

We would like to thank both reviewers for their questions and suggestions. We have addressed these below.

Anonymous Referee #1

The authors use the CESM to investigate the impact of infestations of Western North American forests with two bark beetle types on monoterpene emissions and associated secondary organic aerosol (SOA) formation. As biotic stress in ecosystems is likely to increase with warming climate and the emission of BVOCs affects air quality and climate, the topic is an interesting one and fits the scope of the journal. The paper is overall well written and the study performed thoroughly. The paper can be published after the following aspects have been addressed.

General:

The authors use SOA yields in their model that are likely too high under the conditions prevailing in the ambient atmosphere. The authors state so in section 4 (page 29779, line 25) but do not provide a scenario calculation with results based on more realistic SOA yields, although they announce such a comparison for section 3.4 (see text in section 2, page 29773 “. . .we calculate total SOA formed from all monoterpenes with a yield of 10%....”). Such a calculation could be based on laboratory studies using whole plant emissions as SOA precursors and typically observing SOA yields on the order 5-10% (Mentel et al., 2009; Hao et al., 2011) at atmospherically relevant concentration levels.

We showed the total SOA simply to compare vs. the total observed OA (not to relativize yield accuracy for specific SOA products). Unfortunately, low-loading SOA yields are not available for all our species of interest, thus we cannot present an alternate scenario. We have clarified the disconnect between the (perhaps more accurate) overall 10% yield (citing the suggested studies), and the individual yields used from Lee et al. in the text.

It seems that the model – based on available detailed observational data – takes into account monoterpene emission changes only, when considering the impact of bark beetle infestations. The statement that “scale-up factors could not be calculated for compounds not detected in healthy trees” (page 29771, line 19), implies that such stress induced emissions are not taken into account. While this reviewer understands that the observational data set is sparse, it seems unlikely that under bark beetle infestation typical stress induced emissions such as sesquiterpenes and methyl salicylate do not occur. As the SOA yields of these BVOCs are typically larger (e.g. on the order 20%, Kiendler-Scharr et al., 2012) than those of monoterpenes, an estimate of the effect of such stress induced emissions should be given.

Indeed we expect that other compounds may be affected by beetle kill. However given the limited data, we have chosen only to focus on monoterpenes here (per manuscript title). We have added a sentence to Section 4 to highlight that SOA levels may be modulated by changes to other precursors not considered here.

The discussion of the impact of beetle infestation on aerosol direct effects and visibility on page 29778 only mentions the SOA mass concentrations under bark beetle attack and an estimated natural aerosol level. Visibility in itself is not discussed and no numbers comparing visibility with/without the bark beetle induced SOA are provided. Such numbers would be interesting and should be provided.

The metric for visibility is visual range. The visual range depends on the absolute PM loading (as well as RH conditions and the extinction properties of the aerosol). Therefore an increase in 1 ug/m³ of SOA could decrease the visual range by more than 20km if assuming a 4 ug/m³ OA background, but with double the background concentrations, the change in visual range would be less than 5 km. It is therefore not possible to attach simple visibility numbers to the concentration changes simulated here.

Specific comments:

The abstract and numerous places in the manuscript refer to “beetle mortality” when “beetle induced tree mortality” is meant. This is misleading and should be changed throughout the manuscript.

Corrected.

The text discussing figure 4 (p 29774) is incorrect in order of 4a, b, etc.

Corrected.

Inconsistency in numbers reported for increase of SOA in the pine scenario (up to 30% - p29776, line 5 – versus 43% same page line 10).yt

Corrected to read “37%.”

Figure 10: would be easier to compare the model with observations when OA is given for the three model runs. As far as this reviewer understands, changes in OA would be due to SOA from bark beetle attack only.

We agree, however this is not strictly possible as the yields for total SOA from monoterpenes are not necessarily consistent with the individual precursor yields used here for beetle-impacted species (see discussion of comment #1 above).

Figures 3 would be easier to read when a common concentration scale is applied in the individual panels; same holds for figure 7.

Corrected by placing all on log scales

Figure 9 caption: should read . . .compare to Figs. 5c, f and 8c, f . . .

Corrected

Figure 10 caption: (a) and (b) rather than (b) and (c).

Corrected

Literature: Amin et al., 2012a and 2012b are sometimes mixed up in text.

Corrected

Anonymous Referee #2

General comments:

The paper presents modeling of bark beetle-induced monoterpene emissions and secondary organic aerosol (SOA) formation in western North America in the areas of recent bark beetle outbreaks. The paper does not give any experimental evidence that bark-beetle attack will cause increases in volatile organic compound emissions. The scale-up factor for pine emissions data is taken from a published paper, but for spruce this information is from an unpublished paper. SOA formation rate in the model is based on the published SOA yields of the major monoterpene compounds in reaction chamber experiments found in literature. SOA in the simulations is calculated by applying a fixed yield to the first generation oxidation products of the precursors found in the target conifer species. Although this paper does not provide direct evidence that bark beetle-induced terpene emission increase SOA yield above attacked forests, it is timely and present a potential of biosphere-atmosphere feedback mechanisms which might be functional in ecosystem scale under rapid environmental changes such as high latitude climatic warming. As far as I know, this is the first paper reporting evidence that forest insect outbreaks may to have a direct link to atmospheric SOA formation. The paper is appropriate for ACP and should be published with a minor revision following the suggestions given below.

Specific comments:

Page 29767, line 16. It is well-known fact that in deciduous trees the emission rates of volatile compounds from beetle-damaged foliage is much more diverse and in higher level, when compared to healthy foliage, although monoterpenes are among most responsive compounds. Because the focus of this paper is in conifers, it should be also mentioned here that beetle feeding damage on conifer bark may increase emission rates of highly reactive sesquiterpenes even more than that of monoterpenes (e.g. 7-fold vs. 4-fold as shown by Heijari et al. 2011). Sesquiterpenes may have important role in SOA formation (e.g. Tan et al. 2012) although higher volatility monoterpenes still form the majority of VOCs in forests attacked by bark beetles.

We agree this is an important point to make. We have added a comment in this section on sesquiterpenes, and this is also mentioned on page 29780 in reference to a comment by Reviewer #1.

Page 29768, Line 22, Beetle mortality? This definitely had to be TREE MORTALITY related to bark-beetle outbreaks

Corrected.

Page 29771. Line 7-8 ". . . the mortality effect is the decrease in VOC emissions that occurs after trees are killed". Is there a real documented drop in the local emissions levels? How the monoterpene emissions from logging activities, and the remaining dead wood (branches, stumps and root system) and needle litter left in the forest site were implemented in the model? Some reports (e.g. Räisänen et al. 2008) demonstrates that during logging activities there could be even 2 to 3-fold increase in the local monoterpene emission rates in pine forests. After the removal conifer trees from a forest site the residual effect may corresponds to about 10% of the monoterpene release detected from intact forests (Haapanala et al. 2012).

We were not aware of this effect, and are grateful to the reviewer for bringing it to our attention. We have now indicated that we have assumed that emissions from dead wood and needle litter are zero and this is clearly a simplification.

Page 29779 Line 2, ". . .we have assumed that trees are impacted by beetle attack for a full year,..". Is there any information available of the annual peak periods of emissions? During warm growing season the emission rates should be relatively high compared to other seasons. During the main attack period of bark beetle females the fresh resin flow from entrance holes should be substantial and the monoterpene emissions rates from fresh resin might be at highest. How these peak emission periods may affect the local SOA levels?

As indicated in Section 3.1, the monoterpene emissions peak in the summertime, and our results focus on the emissions & SOA impacts in that season. While we have no specific information on the timing of attack in the dataset we used, evidence suggests that beetle attack generally occurs during the summer months. We have added text to this effect in Section 4.

Page 29779 Line 5, Perhaps authors can discuss here about the potential effects of forestry activities and residues on the monoterpene emissions of beetle killed forests in association with the increased surface temperature effect.

We are not clear exactly what the reviewer is suggesting here. We added a statement that post-outbreak forestry activities could also impact VOC emissions, with a reference to the Raisanen study that the reviewer suggested above.

Page 29780, Line 9 "Two main effects emerge from this study – the mortality effect and the attack effect". I still wonder how the mortality effect emerges from this study, because I understood that tree mortality was just put as a zero value (reduction of monoterpene emitting area) in the model and not any on mortality site specific monoterpene emission data was not given to support it.

To address the reviewer's previous comment above, we have clarified this assumption in the text.

The Impact of Bark Beetle Infestations on Monoterpene Emissions and Secondary Organic Aerosol Formation in Western North America

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Abstract

Over the last decade, extensive beetle outbreaks in ~~Western~~western North America have destroyed over 100,000 km² of forest throughout British Columbia and the ~~Western~~western United States. Beetle infestations impact monoterpene emissions through both decreased emissions as trees are killed (mortality effect) and increased emissions in trees under attack (attack effect). We use 14 yr of ~~beetle mortality~~beetle-induced tree mortality data together with beetle-induced monoterpene ~~concentration~~emission data in the National Center for Atmospheric Research (NCAR) Community Earth System Model (CESM) to investigate the impact of ~~beetle mortality~~beetle-induced tree mortality and attack on monoterpene emissions and secondary organic aerosol (SOA) formation in ~~Western~~western North America.

