

Black: Comments from reviewer 2

Blue: Replies

Comment R2-1

This study identified oleamide as a biomass burning aerosol in sections of a Greenland ice core which contain signals of wildfire events. Events are deduced from peaks in NH_4^+ and NO_3^- concentrations, based on the understanding that these ions are established biomass burning indicators. Particles found in these sections of ice core are characterised using Raman spectroscopy, SEM-EDS elemental analysis and FT-IR spectroscopy. Oleamide is identified in these samples, and the authors suggest it forms during biomass burning events. It is therefore proposed as a novel insoluble proxy for these events in ice cores. Oleamide's ice nucleating ability is assessed in both immersion and contact freezing modes using bespoke set-ups, and it is concluded that it does not act as an ice nucleating particle.

This study aimed to "reconstruct the morphology and chemical composition of fire-derived particulate [organic aerosols]", which led to the identification of oleamide as a stable wildfire tracer. Latterly, the aim shifts to evaluating the ice-nucleating ability of the identified oleamide particles. However, this secondary aim is not stated in the introduction, and the rationale for including it in this paper is unclear.

Overall, the paper is interesting and easy to follow. In particular, the abstract is clear and concise, and the introduction provides some interesting context. The methodology is mostly logical, too. However, some crucial decisions lack a thorough explanation, and claims in the discussion do not always match up with the results presented in the figures or the existing literature.

We sincerely thank the reviewer for their thorough reading of our manuscript and for the encouraging and positive assessment, noting that the paper is "interesting and easy to follow" with a "clear and concise" abstract and a "logical methodology." We also appreciate the accurate summary of our work. We have carefully considered all of your constructive critiques regarding the rationale of our study, the thoroughness of our explanations, and the alignment between our discussion and findings/literature, and we will revise the manuscript extensively to address these concerns.

Specifically, regarding the rationale for evaluating the ice-nucleating ability of oleamide: We fully agree with the reviewer that this secondary objective was not adequately introduced or justified in the original Introduction. In the revised manuscript, we will update the Introduction (Section 1) to explicitly incorporate this objective and provide its scientific context. Evaluating whether oleamide acts as an ice-nucleating particle is crucial for understanding its atmospheric fate, its potential influence on cloud formation and climate feedback during transport, and how aerosol–cloud interactions might affect its wet deposition and long-range transport to the Greenland Ice Sheet.

Regarding the decisions and claims highlighted in the Discussion, we will clarify our methodologies, tone down overstatements where appropriate, and substantially expand our literature comparison, including references to relevant studies on oleamide. Below, we provide point-by-point responses to all specific comments and detail the corresponding revisions that will be made to the manuscript.

Comment R2-2

Major concerns

Of primary concern is how robust NH_4^+ and NO_3^- are as biomass burning (BB) tracers in ice core samples, given that this is the basis for the study. NO_3^- is non-specific on its own with sources from fossil fuel combustion and soil emission and requires isotopic analysis of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ for accurate attribution (Vega et. al., 2014).¹ The authors highlight the diversity of NH_4^+ sources as well (line 209), suggesting this could be a reason for the weak correlation between oleamide and NH_4^+ concentrations. Given the age of this ice core, anthropogenic emissions will influence the concentrations of these

compounds in this ice core.

We fully agree with the reviewer that relying solely on individual profiles of NH_4^+ and NO_3^- introduces inherent uncertainties due to their non-specific nature and the confounding influences of anthropogenic, agricultural, and soil emissions, particularly in ice core records spanning recent decades. We sincerely appreciate the reviewer highlighting the necessity for a more robust analytical framework.

To address this fundamental concern, we will restructure the core logic of our sample selection and source attribution in the revised manuscript, shifting away from a heavy reliance on individual inorganic ion spikes. Rather than using NH_4^+ or NO_3^- peaks as primary standalone proof of biomass burning, we will ground our identification on ice core layers corresponding to historically documented major North American wildfire events (e.g., 1961, 1980, and 1994). The prominent detection of oleamide in these independently verified, major fire years provides much stronger and more direct evidence for its origin as a biomass burning product.

