Editor comment:

Authors appreciate the editor's comments below that improved the quality of the manuscript a lot. In response to each of the comments, a related line number at which each comment is made in the previous manuscript is first shown in black and then our responses are shown in blue. Line numbers in our responses are based on the revised manuscript.

In addition to revision based on the comments here, we also identified words, sentences and paragraphs that we believed required revision, and revised them to make the manuscript concise.

29: Revised also following a reviewer's comment as follows:

(LL27-29 on p2)

Through a modeling framework, this study investigates how a pyrocumulonimbus (pyroCb) event influences water vapor concentrations and cirrus cloud properties near the tropopause, specifically focusing on how fire-produced aerosols affect this role.

67: Revised as follows also following a reviewer's comment:

(LL67 on p3)

studies have been conducted to improve this understanding

88: Corrected also following a reviewer's comment as follows:

(LL84-86 on p3)

More droplets mean more competition among them for available water vapor needed for their condensational growth, decreasing the size of individual droplets (Twomey, 1977; Albrecht, 1989).

101: Revised as follows:

(LL94-96 on p4)

The role of fire-generated aerosols in the development of pyroCbs and their effects on water vapor and cirrus clouds in the UTLS lacks a firm scientific understanding, and hence this paper focuses on that role of those aerosols.

107: Corrected.

127: The corresponding sentence is considered redundant and removed.

137: The last sentence is removed. The corresponding paragraph is shortened.

147: Corrected.

157: The corresponding sentence is revised as follows:

(LL131-132 on p5)

This study examines this dependence, not studied by Kablick et al. (2018).

194: Corresponding sentence is removed.

215: Corrected also following a reviewer's comment as follows:

(LL182-184 on p7)

These surface heat-flux values follow previous studies which adopt boreal forest emissions (Trentmann et al., 2006; Luderer et al., 2006).

242: Corrected.

244: Corrected.

249: Sentences are contracted as follows:

(LL211-213 on p8)

The control run adopts the unimodal lognormal distribution as an initial aerosol size distribution, a reasonable assumption in fire sites (Reid et al., 2005; Knobelspiessel et al., 2011; Lee et al., 2014).

255: The unit of standard deviation is μm and this is indicated in text.

274: The corresponding paragraph is made more concise.

293: The corresponding sentence is revised as follows:

(LL246-249 on p9)

The average cloud-top and cloud-base heights over the life span of the pyroCb are 10.3 km and 3.6 km in the control run, respectively, and these simulated heights are ~7% different from the satellite-retrieved values.

309: The repetitive sentence is removed.

378: **Sentences of corresponding text are removed, since they contain the same information as in Introduction.**

384: **Done.**

396: **Done.**

418: **Sentence is revised as follows:**

(LL349-350 on p12)

This implies that air parcels and associated updrafts in the pyroCb are stronger, reaching higher altitudes.

436: **The corresponding sentence is considered redundant and removed.**

444: **Corrected.**

465: **Wang's work is mentioned as follows:**

(LL377-383 on p13)

In summary, the pyroCb and associated updrafts cause a substantial enhancement of the transport of water vapor to the UTLS at and above the tropopause. Wang (2019) also reported this enhancement. Using modeling work and satellite observation, Wang (2019) indicated that the upward transport of water vapor in deep convective storms was possibly a major pathway through which water substance entered the stratosphere. Wang (2019) showed that the upward transport of water vapor was aided by gravity wave and its breaking in overshooting convective parcels.

469: **We removed an unnecessary sentence and clarified text.**

493: **The sentence pointed out is considered redundant and thus removed.**

520: **CDNC is replaced with "N_d"**

532: **The corresponding sentence is revised as follows:**

(LL443-445 on p15)

Increasing N_d enhances competition among droplets for a given amount of water vapor. Enhanced competition eventually curbs the condensational growth and reduces droplet size (R_v).

537: **The repetitive text is removed.**

540: R_v is defined in the title of Section 4.2.2 (LL431 on p15), N_d is defined in the title of sub-section "a" in Section 4.2.2 (LL433 on p15), and LWC is defined at line number 244 on p9.

We thought that the editor may want us to define or explain the proportionality itself between R_v and (LWC/N_d)^(1/3). To explain the proportionality, Equation (2) and associated text are added.

550: (LWC/N_d)^(1/3), but not R_v, corresponds to numbers shown in text after (LWC/N_d)^(1/3). Since R_v is linearly proportional to (LWC/N_d)^(1/3) according to Equation (2), the larger variation of (LWC/N_d)^(1/3) leads to the larger variation of R_v.

558: The already defined expression is removed.

559: Corrected.

565, 566: The corresponding sentence is revised as follows:

(LL473-478 on p16)

During period 1, as fire intensity weakens and updraft speed decreases, parcel equilibrium supersaturation decreases and thus, the minimum size of activated aerosol particles increases not only among the clean-scenario runs but also among the polluted-scenario runs. When the production of parcel supersaturation by updrafts and the consumption of supersaturation by droplets balance out, parcel supersaturation reaches parcel equilibrium supersaturation (Rogers and Yau, 1991).

579: Khain's HUCM model calculates equilibrium supersaturation at every model time step by considering dynamic effects on supersaturation (e.g., advection and updrafts) and microphysical effects on supersaturation (e.g., condensation and evaporation). For the consideration of dynamic effects and microphysical effects, dynamic and microphysical sub-time steps are considered for each model time step. After a model time step is completed, both of dynamic and microphysical impacts on supersaturation are completed. So, we obtain equilibrium supersaturation when each model time step is completed, and average it over those time steps and areas with positive updraft speed.

583: Corresponding sentences are contracted by removing unnecessary words.

596: (LWC/CDNC)^(1/3) is removed.

606: The corresponding paragraph is revised substantially to make it more concise as follows:

(LL502-505 on p17)

In association with larger aerosol concentration and the assumed aerosol size distribution, a smaller percentage variation of the number of activated aerosols and Nd with fire intensity is simulated in the polluted-scenario runs than in the clean-scenario runs. This smaller variation of Nd aids the greater reduction in Rv among the polluted-scenario runs.

616: The corresponding repetitive sentence is removed.

636: Throughout the manuscript, when what I am comparing is established in a sentence, the following sentences are simplified by shortening parts related to the comparison in each paragraph. For example, look at following sentences in Section 4.2.3:

To satisfy mass conservation, the freezing- and deposition-enhanced updrafts above the freezing level induce more updraft mass fluxes below the freezing level in the polluted-scenario run than in the clean-scenario run for each fire intensity. This leads to more convergence around and below cloud base in the polluted-scenario run than in the clean-scenario run for each fire intensity.

In the first sentence, the comparison between the polluted-scenario run and the clean-scenario run for each fire intensity is established, so, in the second sentence, "than in the clean-scenario run for each fire intensity" is removed.

640: Following a reviewer's comment, "get greater" is replaced with "increase" and the corresponding sentence is revised as follows:

(LL519-521 on p18)

The increasing differences in autoconversion rates between the polluted-scenario and clean-scenario runs increase those differences in the amount of cloud liquid available for freezing with weakening fire intensity (Figure 9a).

642: Following a reviewer's comment, "gets greater" is replaced with "increase" and the corresponding sentence is revised as follows:

(LL521-523 on p18)

Thus, differences in the average rate of cloud-liquid freezing and freezing-related latent heat over the period 2 between the runs increase with weakening fire intensity (Figure 9a).

644: Corresponding sentence is removed, following a reviewer's comment. However, following the comment here, in other parts of text, we removed the redundant statements of period.

648: Following the editor's and a reviewer's comments here, the repetitive text is removed.

650: Following the editor's and a reviewer's comments here, the repetitive text is removed.

654: We removed unnecessary numbers. We believe that even without a table for those numbers, what we intend to deliver can be delivered well. So, we do not add the table.

674: Revised as follows:

The corresponding sentences are substantially revised to make it more concise as follows:

(LL533-536 on p18)

The greater freezing and thus freezing-related latent heat eventually cause updrafts to be stronger in the polluted-scenario run starting at ~21:00 GMT (Figure 10). Then, the stronger updrafts induce deposition to be greater in the polluted-scenario run around 21:10 GMT (Figure 10).

682: Redundant expressions related to comparisons are removed throughout the manuscript.

686: Removed.

688: Also following a reviewer's comment, the text is revised as follows:

(LL544-545 on p19)

The higher mass fluxes and convergence below the freezing level

693: Redundant expressions related to comparisons are removed throughout the manuscript.

703: Revised as follows:

(LL554-557 on p19)

those differences in the average freezing-affected updrafts and subsequently in deposition-related latent heat over period 3 increase with weakening fire intensity (Figures 9a, 9b and 10).

769: Removed.

781: Corrected.

801: Redundant expressions (i.e., words in parentheses) are removed.

806: Revised as follows:

(LL642-645 on p22)

Much greater N_d is in the polluted-scenario run than in the clean-scenario run for each fire intensity. N_d decreases are smaller among the polluted-scenario runs than among the clean-scenario runs with weakening fire intensity. This situation during the initial stage induces R_v to decrease much more among the polluted-scenario runs with weakening fire intensity.

834: Revised.

836: Done.

837: We find Koren et al. (2008) at Ilan Koren's homopage (https://www.weizmann.ac.il/EPS/Koren/publications) as a paper for invigoration. The citation is as follows:

Koren I., Martins J. V., Remer L. A. & Afargan H. (2008). Smoke invigoration versus inhibition of clouds over the Amazon. Science. 321:(5891)946-949.

We add Koren et al. (2008) in the manuscript.

850: Revised.

861: Corrected.

1100: "which is" is removed.

1124: Revised as follows:

(LL957-959 on p32)

Figure 9. Average rates of condensation, deposition and cloud-liquid freezing at all altitudes in cloudy areas and over periods (a) 2, (b) 3 and (c) 4. In panel (a), average autoconversion rates are also shown.

Referee Report 1

Lee et al., Examination of effects of aerosol on a pyroCb and their dependence on fire intensity and aerosol perturbation using a cloud-system resolving model, submitted to Atmos. Chem. Phys., 2019

General comments

In response to comments and requested from both reviewers, the authors shortened the paper and improved the structure. After making the following minor language revisions, many of which will further decrease the length of the text, I recommend the manuscript for publication in ACP. Even after implementing these changes, I strongly recommend that the manuscript is proofread for correct English before publication. Thanks!

Authors appreciate the reviewer's comments below that improved the quality of the manuscript a lot. We revised the manuscript based on the comments and the manuscript was proofread. In addition, we identified words, sentences and paragraphs that we believed required revision, and revised them to make the manuscript concise with the better structure.

In the following, each comment by the reviewer (black) is followed by our response (blue).

Specific comments

299 Please provide examples in the text of "good agreement"

In the corresponding Section 3.1, we compared the simulation to the observation in terms of cirrus cloud, cloud macro-physical (e.g., LWP, IWP, cloud-top and cloud-base heights), and reflectively fields. We believe that these fields are examples of good agreement between the simulation and the observation. These examples are described in Section 3.1 and the last sentence in Section 3.1 is as follows:

(LL258-260 on p9)

These favorable comparisons between the observed and simulated cirrus clouds, cloud macro-physical and reflectivity fields demonstrate that the pyroCb-case simulation reasonably reproduces the event.

We believe that "examples" mentioned by the reviewer here may also mean the description of quantified differences and similarity in the reflectivity field between the simulation and the observation. Hence, the corresponding text is revised as follows:

(LL252-258 on p9)

Compared are the control-run and observed reflectivity fields. Kablick et al. (2018) provide details about the reflectivity field observed by CloudSat. Observations and the control run both show increasing reflectivity up to ~7 km, decreasing reflectivity between ~7 km and ~11 km and insignificant changes in reflectivity above ~11 km in altitude (Figure 3). The average reflectivity over altitudes along the Cloudsat path in the control run is -8.1 dBZe, ~15% different from the observed value. Hence, the reflectivity field is simulated fairly well compared to the observation.

29-30 “specifically focusing on how fire-produced aerosols affect this role via a modeling framework” -> “specifically focusing via a modeling framework on how fire-produced aerosols affect this role”

This corresponding sentence is revised as follows:

(LL27-29 on p2)

Through a modeling framework, this study investigates how a pyrocumulonimbus (pyroCb) event influences water vapor concentrations and cirrus cloud properties near the tropopause, specifically focusing on how fire-produced aerosols affect this role.

67 “studies to improve this understanding has been going on” -> “studies have been conducted to improve this understanding”

Done.

80 Insert "of pyroCB" after “microphysical properties”

Done.

84 “make” -> “decrease”

Done.

85 remove “smaller”

Done.

86-87 remove “or increasing aerosol”

Done.

88-89 replace “this more competition makes” with “decreasing the size of”

Done.

89 remove “smaller”

Done.

90 change “sizes of droplets” to “droplet sizes”

Done.

91 “autoconversion that is” -> “autoconversion,”

Done.

92 “for them to” -> “in which they”, “be” -> “become”

Done.

94-95 “More cloud liquid is thus available for transport to places above the freezing level by updrafts.” -> “More cloud liquid is thus available for transport by updrafts to altitudes above the freezing level.”

Done.

101 “by” -> “of”

Done.

108 “cloud typical properties” -> “cloud properties”

Done.

110 “For the simplicity of the term” -> “For simplicity”

It is replaced with “For simplicity herein” (LL102 on p4)

113 “have shown” -> “showed”, “updrafts

throughout the manuscript, ex. 115: “the pyroCb development and its impacts on the UTLS” -> “pyroCb development and its impacts on the UTLS”

“have shown” is replaced with “demonstrated”

“the pyroCb development and its impacts on the UTLS” is replaced with “pyroCb development and its impacts on the UTLS” throughout the manuscript.

Regarding updrafts the following is added:

(LL102-104 on p4)

For simplicity herein, an “updraft” refers to the general upward motion of convective air, or to the actual updraft speed representing the updraft or convective intensity, depending on the context.

Based on the added sentence above, we generally use “updrafts” in places where the meaning of “updrafts”, which can be the upward motion or the updraft speed or both, is rather obvious to readers based on the context. However, in a few sentences where conventional rules indicate that the use of “updraft speeds” is better than that of “updrafts”, we write “updraft speeds” explicitly.

An example of the sentences is as follows:

(LL478-482 on p16-17)

Mostly due to greater aerosol concentrations, associated average equilibrium supersaturation and minimum size of activated aerosol particles over areas with positive updraft speeds and period 1 are lower and larger, respectively, in the polluted-scenario run than in the clean-scenario run for each fire intensity (Rogers and Yau, 1991).

In this example sentence, simply because the word “positive” is generally coupled with “updraft speed” not but with “updrafts”, we just use this customary expression “positive updraft speed”

125 “impacts” -> “impacts on”

The corresponding sentence is considered redundant and removed.

