Response from authors

Re: Review of Improved estimates of smoke exposure during Australia fire seasons: Importance of quantifying plume injection heights

January 20, 2024

Referee comment 1

General comment:

Most of my remarks have been properly addressed by the authors, and, in my opinion, the manuscript has improved.

The discussion on the issues arising from assuming that fire emissions injected above the Planetary Boundary Layer (PBL) do not affect surface smoke concentrations is essential. Addressing this issue must be a priority in future work.

We thank the reviewer for these helpful comments. We respond to each specific comment in details below. The referee comments are shown in red. Our replies are shown in black and modified text is shown in blue. The annotated page and line numbers refer to the revised copy of the manuscript.

Specific comments

1. ll 362-363: "The large bias is mainly due to the underestimates of .. such as those in 2019." I recommend adding a sentence here discussing the high model bias for small fire emissions. However, I acknowledge that they may not significantly contribute to the overall (negative) bias.

   We have discussed the possible reason for overestimated injection fractions (see details in Line 357-361) in northern and central Australia, which also leads to the high biases in injected emission fluxes with small fire emissions. We have added a sentence to clarify this.

P13, Line 374-376, Section 3.2

For low fire emissions, the climatological method shows high biases in injected emission fluxes above the PBL due to inaccurate climatological plume profiles in north and central of Australia.

2. ll 458-460: "However, there are also large bias ... Albury"
According to Table S1 the biases are also large in Newcastle and Canberra.

We have now implemented the discussion on the low biases in Newcastle (Wallsend) and Canberra. We also clarify this issue in Section 5.

P16, Line 470-472, Section 4.1

However, there are large biases in simulated total $PM_{2.5}$ concentrations from the INJ-RF experiment compared to the observations in Gladstone, Brisbane, Wollongong, Canberra, Newcastle (Wallsend), and Albury.

P16, Line 477-479, Section 4.1

In Brisbane, Canberra, and Newcastle (Wallsend), the low biases are relatively significant during the high-fire years of 2009 and 2019. We speculate that these biases arise from neglect in our model setup from downward mixing of smoke plumes in remote regions (Section 2.3.2).

P22, Line 659-667, Section 5

In Sydney and Newcastle, these two methods generate surface concentrations in better agreement with observations than the control simulation, with NMBs of -4.5% (INJ-RF) to -7.0% (INJ-CLIM) in Sydney and 6.6% (INJ-RF) to 25.5% (INJ-CLIM) in Newcastle.

However, neither method can quantify the possible downward mixing of fire smoke plumes in downwind regions and the subsequent impact on surface air, a process which may be especially important during more severe fire seasons when intense heat lofts greater quantities of smoke above the PBL in source regions. In Brisbane, Gladstone, and Melbourne, the INJ-RF method leads to more pronounced underestimates of surface $PM_{2.5}$ concentrations compared to INJ-CLIM, perhaps because of this shortcoming.

3. ll 478-480: "In particular ... November to January." The agreement is less evident in Sydney, as indicated by comparable statistical metrics between INJ-CLIM and INJ-RF in Figure 7.

   The statistical metrics in Figure 7 are based on the whole fire seasons from August to January in the following year, which yield a smaller RMSE and a higher correlation coefficient in Sydney. During the peak time (November to mid-December) of the fire season in 2019, the NMBs for INJ-CLIM and INJ-RF are 12% and -5.0% in Sydney. In Newcastle, the NMBs for INJ-CLIM and INJ-RF are 77% and 31%. We have clarified this in Section 4.1

   P17, Line 491-494, Section 4.1

   In particular, compared to results from the INJ-CLIM experiment, the peak values of total $PM_{2.5}$ simulated by INJ-RF experiment agree best with observations in Newcastle and Sydney during the megafires of November to Mid-December, with the lowest NMBs of 31% and -5.0%.

