible or at least has not yet been required in dendrochemistry. Increasing the sample depth will aid in statistical analysis but must also be realistic and feasible for time and cost constraints.

An area of development for the future of the field is research into new methods of analyzing tree samples, specifically into non-destructive methods [77]. One possibility is using X-Ray fluorescence (XRF) techniques. XRF is a non-destructive methodology with relatively simple sample preparation and can detect multiple elements simultaneously [149]. XRF can also collect high-resolution data with great degrees of sensitivity and high precision, allowing for the exploration of intra-annual changes in the elemental distribution of samples [150]. Benchtop instruments such as energy dispersive XRF (EDXRF) can provide transect line scans of cores and are reasonably available in laboratories [150,151]. A higher-powered source of X-rays like a synchrotron can also provide multi-element analysis with faster acquisition times and the ability to create 2D maps of elements through synchrotron X-ray fluorescence (SXRF) [14,152–155]. Faster acquisition times allow for whole or even multiple samples to be scanned, increasing the total number of samples in the study or introducing replicates for statistical analysis. Currently, the use of XRF is limited by the time it takes to scan samples [14]. XRF methods provide qualitative results compared to the absolute quantitative concentrations of ICP-MS. However, calibration curves can be established. It is well known through many techniques, including XRF, that the elemental concentrations in wood are highly heterogeneous between trees, within a trunk, and rings [154,156,157]. A full scan of the elements present within an entire cross-section of a tree would give a visual representation of the changes across a section of the tree, something only feasible through the use of SXRF. The combination of more powerful techniques allowing for more samples to be processed to achieve a higher sample depth, as well as the ability of new techniques to provide different perspectives on tree systems, offers new areas for exploration.

4.3.2. Changes over Time and Space (x, y, and z Dimensions)

Dendrochemistry experiments and investigations have been ongoing for over five decades, but there is still considerable debate about the actual reliability of the results and what conclusions or hypotheses can be made. Many studies discussed in this review had only slightly changed the basic assumptions made when the first dendrochemistry experiments were conducted. For example, many studies still only take core samples from one or two locations on the tree, usually at breast height. Some studies have also taken whole cross-sectional samples (disks) and combined single ring-width years from different areas around the tree's circumference into a single bulk sample with enough material to run the mass of the sample through digestive techniques such as ICP-MS. However, few studies have considered the slight (ppm or ppb) changes in the concentrations of elements depending on the physiological structure of the tree that changes along the x, y, and z dimensions of the tree.

A synchrotron dendrochemistry experiment by Person [155] aimed to compare qualitative results from SXRF results to quantitative acid-digested ICP-MS results from the same tree. In brief, Person [155] took two core samples each from the same tree for white spruce and trembling aspen. One set of cores from each species was then cut for acid digestion and ICP-MS analysis into species-specific segments, annual increments for white spruce, and five-year increments for trembling aspen [155]. The second core of the same tree was scanned using SXRF. The white spruce core was scanned once in a single line, whereas the trembling aspen core underwent a more time-intensive 2D-map scan along the entire core [155]. The SXRF results were broken into bark, heartwood, and sapwood for easier comparison to the ICP-MS results. The outcomes from the ICP-MS and the SXRF techniques were compared and showed no correlation between the two techniques, but interestingly, an analysis of the 2D-map scan of the trembling aspen core showed sporadic pockets of high and low concentrations of nearly all elements analyzed across the two-dimensional space of the sample [155].

Other studies have also identified sporadic speckling of elemental hot spots in tree cores. Gavrikov et al. [156] conducted a recent study where four cores from the cardinal directions were taken from six individual Scots Pine (*Pinus Sylvestris* L.) trees from an unpolluted site. Using XRF analysis, cores from each tree were scanned and counts of 13 elements including Al, Cl, Ca, Mn, Cu, and Zn were recorded. Similarly to Person [155], the results were given in counts of fluorescence. When comparing cores from the same trees, Gavrikov et al. [156] saw minimal correlation between the cores. This result is consistent with the results from Peckham et al. [77] who recommended that cores be taken from multiple cardinal directions from each tree when studying atmospheric pollution. The results from Gavrikov et al. [156] also show the need for multiple cores from each tree regardless of the element being studied.

The fluorescent counts of Person [155] and Gavrikov et al. [156] do not describe the actual concentrations of elements in the samples, but rather the qualitative relative amounts of elements based on fluorescence. Rodriguez et al. [66] created control standards to calibrate and validate quantitative results from non-destructive XRF and destructive digestion methods. In brief, discs of loblolly pine (*Pinus taeda*) treated with fertilizers were collected at breast height, and sections of the same disc were either scanned with XRF or digested. Control standards were made from finely ground sawdust of control trees grown with no added fertilizers that were subsequently spiked with known concentrations of elements and pressed into pellets [66]. When tested with quantitative XRF and digestion techniques, the manufactured standards had significantly high correlations. However, even with the standards to help calibrate the quantitative results, the authors only saw a moderate correlation between the XRF and digestion results from the fertilized discs collected at breast height. Both techniques had similar overall trends but significantly different elemental concentrations [66]. Rodriguez et al. [66] hypothesized that this is likely due to the sampling methods, where a quarter of the disc was digested, and the remaining three-quarters of the same disc was used in the XRF analysis (see Rodriguez et al. [66] for further explanation on the methods used).