1 Regionally, beetle infestations may have a significant impact on monoterpene emissions and
2 SOA concentrations, with up to a 4-fold increase in monoterpene emissions and up to a 40%
3 increase in SOA concentrations in some years (following-in a scenario where the attack effect
4 is based on observed lodgepole pine response). Responses to beetle attack depend on the
5 extent of previous mortality and the number of trees under attack in a given year, which can
6 vary greatly over space and time. Simulated enhancements peak in 2004 (British Columbia)
7 and 2008 (US). Responses to beetle attack are shown to be substantially larger (up to a 3-fold
8 localized increase in summertime SOA concentrations) when followingin a scenario based on
9 bark-beetle attack in spruce trees. Placed in the context of observations from the IMPROVE
10 network, the changes in SOA concentrations due to beetle attack are in most cases small
11 compared to the large annual and interannual variability in total organic aerosol which is
12 driven by wildfire activity in Westernwestern North America. This indicates that most beetle-
13 induced SOA changes are not likely detectable in current observation networks; however
14 these changes may impede efforts to achieve natural visibility conditions in the national parks
15 and wilderness areas of the Westernwestern United States.

16

17 **1 Introduction**

18 In the last decade, Westernwestern North America has experienced the largest bark beetle
19 outbreaks in recorded history (Taylor et al., 2006). The main beetle impacting the region is
20 the mountain pine beetle (MPB), with nearly 74% of needleleaf tree mortality associated with
21 this beetle (Man, 2012). The MPB is native to Westernwestern North America and can kill
22 large numbers of pine trees annually, attacking mainly lodgepole pine and ponderosa pine.
23 Mountain pine beetles bore under the bark of host trees, eventually killing the tree as the
24 beetles consume the phloem and introduce a virulent fungus. In the United States, MPB
25 infestation peaked in 2009 with 0.9 million hectares of killed trees (Meddens et al., 2012). In
26 British Columbia, the area of infestation peaked in 2005-2007 with almost 1 million hectares
27 of trees killed annually during these years (Meddens et al., 2012). Although the area infested
28 in both British Columbia and the Westernwestern United States has decreased recently,
29 beetles continue to expand into new areas (Westfall and Ebata, 2011).

30 Geographical expansion of the MPB has been limited in the past by climate (Carroll et al.,
31 2003). Several studies have attempted to model the future impacts and timing of the
32 expansion of the MPB based on climate change effects and changing pine stand

1 characteristics (e.g. Hicke et al., 2006; Bentz et al., 2010). These studies agree that warming
2 over the next century will allow MPB outbreaks to occur at progressively higher elevations,
3 possibly supporting an on-going MPB outbreak for decades. Climate change may also lead to
4 drought and elevated surface ozone concentrations, both of which increase tree stress making
5 them more susceptible to beetle attack (e.g. Raffa et al., 2008; Jones et al., 2004).

6 Insect damage may impact forests by shifting them from a carbon sink to a carbon source (e.g.
7 Kurz et al., 2008; Hicke et al., 2012). Bark beetle infestation alters carbon stocks differently
8 than stand-replacement wildfires or clear-cut harvesting because smaller diameter trees and
9 non-host trees typically survive a beetle infestation, leading to a different pattern of mortality
10 (Pfeifer et al., 2010). Beetle infestation can impact forest fire susceptibility and activity by
11 increasing the risk of active crown fire in dry beetle-killed forest and decreasing the risk of
12 active crown fire when needles fall (e.g. Simard et al., 2011; [Hicke et al., 2012](#)). Insect attack
13 can also alter surface-atmosphere exchanges of heat, water, and momentum through land
14 surface modification (e.g. Wiedinmyer et al., 2012; Edburg et al., 2012). Hais and Kucera
15 (2008) estimate a 3.5 K increase in temperature in a spruce forest after beetle attack. In
16 addition, snowpack in a beetle-killed forest can also be prolonged (Boon, 2007; Perrot et al.,
17 2012).

18 Bark beetle attack can also prompt elevated monoterpene emissions in trees (e.g. Amin et al.,
19 [2012a](#)), with potential implications for local air quality. The emissions enhancement is likely
20 a defense mechanism of the tree that consists of increasing resin flow to remove beetles,
21 increasing emissions of compounds that are toxic to the beetles, and attracting predators of the
22 beetles (Pare and Tumlinson, 1999). A few studies have examined and quantified
23 monoterpene concentration and emission changes under beetle attack for specific tree species
24 including lodgepole pine, ponderosa pine, and Engelmann spruce. These studies include both
25 beetle infestation studies and fungal inoculation studies and have found significant increases
26 in monoterpenes due to beetle attack (e.g. Gara et al., 1993; Litvak and Monson, 1998; Jost et
27 al., 2008; Amin et al., [2012a](#); Prieme et al., 2000; Blande et al., 2007; Brilli et al., 2009).
28 Insect herbivory can induce both substantial increases in total monoterpene emissions from
29 vegetation and changes in the emission profile, with implications for atmospheric
30 composition. [While not the focus of this study, sesquiterpene emission rates are also elevated](#)
31 [in conifers experiencing bark beetle attack \(Heijari et al., 2011\).](#)

1 Monoterpenes are oxidized in the atmosphere to form lower volatility products which may
2 partition into the particle phase, forming secondary organic aerosol (SOA). These oxidation
3 processes can also lead to ozone formation with important implications for global atmospheric
4 composition. SOA formed from monoterpenes and other compounds can impact the Earth's
5 radiative balance through the direct effect of scattering incoming solar radiation and the
6 indirect effect on cloud albedo and lifetime (Lohmann and Feichter, 2005). Atmospheric
7 visibility may also be degraded by SOA formation in forests. National parks and other
8 protected wilderness areas in the ~~Western~~western US are currently impacted by beetle
9 infestation and these are areas where the EPA has mandated an improvement in visibility
10 under the Regional Haze Rule (US EPA, 1999).

11 Despite growing evidence that bark beetle attack can enhance monoterpene emissions from
12 vegetation, in addition to the role that these beetles play in changing land cover and density,
13 no study has quantified the impact of these changes on regional air quality and visibility. In
14 this work, we use beetle-caused tree mortality data from 1997-2010 and beetle-induced
15 monoterpene data from the recent literature in the National Center for Atmospheric Research
16 (NCAR) Community Earth System Model (CESM) to study the impact of beetle infestation
17 on monoterpene emissions and SOA formation in ~~Western~~western North America. We
18 compare two scenarios based on beetle-induced monoterpene data from lodgepole pine and
19 Engelmann spruce and focus on the spatial and temporal evolution of monoterpene emissions
20 and the SOA formed due to cumulative beetle attack and subsequent forest mortality in the
21 model.

22

23 **2 Model description**

24 The NCAR CESM consists of coupled global models for the atmosphere, ocean, land, land
25 ice, and sea ice (Gent et al., 2011). In this work we use v1.1 of the CESM in a configuration
26 where the land and atmosphere are coupled with imposed ocean (sea surface temperature) and
27 sea ice conditions for the present day. The simulations are run at 1.9° x 2.5° horizontal
28 resolution from 1997 to 2010 (with specified meteorological fields from reanalysis, see
29 below) representing 14 yr of cumulative beetle kill in ~~Western~~western North America.

2.1 Land model, land cover, and ~~beetle mortality~~beetle-induced tree mortality

The land model used in this study is version 4 of the Community Land Model (CLM4, Lawrence et al., 2011). ~~The~~ CLM4 describes the physical, chemical, and biological processes of terrestrial ecosystems, including the hydrology and carbon cycling of the terrestrial biosphere. Vegetation is specified by 16 plant functional types (PFTs). Leaf Area Index (LAI) is also specified for each month for each PFT. ~~The~~ PFT distributions are based on Moderate Resolution Imaging Spectroradiometer (MODIS) land surface data sets (Lawrence and Chase, 2007) and a cropping dataset (Ramankutty et al., 2008). Figure 1 shows coverage of needleleaf forests (PFT 1 and PFT 2), the vegetation types susceptible to beetle attack ~~over~~ in Westernwestern North America.

Beetle-caused tree mortality data are from Meddens et al. (2012). These data were created for the ~~Westernwestern~~ United States (US) from the US Forest Service Aerial D~~etection~~ S~~urvey~~ P~~rogram~~ (1997-2010) and for British Columbia (BC) from the BC Ministry of Forests Aerial O~~verview~~ P~~rogram~~ (2001-2010). The data are ~~provided~~ at 1x1 km grid resolution. Mortality area within each grid cell (crown area of killed trees) was converted from information provided by the US and BC aerial surveys by year and by beetle species. Because the MPB has had such a significant impact in ~~Westernwestern~~ North America compared to other beetles, the impacts from the MPB are compared to impacts from the other 12 beetles species combined (OB) in this study. Figure 2 shows the spatial and temporal extent of tree mortality in BC and the ~~Westernwestern~~ US caused by the MPB and the OB. Bark beetle attack has resulted in extensive damage over the last decade, with total vegetated area decreasing by up to 30% in some grid cells.

Meddens et al. (2012) indicate that the US aerial ~~detection~~-survey data underestimates the number of trees killed based on field observations in Idaho, Colorado, and New Mexico. Using remotely sensed imagery of beetle outbreak locations in Idaho, north-central Colorado, and northern New Mexico, they developed and applied factors to adjust the US ~~beetle mortality~~beetle-induced tree mortality data to match the image-derived mortality area. This more realistic upper estimate improves the agreement with field observations and the continuity with BC data across the US-Canada border. The adjusted data are used for the US in this study.

Uncertainties in the mortality data include variability in estimates in space and time due to differing abilities and techniques of different mappers, the mean tree crown area values used

1 | for calculating mortality in a gridcell, ~~limited-incomplete~~ data collection ~~areas~~ (wilderness or
2 | national park areas were not regularly surveyed), and the adjustment factors for the US. The
3 | ~~“jump”sharp increase~~ in the other ~~beetle-mortality~~~~beetle-induced tree mortality~~ seen in 2003
4 | (Fig. 2) is due in part to incomplete surveying of a pinyon ips beetle outbreak in the
5 | Southwest US in 2002. Surveys were not routinely conducted for pinyon pine forests before
6 | 2003 and so pinyon ips mortality added in 2003 includes mortality from the preceding years.