128 “parameterize” -> “parameterizes”

The corresponding sentence is considered redundant and removed.

133 remove “have”

The corresponding sentence is considered redundant and removed, following editor's comment.

135 "cloud-particle" -> "cloud-particle and aerosol"

The corresponding sentence is considered redundant and removed, following editor's comment.

137 "varying with varying" -> "varying with"

The corresponding sentence is considered redundant and removed, following editor's comment.

137-138 "the bulk schemes in general uses" -> "bulk schemes in general use"

The corresponding sentence is considered redundant and removed, following editor's comment.

138-139 remove "which are not able to consider the variation of collection efficiencies and terminal velocities in reality"

The corresponding sentence is considered redundant and removed, following editor's comment.

139-140 "This makes the bin scheme more sophisticated than the bulk scheme." -> "Thus, the bin scheme is more sophisticated than the bulk scheme."

The corresponding sentence is considered redundant and removed, following editor's comment.

147 "are strongly dependent" -> "is strongly dependent"

Done.

152 "are referred to as fire-driven updrafts, henceforth" -> "are henceforth referred to as fire-driven updrafts"

Done.

153-156 "Aerosol effects on clouds are initiated by an increase in aerosol concentration, which can be caused by an increase in aerosol emission at and near the surface, and dependent on how much aerosol concentration increases, or on the magnitude of an increase in aerosol concentration, i.e., aerosol perturbation" -> "Aerosol effects on clouds are initiated by an increase in aerosol concentration, which can be caused by an increase in aerosol emission at and near the surface, and dependent on how much aerosol concentration increases (aerosol perturbation)"

The corresponding text is simplified as follows to make it more concise:

(LL129-131 on p4)

Effects of fire-induced increases in aerosol concentration on pyroCbs are likely to be dependent on how much aerosol concentration increases (aerosol perturbation) (e.g., Rosenfeld et al., 2008; Koren et al., 2012).

157 "has not been" -> "was not"

The corresponding sentence is revised as follows:

(LL131-132 on p5)

This study examines this dependence, not studied by Kablick et al. (2018).

166-167 "Shortwave and longwave radiation parameterizations have been included in all simulations by adopting" -> "Shortwave and longwave radiation is parameterized by"

Done.

169 "To represent the microphysical processes" -> "To represent microphysical processes"

Done.

176 “The cloud-droplet” -> “A cloud-droplet”, “parameterization, which is based” -> “parameterization based”

Done.

177 “Arbitrary aerosol mixing states and arbitrary” -> “Arbitrary aerosol mixing states and”

Done.

190 “case is performed” -> “case was performed”

The corresponding sentence is revised as follows:

(LL161-162 on p6)

The control run for an observed pyroCb case involved a forested site in the Canadian Northwest Territories (60.03 ° N, 115.45° W).

192 “the site and the pyroCb” -> “the site and pyroCb”

Done.

193 remove “between them”

Done.

198 “from Ft. Smith” -> “from the Ft. Smith”

Done.

204-205 “These tendencies are horizontally homogeneous and applied to the control run every time step by interpolation” -> “These tendencies are applied to the control run every time step by interpolation, in a horizontally homogeneous manner”

The corresponding sentence is revised as follows:

(LL169-171 on p6)

Temperature and humidity tendencies at each altitude from sequential soundings are obtained and applied to the control run every time step by interpolation in a horizontally homogeneous manner.

206 “lengths” -> “extents”

Done.

207 “For the simulation, the” -> “The simulation”

Done.

211-212 “simulation, at the center of the simulation domain, a fire spot with a diameter of 40 km is placed” -> “simulation, a fire spot with a diameter of 40 km is placed at the center of the simulation domain”

Done.

215 “the previous studies which are Trentmann et al. (2006) and Luderer et al. (2006) and adopt boreal forest emissions” -> “previous studies which adopt boreal forest emissions (Trentmann et al. (2006) and Luderer et al. (2006))”

Done.

219 “idealized and this enables” -> “idealized, enabling”

Done.

223 remove “aerosol properties that can be represented by”

Done.

235-236 “~50-70% of organic-carbon (OC) compounds, ~5-10% of black-carbon (BC) material, and ~20-45% of inorganic species” -> “~50-70% organic-carbon (OC) compounds, ~5-10% black-carbon (BC) material, and ~20-45% inorganic species”

Done.

247 Insert new paragraph after “in the control run.”

Done.

247-249 “According to Reid et al. (2005), Knobelspiessel et al. (2011), and Lee et al. (2014), it is reasonable to assume that the initial aerosol size distribution follows the unimodal lognormal distribution in fire sites. Hence, the control run adopts the unimodal lognormal distribution as an initial aerosol size distribution.” -> “The control run adopts the unimodal lognormal distribution as an initial aerosol size distribution, a reasonable assumption in fire sites (Reid et al. (2005), Knobelspiessel et al. (2011), Lee et al. (2014))”

Done.

Throughout, e.g. 251-253 “median aerosol diameter and standard deviation of the distribution” -> “median and standard deviation aerosol diameter”

Done.

263 Insert new paragraph after “(Pruppacher and Klett, 1978).”

Done.

264 “captured” -> “capture”

Done.

268 “points” -> “point”

Done.

267, 270 “counterparts” -> “values”

Done.

279-280 **“Figure 1. This field in Figure 2 represents” -> “Figure 1, representing”

Done.

Throughout, e.g. 284, including figures, “averaged” <cloud property> -> “average” <cloud property>

Done.

297 “well as” -> “well”

Done.

Throughout, including figures, anytime multiple rounds are referred to together in a row, e.g. 33,3 “the medium run and the weak run” -> “the medium and weak runs”

Done.

345 remove “or aerosol perturbation”

Done.

353, 356 “intensity” -> “intensities”

Done.

364 “corresponds” -> “correspond”

Done.

374 remove “themselves”

The corresponding paragraph is considered redundant and removed.

377 “(2018) by” -> “(2018),”

The corresponding paragraph is considered redundant and removed.

379-380 “The updraft mass flux is one of the most representative variables that are indicative of the cloud dynamic intensity and the magnitude of convective invigoration." -> ""The updraft mass flux is one of the most indicative variables of cloud dynamic intensity and magnitude of convective invigoration."

The corresponding sentence is revised as follows:

(LL318-319 on p11)

The updraft mass flux is one of the most indicative variables of the upward air motion and magnitude of convective invigoration.

381 "is" -> "was"

Done.

382-383 "17:00 GMT on August 5th and 12:00 GMT on August 6th, and 17:00 GMT on August 5th is a time around which the pyroCb starts to from" -> "17:00 GMT on August 5th, approximately when the pyroCb starts to form, and 12:00 GMT on August 6th, and 17:00 GMT on August 5th is"

Done.

385 "that" -> "which"

Done.

390-393 “Considering that the stratosphere is between the tropopause and its top that is generally ~ 50 km in altitude, the defined lower stratosphere occupies around a quarter of the total vertical extent of the stratosphere.” -> “The defined lower stratosphere occupies around a quarter of the total vertical extent of the stratosphere, the top of which is generally ~ 50 km in altitude.”

The corresponding sentences are simplified as follows:

(LL329-330 on p11)

The defined upper troposphere and lower stratosphere occupy around a quarter of the total vertical extent of the troposphere and stratosphere, respectively.

397 “the cloudy columns and that over” -> “cloudy and”

Done.

399 “in a part” -> “in the part”

Done.

410, 411 remove “those”

Done.

413 remove “or the pyroCb”

Done.

423 remove “both”

Done.

426 “as compared to that in” -> “versus”

Done.

430-433 “These averaged fluxes are over cloudy columns for the simulation period between 17:00 GMT on August 5th and 12:00 GMT on August 6th. The averaged water- vapor fluxes vary from 8.30x10-6 kg m-2 s-1 in the control run to 8.21x10-6 kg m-2 s-1 in the low-aerosol run.” -> “The averaged water-vapor fluxes over cloudy columns for the simulation period between 17:00 GMT on August 5th and 12:00 GMT on August 6th vary from 8.30x10-6 kg m-2 s-1 in the control run to 8.21x10-6 kg m-2 s-1 in the low- aerosol run.”

The corresponding sentences are revised as follows:

(LL356-360 on p12)

The small variation in updraft mass fluxes between the runs results in a small variation in the average water-vapor fluxes at the tropopause from 8.30×10-6 kg m-2 s-1 in the control run to 8.21×10-6 kg m-2 s-1 in the low-aerosol run over cloudy columns for the simulation period between 17:00 GMT on August 5th and 12:00 GMT on August 6th.

435 “only are” -> “are only”

We thought it was better to put “are”, since “only” is used to emphasize cirrus clouds are composed of ice crystals but not other types of hydrometeors. Stated differently, “only” is not used to emphasize cirrus clouds are between 9 and 13 km. In case we use “are only” as recommended by the reviewer here, the corresponding sentence emphasizes cirrus clouds are between 9 and 13 km. This is not what we intend to emphasize and thus we just put “are” here.

436 “there are the presence of” -> “there is the presence of”

The corresponding sentence is considered redundant and removed.

440 “cloud-ice number concentration and cloud-ice size” -> “cloud-ice number concentration and size”

Done.

442 “micron” -> µm

Done.

443-444 “The altitudes between 9 km and 13 km correspond to a part of the UTLS below the troposphere. Henceforth, the UTLS cirrus clouds mean those clouds in a part of the UTLS below the tropopause.” -> “The altitudes between 9 km and 13 km correspond to a part of the UTLS below the tropopause, and henceforth, “UTLS cirrus clouds” refer to clouds in the troposphere.”

The corresponding sentences are simplified as follows:

(LL366-367 on p13)

Henceforth, “the UTLS cirrus clouds” refer to clouds in the upper troposphere.

445-446 “Updrafts in the pyroCb produce supersaturation, which leads to the generation of cloud ice mass and associated cirrus clouds via deposition, the primary source of cloud-ice mass" -> "Updrafts in the pyroCb produce supersaturation, which leads to the primary source of cloud-ice mass and associated cirrus clouds via deposition"

Done.

449 "circus" -> "cirrus"

Done.

452, 453 remove "large"

As a process of making the corresponding paragraph succinct, the last two sentences in the paragraph are removed. In the removed sentences, "significant variations of cloud-ice number concentration and size" are indicated. Although those sentences are removed, we believe that it is important to deliver this indication in the rest of the sentences not removed. For this deliverance, "significant" replaces "large" in the sentence pointed out here by the reviewer.

456-457 "simulations (Figure 7), and thus a negligible variation of the mass of the UTLS cirrus clouds" -> "simulations (Figure 7), and thus a negligible variation of UTLS cirrus cloud mass"

Done.

462 "The role, which is played by" -> "The role of"

The corresponding paragraph is revised substantially following the editor's comment and the corresponding sentence is revised as follows:

(LL384-386 on p13)

The effects of fire-generated aerosols on the pyroCb updrafts, cirrus cloud mass and the enhancement of the water vapor transport are insignificant when fire intensity is strong.

464-465 “the UTLS at and above the tropopause, and in the production of the mass of the UTLS cirrus clouds is not significant for strong fire intensity" -> “the UTLS at and above the tropopause, and in the production UTLS cirrus cloud mass, is not significant for strong fire intensity"

The corresponding paragraph is revised substantially following the editor’s comment and the corresponding sentence is revised as follows:

(LL384-386 on p13)

The effects of fire-generated aerosols on the pyroCb updrafts, cirrus cloud mass and the enhancement of the water vapor transport are insignificant when fire intensity is strong.

470 “the mass of the UTLS cirrus clouds” -> “UTLS cirrus cloud mass”

The corresponding sentence is considered redundant and removed.

478 “updrafts, produced” -> “updrafts produced”

Done.

480-481 “when the fire-generated surface heat fluxes and the fire intensity” -> “when the fire intensity and fire-generated surface heat fluxes”

Done.

485-486 “relative magnitude of aerosol-induced perturbation of latent heat to” -> “relative magnitudes of aerosol-induced perturbation of latent heat and”

Done.

500 “does vary” -> “varies”

Done.

501-503 “Hence, it can be said that percentage differences in updraft mass fluxes mean percentage differences in updraft speed with good confidence.” -> “Hence, it can be said with good confidence that percentage differences in updraft mass fluxes mean percentage differences in updraft speed.”

The corresponding sentence is revised as follows:

(LL319-323 on p11)

Since the updraft mass flux is updraft speed that is multiplied by air density, and air density at each altitude varies negligibly, differences in updraft mass fluxes are mostly explained by those in updraft speeds among the simulations. Hence, with good confidence, differences in updraft mass fluxes mean those in updraft speeds.

505 “run than the clean-scenario run” -> “run and clean-scenario runs”

Done.

508 and similar equations: remove “the”

Done.

517 “gets larger” -> “increases”

Done.

530 “than the” -> “than in the”

Done.

549 "the averaged LWC and the averaged CDNC” -> “the average LWC and CDNC”

Done.

550, 551 “varies” -> “decreases”

Done.

559 “reduce” -> “decreases”

Done.

566 “to increase” -> “increasing”, “lowers” -> “decreases”

The corresponding sentence is revised as follows:

(LL473-478 on p16)

During period 1, as fire intensity weakens and updraft speed decreases, parcel equilibrium supersaturation decreases and thus, the minimum size of activated aerosol particles increases not only among the clean-scenario runs but also among the polluted-scenario runs. When the production of parcel supersaturation by updrafts and the consumption of supersaturation by droplets balance out, parcel supersaturation reaches parcel equilibrium supersaturation (Rogers and Yau, 1991).

569 “averaged associated” -> “associated average”

The corresponding sentence is revised as follows:

(LL478-482 on p16-17)

Mostly due to greater aerosol concentrations, associated average equilibrium supersaturation and minimum size of activated aerosol particles over areas with positive updraft speeds and period 1 are lower and larger, respectively, in the polluted-scenario run than in the clean-scenario run for each fire intensity (Rogers and Yau, 1991).

575 “reduces” -> “decreases”

Done.

577 remove “size in”

Done.

578 “reduces” -> “decreases”

Done.

586 “less close to” -> “further from”

The corresponding sentence is revised as follows:

(LL491-494 on p17)

A smaller portion of the total aerosol concentration is in the size range closer to the right tail of the distribution as long as the range is on the right-hand side of the distribution peak where most of aerosol activation occurs.

588-590 “distribution; most of aerosol activation occurs for aerosol sizes on the right- hand side of the distribution peak, here we are only concerned with the size ranges on the right-hand side.” -> “distribution. Here we are only concerned with the size ranges on the right-hand side of the distribution peak, where most of aerosol activation occurs.”

