Author's Comments on manuscript ACP-2019-479 "Potential Regional Air Quality Impacts of Cannabis Cultivation Facilities in Denver, Colorado"

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Response to our 2 anonymous reviewers

We thank our reviewers for their comments regarding the importance and timeliness of our study. The reviewers' comments are in *grey italics* and our response is given in black.

Anonymous referee #1:

This manuscript is well written and provides an important initial step toward estimating VOC emissions from a growing industrial sector and demonstrates potential air quality impacts that might be anticipated given some of the range in uncertainty related to quantifying these emissions.

I only have a few minor suggestions/potential revisions.

Section 3.2.1 is rather hard to follow. The paragraph starting at line 351 talks about reductions in pollutants but the levels that follow seem to increase. Also, I found this section a little hard to follow since I am working with the presumption there are no photochemical reactions happening and everything being discussed relates to non- photochemical reactions in the model. Is that correct? I was a little surprised that hydroxyl radical production could increase overnight.

Section 3.2.1: We have modified the text to clarify the PA process. L341-350 now read:

"To better understand why ozone increased at night, the PA model output was analyzed to quantify the chemical and physical processes producing ozone. Plume tracking was used so that only grid cells where the increase in ozone (i.e. the plume) occurred were included in our analysis, which ran from July 27th, 9:00 PM to July 28th, 6:00 AM LST. The number of vertical model layers included in the analysis also varied to incorporate the hourly evolution of the PBL. Figure S4 provides snapshots of the horizontal grid cells used and the vertical layers that were aggregated throughout the simulation time period. Fig. S5 shows the changes in final ozone concentrations (compared to the base case) for the grid cells and vertical layers included in the

analysis, as well as the physical and chemical process rates that account for these changes. Figure S5 shows that the process most responsible for increases in ozone concentrations was chemical production."

We have also expanded our explanation of the chemistry involved as the reviewer is correct that there is no photochemistry occurring at night in the model. At night, HO_2 radicals are produced from the reactions of VOCs with nitrate radicals (NO_3), OH radicals and O_3. OH radicals are formed when O_3 reacts with alkenes. We have modified the text to clarify this:

"For the chosen vertical layers and grid cells Table 3A shows the total rate of the oxidation reactions with TERP across the entire period. Throughout this time, the additional TERP emissions lead to an increase in the number of oxidation reactions thereby generating more secondary VOC products and radical species. The chemical losses of TERP increased due to reactions with: OH (from 0.01 ppb to 0.1 ppb; +900%), nitrate radical (NO_3) (from 0.39 ppb to 1.58 ppb; +305%), and O_3 (from 0.04 ppb to 0.2 ppb; +400%). Further analysis confirms that night-time oxidation chemistry leading to changes in ozone concentration are driven by NO_3. In the 3_EC scenario, TERP emissions only increased the annual VOC emission in Denver County by 3.5%, but this is sufficient to increase the VOC + NO_3 reaction rates by 125%. These increases produce more peroxyl radicals (TRO_2=HO_2 + RO_2) driving further oxidation and further radical production. Table 3B also shows that the generation of OH radicals from reactions of TERP with O_3 increased by 267%. Ultimately, these increases in initial TERP reactions with NO_3 and O_3 increase the NO to NO_2 conversions via the TRO_2 pathway by 44%, reducing the availability of NO to react with O_3. Thus, the increased ozone concentration predicted at night is actually due to the 1 ppb (0.8%) reduction in the loss of ozone to reactions with NO rather than an increase in actual production of ozone (Table 3C). The increased TERP emissions also increase production of NO_x termination products (NO_z) by 27% with organic nitrate (NTR; representing ~71% of this NO_z product) increasing from 0.66 ppb to 1.6 ppb (+142%). This increase in NO_z production at night also results in lower NO concentrations and thus lower ozone titration."

Section 3.2.3 line 397 discusses the increase in overnight O3 but I am not sure this would really be relevant in a discussion about implications with policy relevance since overnight O3 levels are usually well below the level of the standard.

We agree with the author concerning the policy relevancy. It is important to note that during the vegetative stage (roughly half the growth cycle of *Cannabis spp.*) CCFs are under lights 24-hours per day so nighttime monoterpene emission rates remain similar to those during the day. Thus, these emissions can affect night-time chemistry in a way that is unique for BVOCs, as was shown in our model simulations. We have modified the text to clarify that our goal for this part of the analysis was the scientific interest of their impact on nighttime chemistry.

In the conclusion section, line 402-403 needs a qualification that the emissions is the first inventory for Colorado and not the United States.

To our knowledge this is the first inventory of terpene emissions for the cannabis industry to have been conducted anywhere. Our statement is ambiguous and L402-403 has been amended to read:

"This study provides the first VOC emission inventory to be compiled for the cannabis industry in Colorado, the first time such analysis has been conducted anywhere in the USA."

Anonymous referee #2:

This manuscript presents a first attempt at compiling state-wide (Colorado) emissions inventory for monoterpenes from cannabis cultivation facilities (CCFs). The new emissions inventory is incorporated into a chemical transport model to evaluate the impact of CCFs on ambient ozone concentrations. The manuscript is well written and the topic is of interest to the ACP research community and the general public, as it is important to know how much CCFs can impact air quality and provide information to decision maker on whether mitigations may be necessary to reduce the impact. Given the interest in the topic, the large gap in data and information, and generally appropriate methodology and analysis, the manuscript is acceptable for publication provided some revisions are made to clarify some points and to not overstate the results.

Because of the large uncertainties in the emissions, the study carried out sensitivity simulations, with emissions spanning a factor of 10, to evaluate the range of potential impacts on ozone. The manuscript states that the study used "realistic bounds on each parameter" for the emissions parameterization, but it does not clearly explain why the factors chosen were considered realistic. For all parameters (EC, DPW, and PC), in- sufficient justification was provided on why parameter values based on leaf enclosures data of Wang et al., 2018 are considered lower bounds. The statement ". . .plants stud- ied by Wang et al., however, were not grown in the optimized conditions found in a CCF and the reported ECs could be conservative" needs support/citations. Optimal growth conditions are not necessarily correlated with magnitude of monoterpene emissions. Even if one considers EC values of Wang et al., 2018 be to lower bounds, what is the basis to say that a multiple of 10 is realistic?

We thank our reviewer for their constructive comments on our sensitivity simulations. As the reviewer themselves points out, there are considerable uncertainties around all of the factors included in our estimated inventory. Here, we further clarify and justify our parameter choices in response and have modified the manuscript accordingly.

"For all parameters (EC, DPW, and PC), insufficient justification was provided on why parameter values based on leaf enclosure data of Wang et al., 2018 are considered lower bounds."

The DPW (plant dry weight) is not based on the enclosure measurement study. Instead, these are based on figures from Washington State Liquor and Cannabis Board and from Colorado Department of Revenue who oversee the licensing of Cannabis Cultivation Facilities.

Figure S1 summarises the available (reliable) data on material harvested from CCFs; here from Washington State. We use the yields of wet and dry buds (the marketable material) to deduce the water content in Cannabis buds and then assume that the water content is the same in the remaining plant material. From this we are able to estimate the dry weight of an average *Cannabis spp.* plant as described in the main text in L148-169. We use this mean of ~750 g (N = 18,257) as the base case value for DPW. The standard deviation in this estimate is of similar magnitude and we take the mean +1 s.d. (1500 g) as the value of DPW for our first sensitivity test. Our final value (2500 g) is considerably higher and represents the maximum yield recorded by Washington State Liquor and Cannabis Board. As the total plant count and

reported yields are 3 and 4 higher respectively in Colorado than Washington state (LCB, 2017; Topshelfdata, 2017; Hartman et al., 2018b), we used this maximum on the assumption that *Cannabis spp.* cultivated in CCFs in Colorado in summer season is grown under more optimal conditions than those grown in Washington State resulting in considerably higher yields.

We have clarified this a little further in the main text. L173-177 have been modified to read:

"The average and standard deviation of DPW was 754 g (1-2,260 g) (Fig. S1e). For the development of these emission inventories, a base value of 750 g was assumed for DPW based on the average calculated from the Washington database. As a sensitivity test, a DPW of 1,500 g representing the mean plus one standard deviation range was chosen. Finally, a DPW of 2,500 g, the maximum yield recorded by Washington State Liquor and Cannabis Board, was taken as the upper statistical boundary as shown in Fig. S1e. As the total plant count and reported yields are 3 and 4 factor higher respectively in Colorado than Washington state (LCB, 2017; Topshelfdata, 2017; Hartman et al., 2018b), we took this maximum on the assumption that *Cannabis spp.* cultivated in CCFs in Colorado in summer season is grown under more optimal conditions than those grown in Washington State resulting in considerably higher yields."

Table S2 shows the maximum number of plants permitted in a CCF for each licence tier in Colorado. This shows that our choice of value for PC is well below the maximum for Tier 1 premises. As explained in the main text (L179-184) our base value of PC is based on the current (June 2018) 1 million "mature" Cannabis plants under cultivation in Denver County, with two sensitivity simulations exploring a doubling in plant numbers (commensurate with continued expansion at the same rate, as explained in the main text in L185-195) and finally, a simulation with each CCF containing the maximum possible number of plants under a Tier 1 licence. These are summarised in Table S1.

We have changed the main text to clarify our parameter values. L179-195 now reads:

"Counts of all plants larger than 8 inches have been recorded by the Colorado DOR on a monthly basis since 2014. As of June 2018, there are a total of 1.06 million plants (Hartman et al., 2018a, b). We therefore used 1 million as the base number for the emission inventory. The DOR data only provides county-level information rather than actual number of plants per CCF. The plants were then distributed equally among the CCFs to calculate an average of 905 plants per facility in Denver County and 521 outside of the county.

Two sensitivity simulations were conducted based on the assumption that the cannabis industry in Colorado will continue to expand at similar rates in the future. From June 2016 to June 2018 the total number of plants recorded by DOR grew from 826,963 to 1,062,765, an annual average increase of 118,000. Assuming this rate of expansion remains constant, there would be 2 million plants in the state of Colorado by 2025 and this value was used in simulation 6_PC. It was assumed in simulation 7_PC that growth would accelerate in the future to the point at which each recreational and medical CCF would contain the maximum number of plants permitted under a Tier 1 license leading to a state-wide total of nearly 4 million plants. The maximum number of plants that can be grown under each licensing tier is shown in supplemental Table S2 (CDOR, 2019). The average plant count per CCF for each PC sensitivity simulation are shown in Table S1."

