This study provides a thorough characterization of airborne INPs from a remote, alpine area at the Mt. Crested Butte study site in the Rocky Mountains. The long-term monitoring of INPs (almost 2 years) allowed for the emergence of trends and conclusions that can only be made from such a comprehensive data set. The study revealed a distinct seasonal variation in INP concentrations with a peak in the summer time. Further, different INP concentrations were correlated with various sources, with organic-containing soil dust dominating the INP population in this area. The parameterizations developed can be useful for predicting INPs in remote continental regions. This paper adds a valuable dataset to the field of ice nucleation and will be of interest to ACP readers. It is recommended that this paper should be accepted for publication after the authors address some minor revisions.

We appreciate the reviewer's positive assessment of our study and the valuable comments. We have carefully revised the manuscript in response to all suggestions.

General comments:

The discussion on the influence of vegetation on the INP population could benefit from additional context and references. Could the authors expand on this a bit more? For example, did the authors consider the influence of pollen? Biological INPs were present in warm seasons and decreased in winter (line 612). Does that line up with the pollen season? Furthermore, it might be helpful if the authors elaborated a bit more on the possible source pathways that link vegetation and soil, since the main conclusion is that organic-containing soil was the dominant INP.

For pollen in the Rocky Mountain region, Fall et al. (1992) found that pollen during the cold season (October to May) accounted for only about 18% of the annual influx, and the remaining 82% occurred during the warmer months. This pattern is consistent with the seasonal variation pattern in biological/heat-labile INP observed in this study, suggesting the possibility that pollen contributes to the biological INPs in the Rocky Mountains.

Bowers et al. (2012) investigated airborne bacteria at the Storm Peak Laboratory, located in the Rocky Mountains, and found that bacterial abundance was lower in winter and increased in fall and spring, indicating that bacteria may also serve as a source of biological INPs in this region. They also found that summer bacteria taxa were likely derived from soil and leaf-surface environments, suggesting that bacteria-related INPs may partly originate from soil dust.

Also, rainfall was suggested as an important source pathway of biological INPs (Prenni et al., 2013; Mignani et al., 2025).

The seasonal pattern provides indirect evidence for the possible sources of biological INPs, and further studies are required to identify the ice-active types of biological aerosols.

To further discuss these points, the following sentences have been added:

"Also, raindrop impact could be an important biological INP emission pathway (Prenni et al., 2013; Mignani et al., 2025)." (Lines 655–656)

"For example, airborne bacteria (Bowers et al., 2012) and pollen (Fall et al., 1992) in the Rocky Mountain region decrease in winter and increase during the warmer seasons. This seasonal pattern is consistent with the variation in biological/heat-labile INPs observed in this study, suggesting that these biological particles may represent potential sources of the biological INPs." (Lines 665–669)

"While bacteria may also be partly related to soil, as summer bacteria taxa were reported to likely originate from soil and leaf surface (Bowers et al., 2012), the specific ice-active taxa among them require further investigation." (Lines 670–673)

Four related references have been added.

"Bowers, R. M., McCubbin, I. B., Hallar, A. G., and Fierer, N.: Seasonal variability in airborne bacterial communities at a high-elevation site, Atmos. Environ., 50, 41-49, https://doi.org/10.1016/j.atmosenv.2012.01.005, 2012."

"Fall, P. L.: Spatial patterns of atmospheric pollen dispersal in the Colorado Rocky Mountains, USA, Review of Palaeobotany and Palynology, 74, 293-313, https://doi.org/10.1016/0034-6667(92)90013-7, 1992."

"Mignani, C., Hill, T. C. J., Nieto-Caballero, M., Barry, K. R., Bryan, N. C., Marinescu, P. J., Dolan, B., Sullivan, A. P., Hernandez, M., Bosco-Lauth, A., van den Heever, S. C., Stone, E. A., Grant, L. D., Perkins, R. J., DeMott, P. J., and Kreidenweis, S. M.: Ice-Nucleating Particles Are Emitted by Raindrop Impact, J. Geophys. Res: Atmos., 130, https://doi.org/10.1029/2024jd042584, 2025."

