A review of research hotspots and trends in biogenic volatile organic compounds (BVOCs) emissions combining bibliometrics with evolution tree methods

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Abstract

As ozone pollution in the troposphere has become increasingly severe, more publications have focused on the emissions of biogenic volatile organic compounds (BVOCs), which are important precursors for ozone formation. However, most reviews describe the research status of certain specific aspects rather than holistically quantifying research hotspots and development trends, which limit the overall understanding of BVOCs emissions. In this paper, bibliometric analysis was used to study the publication output and hotspots of BVOCs emission research from 1991 to 2019. Then, the evolutionary trends in BVOCs emission sources research were explored further by combining evolution tree and Markov chain methods. We found that the USA consistently took the leading position in BVOCs research, which cooperated with Germany and China closely. Environmental Science & Ecology and Meteorology & Atmospheric Sciences were the most active research subject categories. Current literatures mainly focused on the plant stress response, the atmospheric chemistry of BVOCs emissions, and their measurement by field determination and model-based estimation. Most publications researched BVOCs emitted by plants, in particular Pinaceae, while the growth of publications researching microbial volatile organic compounds (mVOCs) was slow. In the future, we should consider the role of mVOCs and combine field observation with model estimation to improve the credibility of BVOCs estimates and provide scientific guidance for air pollution control. And, with climate change, it will be worth exploring the driving variables of BVOCs emissions and its interaction in earth system to unravel how BVOCs emissions will respond to the changing earth system.

1. Introduction

Volatile organic compounds (VOCs) are a class of compounds characterized by small molecular mass (<300 Da), low boiling point (50 °C–260 °C), low octane value, high saturated vapor pressure and large Henry constant (Kaddes et al. 2019). They play important roles in the earth system, not only by mediating the intra- and interspecific interactions of organisms and responding to abiotic and biotic stress, but also by participating in atmospheric chemical processes (Kravitz 2016, Vivaldo et al. 2017, Huang et al. 2018, Kessler 2018, Nagashima et al. 2019). They are the important precursors in the formation of ozone ($O_3$), aerosol particle and particulate matter (Liu et al. 2019).

According to the IPCC report in 2013, annual average surface $O_3$ concentrations shown increasing
trends around the world in recent decades, and they increased significantly in Asia. The summertime daily maximum 8 h average (MDA8) O₃ levels were usually higher than 100 ppbv in 2013–2017 in China, and the MDA8 O₃ levels during 2016–2017 increased by approximately 20% compared to those in 2013–2014 (Lu et al 2019b). Accordingly, there is a growing concern about VOCs emissions, which are the percursor for O₃ production. VOCs can be released by biochemical processes or anthropogenic activities. Bio-genic volatile organic compounds (BVOCs) mainly derive from plants, animals and microorganisms, and anthropogenic volatile organic compounds (AVOCs) are emitted by motor vehicles, solvents, fossil fuel combustion etc. Annual BVOCs emissions are much larger than AVOCs emissions on a global scale (Guenther et al 2012, Sindelarova et al 2014), and BVOCs are more reactive than AVOCs (Chen et al 2018). Therefore, BVOCs make considerable contributions to tropospheric O₃ formation. Furthermore, with climate change, the emission of isoprene, the dominant BVOCs accounting for approximately 50% of the global flux, may increase by 20%–92% by 2052, and surface O₃ will increase by 19%–20% (Zhang et al 2008, Valolahiti et al 2015, Liu et al 2019). In conclusion, it is necessary and worthwhile to pay attention to BVOCs emissions.

Some studies reviewed BVOCs emissions from multiple perspectives, such as emission sources, detection technology, responses to land-use or global change, emission estimates, functions in forest ecosystem, and their roles in O₃ concentration (Dong 2016, Fu et al 2017, Kessler 2018, Simpraga et al 2019, Majchrzak et al 2020, Yanez-Serrano et al 2020). However, previous researches usually focused on certain specific aspects rather than a comprehensive view of BVOCs emissions, and traditional reviews often neglected to explore the development of research trends. Bibliometric analysis of published data and literature was an effective and innovative method for understanding the pattern and development of a specific field. Quantitative analysis could clarify the research hotspots in BVOCs emissions accurately and comprehensively. But scientometric analysis could not clarify the evolution trends specifically. Combining evolution tree and Markov chain methods, we could discover the development trends of research and forecast prospects quantitatively. Further, such a research combining bibliometrics with evolution tree analysis can not only help researchers understand various aspects of BVOCs emissions across multiple disciplines, but also identify its hotspots and further directions.