The monoterpene emission capacity (EC) was based on the enclosure measurements described by Wang et al., 2018. In this study, emission rates from 4 cultivars were found to vary widely among Cannabis spp. strains and across growth stages. The base value for EC was taken as 10

μg gdw^{-1} h^{-1} based on the average emissions from Critical Mass at the vegetative growth stage. It has been reported that during the flowering stage the bud tissues contain a significant amount of monoterpenes. Further, the Spokane Regional Clean Air Agency (SRCAA) and Washington State University measured monoterpene concentrations from indoor cannabis facilities in grow rooms (Southwellb et al., 2017). They found concentrations of monoterpenes in grow room with 80 days old plants (1,660 ppb) to be 10 times higher than the 48 days old plants (150 ppb) suggesting that the emission rate from plants in the flowering state is higher than those measured at the vegetative stage. Since no studies in which emission rates of monoterpenes from buds have been reported, we feel that the range proposed is sufficiently wide to provide useful information regarding possible impact of this uncertainty.

We have modified the main text to incorporate these points. L134-147 now read:

"Wang et al. (2018) only sampled during the vegetative stage, and to our knowledge emission rates of monoterpenes from buds or flowers do not exist. It is not known how much EC will change during these different growth stages, but the grey literature does report that CCFs actively select cultivars to maximise the amount of monoterpenes found in the bud tissues. The Spokane Regional Clean Air Agency (SRCAA), in collaboration with Washington State University (Southwellb et al., 2017; Wen et al., 2017), measured monoterpenes in flowering rooms of CCFs in Washington state. They found concentrations of monoterpenes in grow room with 80 days old plants (1,660 ppb) to be >10 times higher than the 48 days old plants (150 ppb). CCFs in Colorado house a wide variety of strains at both vegetative and flowering stages of growth suggesting that the emission rate of monoterpenes from CCFs is higher than that measured from foliage by Wang et al. (2019). Currently, no database exists that can provide the number of plants by strain and growth stage. For the base case, it was assumed that each CCF grew only one strain and that all plants were at the vegetative growth stage resulting in a single and constant EC for each CCF; taken to be 10 μg gdw^{-1} h^{-1} of total monoterpenes based on the reported EC from the Critical Mass cultivar (Wang et al., 2019). Given the uncertainty in EC, the variety of possible plant stages and cultivars, the EC used in simulation 1_EC was multiplied by a factor of 5 and 10 in simulations 2_EC and 3_EC as a sensitivity analysis."

Even in the sensitivity case with a factor of 10 increase in emissions, the impact of in- creased monoterpenes associated with CCF is less than 0.5 ppb in hourly ozone during the daytime and only ~ 0.14 for maximum daily average 8-hour (MDA8) ozone. This is unsurprising because the percent increase in VOC emissions is only 3.5% for Denver County for the sensitive case that has 10x the base-case CCF emissions (1_EC). Figure 10's axis going up to 4000 ton/year is hardly meaningful as 1000 ton/year increase in Denver is nearly 30 times the base-case CCF emissions, and even then the increase is only 0.38 ppb in MDA8 ozone. Thus, "further data are urgently required regarding CCF-specific information on plant counts. . ." is overstating the urgency of needing to improve quantification of CCF terpene emissions with respective to ambient ozone.

We agree with the reviewer and have changed the language.

The statement now reads

"Further data are needed to reduce uncertainties in emission inventory estimates specifically those regarding CCF-specific information on plant counts, …"

There are 7 sensitivity simulations listed, but in reality there are only 6 sensitivity cases because simulation 6_PC is the same as simulation 4_DPW. Because the values of EC, DPW, and PC are assumed to be constants, the emission increase is uniform across the simulation domain such that: 2_EC = 5 x 1_EC, 3_EC = 10 x 1_EC, 4_DPW = 6_PC = 2 x 1_EC, 5_DPW = 3.33 x 1_EC, 7_PC = 4 x 1_EC. Really only 3 sensitive simulations (2x, 5x, and 10 x 1_EC) was needed to cover the emissions range explored by the 7 sensitivities simulations.

Although the reviewer is correct that the 6_PC and 4_DPW sensitivity tests are effectively the same, the conception of these two scenarios were different. The 4_DPW emission assumed a heavier dry biomass, and 6_PC is for a future plant count estimate. In addition, the 5_DPW and 7_PC represent the upper bounds of DPW and PC showing their relative impacts on the inventory. Assessing the contribution of the individual factors on our emission inventory of, we concluded that it is the emission capacity (EC) of *cannabis spp.* that is the most significant and also the most uncertain.

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Potential Regional Air Quality Impacts of Cannabis Cultivation Facilities in Denver, Colorado

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Abstract

The legal commercialization of cannabis for recreational and medical use has effectively created a new and almost unregulated cultivation industry. In 2018, within the Denver County limits, there were more than 600 registered cannabis cultivation facilities (CCFs) for recreational and medical use, mostly housed in commercial warehouses. Measurements have found concentrations of highly reactive terpenes from the headspace above cannabis plants that, when released in the atmosphere, could impact air quality. Here we developed the first emission inventory for cannabis emissions of terpenes. The range of possible emissions from these facilities was 66-657 metric tons/year of terpenes across the state of Colorado; half of the emissions are from Denver County. Our estimates are based on the best available information and highlight the critical data gaps needed to reduce uncertainties. These realizations of inventories were then used with a regulatory air quality model, developed by the State of Colorado to predict regional ozone impacts. It was found that most of the predicted changes occur in the vicinity of CCFs concentrated in Denver. An increase of 362 metric tons/year of terpene emissions in Denver County resulted in increases of up to 0.34 ppb in hourly ozone concentrations during the morning and 0.67 ppb at night. Model predictions indicate that in Denver County every 1,000 metric tons/year increase of terpenes results in 1 ppb increase in daytime hourly ozone concentrations and a maximum daily 8-hour average (MDA8) increase of 0.3 ppb. The emission inventories developed here are highly uncertain, but highlight the need for more detailed cannabis and CCFs data to fully understand the

possible impacts of this new industry on regional air quality.

Keywords: *Cannabis spp.*; emission inventory; biogenic volatile organic compound; terpene; particulate matter; ozone; air quality

1. Introduction

The rapid expansion of one of the United States' newest industries, the commercial production and sale of recreational cannabis, was recently likened to the millennial "dot com" boom (Borchardt, 2017). With an increasing number of states passing bills to legalize recreational cannabis, the enterprise is set to rival all but the largest of current businesses. The cultivation, sale, and consumption of recreational cannabis annual sales revenues had reached $1.5 billion in the US state of Colorado by 2017 (CDOR, 2018b), exceeding revenues generated by grain farming in the state. The commercial cultivation and sale of cannabis is not subject to the same strict environmental monitoring and reporting procedures as other industries of similar size. While the relaxation of laws has provided certain medicinal and economic opportunities for the states involved, the potentially significant environmental impact on air quality due to the production of cannabis has largely been ignored.

Previous research on the wider impacts of cannabis production has been limited due to its federal status as an illegal or controlled substance (Crick et al., 2013; Eisenstein, 2015; Andreae et al., 2016; Stith and Vigil, 2016). As a result of this status, most studies have focused on the pharmacological and health effects of the psychoactive constituents of *Cannabis spp.* (Ashton, 2001; Borgelt et al., 2013; WHO, 2016), or the societal impacts associated with the illicit nature of the industry (IDCP, 1995; Sznitman and Zolotov, 2015; WHO, 2016). The few assessments to date on the environmental impacts of the production of *Cannabis spp.* have centered on the detrimental effects of outdoor cultivation on ecosystems and watersheds due to land clearance and high water demand (Bauer et al., 2015; Carah et al., 2015; Butsic and Brenner, 2016). Studies have also quantified the energy consumption of the industry and the resulting greenhouse gas emissions associated with indoor cultivation (Mills, 2012). Little attention has been paid to the possible biogenic volatile organic compounds (BVOCs) emitted from the growing of cannabis and its impact on indoor and outdoor air quality.

The only studies that have measured the composition of gaseous emissions from cannabis have been limited to headspace samples above the plants (Hood et al., 1973; Turner et al., 1980; Martyny et al., 2013). These studies have shown high concentrations of VOCs

such as monoterpenes ($C_{10}H_{16}$), sesquiterpenes ($C_{15}H_{24}$), and cannabinoids. These studies also measured thiols, a sulfur-containing compound responsible for the characteristic odor of *Cannabis spp*. (Rice and Koziel, 2015b, a). The principle (trace) components are reported to be: α- and β-pinene, β-myrcene, d-limonene, cis-ocimene, β-caryophyllene, β-farnesene and α-humulene (Hood et al., 1973; Turner et al., 1980; Hillig, 2004; Fischedick et al., 2010; Martyny et al., 2013; Marchini et al., 2014; Rice and Koziel, 2015b). The precise mix of chemical species, however, was strongly dependent on strain and the growing conditions (Fischedick et al., 2010). It should be noted that the pharmacologically active ingredients, e.g., Tetrahydrocannabinol (Δ^9-THC), generally have low volatility and therefore are rarely detected in the gas-phase (Martyny et al., 2013). Measurements in (illicit) CCFs in conjunction with law enforcement raids in Colorado in 2012 found VOC concentrations of terpenes to be 50-100 ppb within growing rooms (Martyny et al., 2013). In these cases, the CCF operation contained fewer than 100 plants, compared with the thousands of plants found in currently licensed premises (CDOR, 2018a). Further, the Spokane Regional Clean Air Agency (SRCAA) study in Washington state measured indoor VOCs in seven flowering rooms and two dry bud rooms across four different CCFs. The average terpene concentration was 361 ppb (27-1,676 ppb) in those facilities (Southwellb et al., 2017). These indoor measurements indicate the presence of BVOCs, but only limited studies have actually determined the chemical profile of gases actually emitted by the growing plants. For comparison, summertime outdoor monoterpene concentrations in forested regions of Colorado are typically less than 4 ppb (Ortega et al., 2014).