"Prenni, A. J., Tobo, Y., Garcia, E., DeMott, P. J., Huffman, J. A., McCluskey, C. S., Kreidenweis, S. M., Prenni, J. E., Pöhlker, C., and Pöschl, U.: The impact of rain on ice nuclei populations at a forested site in Colorado, Geophys. Res. Lett., 40, 227-231, https://doi.org/10.1029/2012gl053953, 2013."

PMF and source apportionment: While some atmospheric scientists are very familiar with PMF, others may not be fully convinced by your claims without having prior knowledge of PMF. Therefore, including a short explanation of PMF targeted for non-experts would make this section more convincing.

An explanation of the PMF model has been added as follows:

"PMF is a receptor model that decomposes an observation matrix into factor profiles and their corresponding contributions. These factors are related to emission sources and/or atmospheric

processes, providing a quantitative assessment of source influences (Paatero and Tapper, 1994)." (Lines 202–206)

Additional information on the PMF procedure and the identification of PMF factors have been added as follows:

"A five-factor solution was selected as the optimal solution based on the Q/Q_{exp} value and interpretation of the physical meanings of the factors (Brown et al., 2015). The corresponding factor profiles and time series are shown in Figures S5 and S6. These factors were identified, based on chemical signatures and previous literature, as coarse dust, fine dust, biomass burning, sulfate-dominated, and nitrate-dominated sources. Coarse and fine dusts had high contributions from Al, Ca, Fe, Mg, and Si, which are the main components of mineral dust (Liu and Hopke, 2003). Coarse dust explained more than 90% of the coarse mass (> $PM_{2.5}$), while there was no contribution from coarse mass in the fine dust factor. The biomass burning factor was strongly associated with organic and elemental carbon, which are mainly from combustion processes, and K, a tracer of biomass burning (Hopke et al., 2020). The other two factors are dominated by nitrate and sulfate, which are related to the formation of secondary aerosols and possibly some primary emissions from regional sources that include energy production and distant urban regions. Some similar factors were also resolved in published PMF analyses using IMPROVE data (Liu and Hopke, 2003; Hwang and Hopke, 2007)." (Lines 219–232)

This may stem from my lack of PMF knowledge, but it was not clear to me how sample classification rules were determined. For example, why was 40% chosen as the cutoff for coarse dust contribution to PM_{10}?

Thank you for this comment, which led us to better explain how we used PMF to classify air mass types. The PMF results showed the fractional contributions of each aerosol type to every sample, and these proportions were used to identify the dominant aerosol sources for each sample. This classification step is independent of the PMF analysis itself. A figure (Figure S8) showing the proportions of source contribution for all samples has been added to the SI to better illustrate how dominant sources were assigned. As shown in Figure S8, each sample was influenced by a variety of sources, and thus identifying a cutoff to assign the major source was somewhat arbitrary. Generally, a source contributing more than 50% is regarded as the dominant source. For coarse dust, if it contributed more than 50% to the total PM_{10} mass concentration or contributed more than 40% and was higher than other sources, this sample was categorized as a coarse dust-dominated sample.

To clarify the categorization, the method has been updated as follows:

"(1) Coarse dust, if coarse dust contributed more than 50% to the total PM_{10} mass concentration or contributed more than 40% and represented the largest contribution among all sources." (Lines 240–242)

The figure for proportions of source contributions:

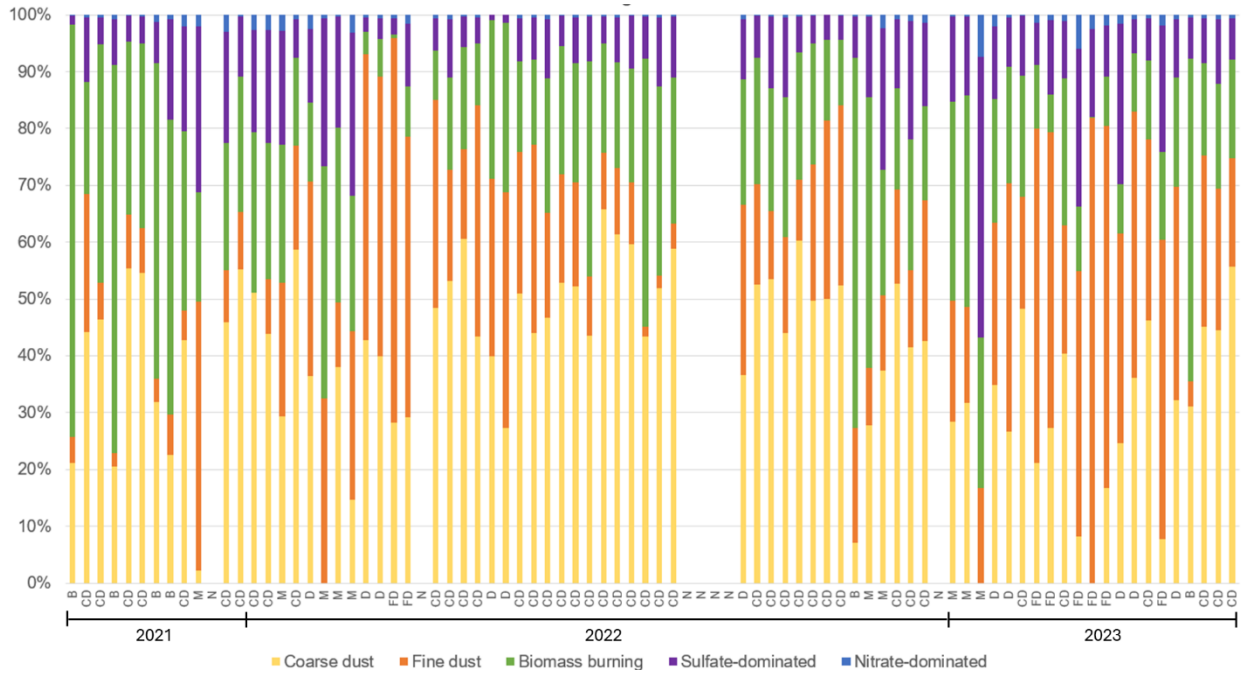


Figure S8. Proportions of source contributions for all samples. The label below each sample indicates its classification: CD (coarse dust), B (biomass burning), D (dust), FD (Fine dust), M (Mixed sources), and N (no source information).

Ulbrich et al., 2009 provides useful PMF guidelines when working with AMS data. Can you provide a similar reference for the technique that was used for the IMPROVE dataset? This will be very useful for scientists who want to learn PMF and reproduce your work, especially considering the interpretation of factors and if that is approached differently for different techniques.

We have added the following references that have applied the PMF model to IMPROVE datasets for source apportionment. We also note that the version of PMF we use was developed by the EPA in the 1990s for source apportionment of environmental datasets and continues to undergo further refinement: https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-data-analyses:

Liu and Hopke (2003) is an early work that applied PMF to resolving the source of $PM_{2.5}$ at two high elevation IMPROVE sites in the western U.S.

Hwang and Hopke (2007) investigated the sources at a west coast IMPROVE site using the PMF model.

Brown et al. (2015) provide methods for evaluating the results using EPA PMF 5.0 with examples.

These studies describe the PMF model and its application to IMPROVE datasets, and they resolved some similar factors as we found in this study, which support our source apportionment results.

These points have been added in manuscript as follows:

"A five-factor solution was selected as the optimal solution based on the Q/Q_{exp} value and interpretation of the physical meanings of the factors (Brown et al., 2015). The corresponding factor profiles and time series are shown in Figures S5 and S6. These factors were identified, based on chemical signatures and previous literature, as coarse dust, fine dust, biomass burning, sulfate-dominated, and nitrate-dominated sources. Coarse and fine dusts had high contributions from Al, Ca, Fe, Mg, and Si, which are the main components of mineral dust (Liu and Hopke, 2003). Coarse dust explained more than 90% of the coarse mass (> $PM_{2.5}$), while there was no contribution from coarse mass in the fine dust factor. The biomass burning factor was strongly associated with organic and elemental carbon, which are mainly from combustion processes, and K, a tracer of biomass burning (Hopke et al., 2020). The other two factors are dominated by nitrate and sulfate, which are related to the formation of secondary aerosols and possibly some primary emissions from regional sources that include energy production and distant urban regions. Some similar factors were also resolved in published PMF analyses using IMPROVE data (Liu and Hopke, 2003; Hwang and Hopke, 2007)." (Lines 219–232)

"Brown, S. G., Eberly, S., Paatero, P., and Norris, G. A.: Methods for estimating uncertainty in PMF solutions: examples with ambient air and water quality data and guidance on reporting PMF results, Sci. Total Environ., 518-519, 626-635, https://doi.org/10.1016/j.scitotenv.2015.01.022, 2015."

