Comparison of biogenic volatile organic compound emissions from broad leaved and coniferous trees in Turkey

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Abstract

Biogenic volatile organic compound (BVOC) emissions from thirty-eight tree species (twenty broad leaved and eighteen coniferous) grown in Turkey were measured. BVOC samples were collected with a specialized dynamic enclosure technique in forest areas where these tree species are naturally grown. In this method, the branches were enclosed in transparent nalofan bags maintaining their natural conditions and avoiding any source of stress. The air samples from the inlet and outlet of the bags were collected on an adsorbent tube containing Tenax. Samples were analyzed using a thermal desorption (TD) and gas chromatography mass spectrometry (GC/MS) system. Sixty-five BVOC compounds were analyzed in five major groups: isoprene, monoterpenes, sesquiterpens, oxygenated sesquiterpenes and other oxygenated VOCs. Emission factors were calculated and adjusted to standard conditions (1000 μmol/m^2 s photosynthetically active radiation-PAR and 30°C temperature). Consistent with the literature, broad leaved trees emitted mainly isoprene while the coniferous trees emitted mainly monoterpenes. Even though fir species are coniferous trees, they emitted significant amounts of isoprene in addition to monoterpenes. Oak species showed a large inter-species variability in their emissions. Pine species emitted mainly monoterpenes and substantial amounts of oxygenated compounds.
Keywords: BVOC emissions, dynamic enclosure system, emission factor, Turkey.

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1 Introduction

Gas exchange between atmosphere and biosphere makes these mediums strongly interlinked components of the Earth's ecosystem while most of the organisms synthesize and emit various biogenic volatile organic compounds (BVOCs). Plant based emissions are the most prominent BVOCs as they have widely investigated by researchers in recent studies [1–5]. It was estimated that the global BVOC emissions from plants accounts for 1,150 Tg C/year, almost twice the anthropogenic VOC emissions (Guenther *et al*. [6]). Forest areas cover 31.1% of the Earth's surface and also 27% of Turkey's (215,370 km^2) [7, 8] making them significant BVOC sources. Therefore, specifying the emission factors of the trees is crucial. Wilske *et al.* [9] investigated isoprenoid emissions of eight tropical tree species of Southeastern Asia, and also Bao *et al.* [10] screened ten Japanese plants for their biogenic emissions and many other species were investigated within numerous of studies and several well-organized BVOC measurement projects such as; ECHO (Emission and Chemical Transformation of Biogenic Volatile Organic Compounds) (Spirig *et al.* [11]), BEMA (Biogenic Emissions in the Mediterranean Area) (Versino [12]). A large number of plants were studied and the emission factors are available on the relevant databases.

BVOCs; including isoprene, terpenes (monoterpenes, sesquiterpenes) and oxygenated VOCs (alcohols, aldehydes, ketones, and acetates) are important for atmospheric chemistry since they contribute to secondary organic aerosol formation and play an important role in the oxidative capacity of the atmosphere [13, 14]. Furthermore, BVOCs have effects on the chemical composition and physical characteristics of the atmosphere as they photochemically react with nitrogen oxides (NO_x) and forming tropospheric ozone [15–17]. Therefore, leaf/needle, branch, canopy or ecosystem level measurements with various methods and equipments are being conducted to determine the biogenic emissions in global or regional scales around the world [18–20].

Vegetative emissions have a large inter-species and temporal/seasonal variability [21]. Thus the reliability and the comparability of the reported values are biased by the diverse experimental conditions used. Previously, researchers have determined biogenic emissions at leaf/needle or branch scale mostly using the dynamic enclosure method [4]. In this method, a leaf/needle, branch or a whole plant is enclosed in a transparent chamber. The main requirements for an enclosure system are maintaining inside-chamber conditions (e.g. temperature, CO_2 concentration, humidity, photosynthetically active radiation [PAR]) similar to ambient air. Tambunan *et al.* [3] measured isoprene emission from 42 indigenous and exotic tropical trees in subtropic Okinawa, Japan, Sabillon and Cremades [22] investigated monoterpene emissions from two of the most typical Mediterranean tree species (*Quercus ilex* and *Pinus pinea*) and Padhy and Varshney [23] measured isoprene emissions of nine commonly growing tree species in Delhi, India using the dynamic enclosure technique. A number of analyzed BVOCs in literature were generally limited because of the experimental and analytical difficulties. Measurement procedures were performed in related studies focusing on variables such as, species, season, plant tissues, measurement