Terpenoids, such as monoterpenes ($C_{10}H_{16}$) and sesquiterpenes ($C_{15}H_{24}$), are highly reactive compounds with atmospheric lifetimes ranging from seconds to hours (Fuentes et al., 2000; Seinfeld and Pandis, 2006). They are primarily biogenic in origin (Fuentes et al., 2000; Guenther et al., 2012) and their reactions alter the atmospheric oxidizing capacity, resulting in a range of low volatility products that can partition into the aerosol phase and, depending on the concentration of nitrogen oxides (NO_x), lead to the formation of ozone (Laothawornkitkul et al., 2009; Guenther et al., 2012). Both ozone and aerosols are climate-relevant components of the atmosphere as well as criteria air pollutants (USEPA, 2016).

In Colorado, the commercial growing of *Cannabis spp.* is restricted to secure and locked premises, resulting in indoor operations in most counties (CDOR, 2018a). Since legalization, the number of cannabis cultivation facilities (CCFs) has risen to 1,400 across the state of Colorado in 2018, including more than 233 registered recreational and 375 medical

CCFs within the Denver city limits alone. In Denver, the CCFs are commonly housed in commercial warehouses and the majority of these are located near transport links such as train hubs and major interstate highways (CDOR, 2019; Mills, 2012). Denver and the Front Range area are currently classified as "moderate" nonattainment of the ozone standard (USEPA, 2017). Due to that status, a federally mandated State Implementation Plan (SIP) was developed and mutually agreed upon between the state of Colorado and the United States Environmental Protection Agency (EPA) (CDPHE, 2009). Under the terms of the SIP, Colorado Air Quality Control Commission (AQCC) developed regulatory models to predict reductions in ozone precursors (CDPHE, 2009). These studies have found that ozone concentrations in Denver are VOC-sensitive, meaning that an increase in VOC concentrations will increase ozone production (UNC-IE and ENVIRON, 2013). The location of CCFs in a VOC sensitive region in Denver suggests a potential emission source that may impact regional air quality (UNC-IE and ENVIRON, 2014). This work used the best available information to produce the first emission inventory of VOCs from CCFs in Colorado. Colorado's regulatory model was then used to determine the extent that these emissions could impact regional air quality.

2. Materials and Methods

2.1 Emission Rate calculation

Figure 1a shows the locations of the licensed 739 recreational and 733 medical CCFs in Colorado as of March 2018 (CDOR, 2018a). Eq. (1) was first used to estimate an emission rate for each CCF, and then all CCFs were used to build a bottom-up BVOC emission inventory.

$$ER_i = \sum_j EC_{ij} \times DPW_{ij} \times PC_{ij} \quad (1)$$

Where, ER_i (μg h^{-1}) is the total emissions rate for CCF i based on the sum of emission rates for all j cannabis strains; EC_{ij} is the emission capacity (μg dwg^{-1} h^{-1}) for cannabis strain j in facility i, DPW_{ij} is the dry plant weight per plant (g) for cannabis strain j, and PC is the plant count number for strain j in facility i.

Since state legalization only occurred in 2014, and given the current federal illicit status of *Cannabis spp.*, there is a lack of available data for the three parameters used in Eq. (1). The following describes the assumptions made for a range of potential values of *EC, DPW*, and *PC* given the best information available.

2.1.1 Emission Capacity (EC)

The only data of *EC* from a leaf enclosure measurement are of three strains namely: Critical Mass, Lemon Wheel and Rockstar Kush, that were 45 days old (Wang et al., 2018). This study found that at this growth stage the *EC* for total monoterpenes varied among strains: 10 μg gdw^{-1} h^{-1} for Critical Mass, 7 μg gdw^{-1} h^{-1} for Lemon Wheel, and 6 μg gdw^{-1} h^{-1} for Rockstar Kush. The Department of Revenue (DOR) in Colorado has classified *Cannabis spp.* in a CCF into four different growth stages: immature (0-24 days old), vegetative (25-79 days old), flowering (80-132 days old), and at harvest (132-140 days old) (Hartman et al., 2018b). Wang et al. (2018) only sampled during the vegetative stage, and to our knowledge emission rates of monoterpenes from buds or flowers do not exist. It is not known how much EC will change during these different growth stages, but the grey literature does report that CCFs actively select cultivars to maximise the amount of monoterpenes found in the bud tissues.

The Spokane Regional Clean Air Agency (SRCAA), in collaboration with Washington State University (Southwellb et al., 2017; Wen et al., 2017), measured monoterpenes in flowering rooms of CCFs in Washington state. They found concentrations of monoterpenes in grow room with 80 days old plants (1,660 ppb) to be >10 times higher than the 48 days old plants (150 ppb). CCFs in Colorado house a wide variety of strains at both vegetative and

flowering stages of growth suggesting that the emission rate of monoterpenes from CCFs is higher than that measured from foliage by Wang et al. (2019). Currently, no database exists that can provide the number of plants by strain and growth stage. For the base case, it was assumed that each CCF grew only one strain and that all plants were at the vegetative growth stage resulting in a single and constant EC for each CCF; taken to be 10 μg gdw^{-1} h^{-1} of total monoterpenes based on the reported EC from the Critical Mass cultivar (Wang et al., 2019). Given the uncertainty in EC, the variety of possible plant stages and cultivars, the EC used in simulation 1_EC was multiplied by a factor of 5 and 10 in simulations 2_EC and 3_EC as a sensitivity analysis.

2.1.2 Dry Plant Weight (DPW)

No published studies report the *DPW* of a *Cannabis spp.* plant. Both the states of Colorado (METRC, 2018) and Washington (LCB, 2017; Topshelfdata, 2017) track the mass of the commercially sold portion of the plant, the "dry bud." The Colorado database, however, is not publicly accessible and was not available for this study. In Washington, using data from all type of facilities (outdoor and indoor) from August–October 2017, it was found that the average dry bud mass per plant was 210 g (0-586 g) shown in Fig. S1a. The Washington database also includes the "wet bud" weight defined as the mass of the bud after it was just harvested (Fig. S1b), but prior to the 7-10 day drying process. The total waste weight, or the remaining mass of the plant after the buds have been harvested, is also recorded. As shown in Eq. (2), the sum of these two masses should equal the total mass of the wet plant.

$$M_{wet\ plant} = M_{wet\ buds} + M_{waste} \quad (2)$$

Where, $M_{wet\ plant}$ is the mass of the entire wet plant (g), and $M_{wet\ bud}$ is the mass of the wet bud (g), and $M_{wet\ waste}$ is the mass of the waste (g).

Data from August-October of 2017 were used with Eq. (2), to estimate the wet plant weight resulting in an average of 3,770 g (6-13,405 g) shown in Fig. S1c. The large range in mass is due to the different growing conditions found in CCFs, and the type of strain being grown. The ratio of the wet and dry bud mass data from Washington was used as a surrogate to determine the percentage of water found in the total plant material as shown in Eq. (3).

$$R_{D/W} = M_{dry\ bud} / M_{wet\ bud} \quad (3)$$

Where, $R_{D/W}$ is the ratio of the masses of the dry to wet bud, and $M_{dry\ bud}$ (g) is the mass of the harvested buds after 7-10 days of drying (Fig. S1d).

It was assumed that the same factor could be applied to the total wet plant weight to estimate the *DPW* as shown in Eq. (4).

$$DPW = M_{wet\ plant} \times R_{D/W} \quad (4)$$

The average of DPW was 754 g (1-2,260 g). For the development of these emission inventories, a base value of 750 g was assumed for DPW based on the average calculated from the Washington database. As a sensitivity test, a DPW of 1,500 g representing the mean plus one standard deviation range was chosen. Finally, a DPW of 2,500 g, the maximum yield recorded by Washington State Liquor and Cannabis Board, was taken as the upper statistical boundary as shown in Fig. S1e. As the total plant count and reported yields are 3 and 4 factor higher respectively in Colorado than Washington state (LCB, 2017; Topshelfdata, 2017; Hartman et al., 2018b), we took this maximum on the assumption that *Cannabis spp.* cultivated in CCFs in Colorado in summer season is grown under more optimal conditions than those grown in Washington State resulting in considerably higher yields.

2.1.3 Plant Count (PC)

Counts of all plants larger than 8 inches have been recorded by the Colorado DOR on a monthly basis since 2014. As of June 2018, there are a total of 1.06 million plants (Hartman et al., 2018a, b). We therefore used 1 million as the base number for the emission inventory. The DOR data only provides county-level information rather than actual number of plants per CCF. The plants were then distributed equally among the CCFs to calculate an average of 905 plants per facility in Denver County and 521 outside of the county.

Two sensitivity simulations were conducted based on the assumption that the cannabis industry in Colorado will continue to expand at similar rates in the future. From June 2016 to June 2018 the total number of plants recorded by DOR grew from 826,963 to 1,062,765, an annual average increase of 118,000. Assuming this rate of expansion remains constant, there would be 2 million plants in the state of Colorado by 2025 and this value was used in simulation 6_PC. It was assumed in simulation 7_PC that growth would accelerate in the future to the point at which each recreational and medical CCF would contain the maximum number of plants permitted under a Tier 1 license leading to a state-wide total of nearly 4 million plants. The maximum number of plants that can be grown under each licensing tier is shown in supplemental Table S2 (CDOR, 2019). The average plant count per CCF for each PC sensitivity simulation are shown in Table S1.

2.2 Emission Inventories for Cannabis Cultivation Facilities (CCF)

Given the large gaps in knowledge, this study will focus only on variabilities in *EC*, *DPW*, and *PC* and will hold other parameters constant. For example, to maximize growing conditions relative humidity, temperatures, CO_2 concentrations, and fertilizer usage are all optimized and vary widely by CCF. Further, this study did not consider other processes such as trimming, harvesting and drying buds which may also release BVOCs.

For this study, it was assumed that all CCFs operated in the same way at a temperature of 30ºC and 1000 μmol m^{-2} s^{-1} of photosynthetically active radiation (PAR). In addition, it was assumed that all emissions from the plants inside a CCF enter the atmosphere. Ventilation to the atmosphere varies widely by the operation, and there are no current regulations or industry-wide practices that are being used to mitigate emissions.