7 | Changes in plant functional type coverage are calculated to correspond with the beetle-caused
8 | mortality datasets, at the 1.9° x 2.5° horizontal resolution used here. Cumulative PFT
9 | reductions are calculated for each year from 1997-2010. Because the PFTs do not break into
10 | specific tree species and maps of sub-PFT species composition are not available, the mortality
11 | data are applied to the two needleleaf PFTs covering ~~Western~~~~western~~ North America (Fig. 1)
12 | to simulate beetle attack. Given that the mortality data are provided as a percentage of area,
13 | total tree mortality is conserved by this approach. As a result of the PFT reductions, total LAI
14 | decreases with bark beetle kill. The original CLM PFT dataset is used as the baseline. In
15 | some gridcells mortality area can exceed PFT coverage, because the PFT coverage and
16 | mortality data are from two different sources. In these cases, mortality is capped at this
17 | maximum PFT coverage.

18 | **2.2 Monoterpene emissions**

19 | The Model of Emissions of Gases and Aerosols from Nature (MEGAN 2.1) is used in CLM4
20 | to estimate emissions from terrestrial vegetation of up to 150 different BVOC compounds,
21 | including isoprene, monoterpenes, sesquiterpenes, and other oxygenated VOCs (Guenther et
22 | al., ~~submitted~~~~2012~~). A VOC flux in units of $\mu\text{mol m}^{-2} \text{h}^{-1}$ is calculated from a baseline
23 | emission that is modulated by an emission activity factor, which accounts for emission
24 | responses to meteorological and phenological conditions, including light, temperature, leaf
25 | age, and LAI. There is a specific emission factor for each PFT for each compound. In CLM4
26 | the BVOC emissions are calculated interactively at every time step.

27 | Bark beetle infestation has two main effects on VOC emissions from trees. First, the attack
28 | effect is the increase in VOC emissions that occurs while a tree is under attack. To simulate
29 | the attack effect in the model, scale-up factors for the monoterpene emissions are calculated
30 | from field observations (see below) and applied to the fraction of needleleaf evergreen that
31 | ~~are-is~~ under attack in each year. The increase in mortality from the current year to the next

1 year corresponds to trees under attack in the current year. We note that some trees attacked
2 by beetles can survive the attack, and that these trees would not be accounted for here as the
3 mortality dataset only includes trees under attack that subsequently died. Second, the
4 mortality effect is the decrease in VOC emissions that occurs after trees are killed. This
5 simplification assumes that monoterpene emission rates from dead wood and needle litter are
6 effectively zero. This effect is implemented through the reduction in evergreen needleleaf
7 PFT coverage.

8 To estimate the attack effect on monoterpene emissions we consider two scenarios based on
9 field observations of infested trees. Amin et al. (2012a, ~~b~~2013) compared sorbent trap
10 concentrations of monoterpenes emitted from ~~healthy Engelmann spruce and lodgepole~~
11 ~~pine~~ healthy lodgepole pine and Engelmann spruce trees to ~~spruce and pine~~ pine and spruce
12 under attack by bark beetles. In the first scenario, we apply relative increases in the
13 monoterpene compounds 3-carene, β -phellandrene, β -pinene, and p-cymene calculated from
14 the lodgepole pine data from Amin et al. (2012a). In the second scenario, we use relative
15 increases in 3-carene, β -phellandrene, β -pinene, p-cymene, α -pinene, and sabinene from the
16 Amin et al. (~~2013~~2012b) Engelmann spruce data (Table 1). Scale-up factors could not be
17 calculated for compounds not detected in healthy trees. Table 1 also includes the baseline
18 simulated total emission of these monoterpenes for ~~Western~~ western North America. Overall,
19 the largest relative increase is for β -phellandrene in the pine study and 3-carene in the spruce
20 study. For each scenario, the factors are applied to PFT 1 and PFT 2 for all locations under
21 bark beetle attack, assuming that all trees would respond the same way to infestation by
22 different bark beetle species. Although numerous studies have demonstrated that the VOC
23 increase ~~effect~~ occurs in many plant species caused by many insect species (e.g. Blande et al.,
24 2007; Brillì et al., 2009; Staudt and Lhoutellier, 2007), ~~the~~ differences between the pine and
25 spruce scale-up factors indicate that different tree species may have a very different response
26 to bark beetle attack. We include these two scenarios here in an effort to characterize this
27 range of response.

28 **2.3 Atmospheric model**

29 The Community Atmosphere Model (CAM4) is a 3D global atmospheric model. We use
30 specified meteorological fields generated from the GEOS-5 product for 2008 (Rienecker et
31 al., 2008). CAM can be run with an interactive atmospheric chemistry scheme based on the
32 MOZART-4 (Model of Ozone and Related Chemical Tracers) chemical transport model, a

1 configuration known as CAM-Chem. Lamarque et al. (2012) describe the features of the
2 CAM-Chem model, as well as validation against observations. The chemical mechanism
3 used here contains extensive tropospheric chemistry, including O₃, NO_x, SO_x, CO, VOC
4 oxidation processes, and a bulk aerosol scheme including sulfate, ammonium nitrate,
5 carbonaceous aerosols, SOA, sea salt, and dust. Major BVOC species or classes are
6 calculated within CLM4 and fed into the chemical mechanism of CAM-Chem (e.g. isoprene,
7 monoterpenes, acetone, etc.). For these bark beetle simulations, the emissions for select
8 speciated monoterpenes (Table 1) are also input from the CLM4.

9 **2.4 SOA formation**

10 SOA in these simulations is produced by applying a fixed yield to the first generation
11 oxidation products of the precursors. We consider SOA formed from both ozonolysis and
12 photooxidation reactions (rates from Atkinson (1997)), following primarily bulk yields
13 measured by Lee et al. (2006a,b) (Tables 2 and 3). There is a large range in measured SOA
14 yields for our species of interest; therefore we choose Lee et al. (2006a,b) for consistency, but
15 note here the uncertainty in these values. Only one study has looked at SOA yields from β-
16 phellandrene (Surratt et al., 2008) finding that SOA yields from β-phellandrene may be
17 similar to limonene SOA yields. Therefore, limonene SOA yields are used here to
18 approximate SOA yields for β-phellandrene. To date, SOA formation from p-cymene has not
19 been investigated. Here we use SOA yields from the structurally similar compound 1-methyl-
20 3-n-propylbenzene from Odum et al. (1997) to approximate SOA yields for p-cymene.

21 The chamber studies of Lee et al. (2006a,b) were conducted at low HC:NO_x ratio, considered
22 representative of a ponderosa pine forest. Temperature dependence of the reaction rates are
23 only available for α-pinene and β-pinene, thus the reaction rates are fixed at 298K; however,
24 given the short lifetimes of these species, differences in oxidation rates at atmospherically
25 relevant temperatures are negligible. We do not include SOA formed from the reaction of
26 monoterpenes with the NO₃ radical due to the difficulty in obtaining yields and reaction rates
27 for the specific monoterpenes considered here. Because monoterpene oxidation by the NO₃
28 radical may be an important source of SOA at night (e.g. Winer et al., 1984; Fry et al., 2009;
29 Fry et al., 2011), the SOA concentrations from monoterpenes simulated here may be a lower
30 limit. SOA from isoprene is also neglected here, but given the relatively low coverage of
31 deciduous trees in ~~Western~~western North America, the contribution of this source to total
32 organic aerosol in the region is likely low (Heald et al., 2008). Formed SOA is treated as non-

1 volatile, a necessary simplification given the lack of volatility parameter measurements for all
2 of the specific SOA precursors under consideration. In addition to our specific SOA products,
3 we calculate total SOA- formed from all monoterpenes with a yield of 10% (in order to
4 compare with total organic aerosol observations, see Sect. 3.4). This total 10% yield may not
5 be consistent with the sum of the individual precursor yields used here, but is in line with the
6 yields estimated from whole plant emission SOA studies (Mentel et al., 2009; Hao et al.,
7 2011).

8

9 **3 Results**

10 **3.1 Impacts on monoterpene emissions in ~~Western~~western North America**

11 Figure 3 shows the simulated summer-mean baseline emissions (i.e., without beetle activity)
12 of the six monoterpenes impacted by bark beetle in this study (totals in Table 1). These
13 emissions peak in the summertime, therefore we focus on that season in our study. Emissions
14 of β -pinene and 3-carene are the largest, whereas emissions of β -phellandrene and p-cymene
15 are much smaller. Emissions of these monoterpenes are large in the ~~Western~~western US,
16 particularly in Washington, Oregon, and Northern California, due to the extensive coverage of
17 needleleaf trees in this region (Fig. 1).

18 We first show the simulated impact of bark beetle kill on monoterpene emissions and SOA
19 formation based on the observed enhancements in lodgepole pine emissions (Amin et al.,
20 2012a; Table 1) (results using spruce scenario are discussed below in Sect. 3.3). Lodgepole
21 pine is the main species under attack by mountain pine beetles (Logan and Powell, 2001;
22 Meddens et al., 2012). The impact of beetle attack on β -phellandrene emissions is shown as a
23 specific example. This monoterpene has the largest scale-up factor for the pine scenario.
24 Figure 4 shows maps of simulated β -phellandrene emissions for three different years under
25 baseline conditions and accounting for MPB kill (Fig. 4a-b, ~~cd~~, e, f) and OB kill (Fig. 4a, ~~ce~~,
26 ~~f~~). Although MPB infestation peaked in BC in 2007 and the ~~Western~~western US in 2009, the
27 MPB had the largest impact on VOC emissions in BC and the ~~Western~~western US in 2004
28 and 2008, respectively. In later years, the stronger attack effect was overcome by the
29 cumulative mortality of the previous years. In 2004, emissions of β -phellandrene decrease
30 locally by up to 10% due to the mortality effect (Fig. 4ba) and increase up to 3-fold when the
31 attack effect is included (Fig. 4ed). In 2008, emissions of β -phellandrene decrease locally by

1 up to 38% due to the mortality effect (Figure 4cb) and increase up to four-fold when the
2 attack effect is included (Fig. 4fe). In 2002, the OB had the largest impact in both the
3 ~~w~~Westernwestern US and BC; however the impact in BC is small (Fig. 4a, de, f).