In this review, we aimed to provide comprehensive information on BVOCs emissions research from 1991 to 2019. Bibliometric analysis was used to investigate the publication output and hotspots based on keywords cluster. Evolution tree was applied to explore the evolution of BVOCs emission sources research, and Markov chain based on the evolution tree was performed to study the probability of transition from 1991–2005 to 2006–2019. And we used meta-analysis to quantify the response of BVOCs emissions to environmental factors. Our findings could lead researchers to insight into key issues and provide theoretical guidance for scientific decisions.

2. Method and data

2.1. Data source and process
To research the development of BVOCs emissions, peer-reviewed scientific publications were retrieved from the Science Citation Index Expanded (SCIE) and Social Sciences Citation Index (SSCI) databases based on Web of Science Core Collection (WOS). The WOS was chosen, because it is considered to be relatively authoritative and representative. Both the SCIE and SSCI databases cover a large number of journals across most scientific disciplines (Cheng et al 2019). Our search query in the databases was 'TS = (volatile organic compound∗ OR VOC∗) AND (biogenic OR plant OR vegetation OR forest OR microbial OR fungal OR bacterial OR animal) AND emission∗'. The terms were contained in titles, abstracts and keywords. The document types were limited to article and review, which accounted for 95.19% of the total publications, and excluded proceedings paper, book chapter etc. The search period began in 1991, because the publication number was <5 prior to that year, and the search reported publications up to 15 November 2019. In total, 4697 raw data items were recorded. There were inevitably irrelevant and duplicative publications, so it was necessary to refine the raw data to increase accuracy of the units of bibliometric analysis (Braun et al 2019, Hossard 2019). First, we excluded irrelevant records by going through the titles and abstracts of all the preliminary records manually, such as ‘power plant’ and ‘industrial wastewater plant’, which mean buildings or places for carrying out industrial processes rather than living organisms growing in the earth. Next, we used BibExcel, which can identify repeated literatures based on the title and the journal field, to eliminate duplicate records (Yu et al 2020). In the end, 3709 publications were obtained and considered for bibliometric and evolution tree analysis.

In addition, meta-analysis was used to quantify the response of BVOCs emissions to environmental factors (strengthen light, warming, drought and elevated O₃). The data processing and analysis method were presented in Supplement S1.

2.2. Bibliometric analysis
Bibliometric analysis is an effective research method in many fields that adopts both qualitative and quantitative analyses to measure patterns in scientific publications as well as to evaluate research hotspots.
Yao et al., used to analyze bibliometric networks and analysis. Then, VOSviewer, a Java programming language, was used to analyze bibliometric networks and to create, visualize and explore maps based on network data (Martinez et al., Miao et al., Yao et al.). We applied item density visualization to create a map of inter-country collaboration. The label size of each country is determined by the weight of the total link strength. The higher the weight is, the larger the label of the item. The more neighboring items there are and the higher the weights of the items in the neighborhood of the point, the closer the color is to red. Inversely, the point color is closer to white. We used network visualization to display co-authorship and keywords occurrences. The size of the circle and label for each author is determined by the weight of the total link strength. The size of the keyword depends on the weight of the item. The color of the item depends on the cluster to which it belongs. Lines between items represent links. The distance between items approximately indicate their relatedness.

2.3. Evolution tree
The evolution tree method was first introduced by Wang et al. (2011) and applied to explore city evolutionary pathways. It can be used to clarify development trends, and express the possible mechanism associations and evolutionary variations in multidimensional data in a simple and clear visual form (Wang et al., Liao, Hu et al.).