In total, seven scenarios of emission inventories were created to explore sensitivities in *EC*, *DPW*, and *PC* as shown in Table 1. In scenarios 1-3, the *PC* was held to a total of 1 million and a 750 g *DPW* was assumed. The *EC* of 10 μg gdw^{-1} h^{-1} as reported by Wang et al. (2018) was used in 1_EC, with a sensitivity that multiplied that rate by a factor of 5 (scenario 2_EC), and 10 (scenario 3_EC). The remaining scenarios in Table 1 kept the *EC* constant at 10 μg gdw^{-1} h^{-1}. Scenarios 4_DPW and 5_DPW explored the sensitivity of increasing *DPW*, and scenarios 6_PC and 7_PC increased the total plant count.

2.3 Model description and analysis tools

2.3.1 Model protocols and evaluation

The Comprehensive Air Quality Model with Extensions, CAMx6.10 (ENVIRON, 2013; ENVIRON and Alpine., 2017b), was used to predict ground-level ozone concentrations. The model and protocols used in this study are based on the Western Air Quality Modeling Study (WAQS) for 2011 (ENVIRON and Alpine., 2017b; Adelman et al., 2016). The WAQS 2011b baseline model simulation period runs from June 15th to September 15th, 2011, and is driven with meteorological data from WRF version 3.3 for the same time period and domain. The model was initialized using Three-State Air Quality Modeling Study standard boundary and initial conditions (ENVIRON and Alpine., 2017b). The model domain is a 2-way nested grid at 12 and 4 km grid cell resolutions (Fig. 1b). Anthropogenic emissions were derived from EPA National Emission Inventory (NEI) version 2011 NEIv2 with updates for point and area sources of oil and gas emissions in the western US. The biogenic emissions inventory was based on the Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGANv2.1) (Guenther et al., 2012). All data and supporting documentation are publicly

238 available via the Intermountain West Data Warehouse (IWDW) website (WAQS, 2017).

239 The revision 2 of Carbon Bond 6 (CB6r2) (Ruiz and Yarwood, 2013) chemistry
240 mechanism was used in all model runs. This groups all monoterpenes as a single compound
241 species, TERP. Thus, the total monoterpenes EC reported in Wang et al. (2018) was converted
242 into the TERP species. TERP undergoes oxidation reactions with the nitrate radical (NO_3), the
243 hydroxyl radical (OH), ozone (O_3), and singlet oxygen. It should be noted that the TERP
244 category includes a wide variety of monoterpenes whose reaction rate constants may differ
245 from TERP ($k_{298} = 6.77 \times 10^{-11}$ molecules cm^{-3} s^{-1}). For example, the rate constant of β-myrcene
246 with OH radical (Hites and Turner, 2009) is 3.35×10^{-10} molecules cm^{-3} s^{-1} (k_{298}), which is 4
247 time higher than TERP and 5.6 times faster than α-pinene (Carter, 2010).

248 The details of the WAQS model setup protocol (ENVIRON and Alpine., 2017b) and
249 model performance (Adelman et al., 2016) can be found in IWDW website. In summary, the
250 model performance evaluation concluded that this simulation had met all performance goals
251 for both maximum daily 1-hour (MDA1) and maximum daily 8-hour average (MDA8) ozone.
252 In the performance review report, it was found that the WAQS model had a positive bias for
253 ozone simulated in a 4 km × 4 km resolution domain, when compared with EPA Air Quality
254 System (AQS) surface monitors (MDA1: 0.8%, MDA8: 0.9%). On days when ozone
255 concentrations higher than 60 ppb were measured, the model had a negative bias of −6.2% for
256 MDA1 and −6.3% for MDA8. The model evaluation result also noted that the model
257 performance was best during the spring and summer months.

258 **2.3.2 Process Analysis**

259 CAMx runs used in this analysis had the process analysis (PA) option enabled
260 (ENVIRON, 2013). The CAMx configuration used here produces two additional files needed
261 for PA: the integrated reaction rate (IRR) and integrated process rate (IPR). These files include
262 the rates of change in concentration of every species due to chemistry and transport for every
263 grid cell and timestep. Python-based Process Analysis (pyPA) and the Python Environment for
264 Reaction Mechanisms/Mathematics (PERMM) (Henderson et al., 2010; Henderson et al.,
265 2011) were then applied to post-processing the CAMx PA output. PERMM was used to
266 aggregate the chemical and physical process rates for selected model grid cells and layers
267 allowing for tracking of plumes within the planetary boundary layer (PBL).

3. Results

3.1 Emissions Inventory

The seven scenarios were used to estimate a range of emissions of BVOCs from CCFs for the entire state of Colorado. As shown in Table 2, the base case (BC) scenario estimates 731,442 ton/year of all VOCs being emitted in Colorado, of which 47% are BVOCs. The BC scenario does not include any emissions from the cannabis industry. Table 2 also shows the seven scenarios that did include CCF emissions ranked in order of their increases in state-wide BVOC emissions. As expected the CCF BVOC emissions scaled linearly with each factor that was changed in Eq. (1). In scenario 3_EC, a 10-fold increase in the emission rate *(*100 μg gdw^{-1} h^{-1}) resulted in a 657 metric tons/year increase. Similarly, scenario 2_EC assumes 50 μg gdw^{-1} h^{-1} and produces 329 metric tons/year. Scenarios 4 and 5 showed the sensitivity of terpene emissions from CCFs to variation in *DPW* while holding *PC* constant and an *EC* of 10 μg gdw^{-1} h^{-1}. It was estimated that an additional 66 ton/year of emissions were produced when a 750 g *DPW* is assumed. This doubles to 131 metric tons/year with a *DPW* of 1500 g and reaches 219 metric tons/year with a *DPW* of 2500 g. Comparing scenario 1_EC with scenario 6 and 7 shows how the growth in *PC* will impact emissions of BVOCs. In Colorado, a doubling of the *PC* increases BVOC emissions by 131 metric tons/year in scenario 6_PC and 261 metric tons/year for the 4 million plants in scenario 7_PC. The largest increases in BVOC emissions were predicted in scenarios 3_EC and 2_EC showing that the total emission rate of BVOCs from CCFs were most sensitive to *EC*.

In March 2018, Denver County housed 41% of CCFs and 55% of all cannabis plants in Colorado (Hartman et al., 2018a). As a result, about 43% of state-wide CCF BVOC emissions occur there (Table 2). Current emission inventories of Denver County show negligible amounts of biogenic emissions accounting for only 0.1% of the total state-wide BVOC emissions. CCF emissions increased BVOC emission rates in Denver Country up to 136% in scenario 3_EC. This changes the total VOC emission rate in Denver County by up to 3.5%. Other cities in Colorado do not have as high a concentration of CCFs, and thus the relative increases were smaller as shown in Table 2.

The introduction of additional cannabis BVOC emissions into model simulations increased the predicted TERP concentrations. Figure 2 shows the maximum increase in TERP concentrations for three scenarios for Denver County over the entire 90-day simulation period. Regardless of the scenario, the largest increases in TERP occurred near the largest

concentrations of CCFs. The absolute maximum changes ranged from 0.5-5.0 ppb located at the Elyria Swansea and Globeville neighborhoods in north-central Denver. Increases in TERP were also predicted to the north due to the dominant wind flows in that direction throughout the simulation period. Figure S2 shows the maximum increase in TERP concentrations for the 1_EC, 5_DPW, and 3_EC scenarios in the 4 km × 4 km domain for the entire 90-day simulation period. As expected substantially lower increases in TERP concentrations were predicted for other cities in Colorado: 0.26 ppb in Colorado Springs and 0.24 ppb in Pueblo. Figure 3 shows the hourly changes in TERP concentrations across the entire 4 km × 4 km domain. The largest increases for all scenarios occurred at night with a peak of 5 ppb at 4:00 AM local standard time (LST). Given that the hourly emissions of terpenes from CCFs were assumed constant for 24 hours, these larger nighttime changes can be primarily ascribed to the lack of photochemistry and a shallow nocturnal PBL. These results suggest that the increases of TERP are highly correlated with locations of CCFs, accumulate at night, and have significant losses during the day.

3.2 Regional Ozone impacts

Predicted increases in hourly ozone concentrations in excess of 0.1 ppb only occurred when terpene emissions were in excess of 219 metric tons per year, with scenarios 4_DPW, 6_PC, and 1_EC having little impact on predicted ozone. Thus, this analysis will focus on two scenarios, 5_DPW, and 3_EC to explore potential regional ozone impacts in the present and future. Figure 4 shows the hourly changes in ozone concentrations across the entire 4 km × 4 km domain for these two scenarios. During the daytime, the increase in TERP emissions results in a peak ozone increase of 0.34 ppb at 9:00 AM LST for 3_EC with only minimal changes in 5_DPW. Figure 5 shows, for Denver County and the Front Range Metropolitan Area, the locations of the daytime (6:00 AM – 6:00 PM LST) maximum increases in hourly ozone concentrations for all 90 days when emissions were added for scenarios 5_DPW and 3_EC. Ozone increases for the entire 4 km × 4 km domain can be found in Fig. S3. The largest predicted ozone concentrations occurred in Denver County with impacts of 0.11 ppb in 5_DPW, and 0.34 ppb in 3_EC as shown in Fig. 5. Both scenarios show that daytime increases in ozone were limited to Denver County and just to the northwest, west, and southwest of Denver County.

There were also night time variations in ozone observed for the modeling domain. In scenario 5_DPW and 3_EC, nighttime increases were more than double the increases predicted

during the day. The largest changes in hourly ozone concentrations of 0.67 ppb occurred at 0:00 AM LST (i.e. midnight) for 3_EC. Figure 6 shows the location and magnitude of the maximum changes in hourly ozone concentrations during the night (6:00 PM – 6:00 AM LST) in 5_DPW and 3_EC. The extent of ozone increases at night are primarily to the north of Denver indicating a northern outflow. The maximum increase in hourly ozone for the whole of Colorado is shown in Fig. S3, with visibly little changes at night in other cities. These model results suggest that the additional emissions of TERP have immediate impacts on local ozone production chemistry during both the day and night, but little wider impact.

A critical metric for the attainment of the NAAQS ozone standard in Denver County is the maximum daily average 8-hour ozone concentration (MDA8). Figure 7 shows the maximum difference in MDA8 for each grid cell centered on Denver County, across the entire 90-day simulation period for the 5_DPW, and 3_EC scenarios. Maximum increases in MDA8 are 0.14 ppb for 3_EC (Fig. 7b) co-located with the maximum increases in TERP concentrations.