techniques, environmental conditions (e.g. temperature, humidity, PAR, CO_2) directly affecting biogenic emissions [24, 25]. Harrison *et al.* [26] determined both monoterpene and isoprene emissions from a fir species (*Abies borisii-regis*), addressing PAR and temperature effects on emissions and Vuorinen *et al.* [27] discussed the effects of ambient CO_2 and O_3 concentrations on BVOC emissions of silver birch trees. As commonly reported in literature, isoprene and monoterpenes are the most abundant BVOCs. Isoprene is mostly produced photosynthetically by broad-leaved trees and its emissions depend on light intensity and temperature while monoterpenes from coniferous species predominantly depend on temperature [10, 28, 29]. However, for several tree species unexpected emission patterns were determined. For instance a fir species (*Abies borisii-regis*) found to be a strong isoprene emitter Harrison *et al.* [26] and Holly Oak (*Quercus ilex*) emitted monoterpenes as much as typical coniferous trees.

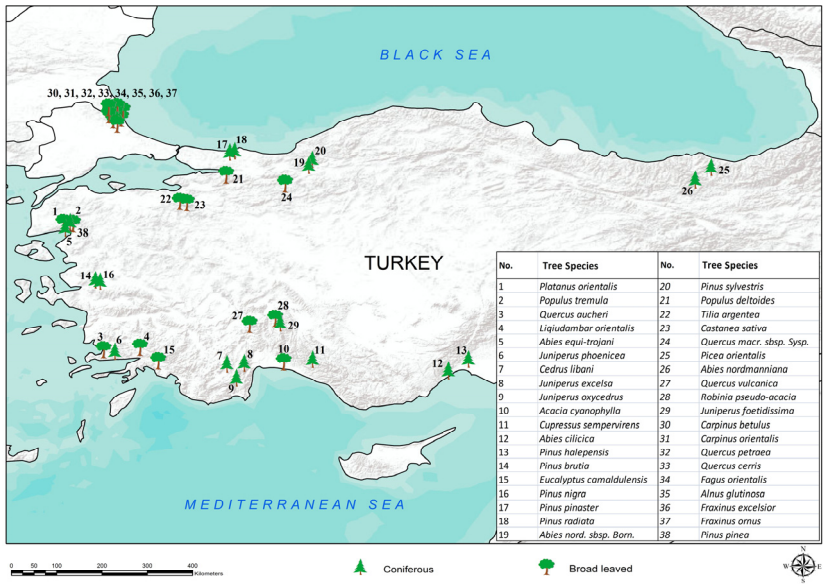
There is no investigation that directly focused on BVOC emissions in Turkey's forests. In this study, 15 field campaigns were performed for 38 tree species in different regions of Turkey during two summers (2011–2012) to measure BVOC emissions. The main objectives of the present study were, (i) to determine the BVOC (isoprene, monoterpenes, sesquiterpenes, oxygenated compounds and others) emission factors from 38 tree species those naturally and widely grown in Turkey and (ii) to investigate the BVOC compositions and compare the emissions from broad-leaved and coniferous species.

2 Materials and methods

2.1 Sampling program and collection

On-site measurements were conducted between June and October 2011, and May and November 2012 in different forest areas where the natural habitats of 38 trees are located. Trees including 31 common and 7 endemic species (18 of coniferous and 20 broad-leaved trees) representing the biological tree diversity of Turkey were sampled (Figure 1). Three middle-aged healthy (i.e., no pest or diseases affected, well-developed trees, not obstructed by neighboring plants, favorable soil conditions and no other biotic/abiotic stress sources) representative-trees of each species were selected for sampling. Measurements were conducted during daytime and collected samples were kept at 4°C until they were analyzed. BVOC samples collected simultaneously from two different branches of each tree. Thus, the emission rates for each species were calculated by averaging six values (from 3 trees x 2 branches) for each BVOC.