To better understand the evolution of BVOCs emission sources research, the research period was split into two time intervals (1991–2005 and 2006–2019) based on the annual publication volume researching on BVOCs emission sources. We used Geotree to visualize the evolution of the number of publications during 1991–2005 and 2006–2019, then established a Markov chain to study the probability of transition from 1991–2005 to 2006–2019 based on the evolution tree. In the evolution tree, the trunk represented the BVOCs emission sources, which were divided into three types, plant, animal and microbe, and were denoted as I, II and III respectively. The branches represented the order of magnitude of publications, which were divided into six sections to better display the evolution in the number of publications, $\{0, 1\}, \{1, 3\}, \{3, 10\}, \{10, 40\}, \{40, 80\}$ and $\{80, 130\}$, respectively. The arrows start at the magnitudes of publication during 1991–2005 and point toward the magnitudes of publication during 2006–2019. The numbers in brackets indicate the number of families in which a change in the order of magnitude took place and the average rate compared with the number of publications during 1991–2005.

3. Publication output
3.1. Distribution of publications and citations by year
The dataset on the topic of BVOCs emissions in this paper consisted of 3709 publications up to 15 November 2019. The number of publications showed an upward trend as a whole, from 12 in 1991–327 in 2018. The literature volume in 2018 was more than 27 times that in 1991. The average number of publications per year was 121. The annual growth rate of publications was 23.75% approximately (figure S1, which is available online at stacks.iop.org/ERL/16/013003/mmedia). The predicted number of publications was 915 in 2050 (figure S2). The 3709 publications were cited 120,870 times until the retrieval date. The number of citations on BVOCs emissions presented an irregular trend from 1991 to 2010. The number of citations in 1995 was significantly higher than that in 1994 and 1996, which may be mainly due to the publication of several highly cited publications in 1995 (table S1). Similar to the citations in 1995, the numbers of citations in 1999 and 2006 were also obviously higher than those in the adjacent years. There was a negative tendency in the number of citations from 2010 to 2018. The number of citations peaked in 2010 with 8619 citations. The average cited times per published item was 33 (figure S1). As a result, we found that research on BVOCs emissions received increasing attention.

3.2. Distribution of publications in countries and inter-country collaborations
There was spatial heterogeneity in the publication distribution among countries. 88 countries contributed to the 3709 publications on BVOCs emissions. Most of these countries were in Europe and Asia, and these accounted for 67.05% of the total countries. The United States of America had the highest publication output, with 1238 publications, accounting for approximately 33.38% of the total. Germany and China contributed approximately 15.31% and 12.08% of the total with 568 and 448 publications respectively. They were followed by Finland, Italy, the United Kingdom, France, Spain, Austria, and Canada (figure S3). The publication numbers of the top 10 most productive countries presented an increasing trend from 1991 to 2018. The growth of Austria and
Canada was slower than that of other countries. Notably, the publication volume from China increased rapidly starting in 2011, while the publications from the USA increased slowly. However, the USA took the leading position consistently in the research field from 1991 to 2018 (figure S4).

Among the 88 countries contributing to the topic of BVOCs emissions, 55 countries collaborated with others. The USA and Germany emerged clearly as current cooperation centers, with 1036 and 799 for their total link strength, respectively. Researchers from the USA cooperated with counterparts from 51 countries, such as Germany and China. When the USA worked with Germany, the researchers published the most related literature, with 116 publications. This was followed by the USA—China collaboration, which resulted in 104 publications. Researchers from Germany cooperated with counterparts from 43 countries, such as the USA, Finland and France. Researchers in Germany and the USA had close collaborations and high publication output, while researchers from Germany had relatively little cooperation with counterparts from China, publishing only 30 publications collaboratively (figure S5).

3.3. Distribution of publications in subject categories and journals

The topic of BVOCs emissions attracted attention from researchers in various research areas. The publications were distributed in 77 subject categories. Environmental Science & Ecology and Meteorology & Atmospheric Sciences were the most active subject categories, with 2048 and 1245 publications respectively. They were followed by Plant Sciences. The publications in Environmental Science & Ecology, Meteorology & Atmospheric Sciences and Plant Sciences increased rapidly from 1991 to 2018, while the publications in Agriculture, Geology and Biotechnology & Applied Microbiology increased slowly. Notably, Environmental Science & Ecology and Meteorology took the leading position consistently from 1991 to 2018 (figure 1).

The 3709 literatures studied on BVOCs emissions were published in 646 journals. The top 10 most active journals on the topic accounted for approximately 39.39% of the studies, with 1461 publications. Atmospheric Environment had the most publications of related research, with 417 publications accounting for about 11.24% of the total. These studies mainly covered BVOCs emissions, chemical processes, and impacts on air quality, ecosystem and human health. The second major international journal was Atmospheric Chemistry and Physics, with 374 publications accounting for 10.08% of the total. These studies covered field measurement, BVOCs models, and biospheric interactions. This was followed by Journal of Geophysical Research-Atmosphere and Science of the Total Environment (figure S6).