"Hwang, I. and Hopke, P. K.: Estimation of source apportionment and potential source locations of PM2.5 at a west coastal IMPROVE site, Atmos. Environ., 41, 506-518, https://doi.org/10.1016/j.atmosenv.2006.08.043, 2007."

"Hopke, P. K., Dai, Q., Li, L., and Feng, Y.: Global review of recent source apportionments for airborne particulate matter, Sci. Total Environ., 740, 140091, https://doi.org/10.1016/j.scitotenv.2020.140091, 2020."

"Liu, W. and Hopke, P. K.: Origins of fine aerosol mass in the western United States using positive matrix factorization, J. Geophys. Res: Atmos., 108, https://doi.org/10.1029/2003jd003678, 2003."

In addition, could the authors please mention the techniques used for the filter and elemental analysis that were used from the IMPROVE network.

The techniques used by the IMPROVE network for chemical analysis are introduced in the IMPROVE Data User Guide 2023 (Version 2), and have been added as follows:

"In the IMPROVE program, elemental analysis was performed on the Teflon filters using X-ray fluorescence (XRF), water-soluble anions were analyzed by ion chromatography (IC), and elemental carbon (EC) and organic carbon (OC) were analyzed using a thermal-optical carbon analyzer (Hand, 2023)." (Lines 210–213)

"Hand, J.: IMPROVE Data User Guide 2023 (Version 2), Interagency Monitoring of Protected Visual Environments (IMPROVE) Program, available at: https://vista.cira.colostate.edu/Improve/data-user-guide/, 2023."

Just recently, a preprint by Lacher et al. 2025 was released, which also studied INP concentrations in the U.S. Rocky Mountains. The sampling period from Lacher et al., 2025 has some overlap with the sampling period in this paper and the sampling sites are close together. It would be very beneficial if you can compare trends in your data set with those in the other study.

Lacher et al. (2025) found that INP concentrations were lowest in winter and increased in spring, and that supermicron particles were the major contributor to INPs in the Rocky Mountains, based on online INP measurements at cold activation temperatures (−22 °C to −32 °C). These results agree well with our findings. Their results are compared and discussed in the manuscript as follows:

"This is comparable to online INP measurements in the Rocky Mountain region (median: 8.2 L−1 at −26 °C; Lacher et al., 2025)." (Lines 314–315)

"Recent online INP measurements for activation temperatures from −22 °C to −32 °C conducted from October 2021 to May 2022 and January to May 2025 at the Storm Peak Laboratory in the Rocky Mountains (Lacher et al., 2025) found a similar seasonal pattern, with the lowest INP concentrations in winter and increased in spring, suggesting that the INP sources could be similar and may dominate INPs across a broaden region of the Rocky Mountains." (Lines 347–351)

"Furthermore, Lacher et al. (2025) provided direct evidence that INPs active at cold temperatures were significantly contributed by supermicrometer particles, which they attributed to dust, in the Rocky Mountains. Their observation site was located near to the IMPROVE site at Mount Zirkel, where our PMF analyses identified similar sources and trends to those near the SAIL (Text S1), suggesting that INPs in both studies were impacted by coarse dust." (Lines 388–393)

Specific comments:

Title: The paper might benefit from a more precise title. Adding in your main finding into the title would make it more targeted.

Thanks for the comment, the main finding regarding the importance of soil dust and biological contributions has been incorporated into the title, as follows:

"Seasonal variability, sources, and parameterization of ice-nucleating particles in the Rocky Mountain region: Importance of soil dust and biological contributions"

Line 363: course dust was found to correlate with INPs at -10 °C (R^2 = 0.43). Is this R^2 value significant for INP characterization? Is this a typical value you expect in INP field studies? I feel like this value is a bit low to suggest a direct correlation.