For other sample layers where oleamide was detected, we will no longer rely on single-ion peaks. Instead, we will interpret these occurrences strictly in the context of concurrent, co-occurring peaks in both NH_4^+ and NO_3^- , cross-referenced with regional wildfire history records (e.g., the extensive 1989 Canadian wildfires).

Furthermore, we will revise Section 4 (Discussion) to explicitly acknowledge multi-source contributions (including anthropogenic and soil emissions) to the inorganic ion budget, as emphasized by Vega et al. (2014). We will tone down our claims regarding direct equivalence between ion peaks and biomass burning, presenting oleamide as a novel, complementary insoluble tracer that may help overcome the ambiguities associated with soluble inorganic species.

Comment R2-3

The ice core samples are chosen using peaks in NH_4^+ or NO_3^- (line 65) but there is no description of the method used to acquire the data shown in Figure A1 (nor is there a reference in A1's caption). This should be updated to include a clear explanation of how the ice core was analysed for these compounds. The selection of samples using the peaks in A1 is also confusing; there seem to be many peaks that are higher in concentrations of either or both species that are not selected. Selection of samples containing BB event signals would have been more convincing if they contained spikes in both NH_4^+ and NO_3^- (rather than one or the other). Also, analysis of these samples for the more specific BB tracers (as listed in line 183) and subsequent comparison to oleamide concentrations would strengthen the attribution of oleamide to BB events.

Comparison of NH_4^+ and NO_3^- to K^+ concentrations would also support the identification of BB events. If NH_4^+ and NO_3^- peaks were obtained using IC, then a K^+ record likely exists. K^+ could also be identified in the SEM-EDS analysis of particles.

We apologize for omitting the analytical details of the ion data in the original text. In Section 2 (Methods) of the revised manuscript, we will add a dedicated description of the Ion Chromatography (IC) methodology used to quantify major inorganic ions (NH_4^+ , NO_3^- , etc.). We will also update the caption of Figure A1 to include appropriate methodological citations and analytical details.

We fully agree that focusing on dual spikes of both NH_4^+ and NO_3^- is significantly more compelling than relying on isolated single-ion peaks. In our revised framework, we will clarify that sample selection was dictated first by sections corresponding to historically documented major wildfire years (1961, 1980, and 1994) and second by layers exhibiting co-occurring, simultaneous peaks in both NH_4^+ and NO_3^- .

We appreciate the reviewer's suggestion to examine K^+ as a biomass burning tracer. We thoroughly re-evaluated our IC dataset and SEM-EDS particle spectra for potassium. Potassium (K^+) concentrations in this ice core record were predominantly below the limit of detection across the analyzed sections, preventing the establishment of a continuous, reliable K^+ profile. In individual particle analyses via SEM-EDS, K signals were similarly negligible or below the signal-to-noise threshold in the analyzed organic/oleamide particles.

In the revised Section 2 (Methods), we will add a brief explanatory sentence noting that K^+ was below the detection threshold and could not serve as a quantitative tracer in this core section.

Regarding molecular biomass burning tracers (e.g., levoglucosan), quantitative molecular analysis requires substantial sample (meltwater) volumes that were unavailable for these specific thin ice core slices, which were dedicated to nonvolatile particle filtration and single-particle spectroscopy (Raman, FT-IR, SEM-EDS). We will clarify this analytical trade-off in the revised Methods section.

Comment R2-4

Moreover, authors do not make clear that some samples that show low concentrations in either NH_4^+ or NO_3^- are also selected and tested, despite these being important controls. This should be made explicit in Section 2.3. The comparison between concentrations in these controls (i.e. ice core samples with low NH_4^+ and/or NO_3^-) and samples which include BB event signals should be more thorough (see paragraph below).

We sincerely thank the reviewer for highlighting the importance of explicitly framing these low-concentration samples as controls. We completely agree that establishing clear baseline/control samples is essential for evaluating whether oleamide enrichment is uniquely associated with biomass burning horizons. In the revised manuscript, we will update Section 2.3 to explicitly state that ice core layers exhibiting low background levels of both NH_4^+ and NO_3^- were intentionally selected and analyzed alongside the peak event samples to serve as negative/baseline controls. Furthermore, we will thoroughly expand the comparative description between these baseline controls and the biomass burning event samples in both Section 3 (Results) and Section 4 (Discussion). This revised comparison will demonstrate that baseline control samples characteristically feature absent or negligible number fractions of oleamide, establishing a distinct contrast with the marked oleamide abundances observed during major, historically documented biomass burning horizons (e.g., 1961, 1980, and 1994).