3.4. Academic impact of publication output and co-authorship

There were 10 760 authors who contributed to the 3709 publications on BVOCs emissions. The top 10 most popular authors were Guenther, Goldstein, etc. It was worth noting that Guenther, an American scientist, was the leader in the related fields, contributing 130 publications with 12 336 citations. Among all authors, his h-index was the highest, at 51. The performances of the nine following authors were not obviously different, with h-indexes ranging from 28 to 34 (table S2). In addition, the most cited literature was published by Guenther in Journal of Geophysical Research -Atmospheres in 1995, with 2628 citations (Guenther 1995). Two of the top ten most cited publications were published in Atmospheric Environment (Sillman 1999, Grell et al 2005), and two of them were published in Atmospheric Chemistry and Physics (Guenther 2006, Ervens et al 2011) (table S1).

Extensive collaborations by the main authors researching on BVOCs emissions were visualized in figure S7. There were five clusters. In the red cluster, Guenther was the most popular author, with 511 for the total link strength. This was followed by Karl and Goldstein, with 339 and 336 for the total link strength respectively. They concentrated on meteorological atmospheric sciences, mainly modeling BVOCs emissions. In the green cluster, Hansel was the most active author, with 321 for the total link strength. He cooperated closely with Graus and Schnitzhofer. They focused on environmental science ecology, mainly researching plant VOCs emissions. In the yellow cluster, Kulmala was the most active author, with 382 for the total link strength. This was followed by Williams, with 276 for the total link strength. They primarily researched the VOCs emitted by boreal forests. In the blue cluster, Hewitt was the most popular author, with 237 for the total link strength. He collaborated closely with Langford and Owen. They concentrated on the monitoring and analysis of BVOCs emissions. In the purple cluster, Holopainen was the most popular author, with 220 for the total link strength. He cooperated closely with Blande and Kivimaenpaa. They mainly researched the stress response and function of plant VOCs emissions.

4. Evolution of BVOCs emission sources research

BVOCs emission sources can be divided into plants, animals and microbes. In both 1991–2005 and 2006–2019, most publications researched BVOCs emitted by Pinaceae (figures 2 and S8). During 2006–2019, there were 736 articles involving BVOCs emission sources in total. Among them, most articles researched BVOCs derived from plants, with 651 articles researching 83 families. They were followed by animals and microorganisms, with 93 and 69 articles researching 43 and 40 families respectively. For
plants, 129 articles, accounting for 19.82%, contributed to research on BVOCs emitted by Pinaceae, mainly *Pinus*, *Picea* and *Abies*. Pinaceae was followed by Gramineae, with 74 articles accounting for 10.05%, on species such as *Zea*, *Triticum* and *Hordeum*. However, only 6.62% of articles researched BVOCs emitted by Gramineae during 1991–2005. In this period, 33.82% of articles contributed to the study of BVOCs emitted by Pinaceae, followed by Fagaceae with 17.65%. For animals, 8 articles during 1991–2005 researched 7 families, such as Noctuidae, Suidae and Bovidae. During 2006–2019, 15 articles contributed to the study of BVOCs emitted by Bovidae, followed by Aphididae and Noctuidae with 12 and 10 articles respectively. In addition, 28 families, accounting for 65.12% of the total families, were studied in only one article each. For microorganisms, five articles researching seven families, such as Pseudomonadaceae and Enterobacteriaceae, were published during 1991–2005. During 2006–2019, Bacillaceae was the most researched, with nine articles accounting for 13.04% of the total articles. They were followed by Enterobacteriaceae and Moniliaceae, with six articles accounting for 8.70% respectively. In addition, 22 families, accounting for 55% of the total families, were studied in only one article each (figures 2 and S8).

From 1991–2005 to 2006–2019, most families showed an increase in their number of publications. For plants, the magnitude of publication for 78 families increased, that for 11 families decreased, and that for 4 families remained the same. Compositae showed the greatest rate of change (figure 3). Compared to that in 1991–2005, the number of publications increased overall during 2006–2019. The average rates of publications researching VOCs emitted by plants, animals and microorganisms were 2.66, 2.93 and 0.64 respectively. To date, the growth in research on microbial volatile organic compounds (mVOCs) is still slow.