4 Figure 5 shows the impact of all beetles combined in 2002, 2004, and 2008 on the sum of
5 monoterpenes affected in the pine scenario. The impact of bark beetle attack is strongest for
6 β -phellandrene, therefore, the summed monoterpene emissions have a lower relative increase.
7 However, the absolute increase is larger due to the inclusion of more abundantly emitted
8 monoterpenes (e.g. β -pinene). With all four monoterpene emissions included, the largest
9 local increase above baseline is 111% in 2002, 70% in 2004, and 104% in 2008. The majority
10 of the impact is from the MPB, except in 2002. Although the impact from MPB alone peaks
11 in later years, the impact of all beetles together for the entire region peaks in 2002 due to the
12 large impact of OB in the US in 2002 and because the cumulative mortality is lower in 2002
13 than in later years (Fig. 5a vs. b, c), causing the attack effect to have a greater impact in 2002.

14 Figure 6 shows the temporal evolution of simulated total regional emissions of the four
15 monoterpenes affected by beetle attack in BC and the ~~Westernwestern~~ US. Because the
16 fraction of trees under attack in a year is calculated based on the change in mortality from the
17 current year to the following year, an attack effect cannot be calculated for the final year of
18 mortality in the dataset (2010). In a given year, the more trees under attack, the stronger the
19 attack effect; however, increasing cumulative mortality in later years offsets more of the
20 attack effect. For BC, the maximum increase in total emissions of these four compounds is
21 7% above baseline in 2004. For the US, the maximum increase in total emissions of these
22 four compounds is 3% above baseline in 2008. Although total monoterpene emissions are not
23 significantly perturbed by bark beetle kill, Fig. 5 shows that local effects can be substantially
24 larger. For OB impacts in the US, there is a peak in monoterpene emissions in 2002
25 associated with the pinyon ips outbreak from 2002-2004 discussed in Sect. 2.1. In other years
26 there is little to no increase in emissions above baseline, and after 2003, emissions return to
27 the baseline level. Overall, OB impacts in BC are small.

28 **3.2 Impacts on SOA formation in ~~Westernwestern~~ North America**

29 Changes in monoterpene emissions throughout ~~Westernwestern~~ North America ~~will~~ impact
30 SOA loading in the region. Figure 7 shows baseline summertime ~~_~~-average simulated
31 concentrations of SOA from each of the six monoterpenes. These SOA distributions largely

1 mimic the spatial patterns of the monoterpene emissions shown in Fig. 3 due to the rapid
2 oxidation and formation of the aerosol products in the model. Both 3-carene and β -pinene
3 continue to be the dominant SOA precursors for the pine scenario, but β -phellandrene
4 emissions make a more important relative contribution to SOA than VOC emissions as a
5 result of the high estimated SOA yields for this compound (Table 2). Here we assess the
6 spatial and temporal changes in SOA concentration caused by the changes in the
7 monoterpenes discussed. We show only the integrated effects of all the monoterpene
8 precursors on SOA concentrations in the pine scenario.

9 The largest overall impact on SOA surface concentration from the attack effect from all
10 beetles (MPB plus OB) for the whole region (~~Western~~western US plus BC) is in the year 2002
11 (Fig. 8) just as for monoterpenes (Fig. 5). During this year, the mortality effect causes
12 widespread decreases in SOA from 1-5%. Including the attack effect, specific areas see
13 increases above baseline of up to 307% with a widespread increase above a baseline of ~10%.
14 The year with the smallest overall attack effect impact is in ~~the year~~ 2009, likely a
15 combination of decreasing infestation of MPB in BC and a decreasing infestation of the OB in
16 the ~~Western~~western US, allowing the mortality effect to overcome the attack effect in this
17 year. Figure 8 also includes the impact of MPB and OB attack on SOA in 2004 and 2008.
18 ~~The largest increases above baseline are 43% in 2002, 37% in 2004 and 36% in 2008.~~

19 3.3 Alternate spruce scenario

20 The impact of bark beetle infestation on observed monoterpene emissions differs considerably
21 in an Engelmann spruce vs. a lodgepole pine, both in magnitude and in speciation (with two
22 additional monoterpenes α -pinene and sabinene affected) (Table 1). Results in Sect. 3.1 and
23 3.2 assumed that emissions from all needleleaf trees respond as the lodgepole pine; we show
24 here the impact of instead assuming a spruce-like response.

25 In this scenario, the maximum increases in monoterpene emissions (3-fold) and SOA (over 2-
26 fold) caused by mountain pine beetle occurs in 2008 (Fig. 9). The mortality effect is the same
27 for both the pine and spruce scenarios, whereas the attack effect, which takes into account the
28 different scale-up factors, is much larger for the spruce scenario (3-4 times the impact seen
29 when employing the pine scenario).

30 This scenario is presented to exemplify a possible range in response. However, evidence
31 suggests that lodgepole pine stands (25% of coverage of BC forests prior to bark beetle

1 infestation) have been preferentially impacted by beetle attack, with little change in
2 Engelmann spruce coverage (Westfall and Ebata, 2011). Thus, the pine scenario results
3 presented in Sect. 3.1 and 3.2 are more likely regionally represented, with possible localized
4 spruce-like response.

5 **3.4 Implications of bark beetle impacts on SOA**

6 We compare simulated SOA to measurements of organic aerosol (OA) from the Interagency
7 Monitoring of Protected Visual Environments (IMPROVE) network to provide some context
8 for the simulated aerosol concentration changes due to beetle kill. The IMPROVE network
9 consists of about 200 sites across the US where filters are collected every three days and
10 analyzed for concentrations of speciated fine particulate matter. Organic aerosol typically
11 makes up 15-70% of total fine particulate matter (PM_{2.5}) measured in summertime in the
12 ~~Western~~western US. Simulated SOA makes up only a fraction of total simulated OA in the
13 ~~Western~~western United States, typically 15-50% in winter and 40-90% in summer, with large
14 primary emissions due to wildfires. The model simulation underestimates total observed OA
15 over the region, consistent with previous model studies around the world (e.g. Heald et al.,
16 2005; Volkamer et al., 2006; Lamarque et al., 2012). The factor of 1.4 used in the conversion
17 of OC to OA in the IMPROVE database may also be too low for many areas of the
18 ~~Western~~western US (El-Zanan et al., 2005), further widening the gap between observed and
19 simulated OA. However, reconciling this OA measurement-model gap is not the objective of
20 this work.

21 Figure 10 shows monthly mean measurements from two sites in Montana and Colorado in
22 2008 that have been impacted by beetle infestation along with baseline and beetle-attack
23 simulated SOA concentrations (for both the pine and spruce scenario) and simulated OA
24 concentrations sampled to site location. These sites are examples of areas that saw some of
25 the highest increases in total SOA concentrations above baseline in the ~~Western~~western US in
26 our simulation. However, the simulated increase in SOA under the pine scenario (maximum
27 increase of 0.2 µg m⁻³) is dwarfed by the seasonal and interannual variability in observed OA
28 which is largely driven by wildfire activity (Spracklen et al., 2007). This suggests that the
29 impacts from beetle infestation may be difficult to detect in surface OA or PM_{2.5}
30 measurements. However, the spruce scenario results in much larger increases in SOA
31 concentrations due to beetle attack, in this case more than a doubling of SOA concentration in
32 summertime (increases of more than 1 µg m⁻³). Although, these changes are unlikely to lead

1 to significant degradation in air quality, the magnitude of these localized changes may be
2 large enough to observe in surface particulate matter measurements, particularly in areas less
3 affected by wildfires. However, due to uncertainties in simulated SOA concentrations and
4 poor overall model agreement for OA concentrations, it is difficult to make this assessment.
5 Furthermore, the spruce scenario is shown to provide an estimate of uncertainty in the
6 diversity of tree-species response to bark beetle attack, but forests surrounding these
7 particular sites are likely composed of pine species.

8 Although the changing impact of beetle infestation may be difficult to discern from
9 observations, these biogenic aerosols may be a significant contributor to the natural aerosol
10 background, particularly in light of the EPA's Regional Haze Rule (US EPA, 1999), which
11 mandates a return to "natural visibility conditions" by the year 2064. Changes in SOA
12 concentrations estimated under the spruce scenario ($\sim 1 \mu\text{g m}^{-3}$) are comparable to the natural
13 aerosol levels estimated for the ~~Western~~western United States ($1.21 \mu\text{g m}^{-3}$ US EPA, 2003),
14 suggesting that achieving "natural visibility" may not be possible in forests impacted by
15 beetle infestation.

17 **4 Uncertainties**

18 The robustness of changes in emissions following beetle infestation is difficult to ascertain.
19 Here we use monoterpene-increase data from only one study location (Colorado) for two
20 different tree species (Amin et al., 2012a, lodgepole pine data, ~~2012b~~2013, Engelmann spruce
21 data). The large differences between the pine and spruce scenarios illustrate the large species-
22 variability in response to beetle attack and the uncertainties that still surround the impact of
23 beetles on atmospheric composition. Duhl et al. (~~submitted~~2013) seek to quantify the impacts
24 of MPB on monoterpene emissions from lodgepole pine as well as changes over time;
25 however, their results for a limited number of trees are inconclusive and appear to be
26 dominated by tree-to-tree variability. Beetle attack generally occurs during the summer
27 months when monoterpene emissions are highest (e.g. Tishmack et al., 2005; Safranyik and
28 Carroll, 2006), therefore we focused on summertime impacts in this study. Although we have
29 assumed that trees are impacted by beetle attack for a full year, it is possible that trees that
30 survive a beetle attack may have an emissions scale-up effect lasting several years, while
31 other trees that succumb quickly to the beetle may only have a scale-up effect lasting a few
32 months. Observational studies done over longer periods of time are required to discern the

1 temporal changes in emissions from a tree under attack. There is also a clear need to study
2 the quantitative change in monoterpene emissions due to other beetles and insect infestations
3 in other tree species (MPB attacking lodgepole pine is by far the most studied combination).