BVOCs were sampled by dynamic branch enclosure method. Selected branches were enclosed into transparent Nalofan bags and air composition in bags was sampled with Tenax-filled adsorbent tubes. Temperature, humidity, CO_2 concentration and PAR were monitored and recorded in ambient air and in the enclosure. Conditions in the enclosure were regulated to provide similar conditions to ambient air. The chamber was purged continuously by an inflow provided by a diaphragm pump and regulated/measured with a mass flow



No.	Tree Species	No.	Tree Species
1	*Platanus orientalis*	20	*Pinus sylvestris*
2	*Populus tremula*	21	*Populus deltoides*
3	*Quercus aucheri*	22	*Tilia argentea*
4	*Liquidambar orientalis*	23	*Castanea sativa*
5	*Abies equi-trojani*	24	*Quercus macr. sbsp. Sysp.*
6	*Juniperus phoenicea*	25	*Picea orientalis*
7	*Cedrus libani*	26	*Abies nordmanniana*
8	*Juniperus excelsa*	27	*Quercus vulcanica*
9	*Juniperus oxycedrus*	28	*Robinia pseudo-acacia*
10	*Acacia cyanophylla*	29	*Juniperus foetidissima*
11	*Cupressus sempervirens*	30	*Carpinus betulus*
12	*Abies cilicica*	31	*Carpinus orientalis*
13	*Pinus halepensis*	32	*Quercus petraea*
14	*Pinus brutia*	33	*Quercus cerris*
15	*Eucalyptus camaldulensis*	34	*Fagus orientalis*
16	*Pinus nigra*	35	*Alnus glutinosa*
17	*Pinus pinaster*	36	*Fraxinus excelsior*
18	*Pinus radiata*	37	*Fraxinus ornus*
19	*Abies nord. sbsp. Born.*	38	*Pinus pinea*

Figure 1: Sampling sites.

controller (MFC) (Aalborg DFC26) at a flow rate of 350 l/h. The excess air was vented through a by-pass line. The inflow air was conditioned using three sequential columns containing silica gel, potassium iodide (KI) and activated carbon to supply VOCs, ozone and humidity-free air to the chamber. All the components in the system in contact with air flows (tubings, connection ports, internal surfaces of the pumps and MFCs) were chemically inert (mostly Teflon or Teflon-coated). Cold water provided by a circulating water bath was circulated in the bag through a spiral Teflon tubing to prevent the increase in temperatures inside the bag. For the first two hours of experiment the enclosure system was purged to provide steady-state conditions. Then, the air was sampled simultaneously from the inlet and outlet lines for an hour. Tenax TA filled sorbent tubes were used to adsorb BVOCs. At the end of the sampling, tubes were removed, tightly closed and placed into their containers. Then, total weight and leaf area of the leaves in the enclosure were measured in field. Dry weight was determined by oven-drying of the leaves for two days at 60°C. Using these data both dry weight and leaf area based emission factors were calculated.

2.2 BVOC analysis

Before sampling, Tenax-filled sorbent tubes were conditioned with a 40 ml/min flow of helium gas at 240°C for 10 minutes and they were kept in their containers.

Samples were analyzed for 65 compounds (isoprene, 21 monoterpenes, 15 sequiterpenes, 3 oxygenated sesquiterpenes, and 25 oxygenated VOCs) with a gas chromatograph (GC) (Agilent 6890N, Agilent, Wilmington, DE, USA)

equipped with a mass selective detector (Agilent 5973 inert MSD, Agilent, Wilmington, DE, USA) and a thermal desorber (Tekmar, Aerotrap 6000, USA). Samples were desorbed for 10 min at 240°C with 40 ml/min helium. Internal trap temperature during sample desorption was 35°C. The trap was desorbed for 1 min at 240°C. Then, it was baked for 10 min at 250°C. Valve oven and transfer line temperatures of the thermal desorber were 200°C.

The chromatographic column was HP5-MS (30 m, 0.25 mm, 0.25 μm) and the carrier gas was helium at 1 ml/min flowrate and 37 cm/s linear velocity. The split ratio was 1:40. The inlet temperature was 240°C. Temperature program for VOCs was: initial oven temperature 40°C, hold 3 min, 40°C to 120°C at 5 C/min, hold 5 min, 120°C to 240°C at 30°C/min, hold 3 min. Ionization mode was electron impact (EI). Ion source, quadrupole, and GC/MSD interface temperatures were 230, 150, and 280°C, respectively. GC/MS was operated at "scan" and "selected ion monitoring" modes simultaneously. Compounds were identified based on their retention times (within ±0.05 minutes of the retention time of calibration standard), target and qualifier ions and were quantified using the external standard calibration procedure.

Six levels of VOC solutions were prepared in methanol as the calibration standards. Thermal desorption tubes used for calibration were loaded by spiking with 1 μl of the calibration standards [30, 31]. Then, these were run at specified conditions to calibrate the Thermal desorber-GC-MS. In all cases, linear fit was good with $r^2>0.999$.