Many studies have examined BVOCs derived from plants and their interactions with herbivores, while few studies have examined the BVOCs emitted by microorganisms. Microbes, including bacteria, fungi, viruses, protists and archaea, were the second largest biomass component, accounting for approximately 17.09% of the global biomass, behind plants, with approximately 82.54% of the global biomass (Bar-On et al. 2018). The microbial number and diversity far exceeded those of other organisms. Globally, microorganisms comprise approximately $10^{30}$ cells and more than $10^{12}$ species (Stolz 2016). Plant surfaces harbor plentiful bacteria and fungi. Each $m^2$ leaf surface was estimated to harbor $10^{11}$ bacteria (Penuelas et al. 2014, Gerard 2016). Some studies have shown that BVOCs of microbial origin may modulate the plant growth patterns and affect plant VOCs emissions (Gerard et al. 2016, Schulz-Bohm et al. 2017, 2018, Schenkel et al. 2018, Tyagi et al. 2018). However, there has been little research on the regulatory mechanisms of mVOCs. It is worth studying mVOCs perception mechanisms and determining their concentrations either as deterrents or as signaling compounds in ecosystems.

5. Hot spots

5.1. VOCs emissions from plant stress response-cluster1

Plant VOCs emissions were caused by mechanical damage or herbivory, by microbial plant pathogens, and by abiotic stresses such as strengthen light,
Figure 2. The evolution tree of publications with research on BVOCs emissions in 2006–2019.

Figure 3. Markov chain of research on BVOCs emission sources.
warming, drought and elevated ozone (Ameye et al 2018) (figures 4 and 5).

VOCs, functioning as pheromones, mediated intra- and interspecific interactions of organisms in ecosystems (Baldwin 2006, Tumlinson 2014). Plant emitted a blend of VOCs from storage structures such as resin canals and glandular trichomes after mechanical damage or feeding damage by herbivores (Murgun et al 2016). The constitutive and induced VOCs provided important information about the status of the damaged plant to surrounding organisms (Simpraga et al 2019). The neighboring undamaged plants may enhance the resistance to herbivores by active or passive plant-plant interactions (Li et al 2015). Many studies indicated that damaged bark tended to increase monoterpene emissions (Heijari et al 2011, Kovalchuk et al 2015). In addition, microbial plant pathogens may suppress or increase plant VOCs emissions by eliciting plant defensive responses (Gerard et al 2016, Sharifi et al 2018). Isoprene emission decreased when Populus balsamifera var. suaveolens (Fisch.) Loudon, infected with the fungal pathogen Melampsora larici-populina Kleb. (Jiang et al 2016), and Populus nigra trees infected with the rust fungus Melampsora laricipopulina reduced their VOCs emissions significantly (Eberl et al 2018); Betula pendula infected with the fungus Marssonina betulae significantly increased their emissions of (Z)-ocimene and (E)-b-ocimene (Vuorinen et al 2007). Among these, terpenoids were emitted in large amounts as defensive VOCs after trees were infected with fungi.