The lack of a correlation between ICP-MS and SXRF in Person [155] and the XRF scans by Gavrikov et al. [156], as well as the moderate correlation of disc samples between XRF and digestion methods in Rodriguez et al. [66], challenges the basic assumption in dendrochemistry that samples from the same tree can be used to validate two separate techniques or assumptions. Many studies have relied on the assumption that two samples taken within centimetres of each other would be similar enough to compare. This could explain the range in results of dendrochemistry studies and why it can be so difficult to reproduce results [77]. Another dimension often neglected is the impact of changes to elements over time, specifically when contamination or sampling occurs, as well as the time between the samples being collected and when they are processed. The aspect of time of sampling has been hinted at before but rarely explored in-depth [25]. In an early dendrochemistry study, Hagemeyer and Schafer [25] examined the radial mobility of Cd, Pb, and Zn at different times of the year in Beech trees (*Fagus sylvatica*). They found that for Pb and Zn, spring had the highest elemental concentration whereas for Cd, winter had the highest concentrations. These studies illustrate the need to understand how interior physiological discrepancies of the tree structure (e.g., damage or disease) can change elemental concentrations, limiting the ability to use different sections of the same sample. Future dendrochemistry studies should also consider the impacts of time on dendrochemistry samples.

4.3.3. A Call for More Lab-Based Experiments

As previously discussed, most dendrochemistry studies find an area of contamination and sample trees naturally growing in the environment. This opportunistic sampling is standard in science, and the data collected help us understand how systems function in the real world. However, if possible, research should also be conducted in a controlled setting to better understand the possible mechanics driving the system. Lab-based experiments are another area of dendrochemistry that should be explored in greater depth. Cutter and Guyette [26] were among the first to fully acknowledge the complexity of the natural environment trees grow in and the many factors that can influence how elements are taken up and retained within the tree system. Most of the studies in this review relied on taking samples of trees from known areas of contamination or environmental change. The samples are commonly used to verify the impacts and presence of contamination or other environmental stresses on the trees and their environment. Many of the factors affecting the uptake of elements by trees that are identified by Cutter and Guyette [26] can be resolved or, at the very least, mitigated through lab-based studies where researchers have greater control over the growing environment and the addition of or exposure to elements for the trees.

One of the first and few lab-based experimental studies in dendrochemistry is from Hagemeyer and Lohrie [158]. Hagemeyer and Lohrie [158] looked at how saplings grown in soil contaminated with Cd and Zn would respond after two years of growth. Two-hundred and fifty total saplings were used in the study, each potted in an individual container with increasing amounts of Cd and Zn added. The saplings were only watered when needed and grown outside until December when they were moved to a cold greenhouse for overwintering until March of the following year [158]. The results from Hagemeyer and Lohrie [158] illustrated a decrease in the radial growth in saplings in the highest levels of Cd and Zn. Levels of Cd and Zn present in the rings and wood of the saplings showed that as the amount of amendment increased, the more Cd and Zn the trees took up into the wood. At the highest levels of amendments of Zn and Cd+Zn, the saplings did not survive, indicating a threshold limit for growth as roughly 7500 µmol/kg of soil of Zn and 90 + 1000 µmol/kg of soil of Cd + Zn [158]. In most contamination studies involving heavy metals and their uptake into the trees, there is often no way of knowing the exact concentrations of heavy metals the trees would have been exposed to over their lifetime. The lack of known concentrations makes researching topics such as translocation and uptake mechanisms difficult. Lab-based studies on trees can be difficult, long-term, and expensive. However, lab-based studies could provide a greater understanding of the underlying environmental conditions and physiological processes impacting elemental uptake.

5. Conclusions

Dendrochemistry has grown substantially as over the last 50 years, and the interest and research in the field have grown considerably. From the past where one of the main focuses was on determining the impacts of forest acidification, to the present studies focusing on quantifying heavy metals and detecting dendroisotopes. However, greater consistency in the research methods and exploring new techniques could further improve the future of the field. We recommend the following measures: (1) promoting the use of new experimental techniques and methods with faster data acquisition time to allow for a greater number of samples to be processed and included in studies to increase statistical significance; (2) that more studies focus on the two and three-dimensional space that trees grow in and consider the complex physiological processes occurring in that space and over time; and (3) more lab-based studies to reduce the variables that cannot be controlled when sampling in situ. If taken into account, these three recommendations for future research can lead to a new era in dendrochemistry research and, hopefully, answer some of the fundamental questions about the processes governing how elements move through the chemical tree systems.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/f14101997/s1, PRISMA Flow diagram of search results for systematic literature review including the number of duplicates in the searches, and reasons for removal of articles from the literature review and number of articles removed. Spreadsheet showing the results of the WoSCC database search using three search terms and after removing results that did not apply to the research. Spreadsheet is organized by the first search of dendrochemistry (ordered oldest to newest) followed by the second search (oldest to newest) and finally the third search (oldest to newest). Other information includes the DOI, first author names, year published, primary affiliated country, journal, species studied, element(s) studied, and methods used.

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