From Table 1 in the manuscript, the p-value of this correlation is lower than 0.01, suggesting that it is statistically significant. However, the R^2 is lower than the correlations observed at colder temperatures (0.50-0.58). To clarify this point, the related sentences have been revised as follows:

"Coarse dust showed good correlations with INPs active at all temperatures (Table 1), with correlation coefficients increasing for colder activation temperatures, suggesting that coarse dust is a major source of INPs, particularly at lower temperatures. This is consistent with previous findings that dust dominates the INPs at temperatures below −20 °C (Beall et al., 2022; Testa et al., 2021; Kanji et al., 2017)." (Lines 384–388)

"Interestingly, coarse dust presented a weaker correlation with INPs at −10 °C (R^2 = 0.43), a temperature range usually associated with biological INPs. This may be due to the large number of coarse dust particles, biological INPs carried on dust particles, and/or the inclusion of biological particles in the coarse dust factor, as biological particles are mostly supermicron in size (Després et al., 2012)." (Lines 393–397)

Line 632-636: Can you give some example methods on how this could be done? Comments like these are very useful when planning future studies and suggesting example methods would be very beneficial to the community.

In addition to the heat treatment method, previous studies have provided indirect evidence of biological INPs based on fluorescence measurement, combining CFDC and mass spectrometer, and microscopy (Cornwell et al., 2023 and 2024; Sanchez-Marroquin et al., 2021). However, from the authors' knowledge, based on currently available methods, it is still very difficult to obtain direct evidence of biological INP identities. Comprehensively investigating the types and abundance of biological aerosols in the Rocky Mountains would be helpful. For directly investigating the biological INPs, new methods that separate INPs, especially biological origins, from other particles need to be developed. To include this information, the sentence has been revised as follows:

"In future studies, identifying the most abundant biological INPs in this region and determining whether they originate from vegetation, soil-associated sources, or from a combination of both would help improve our understanding of biological INP variability and improve their estimation. However, besides heat treatment, current approaches provide only indirect evidence of biological INPs (Cornwell et al., 2023 and 2024; Sanchez-Marroquin et al., 2021). Comprehensive characterization of biological aerosol types and abundance, or developing new analytical approaches, would be highly beneficial for advancing biological INP research." (Lines 673–679)

Line 153: please include the equation for INP concentration directly in the methods section in addition to your reference to Vali, 1971

The equation that calculates the cumulative INP concentrations as a function of temperature has been added as follows:

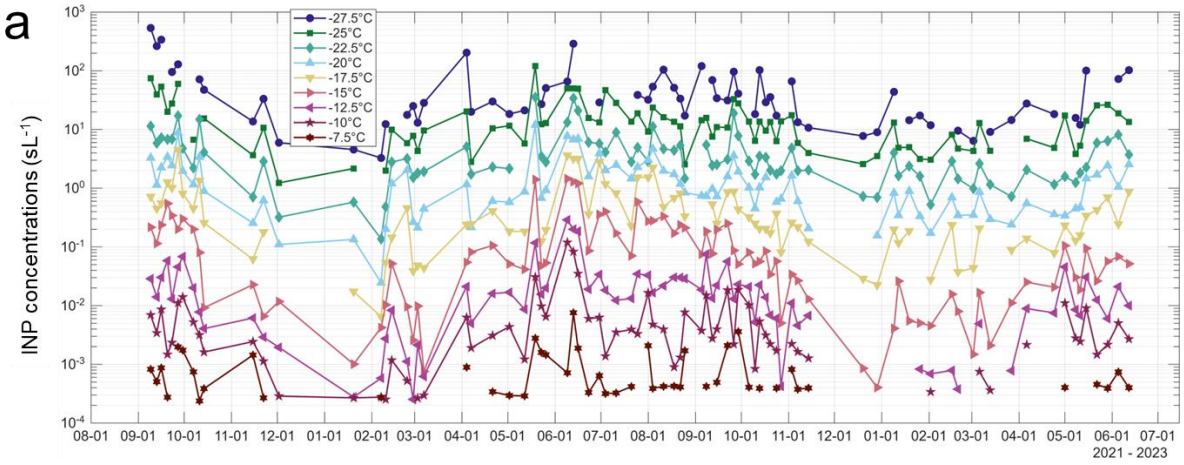
"Cumulative INP concentrations as a function of temperature (n_{INPs}(T), INPs per liter of air) were calculated based on the Vali method (Vali, 1971) using:

$$n_{INPs}(T) = ln(\frac{N_0}{N_0 - N(T)}) \times \frac{V_w}{V_c} \times \frac{1}{V_a}$$

where N_0 is the total number of wells containing aliquots, $N(T)$ is the cumulative number of wells frozen at temperature T, V_w is the volume of water used for particle resuspension, V_c is the aliquot volume added to each well, and V_a is the total sampled air volume." (Lines 148–153)

Figure 3a: This graph is a little difficult to read. Could the lines be thicker to make it easier to follow? Additionally, the data presented in Figure 3a and Figure S3a are very similar. Is it necessary to show both versions? Please also remove the "a" from Figure S3 as there is only one panel.

To improve the readability of Figure 3a, the colors, markers, and size of the figure have been modified as follows:



The "a" in Figure S3 has been removed.

Figure S3 presents all samples measured during the SAIL campaign (in total 113 samples). Samples affected by artificial cloud seeding and snowmaking activities are shown with grey shadows in Figure S3, and these samples were not discussed in this manuscript. Figure 3a shows only the 83 samples that were not affected by cloud seeding and snowmaking activities. The difference has been explained in lines 187–190 and in the caption of Figure 3.

Figure 7: Can you add a line to the figure caption to explain that the y-axis and x-axis is the same for the 2 plots in each column? It was a bit challenging to understand the figure right away.

A label has been added to each panel, and the caption of Figure 7 (Figure 8 in the revision) has been revised as follows:

"**Figure 8**. Correlations between concentrations of (a, b) biological/heat-labile INPs and other organic INPs, (c, d) biological/heat-labile INPs and inorganic INPs, and (e, f) other organic INPs and inorganic INPs, active at either −15 °C (upper row) or −25 °C (lower row). Dashed lines indicate a 10:1 relationship for reference, and the orange rectangles highlight the strongest correlations at each temperature." (Lines 765–769)

Figure S1 and S2: Are the black and orange curves in Figure S1 the same as the curves shown in Figure S2? If so, please consider removing S1 as the data is repeated.

Yes, the black dots and orange dots in both figure S1 and S2 represent the INP spectra of base analyses and heat treatment, respectively. However, Figure S1 shows all samples subjected to heat treatment (43 samples in total), whereas Figure S2 shows only the subset of samples (34 in

total) that underwent H_2O_2 treatment. To clarify this difference, the captions of the two figures have been revised as follows:

"Figure S1. The INP temperature spectra of samples that were subjected to heat treatments (43 samples in total). The base analyses (black dots) are shown along with spectra after heat treatment (orange dots). For clarity, uncertainties are not shown here."

"Figure S2. The INP temperature spectra of samples with base analysis (black dot), heat treatment (orange dot), and H_2O_2 treatment (blue dot; 34 samples in total). For clarity, uncertainties are not shown here."

Figure S12: Please add a marker to show the location of the sampling site. Something like what was done in Figures 1 and S8.

A marker shows the sampling location has been added to Figure S12 (Figure S13 in the revision) as follows:

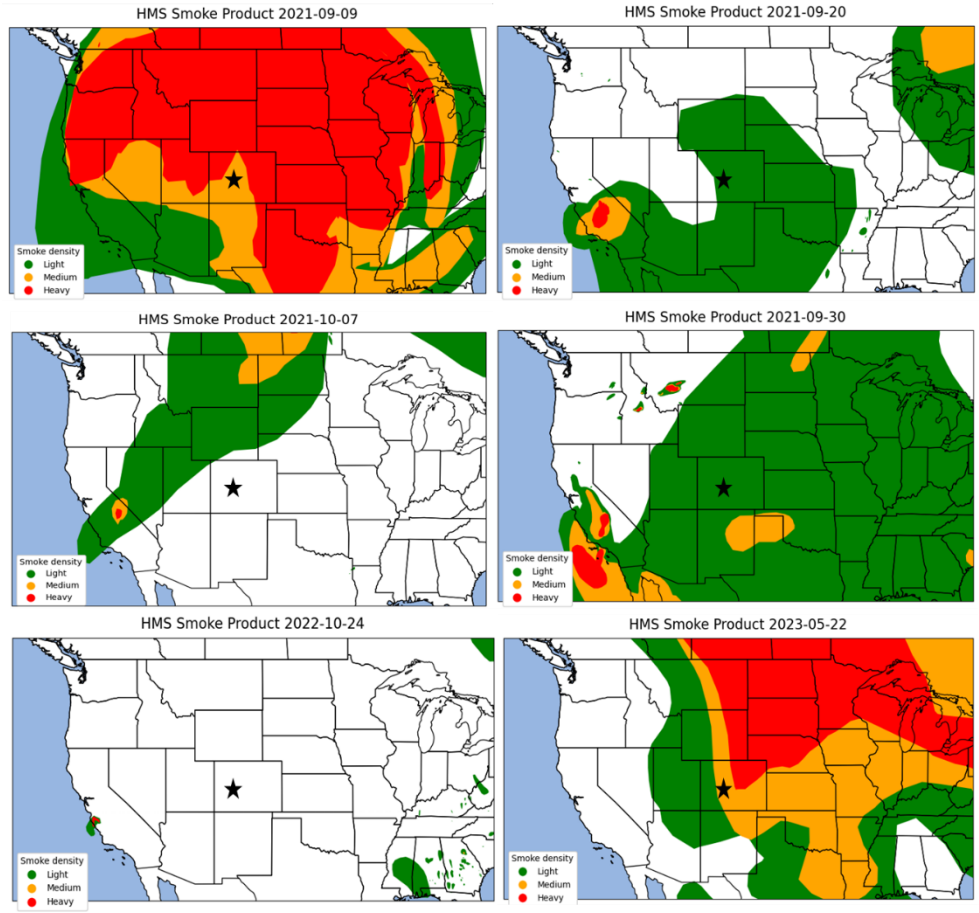


Figure S13. NOAA Hazard Mapping System products for the sample days, which are dominated by biomass burning aerosols. The black stars mark the SAIL sampling location.

Technical corrections:

Figure S8: Please add a legend or a line in caption to explain what the black star represents.

The caption of Figure S8 (Figure S9 in the revision) has been revised as follows:

"**Figure S9.** Residence-time weighted back trajectories for samples that were categorized as dominated by (a) coarse dust, (b) fine dust, (c) dust, and (d) biomass burning. The categorization was based on aerosol source contributions derived from the PMF analysis; details of the categorization method are provided in the Methods section. The black stars indicate the SAIL sampling location."

Figure S10: Please add x-axis label

The x-axis label "Temperature (°C)" has been added.

Please ensure all figures are presented with adequate resolution.

The resolutions of all figures have been checked and are sufficient for publication.

There is inconsistent figure border formatting throughout the paper. And in some cases, the figure border cuts through text (S3, S4, S6, S7, S9, S13).

The borders of these figures have been modified.

Line 210: typo in April

The typo has been corrected.

Text S1: link for the IMPROVE sop #351 says page not found. Please update.

The IMPROVE SOP in Text S1 has been revised as a reference as follows:

"These uncertainties were therefore calculated based on the method introduced in Niño (2021) and the IMPROVE standard operating procedure (SOP 351; IMPROVE, 2021)."

"IMPROVE (2019): IMPROVE Standard Operating Procedure 351: Data Processing and Validation, available at: https://vista.cira.colostate.edu/improve/wp-content/uploads/2019/06/IMPROVE-SOP-351_Data-Processing-and-Validation_06.2019.pdf"

Other revisions

1. Citations including "Mccluskey" corrected as "McCluskey".

2. Sentence in line 242 has been revised as follows:

   "Further details on the PMF analysis and results, as well as support for their applicability over the broad surrounding Rocky Mountain region (IMPROVE sites at Mount Zirkel and Rocky Mountain National Park) are provided in Supplement Text S1 and Figure S7."

3. The figure numbers have been updated due to the inclusion of new figure.

4. A new funding number (DE-SC0021116) that supports this work has been included in the Acknowledgements section.