Environmental factors may regulate VOCs emissions (Penuelas et al 2010). Meta-analysis quantifying the mean effect sizes of strengthen light, warming, drought and elevated O$_3$ on the emissions of monoterpene, isoprene and total VOCs were conducted (figure 6). Strengthen light significantly increased the emissions of monoterpene, isoprene and total VOCs. It suggested that these emissions may depend on stomatal conductance or the rate of de novo synthesis (Lindwall et al 2015). Warming increased the emissions of isoprene and total VOCs significantly. It was likely that temperature affected the saturation vapor and diffusion rates of compounds from storage pools (Tang et al 2018, Faiola et al 2020). However, it was not significant to increase the monoterpene emission. This was largely due to the extent of experimental warming (Valolmatch et al 2015, Kramshoj et al 2016). Drought increased monoterpene emissions significantly. This agreed with the findings of Staudt and Copolovici (Staudt et al 2008, Copolovici et al 2014). Drought decreased the emissions of total VOCs significantly. As plant VOCs biosynthesis depended on metabolites, which were primarily formed by photosynthesis, the emissions decreased when drought stress inhibited primary metabolism (Eller et al 2016, Feng et al 2019). Drought stress had positive or negative effect on the emission of isoprene differing by plant species (Federico et al 2007, Sarah et al 2016, Yuan et al 2016, Saunier et al 2017, Parveen et al 2018). Elevated O$_3$ increased the emissions of total VOCs significantly, while the isoprene emission decreased significantly. This was due to the damage to photosynthesis providing carbon for isoprene biosynthesis by chronic exposure to O$_3$ (Agathokleous et al 2018). Elevated O$_3$ increased the emission of monoterpene, but the increase was not significant. This was because monoterpene emission was usually associated with O$_3$ tolerance, and different plants had different resistances to O$_3$ (Feng et al 2019). The abiotic environment can affect plant physiology, chemical characteristics and enzyme activity and control BVOCs emissions, meanwhile abiotic factors can also affect herbivorous physiology, activity, distribution, feeding behavior and their capacity to induce biotic stress on plants (Simpraga et al 2019). Furthermore, plants can tolerate environmental stress through VOCs catalysis. Highly reactive VOCs can protect plant tissue from environmental stress by destroying free radicals (Okawa and Lerdau 2013).

To date, many articles have reported the plant VOCs emissions under biotic stress and abiotic stress. However, few studies have examined the interactions of plant VOCs emissions with biotic and abiotic factors. What will happen to plant VOCs emissions under long-term interactions? It is necessary to focus on the driving variables of VOCs emissions and its interactions with biotic and abiotic factors, and to unravel the mechanisms underlying plant VOCs perception and signaling.

5.2. Atmospheric chemistry of BVOCs emissions-cluster2
BVOCs play fundamental roles in atmospheric chemical processes. They can form O$_3$, aerosol particles and particulate matters by reacting with anthropogenic NO$_x$ under illumination (Guidolotti et al 2019, Liu et al 2019, Sporre et al 2019). BVOCs, with a high potential to create photochemical O$_3$ form or degrade O$_3$ by altering the NO$_x$ cycle. VOCs react with OH radicals generating peroxide radicals, and continuously produce RO$_2$ and HO$_2$ and oxidize NO to NO$_2$. With the increase in NO$_2$ concentration, the O$_3$ concentration increase consequently. In addition, in the polluted troposphere, O$_3$ reacts with unsaturated VOCs producing OH radicals, and contributes to the RO$_x$ cycle and NO$_x$ cycle (Calfapietra et al 2009, Xue et al 2016, Wang et al 2017) (figure 5).

O$_3$ production has a nonlinear dependence on NO$_x$ and VOCs. The entire range of O$_3$ formation sensitivity can be divided into VOCs-limited, NO$_x$-limited and transition regions. Some researchers have studied the ratio of VOCs/NO$_x$, O$_3$/NO$_x$, or H$_2$O$_2$/NO$_x$ to determine the relationship among O$_3$ and precursors (Lu et al 2019a). At low VOCs/NO$_x$, generally below 4, the intensity of the RO$_x$ cycle is
weaker than that of the NO\textsubscript{x} cycle, hence O\textsubscript{3} formation is VOCs-limited. When the ratio is higher than 15, with high VOCs/NO\textsubscript{x}, the NO\textsubscript{x} cycle becomes the primary limiting factor in O\textsubscript{3} formation, so it is NO\textsubscript{x}-limited. When the levels of NO\textsubscript{x} and VOCs are comparable, O\textsubscript{3} formation is limited by both VOCs and NO\textsubscript{x}. Namely, in the transition region, the relative abundances of both precursors should be controlled (Carrillo-Torres et al 2017, Wang et al 2017, Kashyap et al 2019). These methods are simple and straightforward, but they cannot analyze the mechanism of O\textsubscript{3} formation comprehensively. Several models have been developed to provide sensitivity modeling analysis by assuming the reduction in the precursor concentration, such as observation-based models and emission-based models. These models quantify the sensitivity of O\textsubscript{3} formation to various precursors (Carrillo-Torres et al 2017, Wang et al 2017, Kashyap et al 2019).