The corresponding sentence is revised as follows:

(LL491-494 on p17)

A smaller portion of the total aerosol concentration is in the size range closer to the right tail of the distribution as long as the range is on the right-hand side of the distribution peak where most of aerosol activation occurs.

Throughout the manuscript: <cloud property>“, which is averaged” -> <cloud property>“averaged” e.g. 593 “CNDC, which is averaged” -> “CNDC averaged”

Done.

596 “to greater” -> “to a greater”

Done.

605 “thus the” -> “thus a”

Done.

618 “rates, which are averaged” -> “rates averaged”

Done.

624-629 "The averaged autoconversion rates over period 2 reduce from 3.61x10-6 g m- 3 s-1 in the control run with strong fire intensity to 0.93x10-6 g m-3 s-1 in the weak run with weak fire intensity through 2.01x10-6 g m-3 s-1 in the medium run with medium fire intensity by 74%. Those averaged autoconversion rates reduce from 4.52x10-6 g m-3 s- 1 in the low-aerosol run with strong fire intensity to 3.94x10-6 g m-3 s-1 in the weak-low run with weak fire intensity through 4.43x10-6 g m-3 s-1 in the medium-low run with medium fire intensity by 14%." -> "The averaged autoconversion rates over period 2 decrease by 74% from 3.61x10-6 g m-3 s-1 in the control run with strong fire intensity to 0.93x10-6 g m-3 s-1 in the weak run with weak fire intensity through 2.01x10-6 g m-3 s- 1 in the medium run with medium fire intensity. Those averaged autoconversion rates decrease by 14%from 4.52x10-6 g m-3 s-1 in the low-aerosol run with strong fire intensity to 3.94x10-6 g m-3 s-1 in the weak-low run with weak fire intensity through 4.43x10-6 g m-3 s-1 in the medium-low run with medium fire intensity."

The corresponding sentence is shortened substantially as follows to make it more concise and succinct:

(LL513-516 on p18)

Due to the larger absolute and percentage reduction in Rv among the polluted-scenario runs than among the clean-scenario runs with weakening fire intensity during period 2, the average autoconversion rates decrease by 74% (14%) from the control (low-aerosol) to weak (weak-low) runs (Figure 9a).

631 "get greater" -> "increase"

Done.

633 "smaller" -> "lower"

The corresponding sentence is considered redundant and removed.

635 "freezing, which is averaged" -> "freezing averaged"

The corresponding sentence is considered redundant and removed.

640, 642 “get greater” -> “increase”

Done.

644-648 “When fire intensity is strong, the difference in freezing-related latent heat, which is averaged in cloudy areas and period 2, between the polluted-scenario run, which is the control run, and the clean-scenario run, which is the low-aerosol run,” -> “When fire intensity is strong, the difference in freezing-related latent heat averaged in cloudy areas and period 2 between the polluted-scenario run (control run) and the clean-scenario run (low-aerosol run)”

Following the other reviewer’s comment, the corresponding sentences are simplified as follows:

(LL521-523 on p18)

Thus, differences in the average rate of cloud-liquid freezing and freezing-related latent heat over the period 2 between the runs increase with weakening fire intensity (Figure 9a).

649-650 “while with weak fire intensity, that difference between the polluted-scenario run, which is the weak run, and the clean-scenario run, which is the weak-low run” -> “while with weak fire intensity, that difference between the polluted-scenario run (weak run) and the clean-scenario run (weak-low run)”

Following the other reviewer’s comment, the corresponding sentences are simplified as follows:

(LL521-523 on p18)

Thus, differences in the average rate of cloud-liquid freezing and freezing-related latent heat over the period 2 between the runs increase with weakening fire intensity (Figure 9a).

651-652 “differences, which are calculated by Equation (1),” -> “differences (calculated by Equation (1))”

The corresponding sentence is removed as a process of making the corresponding paragraph more concise and succinct, following the other reviewer's comment.

653-654 "and the clean-scenario run from 9% with strong fire intensity to 83% with weak fire intensity through 51% with medium fire intensity over the period 2." -> "and the clean-scenario run over the period 2 from 9% with strong fire intensity to 83% with weak fire intensity to 51% with medium fire intensity."

The corresponding sentence is removed as a process of making the corresponding paragraph more concise and succinct, following the other reviewer's comment.

657, 674 "deposition, which is averaged" -> "deposition averaged"

It is replaced with "average deposition"

658 "period 2," -> "period 2"

Done.

666 "transportation" -> "transport"

Done.

670 remove "as compared to those before 20:30 GMT"

Done.

673 "from" -> "starting at"

Done.

685 “in polluted-scenario” -> “in the polluted-scenario”

Done.

688 “The more mass fluxes and the more convergence” -> “The higher mass fluxes and convergence”

Done.

690 “be greater” -> “increase”

Done.

695-696 “thus, enhancing freezing, deposition, condensation, and updrafts further.” -> “thus further enhancing freezing, deposition, condensation, and updrafts.”

Done.

708, 710, 713, 727,741 “get greater” -> “increase”

Done.

714-715 “Then, the increases in condensation, in turn, further enhance the increases in updrafts in the polluted-scenario run for each fire intensity.” -> “The increases in condensation further enhance the increases in updrafts in the polluted-scenario run for each fire intensity."

The corresponding sentences are revised as follows:

(LL564-566 on p19)

The greater increases in condensation cause the greater further enhancement of the increases in updrafts in the polluted-scenario run with weaker fire intensity.

720 "induce" -> "induces"

Done.

752-753 "This possibility is not that unrealistic, since stronger fire likely involves more material burnt and more aerosols from it." -> "This possibility is not that unrealistic, since stronger fires likely involve more material burned and higher aerosol emissions."

Done.

757, 763, 778 "reduces" -> "decreases"

Done.

758 "less" -> "lower"

Done.

759-760 "For strong fire, the perturbation-related aerosol concentration is 30000

760 cm-3, for medium fire, it is 15000 cm-3, and for weak fire, it is 7500 cm-3." -> "The perturbation-related aerosol concentration is 30000 cm-3 for strong fire, 15000 cm-3 for medium fire, and 7500 cm-3 for weak fire."

Done.

765 "when fire-induced" -> "when the fire-induced"

Done.

772 "has the" -> "has an"

Done.

773 "run; when" -> "run, and when"

The corresponding sentence is revised as follows:

(LL613-615 on p21)

Based on this, the medium run is repeated with a fire-induced aerosol perturbation of 2000 cm^{-3} down from 15000 cm^{-3} (the medium-2000 run). The weak run is repeated with a fire-induced aerosol perturbation of 1000 cm^{-3} down from 7500 cm^{-3} (the weak-1000 run).

780 "with the weakening" -> "with weakening fire"

Done.

783 "whether those aerosol perturbations vary with varying fire intensity or not" -> "whether or not those aerosol perturbations vary with varying fire intensity"

Done.

791 “transport water vapor to the tropopause and above efficiently” -> “efficiently transport water vapor to the tropopause and above”

Done.

796 “gets more” -> “becomes”

Done.

810 “reduce” -> “decrease”

Done.

813 “makes” -> “results in”

The corresponding sentence is revised as follows:

(LL646-648 on p22)

This reduces autoconversion more among the polluted-scenario runs and increases differences in autoconversion between the polluted-scenario and clean-scenario runs as fire intensity weakens.

814 “increase” -> “increasing”

The corresponding sentence is revised as follows:

(LL646-648 on p22)

This reduces autoconversion more among the polluted-scenario runs and increases differences in autoconversion between the polluted-scenario and clean-scenario runs as fire intensity weakens.

830 “It is true that the” -> “The”

Done.

833 “transportation” -> “transport”

Done.

850 “reduce” -> “decrease”

Done.

858 “came up with” -> “generated”, “preformed” -> “performed”

Done.

861 “revised manuscript based on the reviewers’ comments and perform” -> “revised the manuscript based on the reviewers’ comments and performed”

Table 2 “Meidum-low” -> “Medium-low”

Done.

1172, 1190 “The averaged” -> “Averaged”

“The averaged” is replaced with “average”

Figure 10 “Differences in the averaged values” -> “Differences in average values”

Done.

Referee Report 2

Review of:
"Examination of effects of aerosol on a pyroCb and their dependence on fire intensity and aerosol perturbation using a cloud-system resolving model"

Authors: Seoung Soo Lee, George Kablick III, Zhanqing Li

Recommend minor revisions.

General comment:

This revised manuscript is much improved. The results are clearer and presented more concisely in most areas. Following a few additional clarifications and rewording, I think this paper will be ready for publication.

Authors appreciate the reviewer's comments below that improved the quality of the manuscript a lot. In the following, each comment by the reviewer (black) is followed by our response (blue).

Specific comments:

1.Lines 126: Use of the phrase "has been going on" seems a little awkward.

(LL67 on p3)

It is corrected to be "have been conducted to improve this understanding"

2.Lines 248-251: The sentence beginning with "Aerosol effects on clouds…" is confusing as stated. Please make this clearer.

Revised as follows by reflecting another reviewers' comment as well:

(LL129-132 on p5)

Effects of fire-induced increases in aerosol concentration on pyroCbs are likely dependent on how much aerosol concentration increases (aerosol perturbation) (e.g., Rosenfeld et al., 2008; Koren et al., 2012). This study examines this dependence, not studied by Kablick et al. (2018).

3.Line 373: Please find more recent specific references related to IN observations.

Fan et al. (2014 and 2017) are added.

4.Line 527: Grammar error. "pyro Cb starts to from…"

Corrected.

5.Lines 1356-1367: This paragraph is still quite wordy, and thus, it's difficult to read and easily discern the main take-away point. Please reword to be more concise. The specific numbers related to differences in latent heating are not particularly meaningful; the % change is more meaningful and is the basis of your main point here. You could include the actual numbers in a table if you wish.

Number are remove*d and the corresponding text is revised as follows:*

(LL519-523 on p18)

The increasing differences in autoconversion rates between the polluted-scenario and clean-scenario runs increase those differences in the amount of cloud liquid available for freezing with weakening fire intensity (Figure 9a). Thus, differences in the average rate of cloud-liquid freezing and freezing-related latent heat over the period 2 between the runs increase with weakening fire intensity (Figure 9a).

6.Lines 1611-1612: This sentence is worded in such a way that makes it sound like you are saying that the fire intensity weakens due to reduction in LWC. Please reword to better state your point.

The corresponding text is considered redundant and removed.

Examination of effects of aerosols on a pyroCb and their dependence on fire intensity and aerosol perturbation

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[7]National Institute of Environmental Research, Incheon, South Korea

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Abstract

Through a modeling framework, this study investigates how a pyrocumulonimbus (pyroCb) event influences water vapor concentrations and cirrus cloud properties near the tropopause, specifically focusing on how fire-produced aerosols affect this role. Results from a case study show that when observed fire intensity is high, there is an insignificant impact of fire-produced aerosols on the development of the pyroCb and associated changes in water vapor and cirrus clouds near the tropopause. However, as fire intensity weakens, effects of those aerosols on microphysical variables and processes such as droplet size and autoconversion increase. Due to this, aerosol-induced invigoration of convection is significant for pyroCb with weak-intensity fires and associated weak surface heat fluxes. This leads to a situation where there is a greater aerosol effect on the transport of water vapor to the upper troposphere and the production of cirrus clouds with weak-intensity fires, whereas this effect is muted with strong-intensity fires.

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1. Introduction

Recent studies (e.g., Pumphrey et al., 2011; Kablick et al., 2018) showed that pyrocumulonimbus (pyroCbs) may transport significant amounts of water vapor to the upper troposphere and the lower stratosphere (UTLS) and can possibly have an impact on seasonal UTLS water vapor budgets. Any change in water vapor in the UTLS has an exceptionally strong influence on the global radiation budget and thus Earth's climate (Solomon et al., 2010). PyroCbs develop cirrus clouds associated with overshooting convective tops that reach the UTLS. Changes in cirrus clouds in the UTLS are known to have a strong influence on the global radiation budget (Solomon et al., 2010). The level of our understanding of impacts of pyroCbs on water vapor and cirrus clouds in the UTLS on a global scale is very low and studies have been conducted to improve this understanding (Fromm et al., 2010). However, this paper does not focus on these pyroCb impacts at the global scale. Instead, this paper aims to gain a process-level understanding of mechanisms that control local impacts of individual pyroCbs on water vapor and cirrus clouds in the UTLS. The examination of these mechanisms can provide useful information to parameterize interactions among pyroCbs, water vapor and cirrus clouds in climate models.

PyroCbs initiate over a fire, and the large surface energy release mainly through fire-induced latent- and sensible-heat fluxes at and near the surface affects the dynamic, thermodynamic and microphysical development of pyroCbs (Fromm et al., 2010; Peterson et al., 2017). However, questions remain about what role the large concentration of cloud condensation nuclei (CCN) contained in smoke has on the vertical development and microphysical properties of pyroCbs. Studies (e.g., Koren et al., 2008; Rosenfeld et al., 2008; Storer et al., 2010; Tao et al., 2012) showed that aerosols affected cumulonimbus clouds, thus raising the possibility that fire-generated aerosols may affect pyroCb development. As an example of aerosol impacts on cumulonimbus clouds, these studies have demonstrated that increases in aerosol loading can decrease the size of droplets (i.e., cloud-liquid particles). Individual aerosol particles act as seeds for the formation of droplets, so increasing aerosol loading leads to more droplets formed. More droplets mean more competition among them for available water vapor needed for their condensational growth, decreasing the size of individual droplets (Twomey, 1977; Albrecht, 1989).

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Aerosol-induced smaller droplet sizes reduce the efficiency of the growth of cloud-liquid particles to raindrops via autoconversion, a collection process among cloud-liquid particles in which they become raindrops (Pruppacher and Klett, 1978; Rogers and Yau, 1991). This reduced efficiency leads to less cloud liquid converted to rain and thus more cloud liquid available for transport by updrafts to altitudes above the freezing level. This eventually induces more freezing of cloud liquid, enhanced parcel buoyancy and the invigoration of updrafts and associated convection (Rosenfeld, 2008).

The role of fire-generated aerosols in the development of pyroCbs and their effects on water vapor and cirrus clouds in the UTLS lacks a firm scientific understanding, and hence this paper focuses on that role of those aerosols. To examine the role, this study extends the previous modeling work by Kablick et al. (2018). The modeling work therein showed that the effects of fire-generated aerosols on the development of a specific pyroCb and its impacts on the UTLS water vapor and cirrus clouds were negligible compared to the effects of fire-generated heat fluxes. However, aerosol effects on cloud development vary with cloud properties such as typical updraft speeds (e.g., Khain et al., 2008; Lee et al., 2008; Tao et al., 2012). For simplicity herein, an "updraft" refers to the general upward motion of convective air, or to the actual updraft speed representing the updraft or convective intensity, depending on the context. Typical updrafts are determined by environmental instability as represented by convective available potential energy (CAPE). Lee et al. (2008) demonstrated that different clouds with different typical updrafts showed different sensitivity of cloud microphysical and thermodynamic development to aerosol concentration. Hence, it is hypothesized that aerosol effects on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds vary depending on the pyroCb typical updrafts.