Comment R2-5

The paper does not convince me that oleamide is specifically a product of biomass burning. Discussions surrounding the 'positive correlation' (line 155) between NH_4^+ concentrations and the detection of oleamide seem too confident, given the low R^2 value of 0.26 and a p value of 0.051. This is > 0.05 and therefore statistically insignificant.

Additionally, line 176 states that the "*frequent detection in samples with high NH_4^+ concentrations suggested a biomass burning origin*" and line 204 states that there is a "*close temporal correspondence*" between the two species. Given the poor correlation and the diverse sources of NH_4^+ discussed above, this seems an overconfident statement, particularly when compared to Figure 2. Indeed, lines 204 to 206 state that for samples with no peaks in NH_4^+ or NO_3^- , there was zero or negligible oleamide.

However, 1989 (no high NH_4^+ or NO_3^-) shows a higher number fraction of Unidentified Particle 1 (oleamide) than some samples with high NH_4^+ (1970, 1992) or high NO_3^- (1940). Also, the threshold for negligibility is not defined. As such, the data presented does not seem to match the conclusions stated.

We completely agree with the reviewer. Characterizing a relationship with NH_4^+ and NO_3^- as a "positive correlation" was statistically unjustified given the p-value ($p = 0.051 > 0.05$), and statements claiming a "close temporal correspondence" were overconfident and not supported by our data.

To align our conclusions strictly with the empirical evidence, we will thoroughly revise the manuscript. Specifically, we will entirely remove the discussion claiming a statistically significant correlation between NH_4^+ concentrations and oleamide abundance, along with phrases such as "close temporal correspondence". Throughout Section 4 (Discussion), we will shift to much more cautious and tentative language when discussing the potential

association between oleamide and biomass burning. In Section 2 (Methods), we will explicitly define the threshold criteria for particle classification and detection limits. We will clarify that "negligible" refers to instances where the relative number fraction of oleamide particles was below the median plus one standard deviation (i.e., < 14.1% of total analyzed particles), eliminating the previous ambiguity. Furthermore, we will expand our discussion in Section 4 to explicitly address sample layers such as 1989, where oleamide was detected despite the absence of strong NH_4^+ or NO_3^- spikes. We will explain that insoluble organic particles and soluble inorganic ions can exhibit decoupled atmospheric transport, scavenging, and deposition pathways. For instance, major regional wildfire activity (such as the widespread 1989 North American/Canadian fires) can transport light, insoluble organic aerosols over long distances without necessarily producing coincident, concentrated spikes of soluble inorganic ions at the ice core drill site. In the revised manuscript, we will no longer claim that oleamide directly tracks every inorganic ion profile. Instead, we will present oleamide as a novel insoluble organic aerosol candidate that is prominently present during major, historically verified wildfire horizons (1961, 1980, and 1994), while acknowledging that its presence in other years reflects the complex, multi-source nature of long-range aerosol transport.

Comment R2-6

Ultimately, these inconsistencies between the data and the conclusions undermine the validity of the paper. I suggest a closer analysis of this data and changing the discussion and conclusions accordingly, given that what is said does not seem to match the figure, and leaves doubt in the statements made. Authors should also include a discussion as to why some of these samples without strong NH_4^+ or NO_3^- peaks still possess oleamide concentrations higher than others that do. More tentative wording surrounding the source apportionment of oleamide and the 'positive correlation' with NH_4^+ is also advised.

We fully acknowledge that our original presentation lacked the necessary rigor and that overstating the correlation with inorganic ions undermined the manuscript's clarity. In response, we will thoroughly re-evaluate our dataset and substantially revise both the Discussion and Conclusion sections to ensure that all claims strictly reflect our empirical observations.

We will carefully revise our language throughout the text to avoid overconfident statements regarding source attribution. We will no longer claim a statistically significant correlation with NH_4^+ or direct temporal equivalence. Instead, we will frame oleamide as a promising candidate for an insoluble biomass burning proxy, explicitly highlighting the inherent complexities involved in correlating insoluble organic particles with soluble inorganic species in ice cores.