3.2.1 Ozone impact at night

The maximum hourly ozone increase of 0.67 ppb for the 3_EC scenario occurred on Thursday, July 28th, 2011, at 0:00 AM LST (i.e. midnight) near the largest concentration of CCFs (see Fig. 8). In subsequent hours the plume of ozone moved slowly to the east before being dispersed by the rise of the morning PBL at 6:00 AM LST.

To better understand why ozone increased at night, the PA model output was analyzed to quantify the chemical and physical processes producing ozone. Plume tracking was used so that only grid cells where the increase in ozone (i.e. the plume) occurred were included in our analysis, which ran from July 27th, 9:00 PM to July 28th, 6:00 AM LST. The number of vertical model layers included in the analysis also varied to incorporate the hourly evolution of the PBL. Figure S4 provides snapshots of the horizontal grid cells used and the vertical layers that were aggregated throughout the simulation time period. Fig. S5 shows the changes in final ozone concentrations (compared to the base case) for the grid cells and vertical layers included in the analysis, as well as the physical and chemical process rates that account for these changes. Figure S5 shows that the process most responsible for increases in ozone concentrations was chemical production.

For the chosen vertical layers and grid cells Table 3a shows the total rate of the oxidation reactions with TERP across the entire period. Throughout this time, the additional

TERP emissions lead to an increase in the number of oxidation reactions thereby generating more secondary VOC products and radical species. The chemical losses of TERP increased due to reactions with: OH (from 0.01 ppb to 0.1 ppb; +900%), nitrate radical (NO_3) (from 0.39 ppb to 1.58 ppb; +305%), and O_3 (from 0.04 ppb to 0.2 ppb; +400%). Further analysis confirms that night-time oxidation chemistry leading to changes in ozone concentration are driven by NO_3. In the 3_EC scenario, TERP emissions only increased the annual VOC emission in Denver County by 3.5%, but this is sufficient to increase the VOC + NO_3 reaction rates by 125%. These increases produce more peroxyl radicals (TRO_2=HO_2 + RO_2) driving further oxidation and further radical production. Table 3b also shows that the generation of OH radicals from reactions of TERP with O_3 increased by 267%. Ultimately, these increases in initial TERP reactions with NO_3 and O_3 increase the NO to NO_2 conversions via the TRO_2 pathway by 44%, reducing the availability of NO to react with O_3. Thus, the increased ozone concentration predicted at night is actually due to the 1 ppb (0.8%) reduction in the loss of ozone to reactions with NO rather than an increase in actual production of ozone (Table 3c). The increased TERP emissions also increase production of NO_x termination products (NO_z) by 27% with organic nitrate (NTR; representing ~71% of this NO_z product) increasing from 0.66 ppb to 1.6 ppb (+142%). This increase in NO_z production at night also results in lower NO concentrations and thus lower ozone titration.

3.2.2 Ozone impact during the day

The maximum daytime hourly ozone increase of 0.34 ppb occurred at 9:00 AM on Monday, July 18th, 2011, as shown in Fig. 9. On this day, the meteorological conditions favoured the maximum possible production of ozone. This day featured “upslope flows” that are a common meteorological condition linked to ozone exceedances periods (Pfister et al., 2017). We thus chose to focus on July 18th to understand the daytime changes in chemistry that occur from increased BVOC emissions. As expected, the location of predicted ozone increases coincides with the location of the strongest terpene emissions in the domain as shown in Fig. 9a. For the daytime hours of 6:00 AM – 2:00 PM LST, the PA option was used to quantify changes in chemical processes for the grid cells and model layers shown in Fig. S6. For these grid cells and layers, Fig. S7 shows the changes in final ozone concentrations compared to the base case and the physical and chemical process rates that impact those concentrations. Table S3 sums the key chemical processes for these hours. The increases in CCF emissions resulted in a 100% increase in OH reactions with TERP producing intermediate oxidation products and ultimately increasing OH production by 0.6%. As a result of this oxidation chemistry, there was an increase of 0.9% in NO to NO_2 conversion by TRO_2 pathway, ultimately leading to a

0.1% increase in ozone production.

3.2.3 Ozone impact sensitivity

The maximum modelled daytime hourly ozone increase due to additional CCF emissions occurred on July 18th. Using this day multiple sensitivity simulations were performed, where CCF emissions from Denver County were incrementally increased up to 3,800 ton/year. Figure 10 shows the increase in terpene emissions from Denver County versus the largest daily increase in hourly ozone concentrations. Figure 10a shows a linear relationship, indicative of a VOC limited environment, where hourly ozone concentrations are predicted to increase by 1 ppb for every 1,000 ton/year increase in TERP emissions during the day, and 0.85 ppb at night. Also shown is the sensitivity to the MDA8 ozone where there is a 0.30 ppb increase for every 1,000 ton/year of TERP emissions. According to projected emission inventories provided by the state of Colorado, the ozone non-attainment area was expected to see reductions of 26.4% of NO_x and 24.6% of VOC emissions by the year 2017 (ENVIRON and Alpine., 2017a). Under these reduced anthropogenic emission scenarios, Fig. 10b shows how ozone would then respond to additional CCF TERP emissions. Figure 10b continues to show a linear relationship, where hourly ozone concentrations are predicted to increase by 1.5 ppb for every 1,000 ton/year increase in TERP emissions during the day, and 1.8 ppb at night. In the future case, the MDA8 ozone increases by 0.38 ppb increase for every 1,000 ton/year of TERP emissions. Therefore, Denver will still be VOC-limited and ozone is predicted to more sensitive to CCF emissions of terpenes.

4. Conclusion

This study provides the first VOC emission inventory to be compiled for the cannabis industry in Colorado, the first time such analysis has been conducted anywhere in the USA. Given the current state of knowledge of emission rates and growing practices, there are considerable uncertainties in the basic parameters required to build such an inventory. Using realistic bounds on each parameter, we developed seven scenarios, which resulted in estimated emission rates that ranged over an order of magnitude. The highest emissions occur in Denver County, with rates ranging between 36-362 metric tons/year for the different scenarios, from a total of 66-652 metric tons/year across Colorado as a whole.

We included these additional terpene emissions in the Comprehensive Air Quality Model with Extensions (CAMx), the model used by the state of Colorado for regulatory monitoring and projections. Taking the worst case (3_EC) and median scenario (5_DPW) we consider representative of current uncertainty upper boundary and future industry expansion; we find that these projected increases in emissions lead to maximum increases in terpene concentrations of up to 5.0 ppb. The largest impacts were seen in locations with the highest terpene emissions coming from CCFs, i.e. in Denver County. We further found that these increases in terpene concentrations affected the local atmospheric chemistry and air quality with ground-level ozone concentrations increasing by as much as 0.34 ppb during the day and 0.67 ppb at night. In general, simulated nighttime increases were higher than those during the daytime were, and we take the nighttime of July $27^{th} – 28^{th}$ as a case study to further investigate. By applying process analysis (PA), following the evolving plume of VOCs and ozone, we find that the initial reactions of the additional terpenes with OH, NO_3 and ozone result in increased formation of peroxyl radicals which increases the NO to NO_2 conversion rate; also removes the NO_x to generate more NO_z product. This effectively reduces the loss of ozone by reaction with NO, increasing the total ozone concentration.

We acknowledge, however, the considerable uncertainties that surround our projections and call for the need for continued efforts to reduce these such that a more accurate assessment of the regional air quality implications of this industry can be made. Future studies that include ambient BVOC measurements are critical for comparisons with model predictions. Additionally, in the model chemical mechanism more accurate and mechanistic representation of terpene species is needed that can reflect the current cannabis emission composition. Currently, the model surrogate “TERP”, which represents all monoterpene species in the

mechanisms, may not represent the precise rate constant for BVOC emissions from cannabis. Further data are needed to reduce uncertainties in emission inventory estimates specifically those regarding CCF-specific information on plant counts, and weight by cultivar and growth stage, coupled with information about the agronomical practices of Cannabis cultivation in CCFs. Additional measurements of emission capacities of different cannabis strains at different growth stages are also needed. Further, the emission inventory version is for the year 2011; it may not be suitable to estimate the ozone impacts by the CCF industry.

We chose to focus on ozone, since Denver is a moderate non-attainment area with an ozone State Implementation Plan (SIP) (ENVIRON and Alpine., 2017b, a; Colorado, 2018) in accordance with the EPA regulations. But assessments of the impact of these additional terpene emissions on particulate matter ($PM_{2.5}$) is warranted given the high secondary organic aerosol (SOA) yields of terpenes from 0.3 to 0.8 (Iinuma et al., 2009; Lee et al., 2006; Fry et al., 2014; Slade et al., 2017). It should also be borne in mind that investigations of indoor air quality are needed given the findings of Martyny et al. (2013) and Southwellb et al. (2017) that indoor terpene concentrations reached 50-100 ppb in growth rooms and 30-1,600 ppb in flowering room, likely initiating intense photochemistry under the powerful grow lamps in use in CCFs.

Code availability:

1. The source code of the CAMx6.10 model can be downloaded on the Environ website: http://www.camx.com
2. The process analysis tools and source codes including PseudoNetCDF, pyPA, and PERMM, can be downloaded on GitHub: https://github.com/barronh/pseudonetcdf, https://github.com/barronh/pypa, and https://github.com/barronh/permm
3. Python 2.7 is used to treat the model output and can be downloaded on anaconda python website: https://www.anaconda.com/distribution/

Data availability:

The air quality model input data and output data (~2.3TB) of WAQS2011b episode for Colorado can be downloaded on IWDW website: https://views.cira.colostate.edu/iwdw/

Author contribution

Chi-Tsan Wang and Dr. William Vizuete are lead researchers in this study responsible for research design, experiments, analyzing results and writing the manuscript. Dr. Christine Wiedinmyer and Dr. Kirsti Ashworth are also co-head researchers, and guided the research design, assessed model results, and contributed to writing the manuscript. Dr. John Ortega, and Dr. Peter Harley helped in collecting data and writing the manuscript. Dr. Quazi Z. Rasool helped to analyze model results and contributed in writing the manuscript.

Competing interests

The Authors declare that they have no conflict of interest.