4. Table S2: I agree that the three model experiments successfully reproduce the time series of daily $PM_{2.5}$ at most receptor cities, which is encouraging.
   However, I am concerned about the lack of a clear improvement in INJ-RF compared to INJ-CLIM in many receptors. A brief explanation or discussion about this point would be helpful.

   Thank you for this suggestion. We agree that the improvement in INJ-RF compared to INJ-CLIM are not significant in some receptors during the fire seasons over the last decade. We have added the explanation in Section 4.1.

P17, Line 508-513, Section 4.1

Compared to INJ-CLIM, INJ-RF yields higher correlation coefficients and smaller RMSEs at most receptors, indicating that INJ-RF better captures the daily variability and peak values of total $PM_{2.5}$ concentrations during the fire seasons. However, INJ-RF improves the NMBs only in Darwin, Sydney (Liverpool), Melbourne (Alphington), and Newcastle. In other receptors, the total $PM_{2.5}$ concentrations are more underestimated in the INJ-RF experiment than in INJ-CLIM, possibly due in part to the neglect in our model setup of downward mixing of smoke in remote regions.

Referee comment 2

The authors implemented many of the reviewers' suggestions, and I am really impressed with the improvements made to this manuscript.

We thank the reviewer for the helpful comments and questions. Please refer to the revised manuscript with tracked changes. We respond to each specific comment in details below. The referee comments are shown in red. Our replies are shown in black and modified text is shown in blue. The annotated page and line numbers refer to the revised copy of the manuscript.

I have a few questions after seeing the new Figure S1. There are relatively few MISR plume records in the Southeast box (maybe ~150). How many of these were in the training/testing dataset? The "Top End" has many more and is therefore what the RF model is really trained on. What are the statistics if the results are separated for the two different regions? The seasonality for the two regions differ, and I wonder if the drivers would differ if the model was trained separately for the two regions (although there is not a large enough sample size to actually do this effectively for the Southeast). This also creates a bit of a mismatch in that the surface measurement sites are primarily in the Southeast box. It would be good to mention these limitations as it may also explain why the comparisons with the surface concentrations are not much better.

The reviewer makes a good point. We acknowledge that the RF model is better trained on in northern Australia as the MISR plume records are relatively adequate in this region. In southeastern region, the fire season is shorter and there are only 254 MISR plume records. The training data consist of 244 records, and the testing data consist of 10 records. In the northern region, the correlation between the predicted and observed injected emission fluxes above the PBL is 0.7, the NMB is 7.3%, and the RMSE is 1.6×10^{-10} kg m^{-2} s^{-1}. In the southeast region the correlation is 0.97, the NMB is 12%, and the RMSE is 6.1×10^{-10} kg m^{-2} s^{-1}, showing nearly as good agreement with MISR observations as in the north. We have discussed the limitations of the MISR dataset in the discussion and conclusion section. We have also added a new paragraph to Section 5, in which we highlight two of the main limitation of the RF model.

P10, Line 264-268, Section 2.2.3

A shortcoming of our machine-learning approach is that the MISR dataset used for our study includes relatively few plumes in southeastern Australia compared to northern Australia (Figure S1). The fire season is shorter in this region, and there is much greater interannual variability in fire activity. As a consequence, we have available only 244 training records in the southeast and only 10 for testing there, compared to 1447 records for training and 152 records for testing in the north. We further discuss this limitation in Section 5.

P22-23, Line 674-682, Section 5

The machine learning approach (INJ-RF) has two main limitations. First, the relatively short fire season and interannual variability of fire activity in southeastern Australia means that fewer MISR records are currently available to train and test the INJ-RF model. Digitizing more smoke plume records from MISR, a laborious process, could enhance the training and testing of the INJ-RF model. Future studies could then train the random forest models separately in the two regions – northern and southeastern Australia – and identify the drivers for each region. Second, as noted above, the STILT model cannot capture downward mixing of smoke away from source regions. Future studies could explore the impacts of long-range transport and downward mixing of fire emissions on surface smoke concentrations by applying the estimated injection fractions to 3-D chemical transport models.