To examine the potential variation of aerosol effects on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds with typical updrafts of pyroCbs, numerical simulations are performed. Simulated is the pyroCb case examined by Kablick et al. (2018) using a cloud-system resolving model (CSRM). The CSRM is capable of resolving cloud-scale dynamic and thermodynamic processes. The basic modeling methodology in this study is similar to that used by Kablick et al. (2018). However, this

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study uses a more sophisticated microphysical scheme, i.e., a bin scheme, rather than the two-moment bulk scheme used by Kablick et al. (2018).

Note that Kablick et al. (2018) examined aerosol effects on the convective development of a specific pyroCb case with a typical updraft framework. The present study expands upon that work by performing sensitivity simulations in which typical updrafts in the pyroCb vary, enabling us to ascertain the dependence of aerosol effects on typical updrafts. Note that CAPE, which determines typical updrafts in convective clouds, is strongly dependent on surface latent and sensible heat fluxes (e.g., Houze, 1993), and in the case of pyroCbs, these fluxes are strongly controlled by fire intensity. Therefore, the present sensitivity simulations enable us to study the dependence of those aerosol effects on fire intensity. Since fire intensity is the dominant driver of the pyroCb typical updrafts, these typical updrafts are henceforth referred to as fire-driven updrafts.

Effects of fire-induced increases in aerosol concentration on pyroCbs are likely dependent on how much aerosol concentration increases (aerosol perturbation) (e.g., Rosenfeld et al., 2008; Koren et al., 2012). This study examines this dependence, not studied by Kablick et al. (2018).

2. Modeling framework

We use the Advanced Research Weather Research and Forecasting (ARW) model, a nonhydrostatic compressible model, as the CSRM. Prognostic microphysical variables are transported with a fifth-order monotonic advection scheme (Wang et al., 2009). Shortwave and longwave radiation is parameterized by the Rapid Radiation Transfer Model (RRTM; Mlawer et al., 1997; Fouquart and Bonnel, 1980).

To represent microphysical processes, the CSRM adopts a bin scheme based on the Hebrew University Cloud Model described by Khain et al. (2009). The bin scheme solves a system of kinetic equations for the size distribution functions of water drops, ice crystals (plate, columnar and branch types), snow aggregates, graupel and hail, as well as cloud condensation nuclei (CCN) and ice nuclei (IN). Each size distribution is represented by 33 mass doubling bins, i.e., the mass of a particle m_k in the kth bin is determined as $m_k = 2m_{k-1}$.

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A cloud-droplet nucleation parameterization based on Köhler theory represents cloud-droplet nucleation. Arbitrary aerosol mixing states and aerosol size distributions can be fed to this parameterization. To represent heterogeneous ice-crystal nucleation, the parameterizations by Lohmann and Diehl (2006) and Möhler et al. (2006) are used. In these parameterizations, contact, immersion, condensation-freezing, and deposition nucleation paths are all considered by taking into account the size distribution of IN, temperature and supersaturation. Homogeneous aerosol (or haze particle) and droplet freezing is also considered following the theory developed by Koop et al. (2000).

3. Case description and simulations

3.1 Control run

The control run for an observed pyroCb case involved a forested site in the Canadian Northwest Territories (60.03° N, 115.45° W). Kablick et al. (2018) give details about the site and pyroCb case. The control run is identical to the Full Simulation in Kablick et al. (2018) except for the different microphysical schemes. The period covered by the control run is from 12:00 GMT on August 5^{th} to 12:00 GMT on August 6^{th} in 2014 and captures the initial, mature, and decaying stages of the pyroCb. As described by Kablick et al. (2018), balloon soundings of winds, temperature and dew-point temperature were obtained every 6 hours from the Ft. Smith observation station near the forested site. Sounding data at 12:00 GMT on August 5^{th} prescribe the initial atmospheric conditions. Temperature and humidity tendencies at each altitude from sequential soundings are obtained and applied to the control run every time step by interpolation in a horizontally homogeneous manner. These tendencies represent the impacts of synoptic- or large-scale motion on temperature and humidity (Grabowski et al., 1996; Krueger et al., 1999; and Lee et al., 2018). The control run is performed in a three dimensional domain with horizontal and vertical extends of 300 km and 20 km, respectively. The simulation horizontal and vertical resolutions are 500 m and 200 m, respectively, to resolve cloud dynamic and thermodynamic processes.

Figure 1 shows a satellite image of the observed pyroCb and the fire spot (spatial length is ~40 km) when the cloud is about to advance into its mature stage. To emulate this in the

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simulation, a fire spot with a diameter of 40 km is placed at the center of the simulation domain (Figure 2). In the fire spot, the surface latent and sensible heat fluxes are set at 1800 and 15000 W m^{-2}, respectively. In areas outside of the fire spot, the surface latent and sensible heat fluxes are set at 310 and 150 W m^{-2}, respectively. These surface heat-flux values follow previous studies which adopt boreal forest emissions (Trentmann et al., 2006; Luderer et al., 2006). Following Kablick et al. (2018), the surface heat-flux values are prescribed with no temporal variation and no consideration of interactions between heat fluxes and the atmosphere in the control run. Thus, the setup for the surface heat fluxes is idealized, enabling a better isolation of aerosol effects on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds by excluding effects of interactions between the surface heat fluxes and atmosphere on this development and its impacts.

For the selected pyroCb case, aerosol chemical composition, size distribution and concentration are unknown. Hence, in (outside) the fire spot at the first time step, the concentration of aerosols acting as CCN is prescribed to be 15000 (150) cm^{-3}. This prescription is for the planetary boundary layer (PBL) and the concentration decreases exponentially with height above the PBL top. These prescribed aerosol concentrations are typically observed in fire spots and their background (Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998; Reid et al., 1999; Andreae et al., 2004; Reid et al., 2005; Luderer et al., 2009).

Reid et al. (2005) showed that aerosol mass produced by forest fires was generally composed of (by mass) ~50-70% organic-carbon (OC) compounds, ~5-10% black-carbon (BC) material, and ~20-45% inorganic species. The approximate median value of each of these chemical component percentage ranges determines the aerosol particle composition in the control run, i.e., 60% OC, 8% BC, and 32% inorganic species. OC is assumed to be water soluble and composed of 18 % levoglucosan ($C_6H_{10}O_5$, density = 1600 kg m^{-3}, van't Hoff factor = 1), 41 % succinic acid ($C_4H_6O_4$, density = 1572 kg m^{-3}, van't Hoff factor = 3), and 41 % fulvic acid ($C_{33}H_{32}O_{19}$, density = 1500 kg m^{-3}, van't Hoff factor = 5) based on typically observed chemical composition of OC compounds over fire sites (Reid et al., 2005). In the control run, the inorganic species are assumed to be ammonium sulfate, a representative inorganic species associated with fires (Reid et al., 2005). This chemical

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composition taken for aerosol particles is assumed to be spatiotemporally unvarying in the control run.

The control run adopts the unimodal lognormal distribution as an initial aerosol size distribution, a reasonable assumption in fire sites (Reid et al., 2005; Knobelspiessel et al., 2011; Lee et al., 2014). Those studies reported that in general, median and standard deviation aerosol diameters range from ~0.01 μm to ~0.03 μm and from ~2.0 μm to ~2.2 μm, respectively, for aerosols as CCN. The approximate median values of these ranges determine median and standard deviation diameters of aerosols as CCN in the control run, i.e., 0.02 μm and 2.1 μm, respectively. Assumed identical in the control run are IN and CCN aerosol properties except that at the first time step, (1) their median and standard deviation aerosol diameters differ, and (2) the IN concentration is 100 times lower than the CCN concentration (Pruppacher and Klett, 1978; Fan et al., 2014 and 2017). Following Seinfeld and Pandis (2006) and Phillips et al. (2007), for aerosols as IN, median and standard deviation aerosol diameters are assumed to be 0.1 μm and 1.6 μm, respectively, which are typical values in the continent.

Airflow in clouds diffuses and advects aerosols. After activation or capture by precipitating hydrometeors, aerosols are transported within hydrometeors and removed from the atmosphere once these hydrometeors reach the surface. Once clouds disappear completely at any grid point aerosol size distribution and number concentration recover to the background values at the first time step. This assumption simulates overall aerosol properties and their impacts on clouds and precipitation reasonably well (Morrison and Grabowski, 2011; Lebo and Morrison, 2014; Lee et al., 2016). This assumption means that fire continuously produces aerosols to maintain the initial background aerosol concentration.

Located to the northeast of the fire spot is the observed cirrus cloud at the top of the pyroCb, since winds advect the cloud northeastward (Figure 1). The extent of the observed cirrus cloud is ~100 km. Figure 2 shows the simulated field of cloud-ice mass density at a time that corresponds to the satellite image in Figure 1, representing the simulated cirrus cloud in the control run. Located to the northeast of the fire spot is the simulated cirrus cloud, and the extent of this cloud is ~100 km. The morphologies of the observed and simulated cirrus clouds agree well.

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The average liquid-water path (LWP) over areas with non-zero LWP in the control run is 960 g m^{-2}, and the average ice-water path (IWP) over areas with non-zero IWP in the control run is 202 g m^{-2}. These simulated LWP and IWP are ~10 % different from the satellite-retrieved values. In this study, droplet mass but not raindrop mass is used to obtain liquid-water content (LWC) and LWP, and the mass of ice crystals but not the mass of snow aggregates, graupel and hail is used to obtain ice-water content (IWC) and IWP. Drops with radii smaller (greater) than 20 µm are classified as droplets (raindrops). The average cloud-top and cloud-base heights over the life span of the pyroCb are 10.3 km and 3.6 km in the control run, respectively, and these simulated heights are ~7% different from the satellite-retrieved values. Overall, cloud macro-physical structures, as represented by LWP, IWP, cloud-top and cloud-base heights, are simulated reasonably well compared to the observation.

Compared are the control-run and observed reflectivity fields. Kablick et al. (2018) provide details about the reflectivity field observed by CloudSat. Observations and the control run both show increasing reflectivity up to ~7 km, decreasing reflectivity between ~7 km and ~11 km and insignificant changes in reflectivity above ~11 km in altitude (Figure 3). The average reflectivity over altitudes along the Cloudsat path in the control run is -8.1 dBZe, ~15% different from the observed value. Hence, the reflectivity field is simulated fairly well compared to the observation. These favorable comparisons between the observed and simulated cirrus clouds, cloud macro-physical and reflectivity fields demonstrate that the pyroCb-case simulation reasonably reproduces the event.

3.2 Low-aerosol run

To see the role played by fire-generated aerosols in the development of the pyroCb and its effects on the UTLS water vapor and cirrus clouds, we repeat the control run by reducing aerosol concentration in the fire spot from 15000 cm^{-3} to the background aerosol concentration (i.e., 150 cm^{-3}). This reduction removes fire-generated aerosols in the fire spot. Hence, comparisons between the control run and this repeated run, referred to as the low-aerosol run, will identify the role played by fire-generated aerosols. The low-aerosol

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run is identical to the Low Aerosol Simulation in Kablick et al. (2018) except for the different microphysical schemes between them.

3.3 Additional runs

For the examination of the potential variation of effects of fire-generated aerosols on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds with fire intensity and associated fire-driven updrafts, we repeat the control run by varying fire intensity. Remember that fire intensity controls surface latent and sensible heat fluxes on which fire-driven updrafts are strongly dependent. Therefore, variations in fire-induced surface latent and sensible heat fluxes can represent variations in fire intensity. As a first step, the control run is repeated by reducing fire-induced surface latent and sensible heat fluxes by factors of 2 and 4, respectively. The first repeated run represents a case with medium fire intensity, referred to as "the medium run". The second repeated run represents a case with weak fire intensity, referred to as "the weak run". Relative to these repeated runs, the control run represents a case with strong fire intensity. Then, to see effects of fire-generated aerosols on pyroCb development for each of those different fire intensities, the medium and weak runs are repeated with the identical initial aerosol concentration to that in the low-aerosol run. The repeated medium and weak runs are referred to as "the medium-low run" and "the weak-low run", respectively. The control, medium, and weak runs are the polluted-scenario runs, and the low-aerosol, medium-low, and weak-low runs are the clean-scenario runs.

Effects of fire-generated aerosols on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds can also depend on the magnitude of fire-induced increases in aerosol concentration in a fire spot. To test this dependence, for each fire intensity, we repeat the polluted-scenario run by increasing and decreasing the magnitude by a factor of 2 in the fire spot but not outside of the fire spot. The simulations with the increased magnitude have an aerosol concentration of 30000 cm^{-3} at the first time step over the fire spot in the PBL, referred to as the control-30000, medium-30000, and weak-30000 runs for strong, medium, and weak fire intensities, respectively. The simulations with the decreased magnitude have an aerosol concentration of 7500 cm^{-3} at the first time step over

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the fire spot in the PBL, referred to as the control-7500, medium-7500, and weak-7500 runs for strong, medium, and weak fire intensities, respectively. Motivated by the analysis described in Section 4.3, we additionally repeat the medium and weak runs with aerosol concentrations of 2000 and 1000 cm^{-3}, respectively, at the first time step over the fire spot in the PBL. The repeated medium (weak) run is referred to as the medium-2000 (weak-1000) run. Table 1 summarizes the simulations.

The aerosol concentration of 30000 cm^{-3} (7500, 2000 and 1000 cm^{-3}) over the fire spot corresponds to a situation where fire produces a larger (lower) concentration of aerosols than what is typically observed, i.e., 10000 to 20000 cm^{-3} (Reid et al, 1999; Andreae et al, 2004; Reid et al, 2005; Luderer et al., 2009).

4. Results

4.1 The control and low-aerosol runs

The updraft mass flux was averaged over the simulation period between 17:00 GMT on August 5th, approximately when the pyroCb started to form, and 12:00 GMT on August 6th (Figure 4). The updraft mass flux is one of the most indicative variables of the upward air motion and magnitude of convective invigoration. Since the updraft mass flux is updraft speed that is multiplied by air density, and air density at each altitude varies negligibly, differences in updraft mass fluxes are mostly explained by those in updraft speeds among the simulations. Hence, with good confidence, differences in updraft mass fluxes mean those in updraft speeds.