2.3 Quality assurance/quality control

Instrumental detection limits (IDL) were determined from linear extrapolation, based on the lowest standard in calibration curve and using the area of a peak having a signal/noise ratio of 3. The quantifiable VOC amounts were between 5.7 (p-cymene) and 295 pg (guaiol). Blank Tenax tubes were routinely placed in the field to determine if there was any contamination during sample handling and preparation. The limit of detection of the method (LOD, pg) was defined as the mean blank mass plus three standard deviations. Instrumental detection limit was used for the compounds that were not detected in blanks. LOD ranged between 7.5 (trans-dihydro-b-terpineol)-1455 (crotonaldehyde) pg. Average VOC amounts in blanks were 5% of the amounts found in samples. Samples were blank-corrected by subtracting the average blank amount from the sample amount. Using an average sampling volume of 0.009 m^3, limit of detection of the method ranged between 0.0008 (trans-dihydro-b-terpineol) and 0.16 (crotonaldehyde) $\mu g/m^3$.

The system performance was confirmed daily by analyzing a midrange calibration standard. If the relative standard deviation from the initial calibration was <10%, system was recalibrated. Analytical precision determined from three pairs of duplicate samples ranged between 2–5%.

3 Results and discussions

Emission factors for 65 analyzed compounds were calculated and normalized for 1000 μmol/m^2 s and 30°C as suggested by Guenther *et al.* [32]. BVOCs were grouped as isoprene, monoterpenes, sesquiterpenes, oxygenated sesquiterpenes and oxygenated compounds. Table 1 summarizes the calculated average BVOC emissions for each species.

Isoprene and monoterpenes were the two predominant BVOCs for all species while the other three BVOC groups contributed less to total emissions. *Platanus orientalis* had the highest isoprene emission (27 μg/g h). It was also the strongest BVOC emitter with a total emission factor of 27.1 μg/g h. Two ash species, *Fraxinus ornus* and *Fraxinus excelsior* were the weakest emitters with emission factors of 0.033 and 0.038 μg/g h. The highest monoterpene emitter was *Castanea sativa* with an average monoterpene emission factor of 14.4 μg/g h.

3.1 Emissions from broad-leaved species

Broad-leaved trees are largely known as isoprene emitters due to their high photosynthetic activities [10]. Fifteen out of twenty studied broad-leaved species emitted mainly isoprene, consistent with general broad-leaved emitting characteristics. However, half of them emitted comparatively low total BVOC amounts ranging from 0.033 to 0.946 μg/g h. Sweet Chestnut showed a clearly different emitting pattern as a broad-leaved tree with the highest monoterpene emitting rate of 14.4 μg/g h. Oaks might be considered as the most prevalent broad-leaved trees those largely investigated in the literature and classified as predominant isoprene emitters [33–35]. In Turkey, approximately 30% of all forests are covered by 18 oak species and 5 of them were investigated in the present study. *Quercus macranthera* subsp. *syspirensis*, *Quercus vulcanica* and *Quercus petraea* obviously indicated the typical oak emission characteristics with the isoprene rates of 19.2, 9.79 and 9.63 μg/g h, respectively. However oaks showed a wide inter-species variability in their emissions ranging from 0.157 to 19.4 μg/g h for total BVOCs.

3.2 Emissions from coniferous species

Pine, juniper and fir species were the three main genera comprising 15 of the 18 studied coniferous trees. Monoterpenes were the prominent compounds for all coniferous species except four species (*Abies nordmanniana*, *Abies cilicica*, *Pinus pinea* and *Pinus radiata*). *Abies cilicica* anomalously emitted 14.1 μg/g h isoprene which equals to 91.2% of its total emissions. Pine species were mostly prone to emit monoterpenes and oxygenated compounds while no significant isoprene emissions were observed (Figure 2). *Pinus brutia* had the highest total BVOC emitting rate of 12.3 μg/g h that 10.6 μg/g h of it consisted of monoterpenes. Four juniper species showed regular emission patterns and their emission factors were relatively low ranging from 0.468 to 2.20 μg/g h and their total emissions dominated by monoterpenes. Fir species had completely different

Table 1: Emission factors (μg/g h) for BVOC groups of broad-leaved and coniferous trees (normalized to 1000 μmol/m2 s PAR and 30°C temperature).