A couple other revisions:

1. Line 35: Change to "also leads to better model agreement". I think this should just be softened as that isn't clear in all the metrics at any of the sites.

   Changed as suggested. Thank you.

   P2, Line 35-37, Abstract

   Using the plume behavior predicted by the random forest method also leads to better model agreement with observed surface $PM_{2.5}$ in several key cities near the wildfire source regions, with smoke $PM_{2.5}$ accounting for 5% to 52% of total $PM_{2.5}$ during fire seasons from 2009 to 2020.

2. Line 53-55: Change to "Smoke $PM_{2.5}$ is harmful to human health and the ambient environment..." OC and BC are part of $PM_{2.5}$ and there's also a lot of HAPs in smoke that are more toxic but not well-measured and likely in lower abundance.

   Done.

   P3, Line 53-54, Introduction

   Smoke $PM_{2.5}$ is harmful to human health and the ambient environment (Reid et al., 2016; Aguilera et al., 2021; Johnston et al., 2021).

3. Line 82: change to "utilized for a long-term study."

   Done. Thank you for pointing this out.

   P4, Line 79-81, Introduction

The plume heights retrieved from TROPOMI offer daily global coverage, but TROPOMI data are available only from 2018 onwards and so cannot be utilized for a long-term study.

4. Line 88-89: The sentence "Besides these two studies…" feel out of place here. I'd reword to make it flow better.

Done.

P4, Line 87-89, Introduction

Besides these two methods for estimating injection height, Yao et al. (2018) used a machine learning model (random forest) and CALIOP data to predict the minimum heights of forest fire smoke in Canada.

5. Line 90: change to "represent"

Done.

P4, Line 89-91, Introduction

These three datasets represent the vertical extent of smoke plumes with high-resolution single parameters that specified the top and bottom heights of plumes, as well as the mean height of maximum injection (MHMI).

6. Line 91: add more explanation of MHMI

We have now included the definition and description of MHMI in Supplement Information.

P4, Line 89-91, Introduction

These three datasets represent the vertical extent of smoke plumes with high-resolution single parameters that specified the top and bottom heights of plumes, as well as the mean height of maximum injection (MHMI). The definitions of these variables are described in appendix S1.

7. Line 114: Change to “Alternatively, some studies use”

   Done.

   P5, Line 114-116, Introduction

   Alternatively, some studies use atmospheric chemistry models to explicitly simulate smoke $PM_{2.5}$ concentrations from open fires and their impacts on air quality and health in Australia (Rea et al., 2016; Nguyen et al., 2020, 2021; Graham et al., 2021).

8. Line 117: Change to “of smoke air quality but may focus only on…”

   Done.

   P5, Line 116-118, Introduction

   These studies can provide more accurate spatiotemporal variability of smoke air quality but may focus only on short-term simulations due to computational expense.

9. Line 138: Can likely remove this sentence about MERRA because it isn’t really essential here (it focuses on the fact that it uses aerosols, but the authors are only using it for PBL).

   Removed it as you suggested. Thanks.

   P6, Line 136-139, Section 2.1

   Daily mean PBL heights across Australia are obtained from the Modern-Era Retrospective Analysis for Research and Applications Version 2 (MERRA-2, Gelaro et al., 2017) at a spatial resolution of 0.5° latitude × 0.625° longitude. This reanalysis is often used to drive chemical transport models such as GEOS-Chem (Bey et al. 2001; Keller et al., 2014; Kim et al., 2015).

10. Line 349. I’d add something to remind the readers of what the first method described in Section 2.1 entails.

Done.

P13, Line 356-358, Section 3.2

Figure 2a compares the plume injection fractions above the PBL ($f_{abovePBL}$) derived from the MISR plume records with those calculated using the climatological plume profiles with assimilated PBL data (first method described in Section 2.1).

11. Figure 6: Can the pie carts just be made a bit smaller? I find the size visually unappealing.

Changed it as you suggested.