4 Increased surface temperatures measured in beetle-killed forests by Hais and Kucera (2008)
5 and modeled by Wiedinmyer et al. (2012) may also increase VOC emissions beyond what we
6 show here, as VOC emissions increase exponentially with increasing temperature (Guenther
7 et al., 1993). Similarly, post-outbreak forestry activity such as salvage logging could impact
8 VOC emissions (e.g. Räisänen et al. 2008).

9 Following a beetle infestation, there may also be changes in the VOC emissions profile of
10 forests due to recovery that includes establishment and dominance of different tree species.

11 Although ~~re-postdisturbance~~ vegetation dynamics are not fully understood, Collins et al.
12 (2011) find that in remote forest in Colorado, lodgepole pine stands can be replaced with
13 deciduous trees such as aspen, which emit mainly isoprene. However, in other areas,
14 coniferous trees such as subalpine fir or Engelmann spruce may initially replace lodgepole
15 pine (Collins et al., 2011). Any transformation of vegetation will cause a change in the
16 amounts and types of VOCs emitted, as well as subsequent SOA loadings in the region.

17 Changes in species composition following beetle outbreaks will likely impact the longer term
18 emission characteristics of forests in western North America. ~~Free succession is expected to~~
19 ~~occur on longer timescales than the decade of beetle infestation investigated here, but will~~
20 ~~likely impact the longer term emission characteristics of forests in Westernwestern North~~
21 ~~America.~~

22 SOA yields are loading-dependent (e.g. Presto and Donahue, 2006; Shilling et al., 2008).
23 Typical total OA concentrations in the atmosphere are less than or equal to $5 \mu\text{g m}^{-3}$ (Presto
24 and Donahue, 2006). However, most SOA chamber experiments are conducted with much
25 higher organic mass concentrations, including Lee et al. (2006a,b). It is therefore likely that
26 yield parameters used in this study overestimate SOA formation in ambient conditions,
27 however yield estimates obtained at low loading conditions are not available for all our
28 monoterpene precursors of interest. ~~However,~~ ~~†~~ This overestimate in our simulated SOA is
29 somewhat offset by our neglect of SOA formed via NO_3 oxidation and from ~~isoprene~~
30 ~~oxidation~~ other precursors (i.e. isoprene, sesquiterpenes, methyl salicylate), emissions of
31 which may also be affected by bark beetle attack. ~~although~~ ~~u~~ncertainties in all these yields
32 preclude any firm estimates of the degree of compensation.

1

2 **5 Conclusions**

3 This is the first modeling study to assess the impact of bark beetle kill on monoterpene
4 emissions and air quality. We show that at least locally, beetle infestation may have a
5 significant impact on atmospheric composition in ~~Western~~western North America.

6 Two main effects emerge from this study – the mortality effect and the attack effect.
7 Spatially, responses to beetle attack can vary greatly with smaller-scale areas showing
8 relatively large changes in monoterpene emissions and SOA concentrations (up to four-fold),
9 while for ~~Western~~western North America as a whole, the mortality effect can overcome or
10 greatly mute the attack effect. Any compensation between the mortality and attack effect at
11 the regional scale will depend on the spatial and temporal patterns of tree mortality:
12 compensation will be greater when different areas experience beetle outbreaks in different
13 years. The response to beetle attack also varies from year-to-year, depending on the number
14 of trees under attack and the magnitude of the cumulative mortality effect.

15 In both the pine and spruce scenario explored here, MPB has the largest impact in BC in 2004
16 and the ~~Western~~western US in 2008, with a much larger impact when the simulated attack
17 effect is based on the response of Engelmann spruce. OB have the largest impact in both BC
18 and the ~~Western~~western US in 2002, although the OB impact in BC is small in all years.
19 Although many of the large relative increases in monoterpene emissions and SOA
20 concentrations we see across ~~Western~~western North America are not likely observable in
21 measurements of OA or PM_{2.5} due to the large annual and interannual variability in these
22 measurements, in areas of spruce under attack and lower OA variability, beetle-induced SOA
23 changes may be observable over time. Furthermore, these changes in natural aerosol, and
24 similar changes resulting from future beetle attacks, may impact the achievement of natural
25 visibility objectives set forth by the U. S. EPA Regional Haze Rule.

26 This initial modeling study captures the general picture of how beetles may affect
27 monoterpenes and SOA in ~~Western~~western North America. MPB outbreaks in
28 ~~Western~~western North America are severe and still spreading. Furthermore, other regions in
29 the world also experience large-scale herbivory attacks such as the southeast US (Duehl et al.,
30 2011) and Europe (Seidl et al., 2011). Thus, further experimental work characterizing the
31 effect of herbivory on BVOC emissions is needed. Further work is also required to couple the

1 aerosol impacts examined here to beetle-associated changes in meteorology, ozone and fire
2 susceptibility to attain a complete picture of beetle infestation impacts on air quality.

3

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1 Table 1. Scale-up factors for bark beetle-induced changes in monoterpene emissions
 2 calculated from Amin et al. (2012a, 2013b) lodgepole pine and Engelmann spruce data.

Monoterpene	Scale-up Factor		Baseline simulated total emissions
	Pine	Spruce	(Western western North America) TgC yr ⁻¹
β-pinene	7.7	16	0.36
3-carene	7.3	65	0.19
β-phellandrene	33	5.3	0.036
P-cymene	5.4	42	0.014
α-pinene	-	3.6	0.63
Sabinene	-	18	0.099

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4

1 Table 2. Photooxidation SOA yields from Lee et al. (2006b, or other studies as noted) and
 2 reaction rates from Atkinson et al. (1997). Primary oxidant OH.

Monoterpene	SOA Mass Yield (%)	Reaction Rate at 298K and 1 atm cm ³ molecule ⁻¹ s ⁻¹
Limonene ^a	58	1.7x10 ⁻¹⁰
β-pinene	31	7.9x10 ⁻¹¹
3-carene	38	8.8x10 ⁻¹¹
1-methyl-3-n-propylbenzene ^b	5.6	1.4x10 ⁻¹¹
α-pinene	32	5.4x10 ⁻¹¹
Sabinene ^c	10.2	1.2x10 ⁻¹⁰

a Used for β-phellandrene
 b Used for p-cymene (Odum et al., 1997)
 c Griffin et al. (1999)

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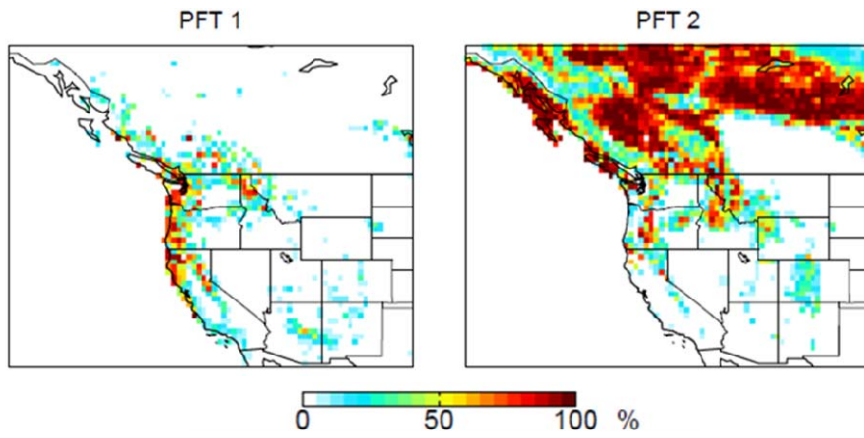
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1 Table 3. Dark ozonolysis SOA yields from Lee et al. (2006a, or other studies as noted) and
2 reaction rates from Atkinson et al. (1997). Primary oxidant O₃.

Monoterpene	SOA Mass Yield (%)	Reaction Rate at 298K and 1 atm cm ³ molecule ⁻¹ s ⁻¹
β-pinene	17	1.5x10 ⁻¹⁷
3-carene	54	3.7x10 ⁻¹⁷
α-pinene	41	8.7x10 ⁻¹⁷
Sabinene ^a	3.0	8.6x10 ⁻¹⁷

a Griffin et al. (1999)