	Emission factors (μg/g h)					
	Isoprene	**MT[a]**	**SQ[b]**	**OS[c]**	**OC[d]**	**Total**
Broad leaved trees						
Fagus orientalis	0.047	0.020	0.003	0.00156	0.023	0.095
Quercus petraea	9.63	0.012	0.007	0.00169	0.037	9.69
Quercus cerris	0.079	0.020	0.005	0.01035	0.042	0.157
Quercus vulcanica	9.79	0.063	0.022	0.00008	0.060	9.93
Quercus aucheri	0.028	0.879	0.006	0.00014	0.033	0.946
Quercus macr. sbsp. *sysp.*	19.2	0.070	0.067	0.00032	0.090	19.4
Carpinus orientalis	0.102	0.009	0.006	n.d	0.034	0.150
Carpinus betulus	0.060	0.061	0.013	0.0005	0.136	0.270
Alnus glutinosa	0.018	0.133	0.003	n.d	0.054	0.209
Populus tremula	22.4	0.235	0.015	0.00074	0.034	22.7
Populus deltoides	4.72	0.070	0.022	0.00021	0.022	4.83
Castanea sativa	0.378	14.4	0.033	0.00027	0.281	15.1
Fraxinus excelsior	0.011	0.011	0.0007	0.00002	0.014	0.038
Fraxinus ornus	0.014	0.005	0.006	0.00003	0.008	0.033
Tilia argentea	0.077	0.483	0.005	0.00037	0.089	0.655
Eucalyptus camaldulensis	5.14	0.169	0.078	0.00091	0.414	5.80
Acacia cyanophylla	0.196	0.099	0.014	0.00070	0.042	0.351
Robinia pseudo-acacia	12.4	0.075	0.013	0.00354	0.041	12.5
Platanus orientalis	27.0	0.027	0.029	0.00040	0.102	27.1
Liqiudambar orientalis	15.0	1.31	0.061	n.d	0.164	16.5
Coniferous trees						
Pinus brutia	0.022	10.6	0.166	0.00630	1.48	12.3
Pinus nigra	0.051	3.48	0.017	0.00017	0.344	3.89
Pinus sylvestris	0.183	1.57	0.027	0.00043	0.734	2.51
Pinus halepensis	0.046	3.81	0.049	0.00575	0.590	4.50
Pinus pinaster	0.015	0.583	0.022	0.00224	0.171	0.793
Pinus radiata	0.108	0.329	0.007	0.00068	0.361	0.805
Pinus pinea	0.084	1.11	0.006	0.00171	2.20	3.40
Picea orientalis	2.37	3.90	0.098	0.00014	0.439	6.81
Cedrus libani	0.010	0.526	0.045	0.00008	0.363	0.945
Juniperus excelsa	0.025	2.03	0.032	n.d	0.105	2.20
Juniperus phoenicea	0.005	0.611	0.019	0.00012	0.059	0.695
Juniperus oxycedrus	0.010	0.502	0.047	0.00054	0.057	0.616
Juniperus foetidissima	0.028	0.398	0.005	0.00003	0.037	0.468
Cupressus sempervirens	0.049	0.403	0.039	0.00064	0.278	0.769
Abies nordmanniana	2.26	0.329	0.089	0.00008	0.190	2.87
Abies cilicica	14.1	1.16	0.074	0.00011	0.135	15.5
Abies equi-trojani	1.35	1.66	0.009	0.00024	0.554	3.57
Abies nord. sbsp. *born.*	1.50	2.30	0.036	0.00012	0.502	4.34

n.d.: not detected, [a]Monoterpenes, [b]Sesquiterpenes, [c]Oxygenated sesquiterpenes, [d]Oxygenated compounds.

emitting behaviours as coniferous trees with their high isoprene emissions, such that 91.2 and 78.8% of their total emissions were isoprene for *Abies cilicica* and *Abies nordmanniana*, respectively (Figure 3).

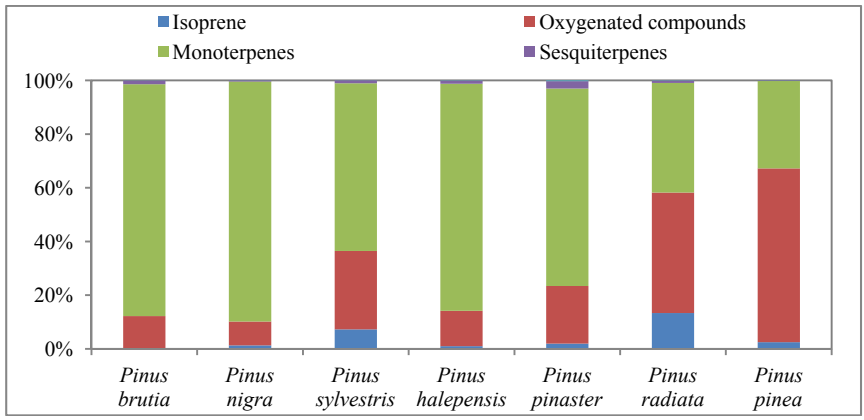


Figure 2: Percentage distribution of BVOC emissions for pine species.