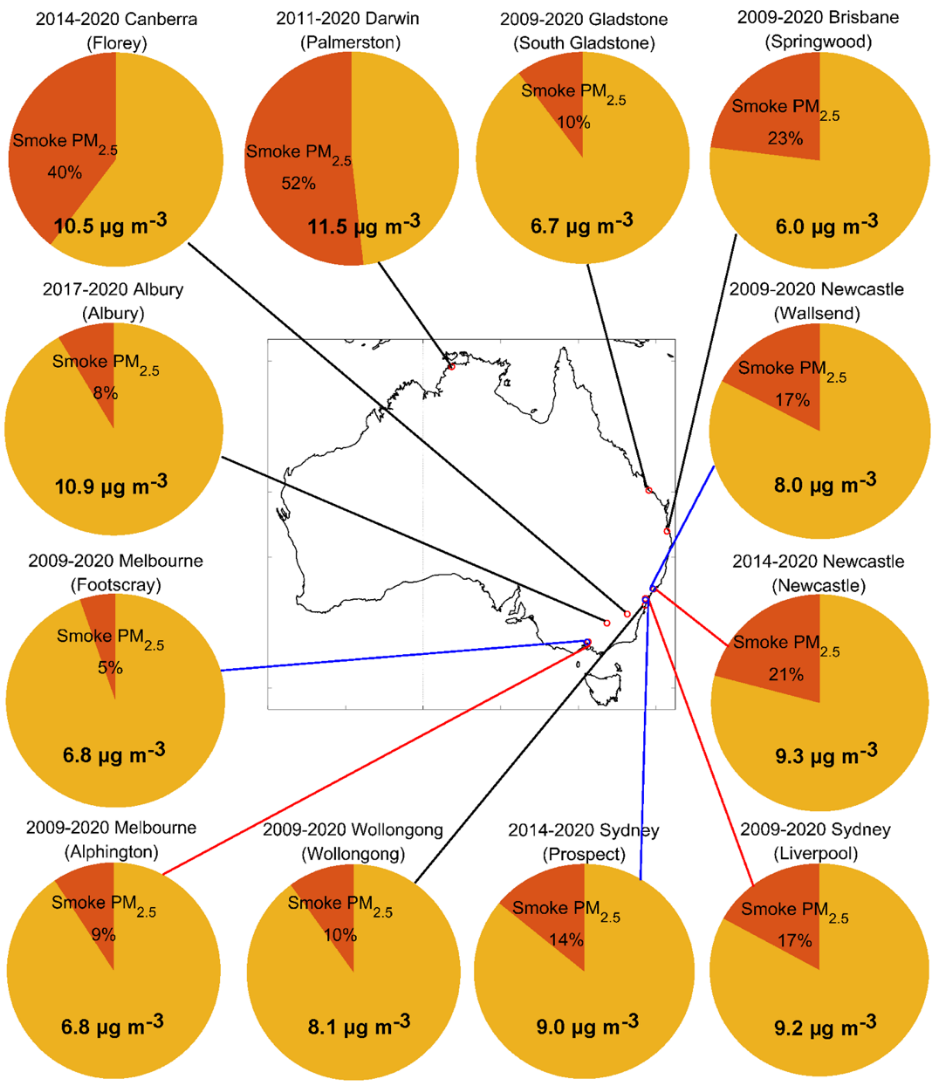

Figure 6. Contributions of simulated smoke $PM_{2.5}$ concentrations from the INJ-RF experiment to the observed total $PM_{2.5}$ concentrations (numbers on the pie charts) at 12 receptors averaged over the fire seasons of their respective observation periods (Table 3). Names of the observation sites are given in parentheses. Red sectors represent smoke contributions, while dark yellow sectors signify the differences between observed total $PM_{2.5}$ and simulated smoke $PM_{2.5}$ concentrations – i.e., the non-fire $PM_{2.5}$. Small circles on map represent the locations of these receptors. Different colors (red, blue, and black) are used to distinguish adjacent receptors.