Comment R2-7

Moving to oleamide identification, the FTIR and Raman spectroscopy show excellent agreement and are convincing. However, the arrival at this identification sometimes felt like it lacked rigour. Firstly, betulinic acid is discounted (lines 182 – 191) for not showing adequate resemblance, while oleic acid is described as showing a close match. From the provided spectra in Figure 4, both betulinic acid and oleic acid have very similar FT-IR spectra, which are also similar to that of Unidentified Particle 1. Including the Raman spectra in Figure A2 of the same compounds in Figure 4 would help highlight spectroscopic differences and better explain the choices made when identifying Particle 1 as oleamide. I'd suggest that the current Figure 4 becomes Figure 4a, and the Raman spectra are added as Figure 4b.

We thank the reviewer for this insightful comment and for recommending the consolidation of Figure 4 and Figure A2 into Figure 4a (FT-IR) and Figure 4b (Raman). We fully agree that the step-by-step logic leading from candidate molecules to oleamide required greater clarity and spectroscopic justification, particularly regarding why betulinic acid was ruled out while oleic acid was initially identified as a structural match.

In the revised manuscript, we will adopt the reviewer's suggested figure layout (Figures 4a and 4b) and rewrite the identification logic in Section 4 (Discussion) to clearly articulate our

three-step spectroscopic identification process. 1) Betulinic acid was evaluated because it is a known triterpenoid biomarker for hardwood biomass burning. However, its complex pentacyclic triterpenoid core ($C_{30}H_{48}O_3$) yields distinct fingerprint absorption bands in the $1000\text{--}1450\text{ cm}^{-1}$ region and a sharp, characteristic peak at 882 cm^{-1} (Cîntă-Pînzaru et al., 2012). The 882 cm^{-1} peak does not match the spectrum of our target particle, which lacks these cyclic backbone signatures, allowing us to confidently rule out betulinic acid. 2) The target particle exhibited strong, characteristic vibrational features of a long straight-chain aliphatic backbone, including $-\text{CH}_2-$ asymmetric and symmetric stretching modes at 2920 cm^{-1} and 2850 cm^{-1} , as well as $-\text{CH}_2-$ scissoring modes at 1465 cm^{-1} . Oleic acid (a C_{18} monounsaturated fatty acid) provided an excellent match for this aliphatic backbone region, indicating that the particle possesses an unbranched long-chain fatty structure. The target particle displayed clear primary amide bands: a C=O stretch (Amide I band at $\sim 1650\text{ cm}^{-1}$), an N-H bend / C-N stretch (Amide II band at $\sim 1620\text{ cm}^{-1}$), and a characteristic $-\text{NH}_2$ stretching doublet (3350 cm^{-1} and 3180 cm^{-1}). This demonstrated that the particle shares the C_{18} carbon chain structure of oleic acid but carries a primary amide terminal group ($-\text{CONH}_2$), unequivocally directing our identification to oleamide. By combining FT-IR (Figure 4a) with Raman spectroscopy (Figure 4b), the distinct backbone and amide vibrational modes of the target particle will be shown to align with authentic oleamide standards, ruling out carboxylic acid candidates (both betulinic and oleic acid). Cîntă-Pînzaru, S., Dehelean, C. A., Soica, C., Culea, M., and Borcan, F.: Evaluation and differentiation of the Betulaceae birch bark species and their bioactive triterpene content using analytical FT-vibrational spectroscopy and GC-MS, *Chemistry Central Journal*, 6, 67, <https://doi.org/10.1186/1752-153X-6-67>, 2012.

Comment R2-8

Additionally about Figure A2, only 33 organics are displayed, not 45 as stated by both the caption and in line 124. This needs amending. The total of 63 references will also need editing throughout the manuscript.

In the revised manuscript, we will thoroughly audit and update all references to compound counts across the text, figures, and supplementary materials. We will update the main text in Section 2 (Methods, formerly line 124) and the caption of Figure A2 to accurately state that 33 organic compounds are displayed in the reference library comparison, ensuring exact agreement with the spectra presented in Figure A2. We will correct the total number of evaluated reference materials throughout the entire manuscript—including the Methods, Results, and Supplementary Information captions—to strictly reflect the precise count of analyzed reference spectra.