The upper troposphere is defined here to be between ~9 km in altitude and the tropopause which is ~13 km in altitude. The equilibrium level where the buoyancy of a rising air parcel becomes zero above the level of free convection is the tropopause (Emanuel, 1994). The lower stratosphere is defined here to be between the tropopause and an altitude 10 km above the tropopause. The UTLS is thus between ~9 km and ~23 km in this study. The defined upper troposphere and lower stratosphere occupy around a quarter of the total vertical extent of the troposphere and stratosphere, respectively.

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Updraft mass fluxes in the control run are only ~3% greater than those in the low-aerosol run (Figure 4 and Table 2). Given the hundredfold difference in aerosol loading over the fire spot between the runs, this 3% difference in updraft fluxes is negligible. The comparison between water-vapor mass density over the cloudy and non-cloudy columns in the control run demonstrates that there is a substantial 5-fold increase in the amount of water vapor in the part of the UTLS at and above the tropopause due to the pyroCb (Figure 5 and Table 2). Henceforth, the UTLS water vapor means water vapor in the part of the UTLS at and above the tropopause.

For the simulation period between 17:00 GMT on August 5th and 12:00 GMT on August 6th, the average water-vapor mass fluxes at the tropopause over cloudy and non-cloudy grid columns are 8.30 $\times 10^{-6}$ and 0.57$\times 10^{-6}$ kg m^{-2} s^{-1}, respectively. Due to the presence of the pyroCb and associated updrafts in cloudy grid columns, there are substantial increases in fluxes at the tropopause over cloudy grid columns compared to fluxes in the background over non-cloudy grid columns. This explains the larger amount of the UTLS water vapor over the pyroCb than in the background in the control run. The vertical extent of water vapor reaches further up to ~16 km by the pyroCb beyond ~14 km in the background (Figure 5). This means that air parcels that include water vapor overshoot the tropopause by ~3 km in the pyroCb, while those parcels in the background do so by ~ 1 km. This implies that air parcels and associated updrafts in the pyroCb are stronger, reaching higher altitudes. Those stronger air parcels enable water-vapor layers to be deepened in the lower stratosphere. These deepened layers and their greater water-vapor mass contribute to more interception of longwave radiation by water vapor in the UTLS over the pyroCb.

Similar to the situation with updraft mass fluxes, there is only a small (~2%) increase in the average mass of the UTLS water vapor in the control run versus the low-aerosol run for strong fire intensity (Figure 5 and Table 2). The small variation in updraft mass fluxes between the runs results in a small variation in the average water-vapor fluxes at the tropopause from 8.30$\times 10^{-6}$ kg m^{-2} s^{-1} in the control run to 8.21$\times 10^{-6}$ kg m^{-2} s^{-1} in the low-aerosol run over cloudy columns for the simulation period between 17:00 GMT on August 5th and 12:00 GMT on August 6th.

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The altitude of homogeneous freezing is 9 km, so cirrus clouds composed of ice crystals (or cloud ice) are between 9 km and 13 km in the control run (Figure 6). The amount of cirrus clouds in the control run, represented by the average cloud-ice mass density, ranges from 0.028 to 0.037 g m^{-3} between 9 km and 13 km (Figure 6). The average cloud-ice number concentration and size, represented by ice-crystal volume mean radius, between 9 km and 13 km range from 6 to 20 cm^{-3}, and from 10 to 20 µm, respectively. Henceforth, “the UTLS cirrus clouds” refer to clouds in the upper troposphere.

Updrafts produce supersaturation, which leads to the primary source of cloud-ice mass and associated cirrus clouds via deposition. Due to the negligible variation of updraft mass fluxes, there are negligible variations of supersaturation and deposition (Figure 7). So, there is only a negligible increase (~4%) in UTLS cirrus cloud mass in the control run compared to that in the low-aerosol run (Figure 6 and Table 2). However, mainly due to the larger aerosol concentrations, and associated greater homogeneous aerosol and droplet freezing, there is a significant ~20-fold increase in cloud-ice number concentration and associated with this, there is a significant ~2-fold decrease in cloud-ice size in the control run between 9 km and 13 km.

In summary, the pyroCb and associated updrafts cause a substantial enhancement of the transport of water vapor to the UTLS at and above the tropopause. Wang (2019) also reported this enhancement. Using modeling work and satellite observation, Wang (2019) indicated that the upward transport of water vapor in deep convective storms was possibly a major pathway through which water substance entered the stratosphere. Wang (2019) showed that the upward transport of water vapor was aided by gravity wave and its breaking in overshooting convective parcels. The pyroCb and its updrafts also produce cirrus clouds. The effects of fire-generated aerosols on the pyroCb updrafts, cirrus cloud mass and the enhancement of the water vapor transport are insignificant when fire intensity is strong.

4.2 Dependence of aerosol effects on fire intensity

When fire-generated surface heat fluxes and fire intensity increase, in-cloud latent heat is also likely to increase because a major source of in-cloud latent heating is surface heat flux.

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Therefore, aerosol-induced perturbations of latent heating may be relatively small compared with large in-cloud latent heat contributed by surface fluxes with very intense burning. Thus, aerosol-induced increases in parcel buoyancy, updrafts and their impacts on water vapor and the amount of cirrus clouds are relatively small compared with the large buoyancy, strong fire-driven updrafts produced by strong fire intensity and their associated impacts on water vapor and the amount of cirrus clouds.

When fire intensity and fire-generated surface heat fluxes decrease, in-cloud latent heat is also likely to be smaller. Here, we are interested in how the magnitude of an aerosol-induced perturbation of latent heating for a pyroCb with weak fire intensity compares to that with strong fire intensity. This is to evaluate the possibility that with background in-cloud latent heat varying with fire intensity, the relative magnitudes of aerosol-induced perturbations of latent heat and surface flux-dominated latent heat may vary.

4.2.1 Effects of Updrafts on the UTLS water vapor and cirrus clouds

The average updraft mass fluxes in the low-aerosol, medium-low, and weak-low runs represent fire-driven updrafts for strong, medium and weak fire intensities, respectively (Figure 4). Due to different fire intensity and associated CAPE, fire-driven updrafts vary between these runs. All weak, medium and strong fire intensity cases show aerosol-induced increases in updraft mass fluxes (Figure 4 and Table 2). Of interest is that the greatest percentage increase in updraft mass flux is in the case of weak fire (weak-low to weak runs), smallest in the case of strong fire (low-aerosol to control runs), and intermediate in the case of medium fire (medium-low to medium runs) (Figure 4 and Table 2). Here, the percentage difference, including both the percentage increase and decrease, is the relative difference in the value of variables between the polluted-scenario and clean-scenario runs for each fire intensity. The following equation determines this percentage difference for the strong fire intensity case:

$$\frac{Control\ run\ minus\ low{-}aerosol\ run}{low{-}aerosol\ run} \times 100\ (\%) \qquad (1)$$

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Replacing the control run with the medium (weak) run, and the low-aerosol run with the medium-low (weak-low) run in Equation (1) determines the percentage difference for the medium (weak) fire intensity case. Associated with the greater increases in updraft mass fluxes, the percentage increases in the UTLS water vapor and cloud-ice mass (Equation 1) are greater in the case of weaker fire (Figures 5 and 6 and Table 2).

In this section, we see that although fire-produced aerosols invigorate updrafts in all three types of fire intensity, the invigoration-induced increases in the UTLS water-vapor and cloud-ice mass increase as fire intensity weakens.

4.2.2 Volume mean radius of droplets (R_v)

a. Cloud droplet number concentration (N_d) and LWC

The simulation period is divided into four sub-periods for this next analysis: period 1 between 17:00 and 19:00 GMT on August 5th (initial formation of the pyroCb), period 2 between 19:00 and 21:00 GMT on August 5th, and period 3 between 21:00 GMT and 23:00 GMT on August 5th (initial stages of cloud development), and period 4 between 23:00 GMT on August 5th and 12:00 GMT on August 6th (mature and decaying stages). The average N_d over period 1 decreases as the fire intensity and updrafts decrease (Figure 8). The polluted-scenario run has higher aerosol concentrations over the fire spot (Table 1), leading to the much higher average N_d in the polluted-scenario run than in the clean-scenario run for each fire intensity. Increasing N_d enhances competition among droplets for a given amount of water vapor. Enhanced competition eventually curbs the condensational growth and reduces droplet size (R_v). This explains why the average R_v over period 1 is smaller in the polluted-scenario run (Figure 8). Of interest is that as fire intensity weakens, although the average N_d decreases, the average R_v decreases not only among the polluted-scenario runs but also among the clean-scenario runs over the fire spot (Figure 8). This is because R_v is proportional to $(\frac{LWC}{N_d})^{\frac{1}{3}}$ based on the following Equation (2):

$$R_v = \left(\frac{3}{4\pi\rho_w}\right)^{\frac{1}{3}} \left(\frac{LWC}{N_d}\right)^{\frac{1}{3}} \qquad (2)$$

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Here, ρ_w represents water density at a constant value of 1000 kg m^{-3}, so $(\frac{3}{4\pi\rho_w})^{\frac{1}{3}}$ has a constant value. LWC represents the given amount of water available for the condensational growth of droplets. This proportionality means that for a given N_d, a decrease in LWC decreases R_v. The average LWC over period 1 also decreases with weakening fire intensity not only among the polluted-scenario runs but also among the clean-scenario runs (Figure 8). Effects of LWC on R_v outweigh those of N_d, leading to the decrease in the average R_v with weakening fire intensity (Figure 8).

Using the average LWC and N_d from Figure 8, $(\frac{LWC}{N_d})^{\frac{1}{3}}$ decreases by 1.50×10^{-5} kg from 3.50×10^{-5} kg in the control run to 2.00×10^{-5} kg in the weak run, while it decreases by 9.80×10^{-6} kg from 1.03×10^{-4} kg in the low-aerosol run to 9.32×10^{-5} kg in the weak-low run. Associated with this, the average R_v shows a 47 % reduction from the control to weak runs, while it shows a 10 % reduction from the low-aerosol to weak-low runs during period 1 (Figure 8).

In summary, the simulated LWC, N_d, their variations with fire intensity, and the functional relation between LWC, N_d and R_v lead to a situation where R_v decreases much more among the polluted-scenario runs than among the clean-scenario runs during the period when the pyroCb initially forms.

b. Equilibrium supersaturation

During period 1, as fire intensity weakens and updraft speed decreases, parcel equilibrium supersaturation decreases and thus, the minimum size of activated aerosol particles increases not only among the clean-scenario runs but also among the polluted-scenario runs. When the production of parcel supersaturation by updrafts and the consumption of supersaturation by droplets balance out, parcel supersaturation reaches parcel equilibrium supersaturation (Rogers and Yau, 1991). Mostly due to greater aerosol concentrations, associated average equilibrium supersaturation and minimum size of activated aerosol particles over areas with positive updraft speeds and period 1 are lower and larger,

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respectively, in the polluted-scenario run than in the clean-scenario run for each fire intensity (Rogers and Yau, 1991).

The average equilibrium supersaturation decreases from 0.21% in the control run to 0.10% in the weak run. Associated with this, the average minimum diameter increases from 0.09 μm in the control run to 0.12 μm in the weak run over period 1. The average equilibrium supersaturation decreases from 0.55% in the low-aerosol run to 0.31% in the weak-low run, and the average minimum size increases from 0.04 μm in the low-aerosol run to 0.07 μm in the weak-low run over period 1.

The increase in the minimum-activation size with weakening fire intensity occurs closer to the right tail of the assumed unimodal aerosol size distribution among the polluted-scenario runs than among the clean-scenario runs. A smaller portion of the total aerosol concentration is in the size range closer to the right tail of the distribution as long as the range is on the right-hand side of the distribution peak where most of aerosol activation occurs. So, a similar increase in the average minimum-activation size for a weakened fire results in a smaller percentage reduction in the total activated aerosol concentration and thus N_d among the polluted-scenario runs during period 1. The average N_d over period 1 decreases by 8% from the control to weak runs. The average N_d decreases by 76% from the low-aerosol to weak-low runs (Figure 8). This contributes to the greater reduction in R_v as fire intensity weakens among the polluted-scenario runs during period 1. This is for a similar LWC between the polluted-scenario and clean-scenario runs for each fire intensity (Figure 8).

In association with larger aerosol concentration and the assumed aerosol size distribution, a smaller percentage variation of the number of activated aerosols and N_d with fire intensity is simulated in the polluted-scenario runs than in the clean-scenario runs. This smaller variation of N_d aids the greater reduction in R_v among the polluted-scenario runs.

4.2.3 Autoconversion, freezing, deposition and condensation

Autoconversion is proportional to the size of cloud droplets (Pruppacher and Klett, 1978; Rogers and Yau, 1991; Khairoutdinov and Kogan, 2000; Liu and Daum, 2004; Lee and Baik, 2017). Due to the larger R_v during period 1, the subsequent average autoconversion

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rates over period 2 are higher in the clean-scenario run than in the polluted-scenario run for each fire intensity (Figure 9a). Due to the larger absolute and percentage reduction in R_v among the polluted-scenario runs than among the clean-scenario runs with weakening fire intensity during period 2, the average autoconversion rates decrease by 74% (14%) from the control (low-aerosol) to weak (weak-low) runs (Figure 9a). Associated with this, differences in the average autoconversion rates between the polluted-scenario and clean-scenario runs increase as fire intensity weakens during period 2 (Figure 9a).

The increasing differences in autoconversion rates between the polluted-scenario and clean-scenario runs increase those differences in the amount of cloud liquid available for freezing with weakening fire intensity (Figure 9a). Thus, differences in the average rate of cloud-liquid freezing and freezing-related latent heat over the period 2 between the runs increase with weakening fire intensity (Figure 9a). Enhanced freezing-related latent heat strengthens updrafts in places where freezing occurs and this, in turn, enhances deposition and deposition-related latent heat (Lee et al., 2017). Although the average deposition over period 2 is slightly lower, those strengthened updrafts enable the average deposition and deposition-related latent heat to be greater in the polluted-scenario run than in the clean-scenario run for each fire intensity during period 3 (Figures 9a and 9b). Differences in the average freezing rate (and thus the average freezing-related latent heating) between the runs do not change much up to ~20:30 GMT (Figure 10). However, after ~20:30 GMT, these differences start to increase as time goes by for each fire intensity. This is because as convection intensifies, the transport of cloud liquid to places above the freezing level starts to be effective around 20:30 GMT. The greater freezing and thus freezing-related latent heat eventually cause updrafts to be stronger in the polluted-scenario run starting at ~21:00 GMT (Figure 10). Then, the stronger updrafts induce deposition to be greater in the polluted-scenario run around 21:10 GMT (Figure 10). Note that deposition-related latent heat is about one order of magnitude greater than freezing-related latent heat for a unit mass of hydrometeors involved in phase-transition processes. This contributes to much greater differences in deposition-related latent heat during period 3 than those in freezing-related latent heat between the runs during period 2 or 3 (Figures 9a and 9b).