Up to now, the O\textsubscript{3} formation processes with VOCs as precursors has been well understood. And, many publications studied the influences of climate change on BVOC emissions by model simulation. However, few studies have reported on what extent BVOCs contribute to ozone formation with climate change. In the future, BVOCs emissions and photochemical O\textsubscript{3} creation potentials in different areas, such as forest, thicket and herbaceous vegetation, will need to be considered. This will be helpful for air pollution control by clarifying the spatial heterogeneity of the limiting factors of O\textsubscript{3} formation.

5.3. BVOCs determination and identification-cluster3

BVOCs emissions measurements are primarily based on BVOCs determination and identification. Usually, enclosure techniques are used to treat samples, mainly including static, dynamic and semi-static enclosure methods (Li et al 2019). In the 1960s–1980s, static enclosure techniques were used to measure BVOCs emissions. In a static chamber without airflow, the concentration of BVOCs changes over time. In general, at several time points, air samples are determined by solid-phase microextraction (SPME). Using the SPME technique, VOCs can be adsorbed and concentrated on a solid or liquid polymer layer by special devices. SPME is simple and rapid relatively, but its devices were too expensive to be widely used (Barreira et al 2015, Misharina et al 2017). Dynamic enclosure system is recommended for accurate measurement. This system has an inlet and outlet to flush the enclosure with clean air continually, which is more representative of the natural environment. The emission samples are usually collected by adsorbents, such as Tenax TA, Tenax GR, Carbotrap and Carbograph. Most studies used one or two adsorbents, so it may sample part of the VOCs and impact emission results (Ortega et al 2008, Materic et al 2015). Li et al (2019) proposed the semi-static system firstly, which inputs a large amount of zero air into the enclosure chamber but has no outflow in order to minimize the environmental variations within the chamber. The sample from the enclosure is collected into bottle canister instantly. This method has the advantages of simplicity and short performance time. In addition, micrometeorological techniques, such as eddy covariance and surface layer gradient, can measure the BVOCs flux at the ecosystem scale over a long period (Park et al 2013, Karl et al 2014, Rinne et al 2016).

BVOCs can be identified by gas chromatography (GC), GC-mass spectrometry (GC-MS) or proton transfer reaction-MS (PTR-MS). Usually, GC or GC-MS are used to identify VOCs. GC analyzes BVOCs qualitatively and quantitatively, and GC-MS separates terpene isomers quantitatively and it possessed of high specificity and sensitivity. However, they are not real-time instruments. In the early 1990s, Lindering
Figure 5. Schematic diagram of BVOCs emissions and their effects on the biosphere and atmosphere.

Figure 6. Mean effect sizes of environmental factors (strengthen light, warming, drought and elevated O$_3$) on the emissions of monoterpene, isoprene and total VOCs.
et al (1998) developed a PTR-MS system based on the reactions of \( \text{H}_3\text{O}^+ \). PTR-MS is suitable for the online monitoring of BVOCs with high mass and time resolution (Park et al 2013, Barreira et al 2015, Materic et al 2015, Mermet et al 2019).

BVOCs determination and identification techniques have developed continuously. Notably, the measurement method should be chosen carefully considering the research goal and the experimental conditions.