3.3 Comparison of measured emission factors with the literature

Isoprene is a photosynthetic compound that is mostly produced by broad-leaved trees and has an emission dependence on light intensity and temperature while monoterpenes predominantly depend on temperature in coniferous species [10, 28, 29]. In this study, results were clearly comparable with previously reported values and emitting patterns. Isoprene and monoterpenes dominated the total BVOC compositions of broad leaved trees. However, emissions of coniferous species might consist of different variety of BVOCs. Padhy and Varshney [23] reported 9.90 µg/g h isoprene emission for *Eucalyptus globulus* while in the present study isoprene emission for *Eucalyptus camaldulensis* was found as 5.14 µg/g h. There were two oak species that showed unexpectedly low isoprene emissions. Similarly *Quercus myrsinaefolia* was studied by Bao *et al.* [10] and they reported no significant isoprene emissions. *Quercus ilex* is one of the most frequently studied oak in the literature and largely characterized as a strong monoterpene emitter compared to the other oak species and broad-leaved trees [22, 36]. *Quercus aucheri* in this study had a monoterpene emission of 0.879 µg/g h consisting 92.9% of the total emissions making it a monoterpene emitter. Hakola *et al.* [37] measured isoprene and monoterpene emissions of a poplar species (*Populus tremula*) as 13.6 and 0.73 µg/g h, respectively, indicating a similar emission pattern observed for the poplar species investigated in the present study (Table 1).

Komenda and Koppmann [38] reported that monoterpene emissions ranged between 0.06 to 3.70 µg/g h for *Pinus sylvestris*. Kempf *et al.* [39] measured 3.15 µg/g h monoterpene from *Picea abies*. Owen *et al.* [40] reported monoterpene and isoprene emission rates of 0.96 and 0.08 µg/g h, respectively

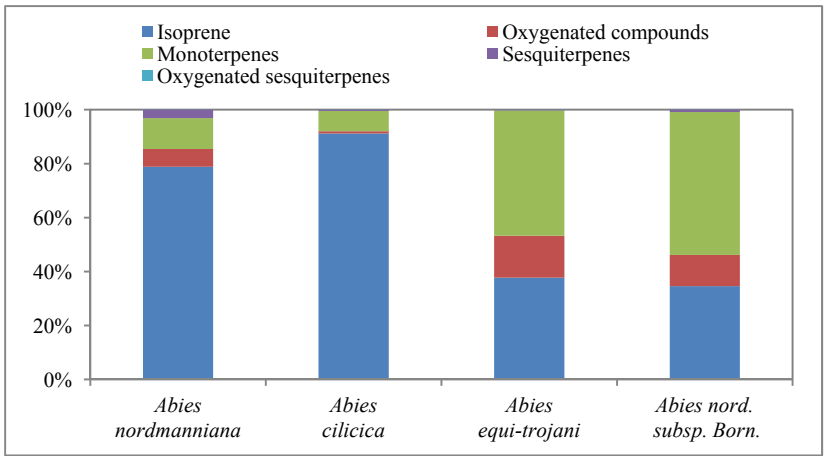


Figure 3: Percentage distribution of BVOC emissions for fir species.

for *Juniperus oxycedrus*. Most of the values in the present study were clearly comparable to those given in previous studies for coniferous trees. Some coniferous trees having an atypical emitting behavior such as *Abies boriisi-regis* with a high isoprene emission rate of 18.4 µg/g h were also reported in literature [26] similar to *Abies cilicica* subsp. *isaurica* included in the present study.

4 Conclusions

Results of this study indicated that vegetative emissions are mostly composed of isoprene and monoterpenes while sesquiterpenes, oxygenated sesquiterpenes and oxygenated compounds were emitted relatively in low quantities. Total emissions 17 of the 38 investigated tree species were <1 µg/g h (Table 1). According to BVOC emission compositions, isoprene and monoterpenes had the largest portion for broad leaved and coniferous species respectively. However, unexpected emitting patterns observed for some species. Characteristically ash and juniper species were weak BVOC emitters and oak species showed the largest inter-species variability. Emission factors determined in this study have an implication for the estimation of total BVOC emissions for Turkey. These emissions will be used as an input for future air quality modelling studies.

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