12. I think Table S2 should be in the main text.

We have moved Table S2 to main text as new Table 3. Thank you.

Table 3. Statistics for daily mean $PM_{2.5}$ concentrations simulated by CTL, INJ-CLIM, and INJ-RF experiments, compared to the ground-based observations at 12 receptors. The daily mean concentrations are calculated over each receptor's observing period. Shown are the temporal correlation coefficients *R*, NMBs, and RMSEs of daily total $PM_{2.5}$ concentrations compared to the surface measurements.

Cities (site)	**Observation periods (Locations)**	***R*** [a]			**NMB**			**RMSE (µg m^{-3})**		
		CTL	***INJ-CLIM***	***INJ-RF***	***CTL***	***INJ-CLIM***	***INJ-RF***	***CTL***	***INJ-CLIM***	***INJ-RF***
Darwin [b] (Palmerston)	2011-2020 (130.94°E, 12.50°S)	0.76	0.76	0.76	17.1%	-17.8%	-2.1%	5.9	3.9	4.0
Gladstone [c] (South Gladstone)	2009-2020 (151.27°E, 23.86°S)	0.57	0.55	0.55	-5.4%	-11.0%	-11.2%	1.9	1.7	1.7
Brisbane [c] (Springwood)	2009-2020 (153.13°E, 27.61°S)	0.24	0.17	0.40	13.2%	2.2%	-4.8%	2.2	1.9	1.2
Newcastle [d] (Wallsend)	2009-2020 (151.66°E, 32.89°S)	0.53	0.48	0.52	16.1%	2.3%	-6.0%	6.6	3.9	2.4
Sydney [d] (Liverpool)	2009-2020 (150.90°E, 33.93°S)	0.40	0.38	0.37	14.8%	6.8%	0.4%	4.8	3.4	2.8
Wollongong [d] (Wollongong)	2009-2020 (150.88°E, 34.41°S)	0.27	0.28	0.27	-6.3%	-11.6%	-14.7%	1.5	1.4	1.6
Melbourne [e] (Footscray)	2009-2020 (144.87°E, 37.80°S)	0.25	0.25	0.25	-9.5%	-10.1%	-14.6%	4.6	4.6	3.6
Melbourne [e] (Alphington)	2009-2020 (145.03°E, 37.77°S)	0.41	0.39	0.40	23.7%	22.9%	14.4%	2.0	2.0	1.2
Albury [d] (Albury)	2017-2020 (146.93°E, 36.05°S)	0.93	0.93	0.93	-22.2%	-23.7%	-31.7%	5.6	5.8	7.3
Canberra [f] (Florey)	2014-2020 (149.04°E, 35.22°S)	0.67	0.63	0.68	19.3%	-8.6%	-16.0%	22.5	17.7	15.3
Sydney [d] (Prospect)	2014-2020 (150.91°E, 33.79°S)	0.72	0.69	0.71	7.5%	-1.2%	-7.2%	2.2	1.5	1.4
Newcastle [d] (Newcastle)	2014-2020 (151.75°E, 32.93°S)	0.59	0.50	0.55	28.9%	9.2%	-1.6%	6.9	4.0	2.7

[a] Temporal correlation coefficient between the observed and simulated annual mean total $PM_{2.5}$ concentrations during the fire seasons (April to December for Darwin and Gladstone; August to December for other cities).
[b] Observation data source: Northern Territory Environment Protection Authority (http://ntepa.webhop.net/NTEPA/Default.ltr.aspx)
[c] Queensland Government Open Data Portal (https://apps.des.qld.gov.au/air-quality/download/)
[d] New South Wales Department of Planning and Environment (https://www.dpie.nsw.gov.au/air-quality/air-quality-data-services/data-download-facility)
[e] Victoria Environment Protection Authority (https://www.epa.vic.gov.au/for-community/airwatch)

[f] Australian Capital Territory Government Open Data Portal (https://www.data.act.gov.au/Environment/Air-Quality-Monitoring-Data/94a5-zqnn