To satisfy mass conservation, the freezing- and deposition-enhanced updrafts above the freezing level induce more updraft mass fluxes below the freezing level in the polluted-

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As shown in Lee et al. (2017), …e…hanced freezing-related latent heat strengthens updrafts in places where freezing occurs and this, in turn, enhances deposition and deposition-related latent heat (Lee et al., 2017). Hence, …a…though the average deposition , which is …ver averaged in cloudy areas and …eriod 2,…is slightly lower, those strengthened updrafts enable due to those strengthened updrafts, …he averaged…erage deposition and deposition-related latent heat to beare…greater in the polluted-scenario run than in the clean-scenario run for each fire intensity during period 3 (Figures 9a and 9b). Differences in the averaged…erage freezing rate (and thus the averaged…erage freezing-related latent heating) in cloudy areas between the polluted-scenario run and the clean-scenario …uns for each fire intensity …o not change much up to ~20:30 GMT after start to appear around 18:30 GMT …Figure 10). However, after ~20:30 GMT, these differences start to increase as time goes by for each fire intensity. This is because as convection intensifies, the ... [54]

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Due to the increasing differences in freezing-related latent heat between the polluted-scenario and clean-scenario runs with weakening fire intensity during period 2, those differences in the average freezing-affected updrafts and subsequently in deposition-related latent heat over period 3 increase with weakening fire intensity (Figures 9a, 9b and 10). Those differences, calculated using Equation (1), in deposition-related latent heat are 16%, 181%, and 417 % for strong, medium, and weak fire intensities, respectively (Figures 9b and 10). Since percentage increases in deposition-related latent heat increase, the subsequent percentage increases in updrafts in the polluted-scenario run increase with weakening fire intensity, particularly during period 3 (Figure 10). During period 4, due to these greater increases in updrafts in the polluted-scenario run with weaker fire intensity, the percentage increases in condensation in the polluted-scenario run increase with weakening fire intensity (Figures 9c and 10). The greater increases in condensation cause the greater further enhancement of the increases in updrafts in the polluted-scenario run with weaker fire intensity. This leads to the overall greater effects of fire-produced aerosols on the UTLS water vapor and ice with weaker fire intensity.

This section shows that the smaller R_v leads to lower autoconversion rates and a larger amount of cloud liquid as a source of freezing, which in turn induces higher freezing rates and stronger feedbacks between freezing, deposition, condensation and updrafts in the polluted-scenario run than in the clean-scenario run for each fire intensity. This results in stronger updrafts and their impacts on the UTLS water vapor and ice in the polluted-scenario run. The greater R_v reduction among the polluted-scenario runs than among the

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clean-scenario runs with weakening fire intensity increases the differences in autoconversion, freezing and the feedbacks between the polluted-scenario and clean-scenario runs as fire intensity weakens. This results in the greater impacts of aerosol-induced stronger updrafts on the UTLS water vapor and ice with weaker fire intensity.

4.3 Dependence of aerosol effects on the magnitude of aerosol perturbation

Table 3 shows that for each of the strong-, medium-, and weak-fire cases, there are increases in the UTLS water-vapor and cirrus-cloud mass in the run with fire-induced aerosol perturbations of 30000 or 7500 cm^{-3}. These increases are relative to the mass in the low-aerosol run for the strong-fire case, in the medium-low run for the medium-fire case, and in the weak-low run for the weak-fire case. Note that for each of the three types of fire-induced aerosol perturbations of 30000, 15000 and 7500 cm^{-3}, aerosol-perturbation-induced percentage increases in the UTLS water-vapor and cirrus-cloud mass increase as fire intensity weakens (Tables 2 and 3). The qualitative nature of results regarding the dependence of the percentage increases in the UTLS water-vapor and cirrus-cloud mass on fire intensity thus does not depend on the magnitude of the fire-induced aerosol perturbation.

Until now, we have taken interest in the sensitivity to fire intensity of an aerosol perturbation on pyroCb development, the UTLS water vapor and cirrus clouds. To isolate the sensitivity, we have shown comparisons among sensitivity simulations by varying only fire intensity while maintaining a constant aerosol perturbation. While working well for the isolation aspect, this strategy does not reflect reality well. It may be that weaker fire intensity produces a lower aerosol concentration. This possibility is not that unrealistic, since stronger fires likely involve more material burnt and higher aerosol emissions.

With this in mind, we make comparisons among three pairs of simulations: the low-aerosol and control-30000 runs for strong fire vs. the medium-low and medium runs for medium fire vs. the weak-low and weak-7500 runs for weak fire. Among these three pairs, the magnitude of the fire-induced aerosol perturbation decreases with weakening fire, emulating the possibility that weaker fire intensity involves a lower amount of aerosols. The perturbation-related aerosol concentration is 30000 cm^{-3} for strong fire, 15000 cm^{-3} for

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medium fire, and 7500 cm^{-3} for weak fire. Comparisons among these three pairs show that relative importance of aerosol effects on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds increases for weaker fires, and that it does not matter if the aerosol perturbation decreases or stays constant with weakening fire intensity (Tables 2 and 3). In these comparisons, it is also possible that when the fire-induced aerosol perturbation is very low for medium or weak fire intensity, the latent heat perturbation by aerosol perturbation can be very low. This very low latent heat is not large enough to increase the relative importance of those aerosol effects with weakening fire intensity. Based on this, the medium run is repeated with a fire-induced aerosol perturbation of 2000 cm^{-3} down from 15000 cm^{-3} (the medium-2000 run). The weak run is repeated with a fire-induced aerosol perturbation of 1000 cm^{-3} down from 7500 cm^{-3} (the weak-1000 run). The percentage increases in the UTLS water-vapor and cirrus-cloud mass from the medium-low to medium-2000 runs or from the weak-low to weak-1000 runs are smaller than the increases from the low-aerosol to control-30000 runs for the case of strong fire. This indicates that when the fire-induced aerosol perturbation decreases too much with weakening fire intensity, the relative importance of aerosol effects on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds no longer increases with weakening fire intensity.

Results in this section show that the increasing impacts of the fire-induced aerosol perturbations on the UTLS water vapor and cirrus clouds with weakening fire intensity are robust whether or not aerosol perturbations vary with fire intensity unless their variation is extremely high.

5. Conclusions

This study investigates an observed case of a pyroCb using a modeling framework. In particular, this study focuses on effects of fire-produced aerosols on pyroCb development and its impacts on the UTLS water vapor and cirrus clouds. Results show that the pyroCb efficiently transports water vapor to the tropopause and above. This leads to a much greater amount of water vapor around and above the tropopause (i.e., the UTLS) over the pyroCb compared to that outside the pyroCb. The pyroCb also generates a deck of cirrus clouds

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around the tropopause. The role of fire-produced aerosols or the fire-induced aerosol perturbation in the water-vapor transport to the UTLS and the production of cirrus clouds becomes significant as fire intensity weakens.

During the initial stage, there is a similar LWC between the polluted-scenario and clean-scenario runs for each fire intensity. The reduction in LWC with weakening fire intensity among the polluted-scenario runs is similar to that among the clean-scenario runs. Much greater N_d is in the polluted-scenario run than in the clean-scenario run for each fire intensity. N_d decreases are smaller among the polluted-scenario runs than among the clean-scenario runs with weakening fire intensity. This situation during the initial stage induces R_v to decrease much more among the polluted-scenario runs with weakening fire intensity. This reduces autoconversion more among the polluted-scenario runs and increases differences in autoconversion between the polluted-scenario and clean-scenario runs as fire intensity weakens. The increasing differences in autoconversion between the runs cause greater differences in freezing-related latent heat as fire intensity weakens. Through feedback between freezing, deposition, updrafts, and condensation, differences in freezing-related latent heat induce those in updrafts between the runs. Those greater differences in freezing-related latent heat lead to greater differences in updrafts, producing the greater differences in the UTLS water vapor and cirrus clouds between the runs with weaker fire intensity. This means that the role of fire-produced aerosols in the water-vapor transport to the UTLS and the production of cirrus clouds becomes more significant as fire intensity weakens. This more significant role of fire-produced aerosols with weaker fire intensity is robust to the magnitude of a given fire-induced aerosol perturbation, and the variation of the fire-induced aerosol perturbation with fire intensity unless the variation is very high.

The level of understanding of the role played by fire-produced aerosols in the development of pyroCbs and their impacts on the UTLS water vapor and cirrus clouds has been low. This study shows that fire-produced aerosols can invigorate updrafts and convection and thus enhance the transport of water vapor to the UTLS and the formation of cirrus clouds. We find that the mechanism that controls the invigoration of convection by aerosols in the pyroCb is consistent with the traditional invigoration mechanism proposed and described by Koren et al. (2008) and Rosenfeld et al. (2008). However, this study shows that for pyroCbs produced by strong fires, the aerosol-induced invigoration

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…hise…more significant role of fire-produced aerosols in water-vapor transport to the UTLS and the production of cirrus cloud in the pyroCb …ith weaker fire intensity is robust to the magnitude of athe…given fire-induced aerosol perturbation, which was assumed not to vary with varying fire intensity…and . This more significant role with weaker fire intensity is also robust to …he variation of the fire-induced aerosol perturbation with the varying ... [60]

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and its effects on the UTLS water vapor and cirrus clouds are insignificant. Note that traditional understanding generally focuses on effects of fire-produced heat and water vapor and their associated surface fluxes on the pyroCb and does not consider effects of fire-produced aerosols on the pyroCb. This understanding adequately explains the mechanics for pyroCbs in association with strong fires. This study suggests that when pyroCbs form over weak-intensity fires, those effects of fire-produced aerosols require consideration.

Note that when fire-induced aerosol perturbations are strongly reduced for cases of weaker-intensity fires compared with strong-intensity fires, the significance of the role of the fire-produced aerosol perturbation no longer increases and starts to decrease with weakening fire. This suggests that there may be a critical level of aerosol perturbation below which the increase in this significance with weakening fire intensity ceases.

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Author contributions

SSL generated the research ideas and goals, performed the simulations, and wrote the manuscript. GK and ZL selected the case, analyzed observations, and provided data to set up the simulations while reviewing and providing comments on the manuscript. CHJ, YSC, JU and WJC revised the manuscript based on the reviewers' comments and performed associated analyses of simulation and observation data.

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Acknowledgements

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FIGURE CAPTIONS

Figure 1. Visible image of the fire, smoke and cirrus cloud in association with the selected pyroCb. This image was taken by the visible infrared imaging radiometer suite onboard the Suomi spacecraft. Bright white represents cirrus (anvil) at the top of the pyroCb, and the red circle marks the fire spot. Dark white represents smoke produced by the fire. Adapted from Kablick et al. (2018).

Figure 2. The simulated fire spot (red circle) and the field of cloud-ice mass density (cirrus cloud) at the top of the simulated pyroCb when it is about to enter its mature stage.

Figure 3. The vertical distribution of the radar reflectivity averaged along the Cloudsat path.

Figure 4. Vertical distributions of the average updraft mass fluxes at all altitudes in cloudy areas (i.e., where the sum of LWC and IWC is non-zero) over the simulation period between 17:00 GMT on August 5^{th} and 12:00 GMT on August 6^{th}.

Figure 5. Vertical distributions of the average water-vapor mass density at altitudes above 13 km and over the simulation period between 17:00 GMT on August 5^{th} and 12:00 GMT on August 6^{th}. Colored lines represent the average values over cloudy grid columns (non-zero sum of LWP and IWP). The black line represents those values over non-cloudy columns (zero sum of LWP and IWP) in the control run.

Figure 6. Vertical distributions of the average cloud-ice mass density at all altitudes in cloudy areas (non-zero sum of LWC and IWC) over the simulation period between 17:00 GMT on August 5^{th} and 12:00 GMT on August 6^{th}.

Figure 7. Same as Figure 6 but for deposition rate.

Figure 8. Average N_d, R_v, and LWC at all altitudes in cloudy areas over the period between 17:00 and 19:00 GMT on August 5th.

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Figure 9. Average rates of condensation, deposition and cloud-liquid freezing at all altitudes in cloudy areas and over periods (a) 2, (b) 3 and (c) 4. In panel (a), average autoconversion rates are also shown.

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Figure 10. Time series of differences in average values of variables related to aerosol-induced invigoration of convection at all altitudes in cloudy areas between the (a) control and low-aerosol runs for strong fire intensity, (b) medium and medium-low runs for medium fire intensity and (c) weak and weak-low runs for weak fire intensity.

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Simulations	Surface sensible heat fluxes in the fire spot (W m^{-2})	Surface latent heat fluxes in the fire spot (W m^{-2})	Aerosol concentration in the PBL over the fire spot (cm^{-3})
Control run	15000	1800	15000
Low-aerosol run	15000	1800	150
Control-30000	15000	1800	30000
Control-7500	15000	1800	7500
Medium run	7500	900	15000
Medium-low run	7500	900	150
Medium-30000	7500	900	30000
Medium-7500	7500	900	7500
Medium-2000	7500	900	2000
Weak run	3750	450	15000
Weak-low run	3750	450	150
Weak-30000	3750	450	30000
Weak-7500	3750	450	7500
Weak-1000	3750	450	1000

Table 1. Summary of simulations

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Backg-round ... [62]

	Backg-round	Control	Low-aerosol	Differe-nce (%)	Medium	Medium-low	Differe-nce (%)	Weak	Weak-low	Differe-nce (%)
Updraft mass fluxes (kg m^{-2} s^{-1})		1.23	1.19	3	0.89	0.70	27	0.42	0.21	100
Water-vapor mass density between 13 and 16 km (10^{-3} g m^{-3})	0.46	2.31	2.26	2	1.61	1.32	22	0.93	0.58	60
Cirrus-cloud mass density between 9 and 13 km (g m^{-3})		0.024	0.023	4	0.017	0.012	42	0.008	0.004	100

Table 2. Average updraft mass fluxes at all altitudes in cloudy areas, the average water-vapor mass density over altitudes between 13 and 16 km and over cloudy columns except for the average background water-vapor mass density which is also over altitudes between 13 and 16 km but over non-cloudy columns, and the average cirrus-cloud mass density between 9 and 13 km in cloudy areas. 16 km is the altitude to which the non-zero water-vapor mass density over cloudy columns extends (Figure 5). These average values are obtained over the simulation period between 17:00 GMT on August 5th and 12:00 GMT on August 6th. "Difference" is the percentage difference between the polluted-scenario and clean-scenario runs for each fire intensity ($\frac{Polluted-scenario\ run\ minus\ clean-scenario\ run}{Clean-scenario\ run} \times$ 100 (%)).