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694

695 **Tables**

696 **Table 1.** Simulation scenarios and assumed values for emission capacity (*EC*) rate, dry plant
697 weight (*DPW*), and the plant count (*PC*) for Colorado and Denver County. The base case (BC)
698 scenario has no cannabis emissions.

Name	*EC*	*DPW*	*PC*	
	(ug gdw^{-1} hr^{-1})	(gdw $plant^{-1}$)	Colorado	Denver County
BC	0	0	0	0
1_EC	10	750	1.0×10^6	5.5×10^5
2_EC	50	750	1.0×10^6	5.5×10^5
3_EC	100	750	1.0×10^6	5.5×10^5
4_DPW	10	1,500	1.0×10^6	5.5×10^5
5_DPW	10	2,500	1.0×10^6	5.5×10^5
6_PC	10	750	2.0×10^6	1.1×10^6
7_PC	10	750	4.0×10^6	2.2×10^6

699

Table 2. The estimated BVOC and total VOC emission rates (metric tons/year) for the base case (BC) scenario. Also shown are the increases in VOC emissions for all scenarios shown in Table 1 for Colorado, Denver County, Colorado Springs, Pueblo, and Boulder. The numbers in parenthesis are the percentage increases compared with the BC scenario.

Name	Colorado		Denver County		Colorado Springs		Pueblo		Boulder	
	BVOC	Total VOC	BVOC	Total VOC	BVOC	Total VOC	BVOC	Total VOC	BVOC	Total VOC
BC	340,268	731,442	265	10,465	5,184	15,143	5,870	9,184	3,677	9,820
3_EC	657 (+0.19%)	+0.09%	362 (+136%)	+3.5%	60 (+1.20%)	+0.40%	53 (+0.90%)	+0.58%	26 (+0.70%)	+0.26%
2_EC	329 (+0.10%)	+0.04%	181 (+68%)	+1.7%	30 (+0.58%)	+0.20%	27 (+0.45%)	+0.29%	13 (+0.35%)	+0.13%
7_PC	261 (+0.08%)	+0.04%	116 (+44%)	+1.1%	42 (+0.80%)	+0.27%	22 (+0.38%)	+0.24%	12 (+0.33%)	+0.12%
5_DPW	219 (+0.06%)	+0.03%	121 (+45%)	+1.2%	20 (+0.39%)	+0.13%	18 (+0.30%)	+0.19%	9 (+0.23%)	+0.09%
4_DPW	131 (+0.04%)	+0.02%	72 (+27%)	+0.69%	12 (+0.23%)	+0.08%	11 (+0.18%)	+0.12%	5 (+0.14%)	+0.05%
6_PC	131 (+0.04%)	+0.02%	72 (+27%)	+0.69%	12 (+0.23%)	+0.08%	11 (+0.18%)	+0.12%	5 (+0.14%)	+0.05%
1_EC	66 (+0.02%)	+0.01%	36 (+14%)	+0.35%	6 (+0.12%)	+0.04%	5 (+0.09%)	+0.06%	3 (+0.07%)	+0.03%

Table 3. All data summed from July 27th, 9:00 PM LST to July 28th, 5:00 AM LST for grid cells and layers shown in Fig. S4. The base case (BC) scenario column shows the absolute predicted values and, the subsequent columns show the predicted changes due to emissions from the 3_EC scenario. Percentages in parenthesis are the changes in 3_EC relative to BC. Shown are the **(a)** total amount of VOC and TERP consumed due to oxidation (ppb), the **(b)** total amount of hydroxyl radical (OH) and total peroxyl radicals (TRO_2) that were generated and their sources (ppb), and the **(c)** total amount of Nitrogen Dioxide (NO_2) and NOx termination products (NO_z) produced and their sources (ppb).

(a)

	BC	3_EC
VOC + OH	1.36	1.68 (+23.5%)
TERP + OH	0.01	0.10 (+900%)
VOC + NO_3	0.91	2.05 (+125%)
TERP + NO_3	0.39	1.58 (+305%)
VOC + O_3	1.80	1.97 (+9.40%)
TERP + O_3	0.04	0.20 (+400%)

(b)

	BC	3_EC
OH generation (from VOC + O_3)	1.00	1.10 (+10.0%)
from TERP + O_3	0.03	0.11 (+267%)
TRO_2 generation	34.2	42.8 (+25.1%)
from VOC initial reactions	3.25	5.03 (+54.8%)
from TERP initial reactions	0.47	1.98 (+321%)

(c)

	BC	3_EC
NO to NO_2	198	197 (-0.70%)
NO + O_3	158	157 (-0.80%)
NO + TRO_2	3.50	5.04 (+44.0%)
NO_z generation	4.91	6.24 (+27.1%)
NTR generation	0.66	1.60 (+142%)
PAN generation	1.54	1.56 (+1.30%)
PANX generation	0.54	0.66 (+22.2%)
HNO_3 generation	2.17	2.42 (+11.5%)

Figures

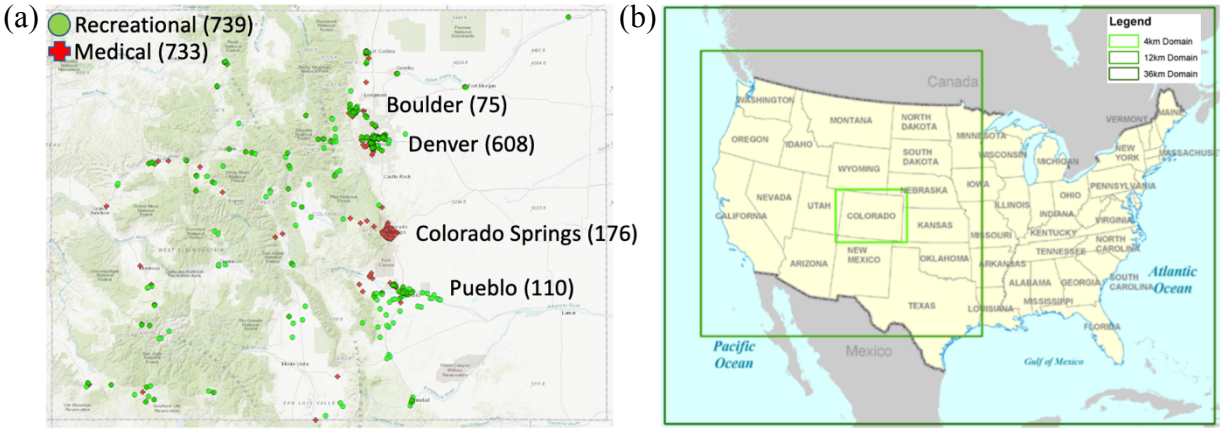
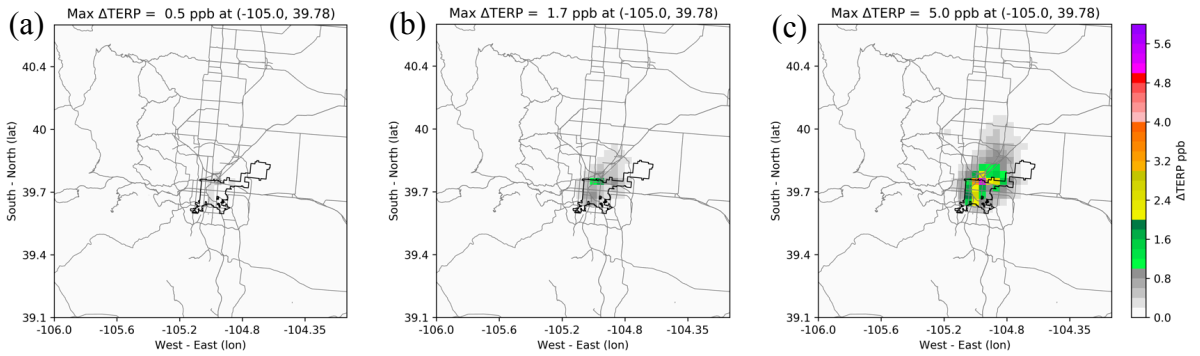


Figure 1. (a) The locations of medical (red) and retail (green) Cannabis cultivation facilities (CCFs) in Colorado as of March 1, 2018. The corresponding values are the number of CCFs found within each city. **(b)** The 36km × 36km resolution of Western Air Quality Model Study (WAQS) and nested inner 12km × 12km resolution domains and 4km × 4km resolution domain used by the Comprehensive Air Quality Model with Extensions (CAMx) (ENVIRON and Alpine., 2017b).



728

729 **Figure 2.** The maximum increase in TERP concentrations (ppb) for Denver County and Front
730 Range over the entire 90-day simulation for the **(a)** 1_EC, **(b)** 5_DPW, and **(c)** 3_EC scenarios.
731 The black outlines Denver County and the grey lines are state and interstate highways.

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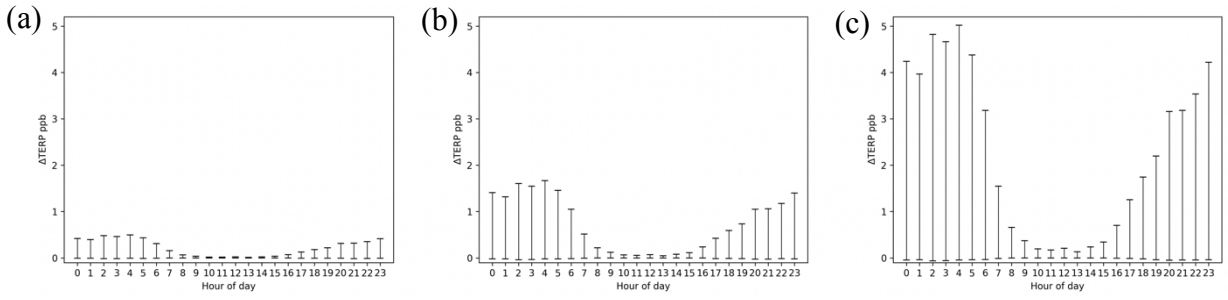

733

734 **Figure 3.** The hourly changes in TERP concentrations across the entire 4 km × 4 km domain,
735 over the 90 days simulation for the **(a)** 1_EC, **(b)** 5_DPW and **(c)** 3_EC scenarios.