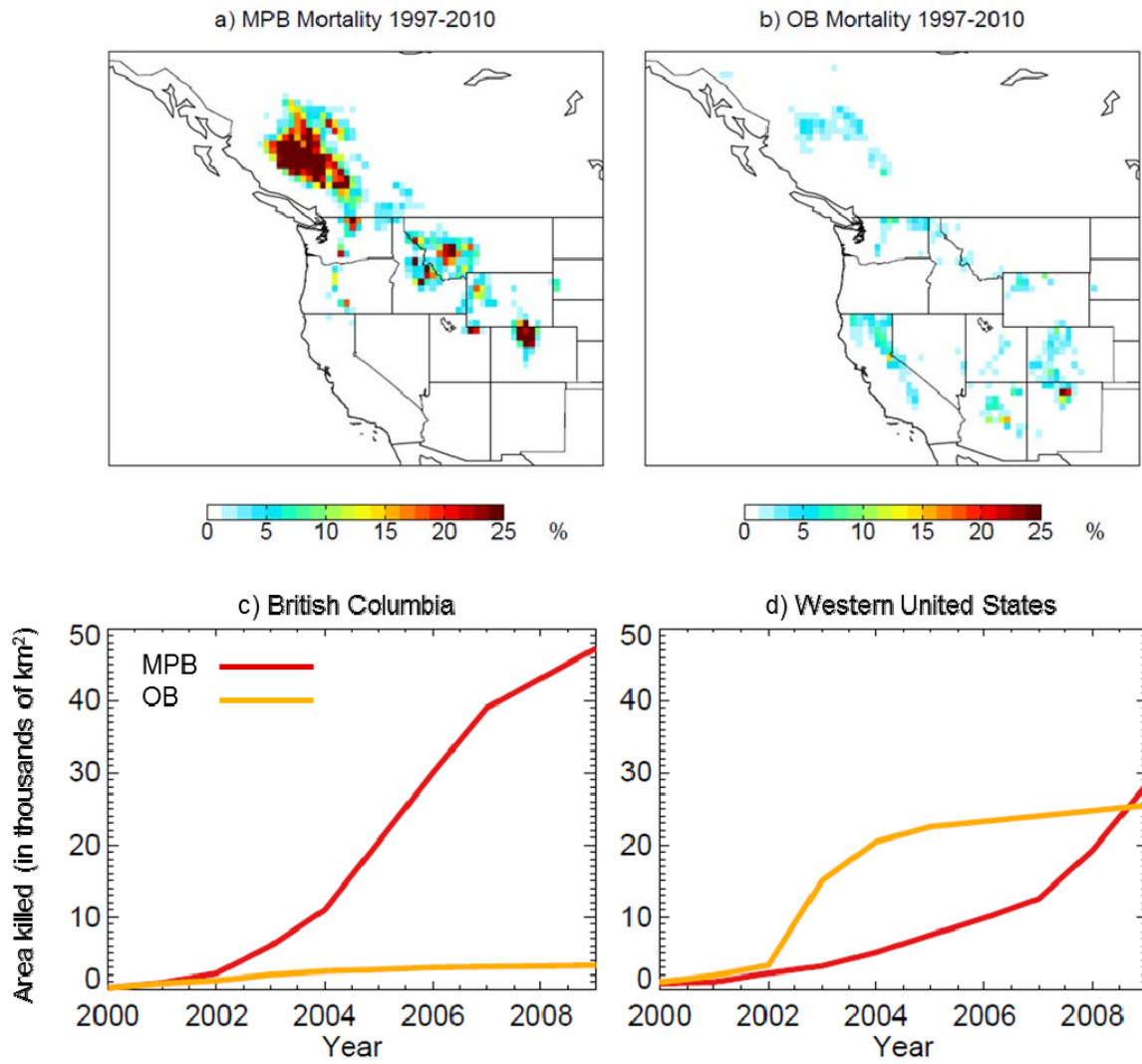
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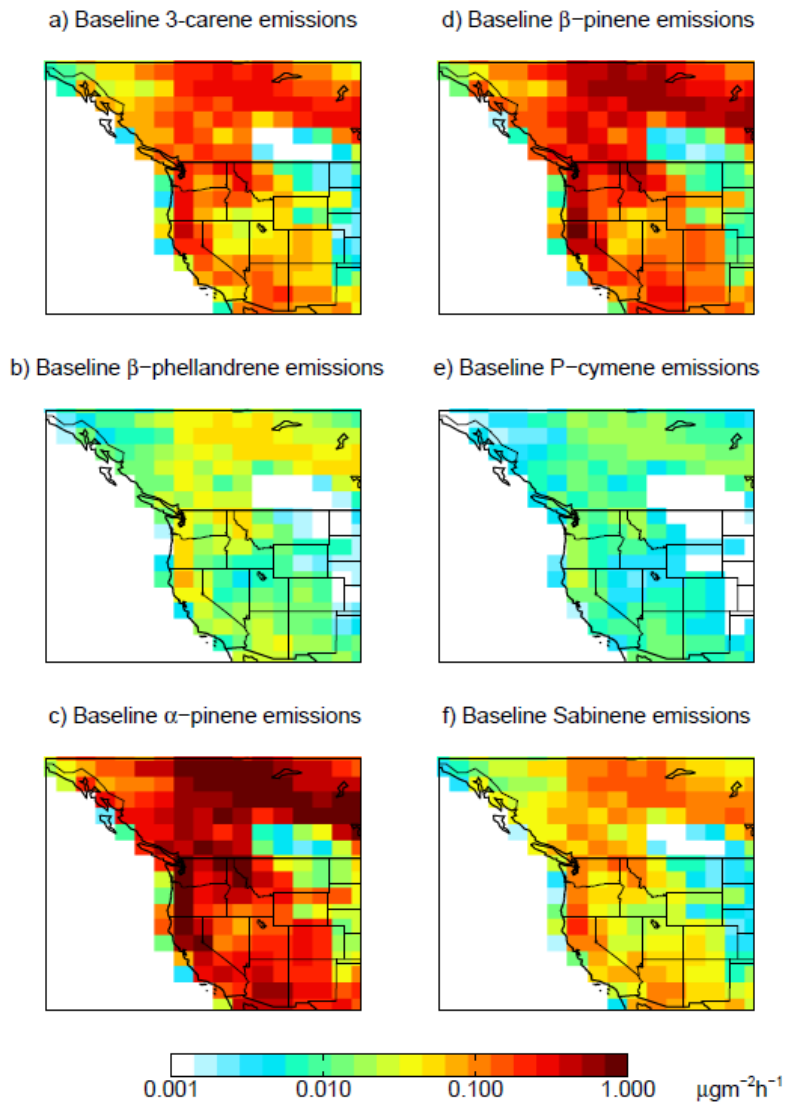
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3 Fig. 1. Percent of total surface area occupied by PFT 1 (needleleaf evergreen temperate tree)
4 | and PFT 2 (needleleaf evergreen boreal tree) in ~~Western~~western North America.



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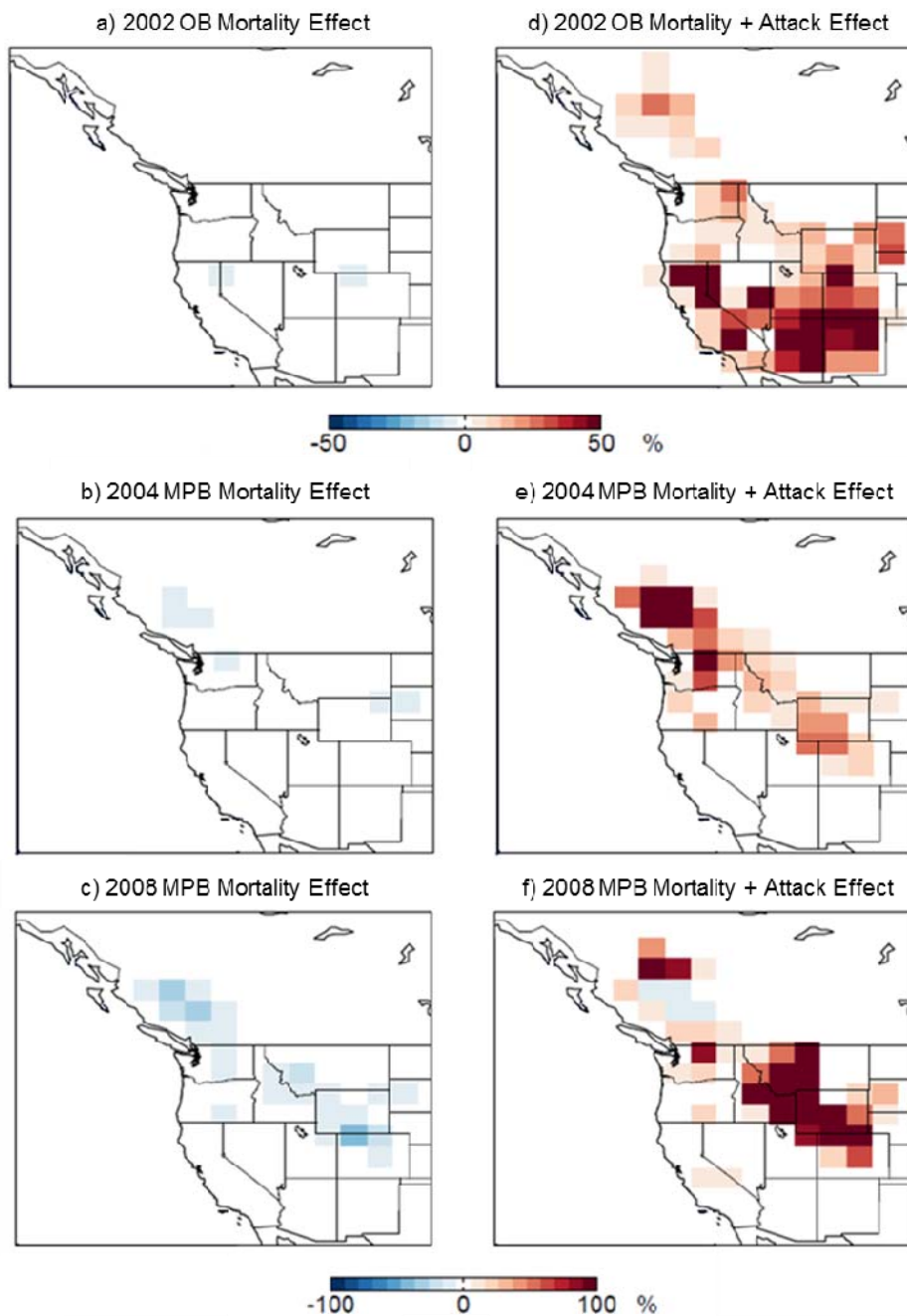
Fig. 2. Cumulative percent of area killed in 2010 in the BC and the ~~Western~~western US due to (a) mountain pine beetle (MPB) and (b) other beetles (OB (cColor bar saturated at 25%) and cumulative ~~beetle mortality~~beetle-induced tree mortality over time in (c) BC and (d) ~~Western~~western US caused by the mountain pine beetle and other beetles.