5.4. BVOCs emissions models-cluster4

The models of BVOCs emissions continue to develop. In the early stages, some studies described the linear relationship between light, temperature and BVOCs emissions. In 1960, Went quantified BVOCs emissions into the atmosphere and first estimated global annual monoterpene emissions. Soon after, the BVOCs emissions models developed at multiple scales and improved of the emission mechanisms constantly. These models include the Guenther model, the biosphere emissions inventory system (BEIS), the model of emissions of gases and aerosols from nature (MEGAN) and its derivative models. The Guenther model has been widely used to estimate the natural source VOCs emissions. In 1991 and 1993, Guenther introduced leaf-scale BVOCs emissions models, which were known as G91 and G93 (Guenther et al 1991, 1993). Then, he developed the first global BVOCs emissions model (G95) in 1995, which focused on the emissions of monoterpene and isoprene, and estimated other BVOCs emissions roughly. G95 include the basal emission rate, the mean leaf biomass monthly and the activity factor related to light and temperature. However, G95 does not consider other controlling factors, such as drought and \( \text{CO}_2 \). Moreover, G95 inputs monthly average data, which are too rough to show patterns in atmospheric chemistry (Guenther et al 1995). BEIS estimates the BVOCs emissions over the Americas and was developed by the US Environmental Protection Agency (EPA). Furthermore, BEIS was developed for the global biosphere emissions and interactions system (GloBEIS) by the US EPA and the National Center of Atmospheric Research (NCAR). GloBEIS considers the basal emission rate, the fraction and annual peak of leaf biomass, and the activity factor that includes photosynthetic photon flux density, temperature, leaf development status and chemical transformation and loss. Considering the BVOCs transformation and loss through the canopy, GloBEIS developed the canopy radiative transfer model based on G95. However, the input of GloBEIS is not easy to prepare, in particular as it includes data on 270 vegetation types. Accordingly, GloBEIS has not been used widely (Wang et al 2016). MEGAN, developed by Guenther in 2002, estimates BVOCs emissions regionally and globally. MEGANv2, proposed in 2006, included the basal emission rate, the activity factor of canopy chemical transformation and loss and the emission activity factor related to the photosynthetic photon flux density, temperature, leaf development status, soil moisture and leaf area index (Guenther et al 2006, 2012). MEGANv3, the latest version released in 2018, considered additional VOCs, emission categories and controlling processes based on MEGANv2. MEGAN has been used widely and its algorithms have been coded into some atmospheric chemistry and transport models, such as the Weather Research Forecast (WRF-Chem), the Goddard Earth Observing System with Chemistry Model (GEOS-Chem), and Community Multi-scale Air Quality Model (CAMQ) (Bauwens et al 2018, Chen et al 2018, Wang et al 2018, Liu et al 2019).

BVOCs emissions modeling is a key approach to study BVOCs emissions. These models estimate the BVOCs emission rates to improve control strategies for air quality. However, most researches have modeled the BVOCs emission rates in global and regional scale, in large part due to the absence of high-resolution datasets, while the research results are difficult to provide guidance for localized air pollution control. And, many studies have investigated only a portion of tree species to measure the basal emission rate for models. This increases the uncertainty of the models. Therefore, more BVOCs measurements may need to be conducted accurately to improve model credibility. In addition, plants are ubiquitously exposed to a variety of microorganisms. mVOCs emitted by microbes, a kind of BVOCs, interact with plants. Therefore, it was necessary to consider the effects of microorganisms and mVOCs on BVOCs emission models.

6. Summary and outlook

BVOCs emissions have attracted increasing attention. Environmental Science & Ecology and Meteorology & Atmospheric Sciences are the most active research subject categories. The USA, which has cooperated with Germany and China closely, consistently produces the most published research in this research field. In particular, Guenther in the USA is the most popular scientist, and concentrates on meteorological atmospheric science and modeling BVOCs emissions.

The emission sources for BVOCs are plants, animals and microorganisms. Most scientists focus on BVOCs emitted by plants, in particular Pinaceae. From 1991–2005 to 2006–2019, publications on most plant families showed growth, while the growth of mVOCs research was slower than that of research on BVOCs derived from plants and animals. Considering that microbes possess enormous biomass and diversity, and that mVOCs may modulate plant growth patterns and affect plant VOCs emissions, it is worth considering mVOCs and their regulatory mechanisms.
BVOCs emissions measurements can be conducted by field determination and by model-based estimation. In field measurements, dynamic enclosure techniques are often used to treat samples, and the BVOCs are identified by GC, GC-MS or PTR-MS. Many models can estimate BVOCs emissions, such as G95, BEIS and MEGAN. However, most studies estimated the BVOCs emissions based on unobservant basal emission rate and less-detailed vegetation distribution, which caused model uncertainty largely. Therefore, more field observations should be conducted to improve model credibility. Further, model-based BVOCs emissions accurately could provide theoretical guidance for air pollution control.

BVOCs play important roles in the biosphere and atmosphere. They can be emitted to mediate intra- and inter-specific interactions in the ecosystem, when plants are subjected to mechanical damage, herbivory or microbial plant pathogens, as well as abiotic stresses such as strengthen light, warming, drought and elevated ozone. And, BVOCs take part in atmospheric chemical processes, and are the important precursors of O₃ and particulate matters. Both VOCs and NOₓ nonlinearly determine O₃ formation. In the future, we should characterize the driving variables and controlling factors of BVOCs emissions in earth system, and determine how BVOCs emissions will respond to the changing earth system.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary information files).

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Conflict of 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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