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	Control-30000	Control-7500	Medium-30000	Medium-7500	Medium-2000	Weak-30000	Weak-7500	Weak-1000
Water vapor mass density between 13 and 16 km (10^{-3} g m^{-3})	2.38 (5%)	2.28 (0.9%)	1.87 (42%)	1.50 (14%)	1.36 (3%)	1.31 (125%)	0.75 (29%)	0.60 (3%)
Cirrus cloud mass density between 9 and 13 km (g m^{-3})	0.025 (9%)	0.023 (0.2%)	0.023 (92%)	0.014 (17%)	0.012 (3%)	0.013 (225%)	0.006 (50%)	0.004 (8%)

Table 3. Average water-vapor mass density between 13 and 16 km over cloudy columns and the average cirrus-cloud mass density between 9 and 13 km in cloudy areas over the simulation period between 17:00 GMT on August 5th and 12:00 GMT on August 6th. The numbers in parentheses are the percentage differences: $\frac{Control-30000\ (or\ control-7500)\ run\ minus\ low-aerosol\ run}{low-aerosol\ run} \times 100$ (%) for strong fire intensity, $\frac{Medium-30000\ (or\ medium-7500\ or\ medium-2000)\ run\ minus\ medium-low\ run}{Medium-low\ run} \times 100$ (%) for medium fire intensity, and $\frac{Weak-30000\ (or\ weak-7500\ or\ weak-1000)\ run\ minus\ weak-low\ run}{Weak-low\ run} \times 100$ (%) for weak fire intensity.

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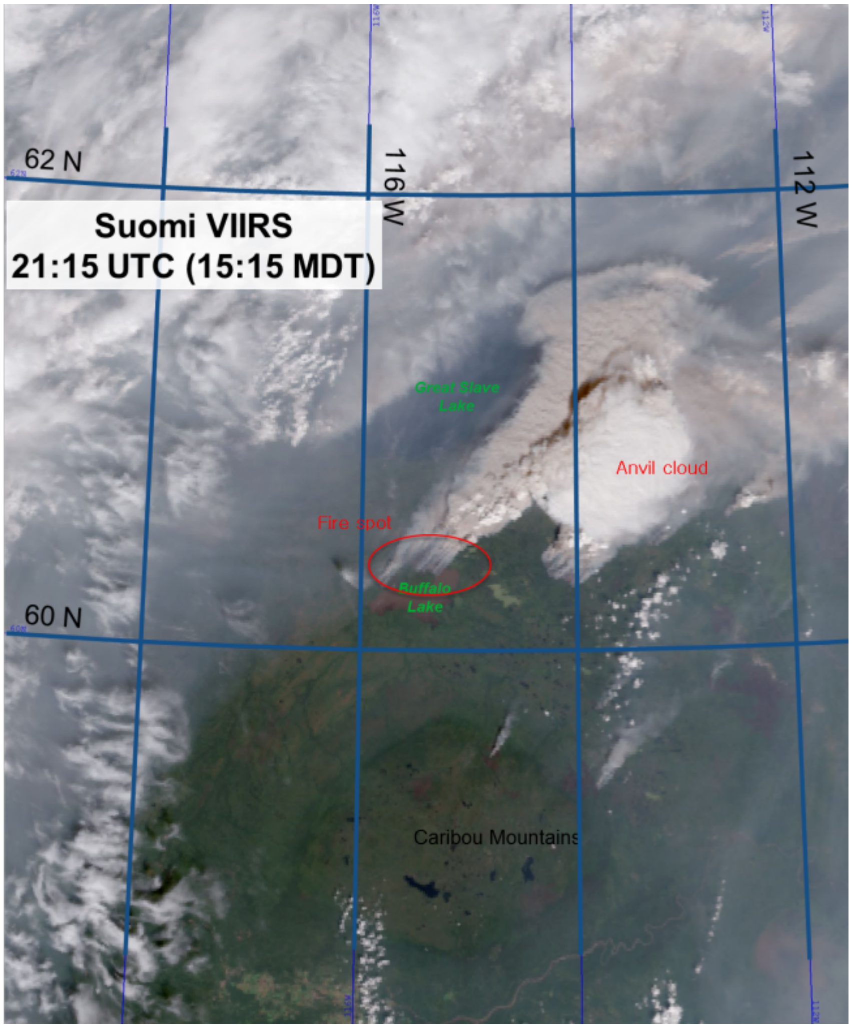
62 N
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Suomi VIIRS
21:15 UTC (15:15 MDT)
Great Slave Lake
Anvil cloud
Fire spot
Buffalo Lake
60 N
Caribou Mountains

Figure 1

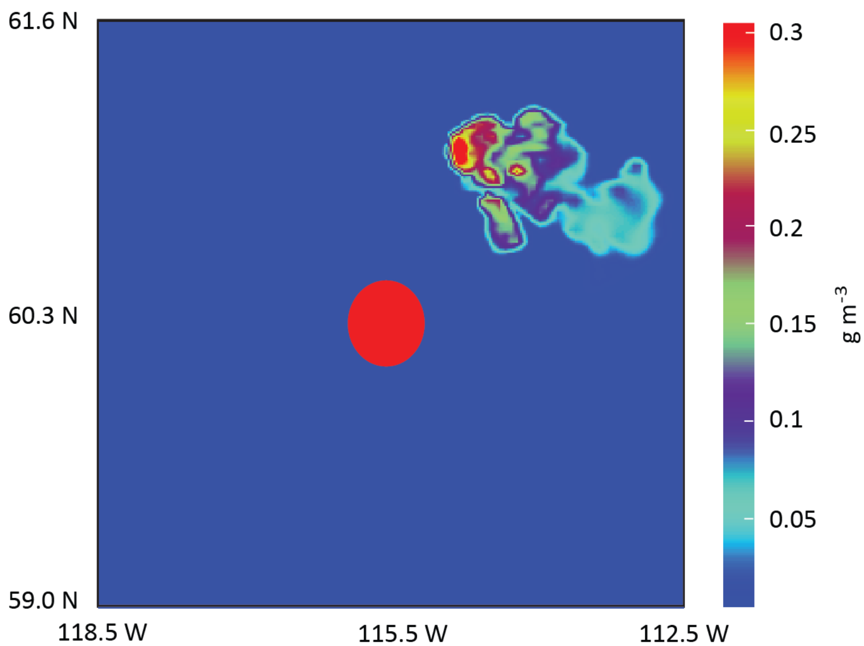
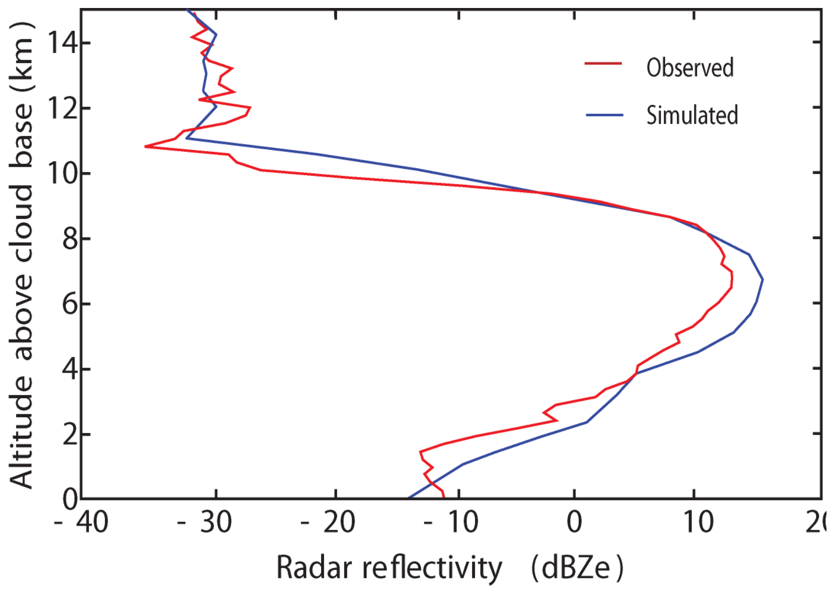


Figure 2


Observed
Simulated
Altitude above cloud base (km)
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- 40
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- 20
- 10
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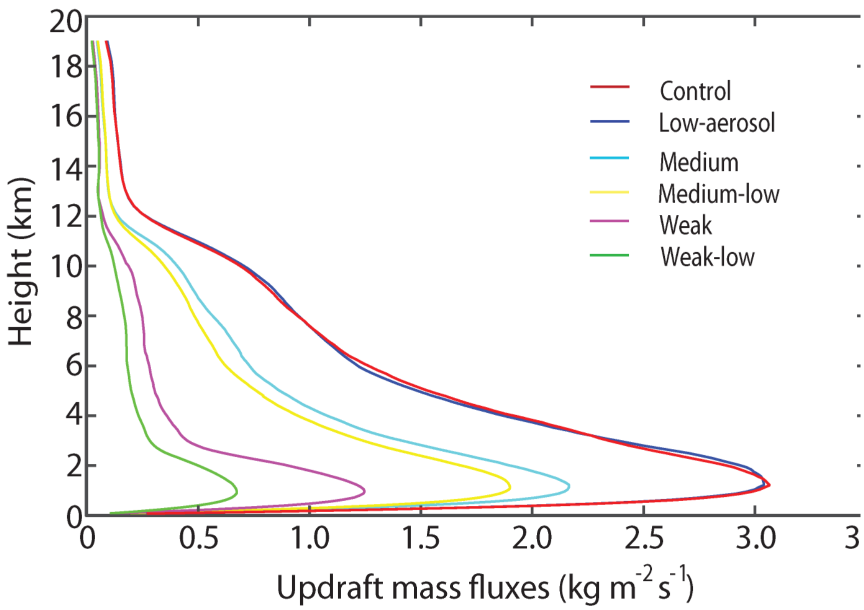
Radar reflectivity (dBZe)

Figure 3


Control
Low-aerosol
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Medium-low
Weak
Weak-low
Height (km)
0
2
4
6
8
10
12
14
16
18
20
0
0.5
1.0
1.5
2.0
2.5
3.0
3
Updraft mass fluxes (kg m-2 s-1)

Figure 4

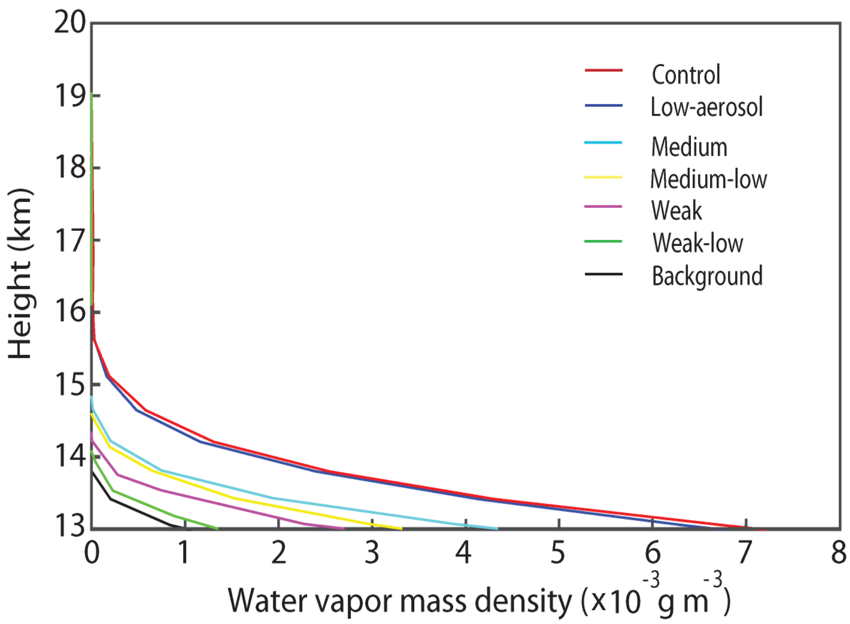

Control
Low-aerosol
Medium
Medium-low
Weak
Weak-low
Background
Height (km)
Water vapor mass density (x10^-3 g m^-3)
20
19
18
17
16
15
14
13
0
1
2
3
4
5
6
7
8

Figure 5

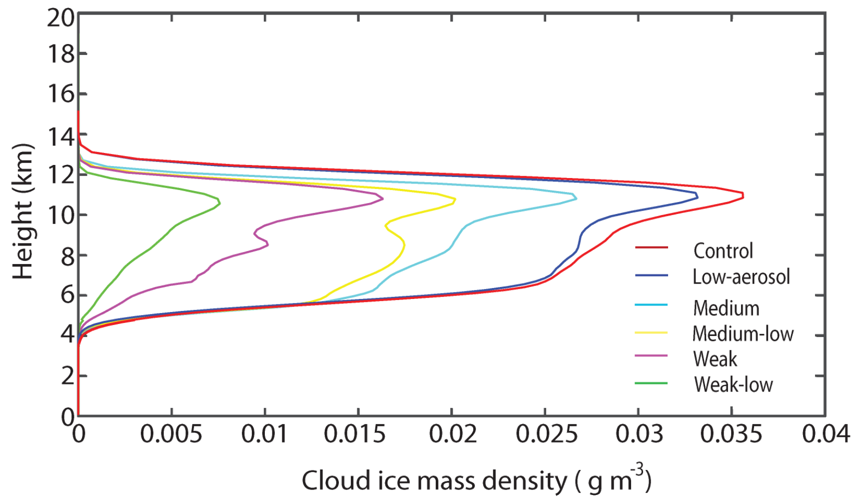

Height (km)
Cloud ice mass density (g m-3)
Control
Low-aerosol
Medium
Medium-low
Weak
Weak-low
0
2
4
6
8
10
12
14
16
18
20
0
0.005
0.01
0.015
0.02
0.025
0.03
0.035
0.04