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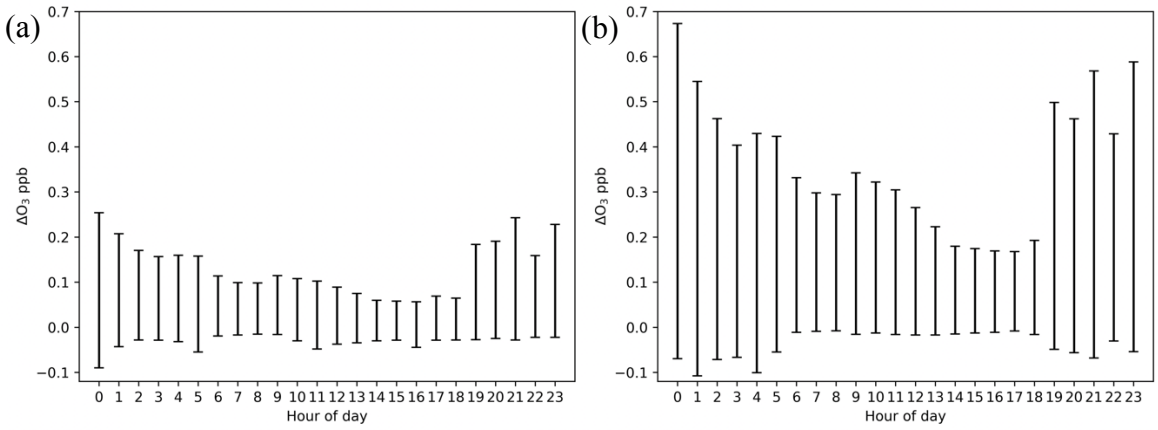

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738 **Figure 4.** The predicted differences in hourly ozone concentrations (ppb) across the entire
739 Colorado domain, over the 90 days simulation for the **(a)** 5_DPW and **(b)** 3_EC scenarios.

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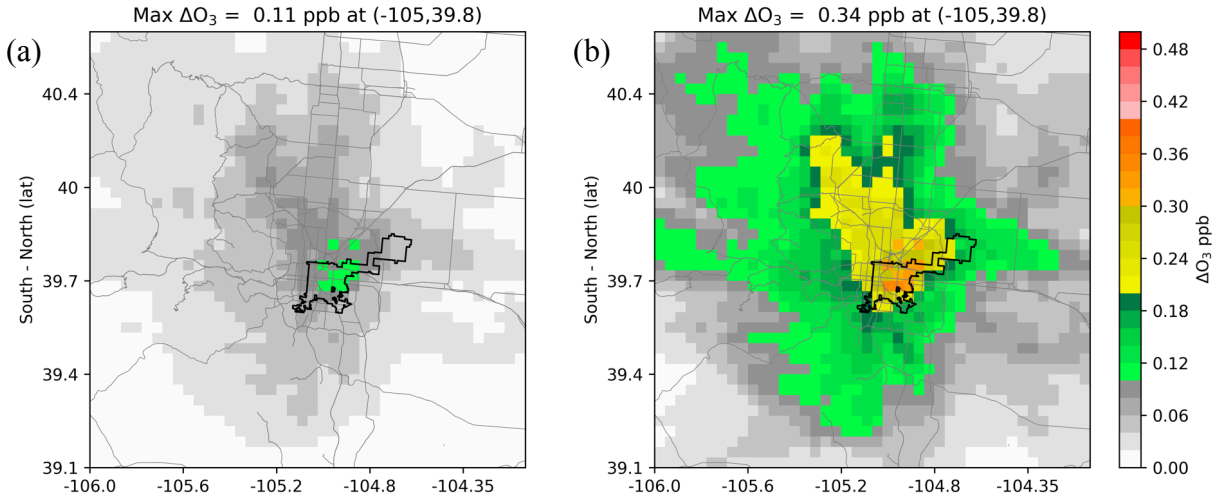
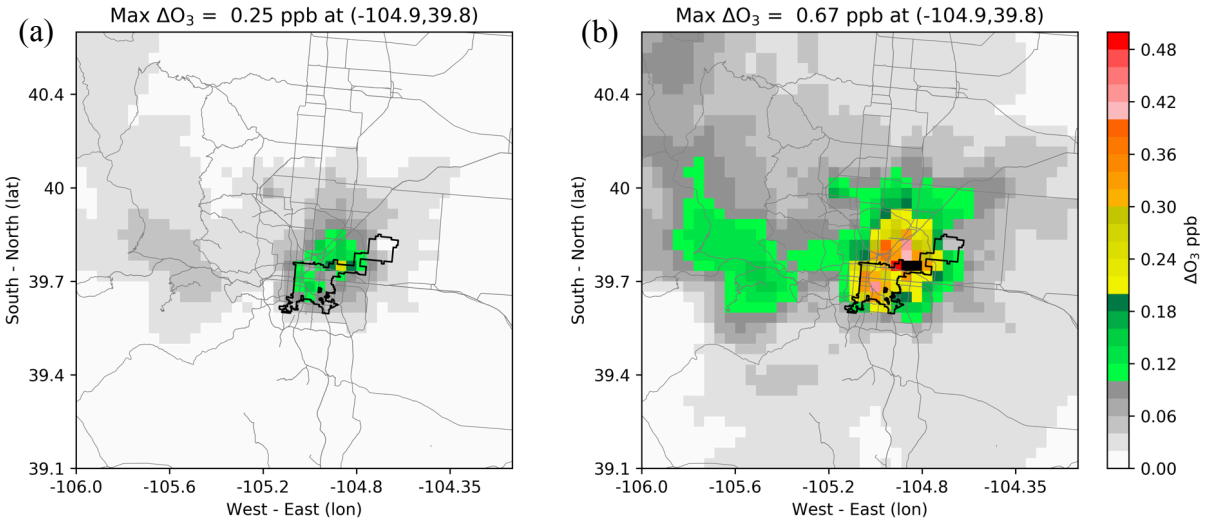


741

742 **Figure 5.** The predicted changes in hourly ozone concentrations for the Denver region from 6
743 AM – 6 PM LST for all 90 days of the simulation for the **(a)** 5_DPW and **(b)** 3_EC scenarios.
744 The grey lines indicate major highways and the black line outlines Denver County**.**

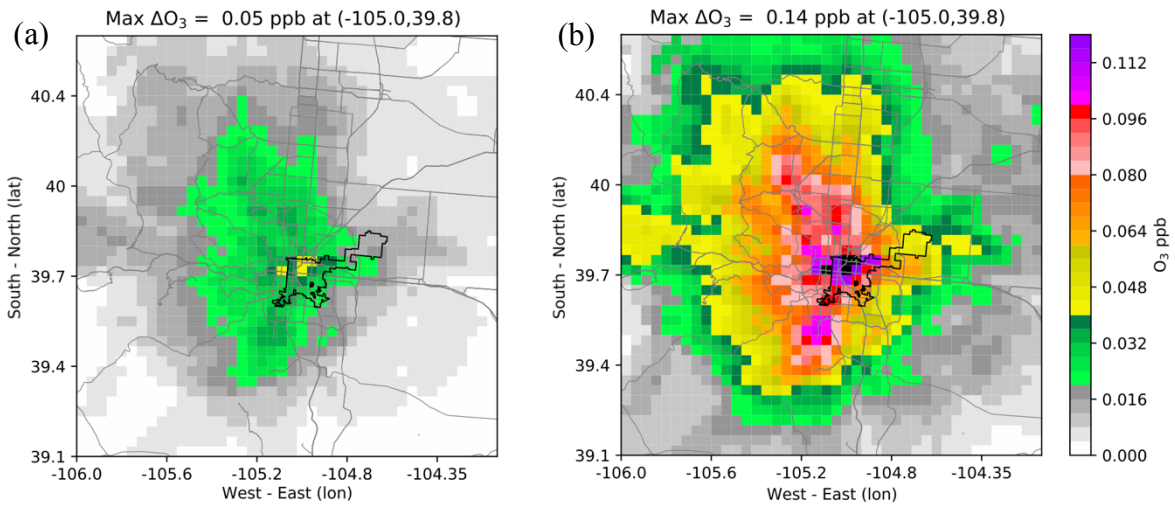
745



746

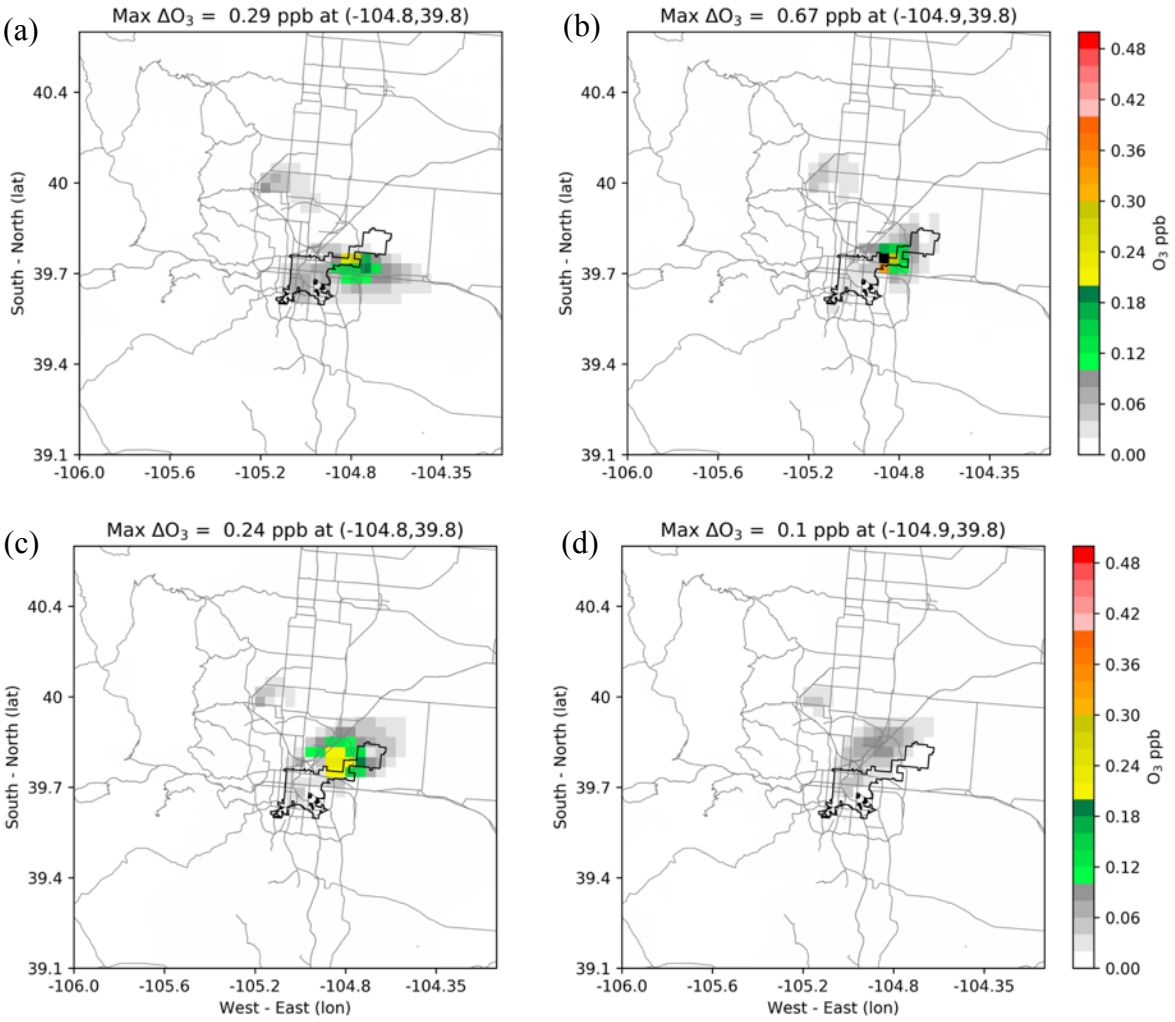
747 **Figure 6.** The predicted changes in hourly ozone concentrations for the Denver region from 6
748 PM – 6 AM LST for all 90 days of the simulation for the **(a)** 5_DPW and **(b)** 3_EC scenarios.
749 Black regions within the map indicate ozone increase values greater than 0.5 ppb. The grey
750 lines indicate major highways and the black line outlines Denver County**.**