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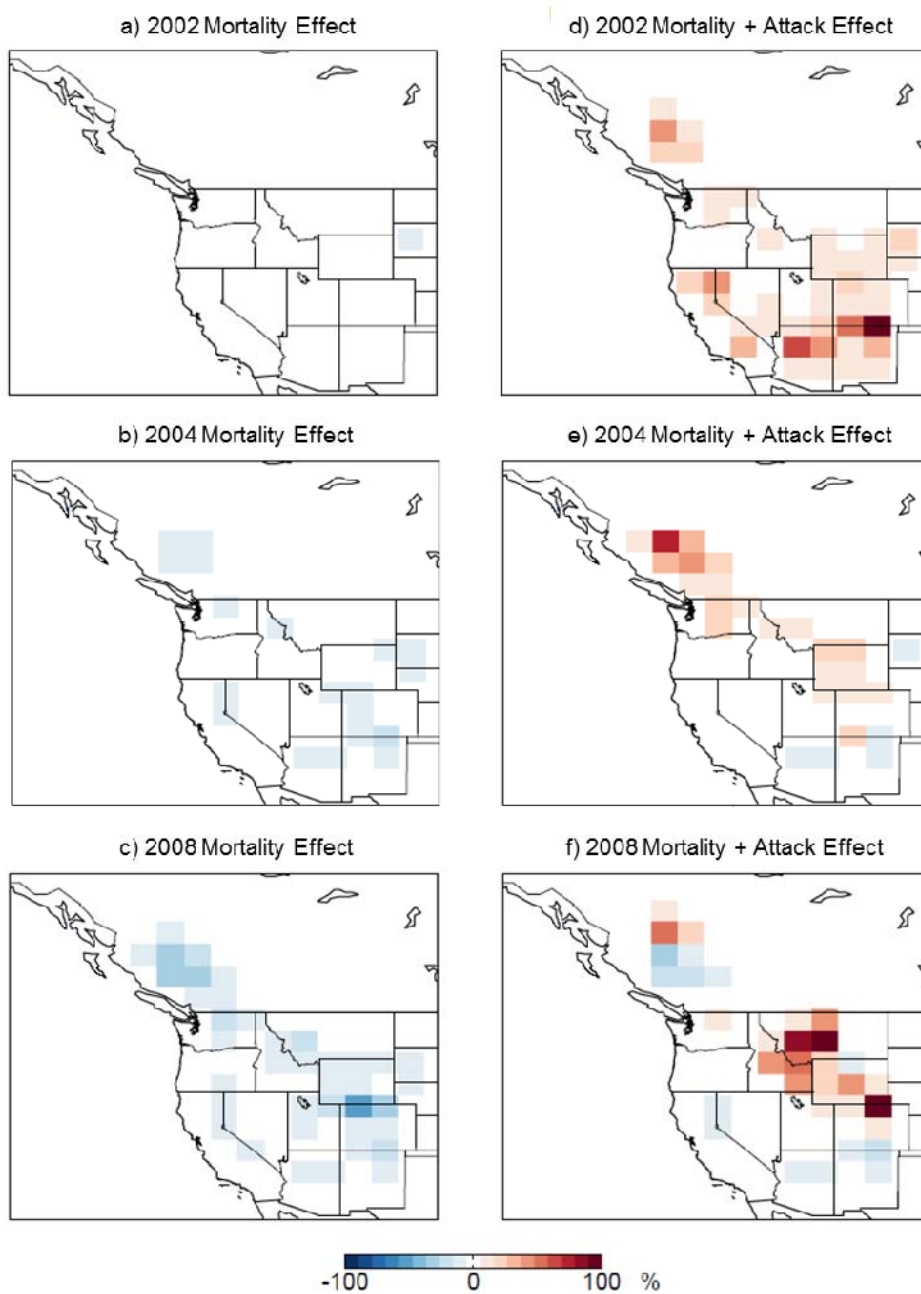
3 Fig. 3. Simulated summer-mean simulated baseline emissions of monoterpenes potentially
 4 influenced by beetles: a) 3-carene, b) β -phellandrene, c) α -pinene, d) β -pinene, e) p-cymene,
 5 and f) sabinene. Color bars are saturated at their respected values $1 \mu\text{g m}^{-2}\text{h}^{-1}$.



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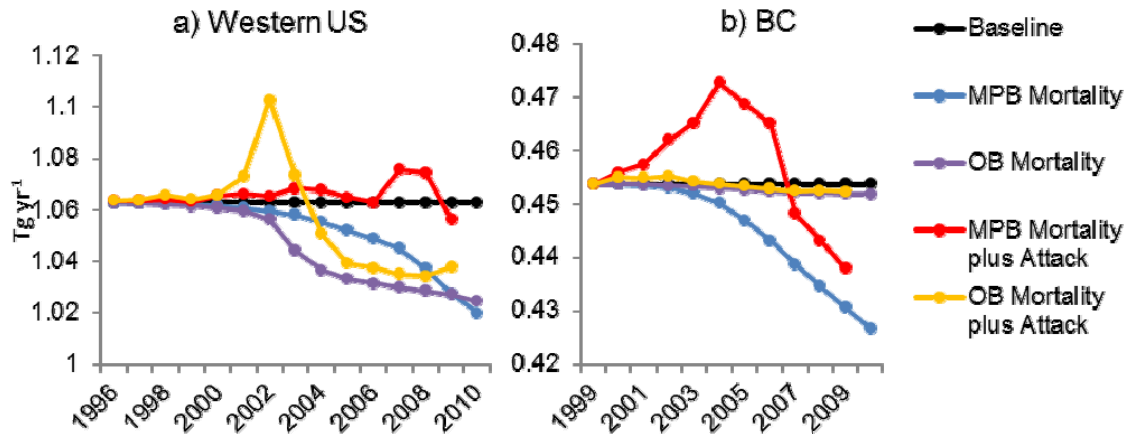
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3 Fig. 4. Change in simulated summer-mean β -phellandrene emissions due to other beetle attack
 4 in 2002 and mountain pine beetle attack in 2004 and 2008 following the pine scenario.
 5 Change in emissions due to the mortality effect alone (a-c) and the mortality plus attack effect
 6 (d-f). Color bars are saturated at their respected values.



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3 Fig. 5. Change in simulated summer-mean emissions of the four monoterpenes impacted by
4 all beetles in 2002, 2004, and 2008. Change in emissions due to the mortality effect alone (a-
5 c) and the mortality effect plus the attack effect (d-f). Color bar is saturated at 100%.



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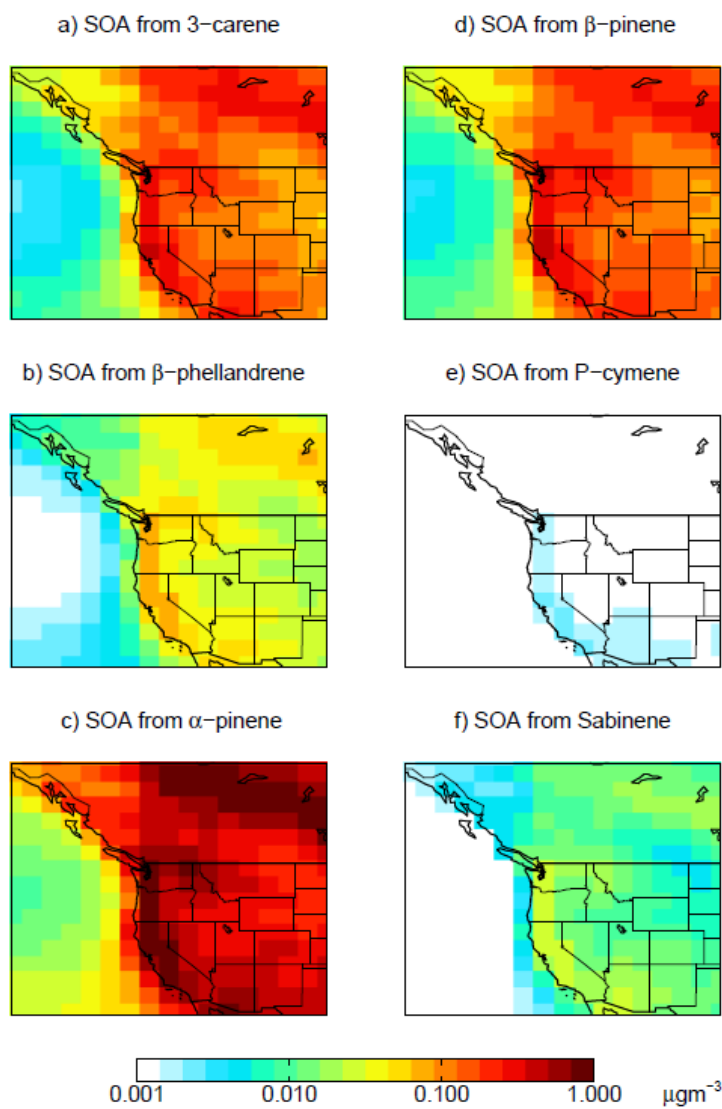
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3 Fig. 6. Time series showing evolution of the simulated total regional monoterpene emissions

4 (sum of 3-carene, β -pinene, β -phellandrene, p-cymene, α -pinene, and sabinene) affected by

5 mountain pine beetle (MPB) and other bark beetle (OB)-caused tree mortality ~~kill~~ in a)