Figure 6

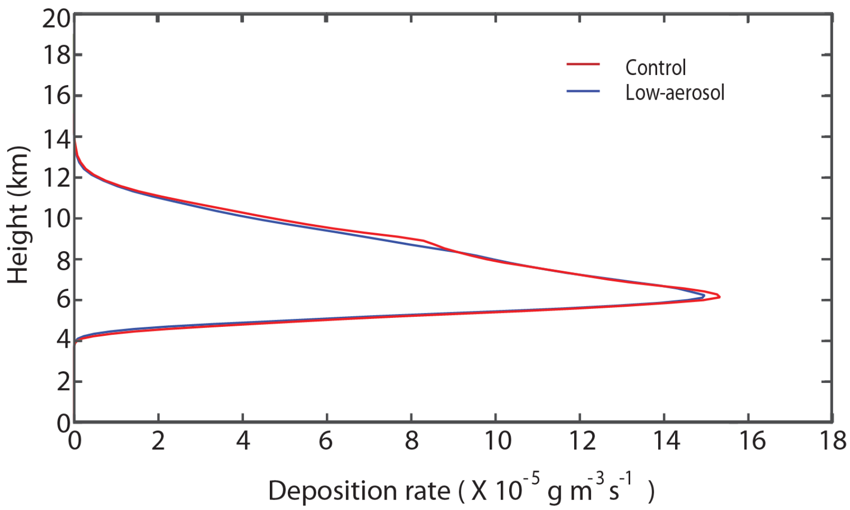
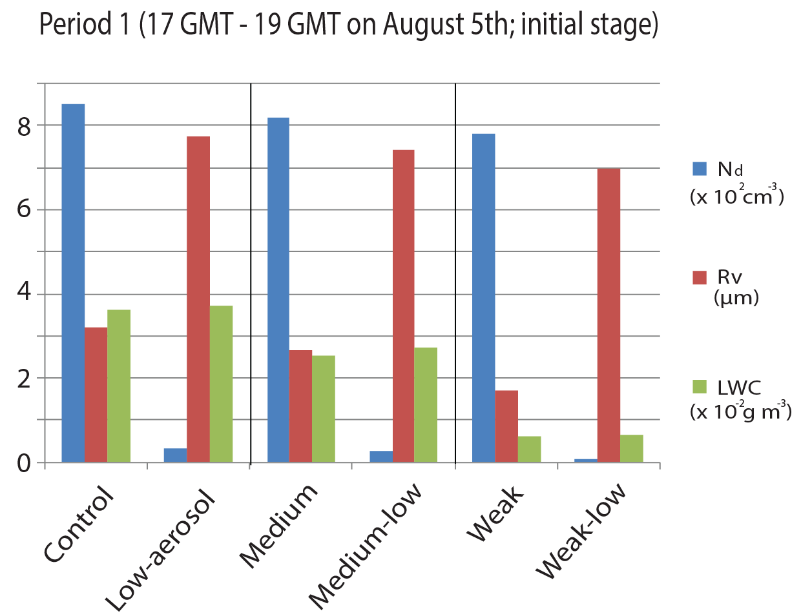
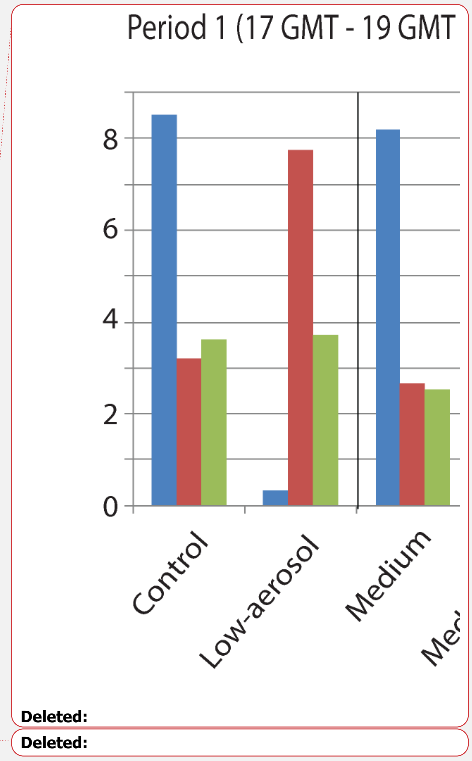

Control
Low-aerosol
Height (km)
Deposition rate (X 10^{-5} g m^{-3} s^{-1})
0
2
4
6
8
10
12
14
16
18
20

Figure 7



Period 1 (17 GMT - 19 GMT on August 5th; initial stage)
8
6
4
2
0
Nd
(x 10²cm⁻³)
Rv
(μm)
LWC
(x 10⁻²g m⁻³)
Control
Low-aerosol
Medium
Medium-low
Weak
Weak-low

Figure 8



Period 1 (17 GMT - 19 GMT
8
6
4
2
0
Control
Low-aerosol
Medium
Deleted:
Deleted:

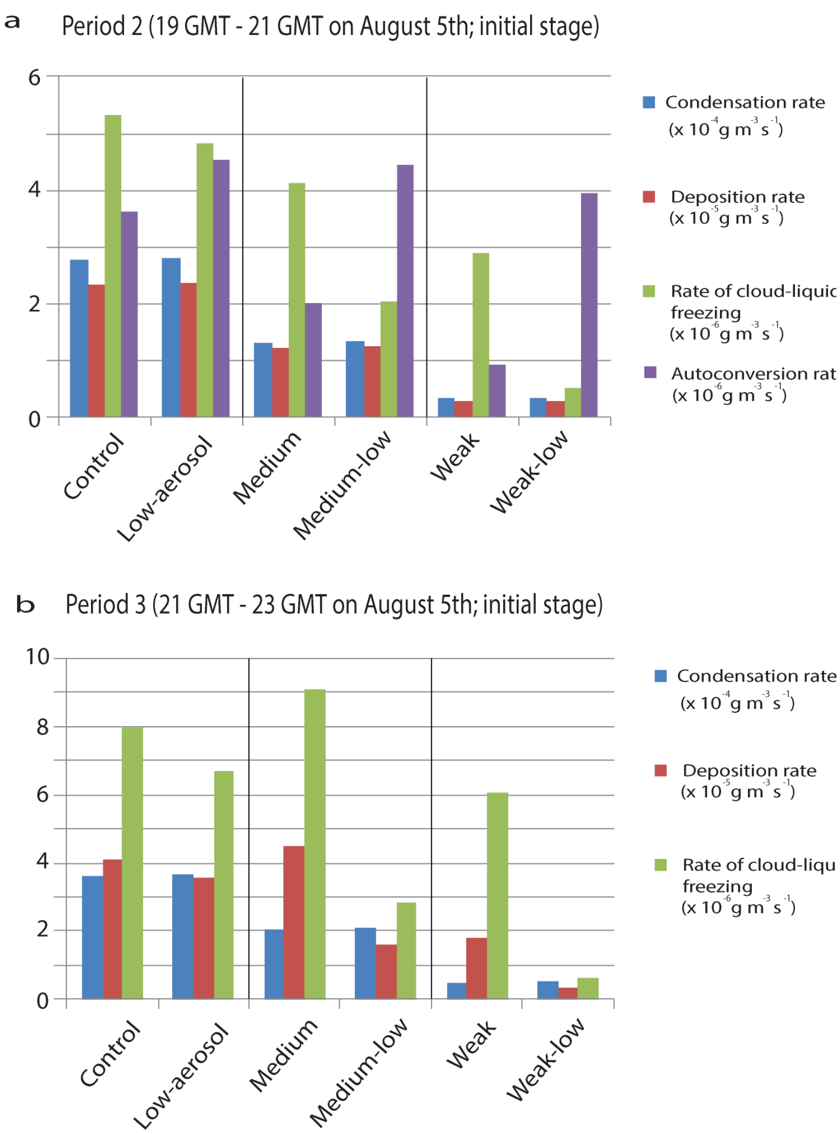

a
Period 2 (19 GMT - 21 GMT on August 5th; initial stage)
6
4
2
0
Control
Low-aerosol
Medium
Medium-low
Weak
Weak-low
Condensation rate
(x 10-4 g m-3 s-1)
Deposition rate
(x 10-5 g m-3 s-1)
Rate of cloud-liquic
freezing
(x 10-6 g m-3 s-1)
Autoconversion rat
(x 10-6 g m-3 s-1)
b
Period 3 (21 GMT - 23 GMT on August 5th; initial stage)
10
8
6
4
2
0
Control
Low-aerosol
Medium
Medium-low
Weak
Weak-low
Condensation rate
(x 10-4 g m-3 s-1)
Deposition rate
(x 10-5 g m-3 s-1)
Rate of cloud-liqu
freezing
(x 10-6 g m-3 s-1)

Figure 9

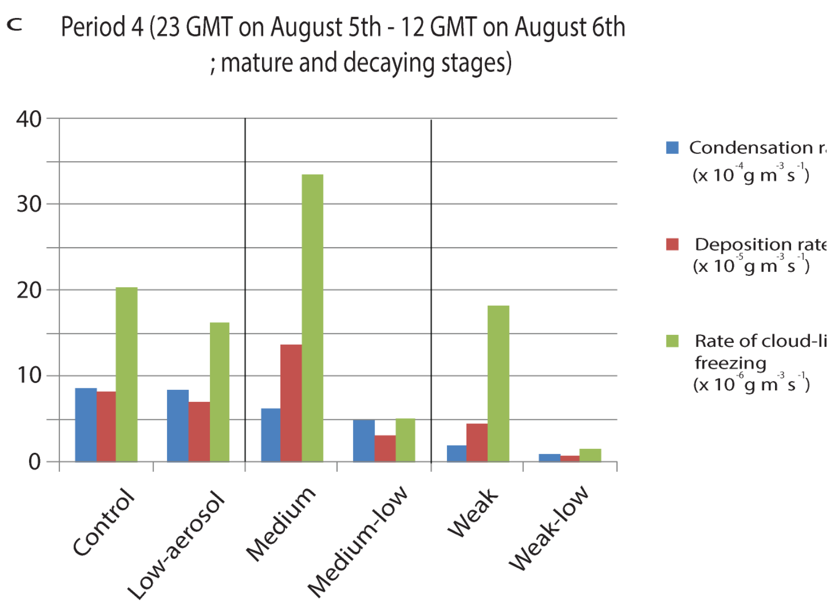

c
Period 4 (23 GMT on August 5th - 12 GMT on August 6th ; mature and decaying stages)
40
30
20
10
0
Condensation r
(x 10^{-4} g m^{-3} s^{-1})
Deposition rate
(x 10^{-5} g m^{-3} s^{-1})
Rate of cloud-li
freezing
(x 10^{-6} g m^{-3} s^{-1})
Control
Low-aerosol
Medium
Medium-low
Weak
Weak-low

Figure 9

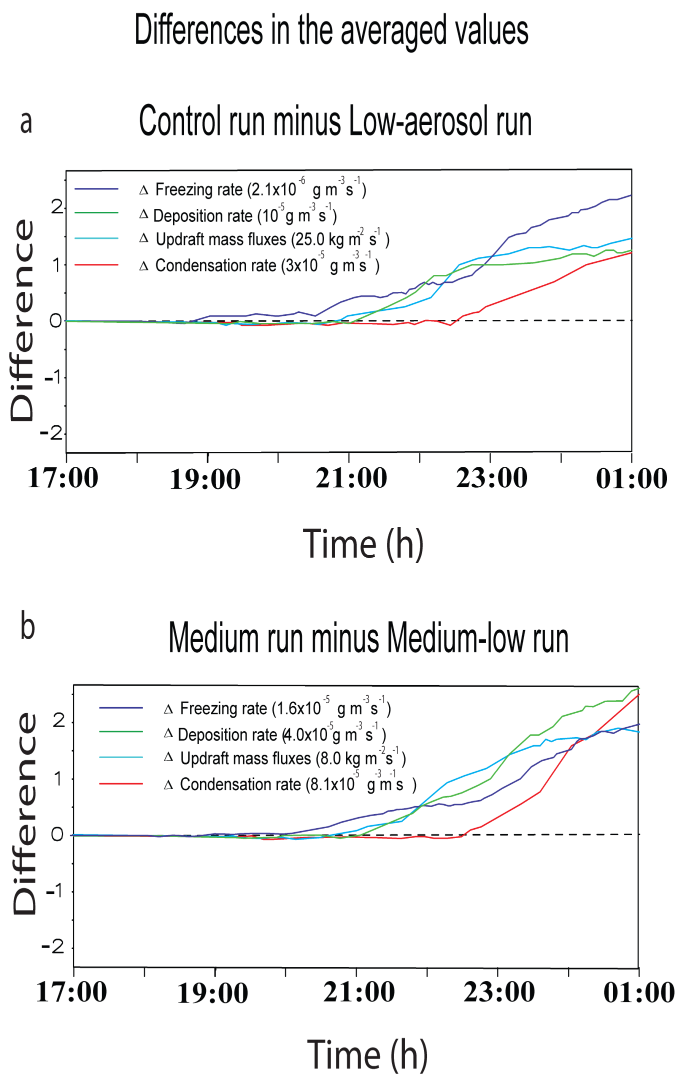

Differences in the averaged values
a
Control run minus Low-aerosol run
Δ Freezing rate (2.1x10^{-6} g m^{-3} s^{-1})
Δ Deposition rate (10^{-5} g m^{-3} s^{-1})
Δ Updraft mass fluxes (25.0 kg m^{-2} s^{-1})
Δ Condensation rate (3x10^{-5} g m^{-3} s^{-1})
Difference
2
1
0
-1
-2
17:00
19:00
21:00
23:00
01:00
Time (h)
b
Medium run minus Medium-low run
Δ Freezing rate (1.6x10^{-5} g m^{-3} s^{-1})
Δ Deposition rate (4.0x10^{-5} g m^{-3} s^{-1})
Δ Updraft mass fluxes (8.0 kg m^{-2} s^{-1})
Δ Condensation rate (8.1x10^{-5} g m^{-3} s^{-1})
Difference
2
1
0
-1
-2
17:00
19:00
21:00
23:00
01:00
Time (h)

Figure 10

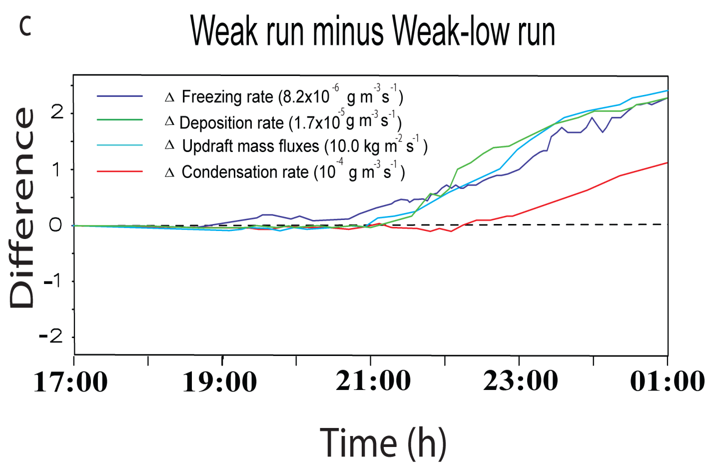

c
Weak run minus Weak-low run
Δ Freezing rate (8.2x10-6 g m-3 s-1)
Δ Deposition rate (1.7x10-5 g m-3 s-1)
Δ Updraft mass fluxes (10.0 kg m-2 s-1)
Δ Condensation rate (10-4 g m-3 s-1)
Difference
2
1
0
-1
-2
17:00
19:00
21:00
23:00
01:00
Time (h)

Figure 10

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