751



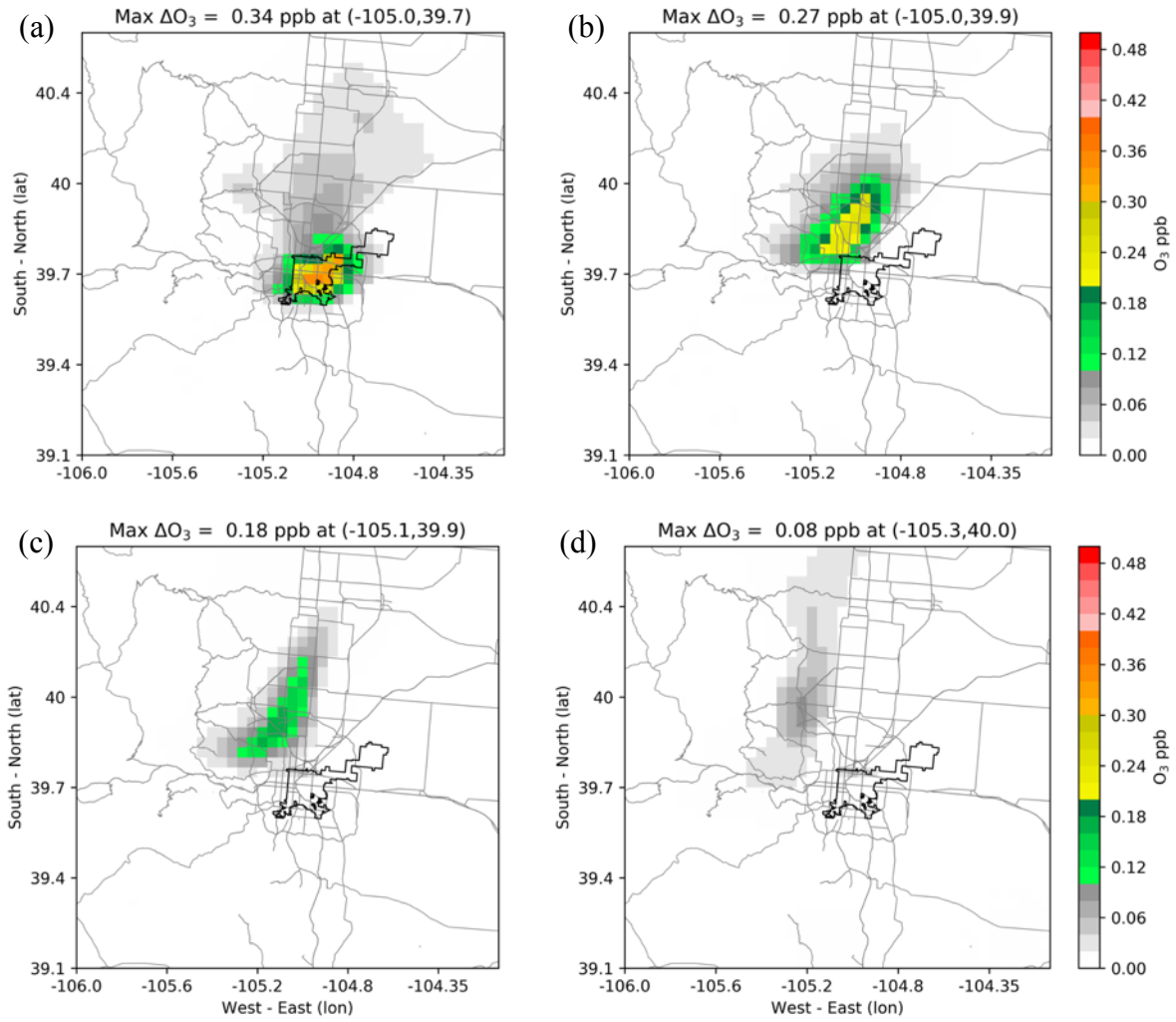
752

753 **Figure 7.** The predicted maximum increases in the maximum daily average 8-hour (MDA8)
754 ozone concentration (ppb) for the **(a)** 5_DPW and **(b)** 3_EC scenarios for the Denver region
755 over the 90-day simulation period. The black indicates ozone increase values greater than
756 0.12 ppb.



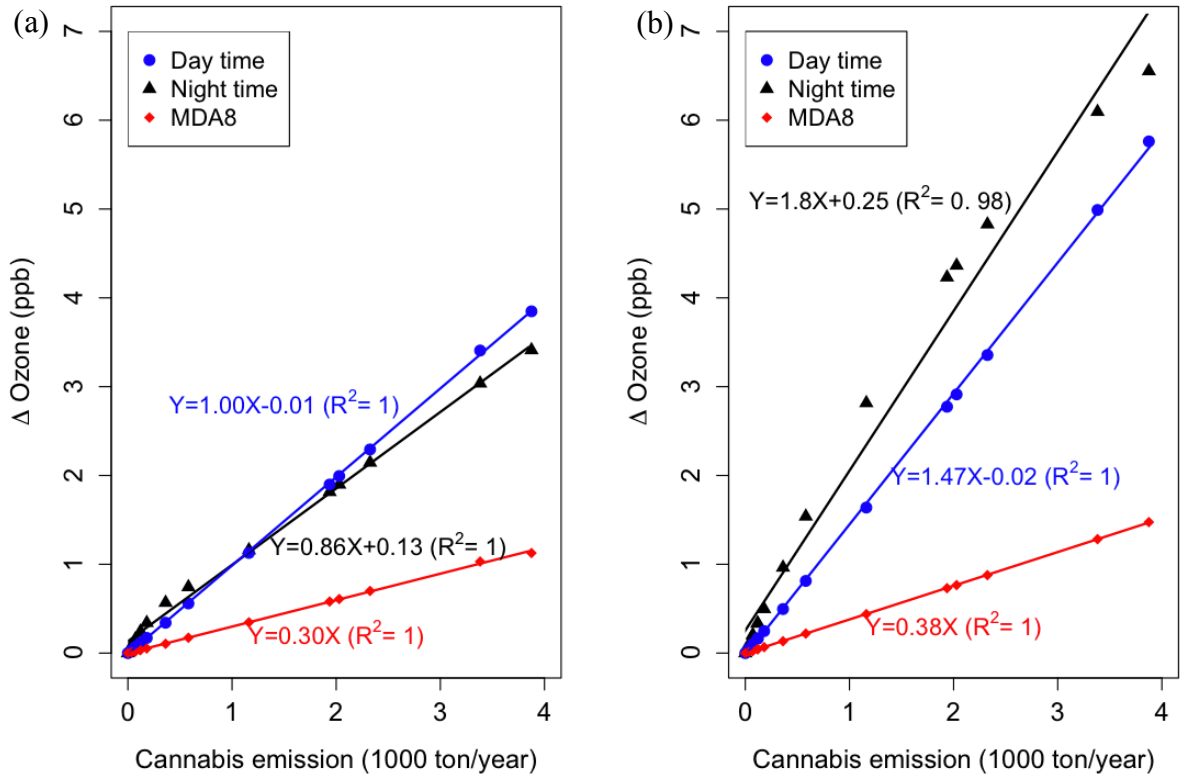
757

758 **Figure 8.** For the 3_EC scenario on July 28th, 2011, the largest hourly predicted ground level
759 ozone increases at **(a)** July 27th, 9 PM LST, and for July 28th, at **(b)** 0 AM LST (i.e. midnight),
760 **(c)** 3 AM LST and **(d)** 6 AM LST.



761

762 **Figure 9.** For the 3_EC scenario on July 18th, 2011 the largest hourly predicted ground level
763 ozone increases at **(a)** 9 AM LST, **(b)** 12 PM LST (i.e. noon), **(c)** 2 PM LST, and **(d)** 5 PM
764 LST. The maximum of 0.34 ppb occurred at 9 AM LST.



765

766 **Figure 10.** For July 18th during **(a)** 2011 and **(b)** 2017 the predicted maximum increase in
767 hourly ozone concentrations during daytime hours (6 AM – 6 PM LST) in blue, and nighttime
768 hours (6 PM– 6 AM LST) in black versus additional terpene emissions in Denver County. Also
769 shown is the response in maximum daily average 8-hour ozone concentration (MDA8) in red.

770