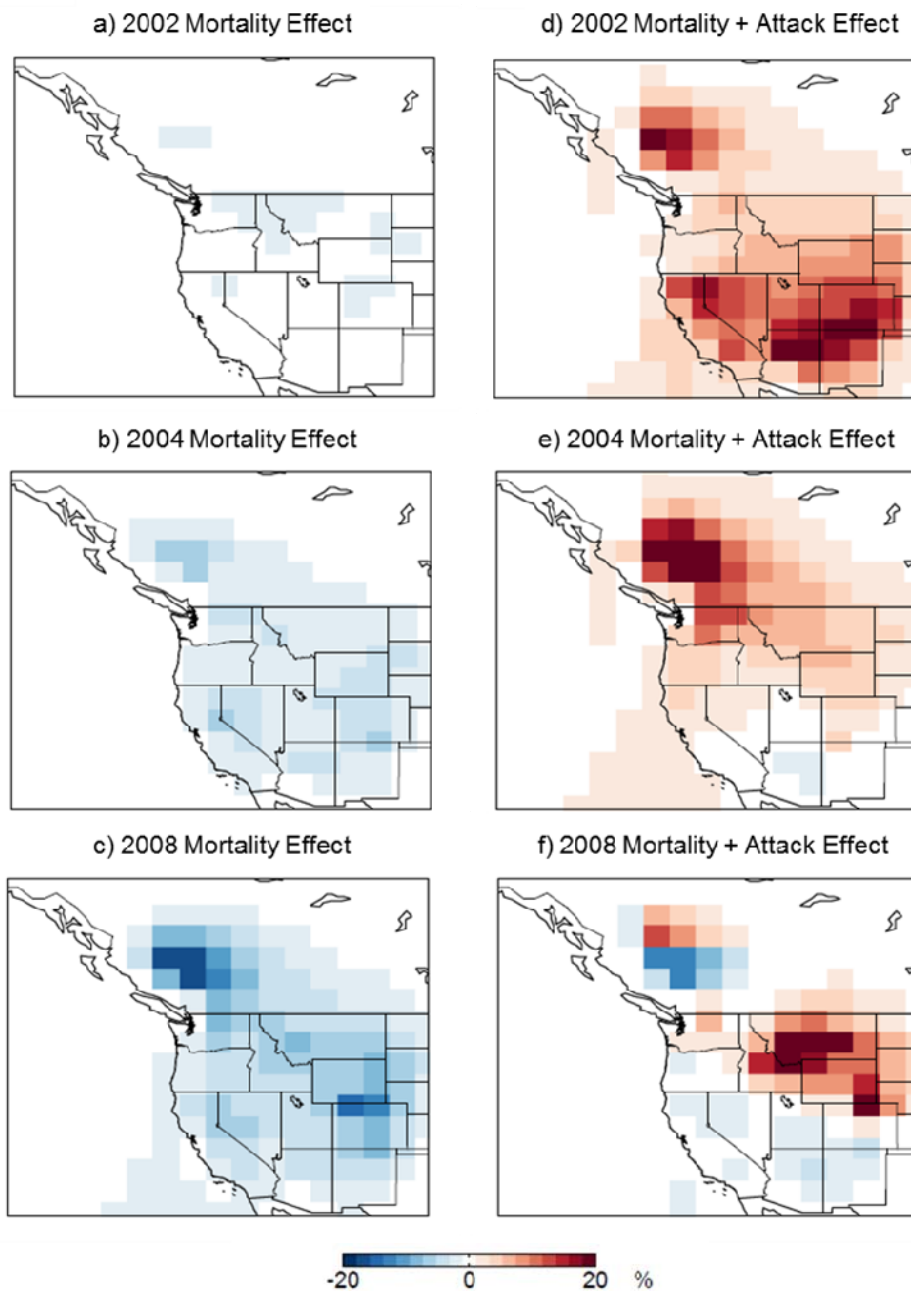
6 wWesternwestern US and b) BC.



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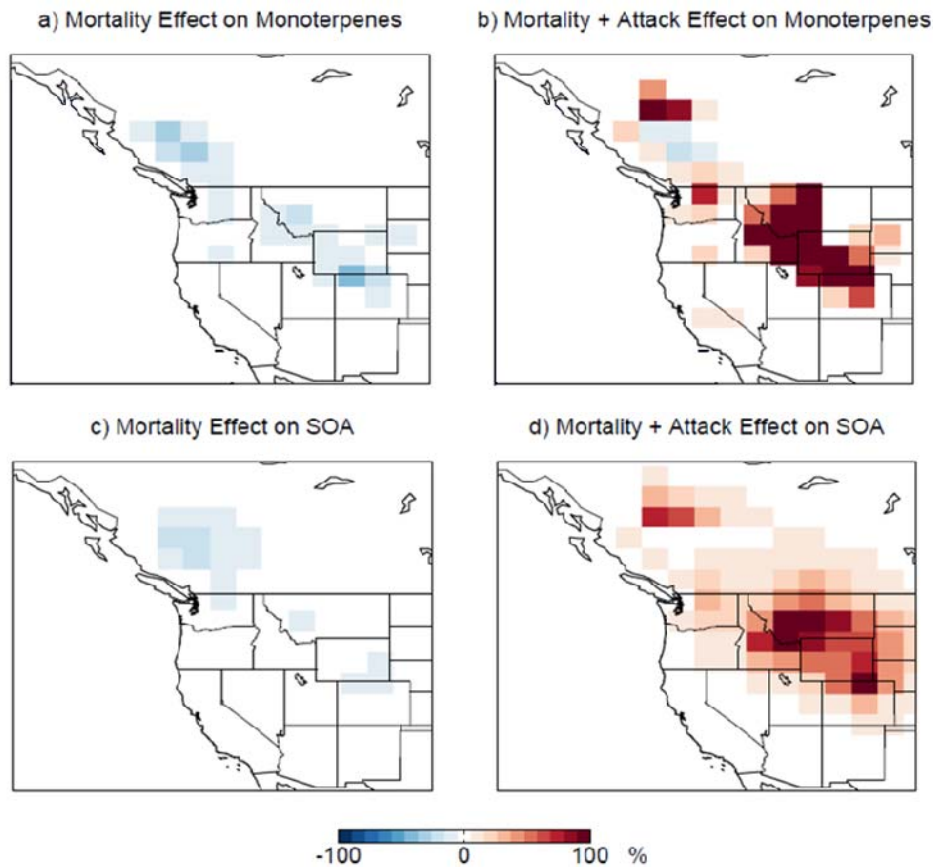
3 Fig. 7. Baseline summer-mean simulated concentrations of SOA from a) 3-carene, b) β -
 4 phellandrene, c) α -pinene, d) β -pinene, e) p-cymene, and f) sabinene. Color bars ~~are~~is
 5 saturated at ~~their respected values~~1 μgm^{-3} .



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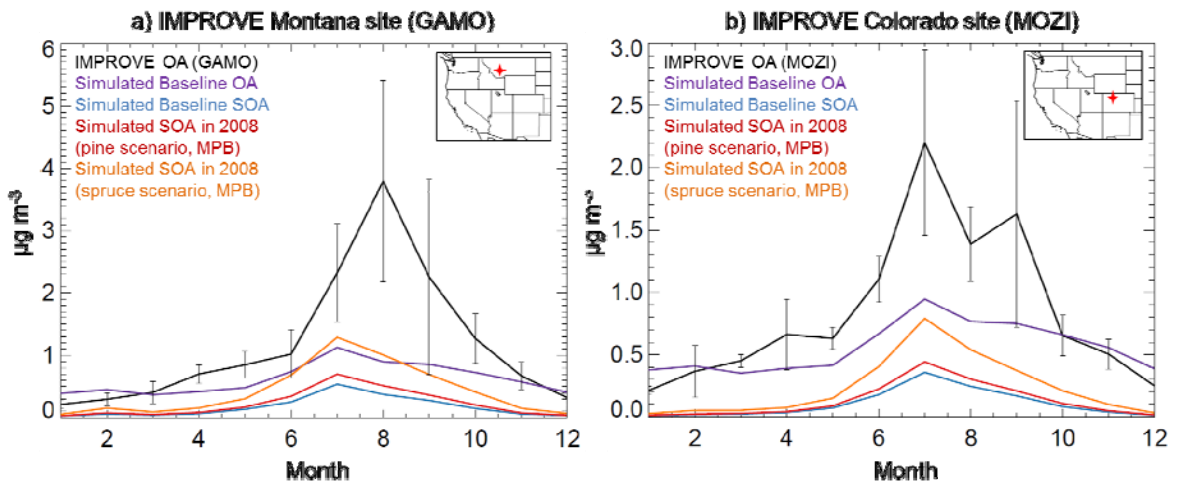
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3 Fig. 8. Change in simulated summer-mean SOA concentration from all six monoterpenes due
 4 to the impact of both mountain pine beetle and other beetle attack in 2002, 2004, and 2008
 5 | (following-in the pine scenario). Change due to the mortality effect alone (a-c) and mortality
 6 effect plus the attack effect (d-f). The color bar is saturated at 20%.



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Fig. 9. Change in simulated summer-mean monoterpene emissions and summer-mean SOA concentrations from all four monoterpenes due to the of impact mountain pine beetle attack in 2008 for the spruce scenario (compare with Figs. 5c, 5f, 8c and 8fd and 8e, d. a) Change in monoterpene emissions due to mortality effect alone. b) Change in monoterpene emissions due to mortality effect plus the attack effect. c) Change in SOA concentrations due to the mortality effect. d) Change in SOA concentrations due to the mortality effect plus the attack effect. The color bar is saturated at 100%.



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3 Fig. 10. Comparison between observed baseline IMPROVE OA concentrations from 2008 to
 4 simulated baseline OA and SOA at a site in **a)** Montana (GAMO) and **b)** Colorado (MOZI).
 5 Simulated SOA concentrations under the pine and spruce scenario are also shown. Error bars
 6 for IMPROVE observations are standard deviation of monthly means.