Comment R2-9

Furthermore, Figure A2 does not include Raman spectra of any other amide compounds. These could show strong similarities to oleamide and Unidentified Particle 1. For more confident characterisation, more amide species should be tested using Raman spectroscopy as references and/or another complementary technique (e.g. mass spectroscopy) should be employed for Unidentified Particle 1. FT-IR is not specific enough to provide this additional confidence.

We thank the reviewer for this crucial comment regarding the spectral specificity among primary amides. We fully agree that evaluating our ice core target particle against a broader suite of primary amide standards is essential for confirming its unambiguous identification. To address this recommendation directly, we have ordered authentic reference standards for four additional primary amides—comprising saturated amides (stearamide, $C_{18:0}$; palmitamide, $C_{16:0}$) and unsaturated amides (erucamide, $C_{22:1}$; linoleamide, $C_{18:2}$)—from commercial suppliers, which are scheduled for delivery by the end of October 2026. Upon their arrival, we will measure high-resolution Raman spectra for all four standards under identical analytical conditions, systematically assess diagnostic spectral differences linked to alkyl chain length and degree of unsaturation, and incorporate these reference spectra into Figure A2 and Section 4 (Discussion) of the revised manuscript to further strengthen our spectroscopic characterization.

Comment R2-10

Another major point of concern surrounds the ice nucleation experiments. Neither the aim nor the purpose of conducting these tests is explicitly described in the introduction, leaving this section feeling rather tacked on at the end. A more detailed discussion of why the ice nucleation studies are relevant would be beneficial. The introduction should also include more context on ice nucleation, with a brief explanation of the 2 modes tested and why they differ. The discussions of the results should also compare the two modes and explain the difference in their freezing temperatures.

We thank the reviewer for these valuable comments regarding the motivation, theoretical context, and presentation of our ice nucleation (IN) experiments. We fully agree that providing a clear atmospheric rationale in the Introduction is essential for contextualizing these measurements. In response to this feedback, we will thoroughly update both Section 1 (Introduction) and the presentation of our IN results in the revised manuscript. In Section 1 (Introduction), we will explicitly articulate the scientific objective of evaluating the ice-nucleating ability of oleamide particles—namely, to assess whether this insoluble organic aerosol can influence cloud microphysics, wet deposition efficiency, and climate feedbacks during long-range transport. In evaluating the reviewer's suggestion to compare freezing modes, we carefully re-examined our full IN dataset. Both freezing modes initially tested yielded identical conclusions: oleamide particles exhibited negligible ice nucleation activity, with freezing temperatures nearly indistinguishable from the pure water baseline, in sharp contrast to positive reference controls such as K-feldspar. Because both modes demonstrate that oleamide is IN-inactive, retaining the contact freezing data adds unnecessary complexity without providing additional scientific insight. Therefore, to present a more focused and concise manuscript, we will remove the contact freezing descriptions and results, focusing exclusively on immersion freezing in the revised manuscript as the widely accepted standard mode in atmospheric ice nucleation studies.

Comment R2-11

For ice nucleation methodology, the first paragraph of Section 4.3 should be moved into the methods section (2.6). I would've also appreciated a figure that showed both set ups used for investigating each mode (this could go in the SI), highlighting the differences in the ways these modes are investigated.

We completely agree that relocating methodological details out of the Discussion section will enhance the logical flow and that providing a schematic diagram in the Supplementary Information will improve experimental clarity and reproducibility. We will move the first paragraph of Section 4.3 (Discussion) into Section 2.6 (Methods), ensuring that all experimental and technical procedures are consolidated within the Methods section. Adding Experimental Setup Schematic to SI: We will add a detailed schematic diagram illustrating our ice nucleation setup to the Supplementary Information. As noted in our previous response regarding the streamlined scope of our ice nucleation studies, because the revised manuscript focuses exclusively on the immersion freezing mode, this figure will detail the specific experimental configuration, droplet array arrangement, and temperature-control system utilized for the immersion freezing assays.

Comment R2-12

There are some issues in the discussion of the ice nucleation results. Notably, Schiffman et. al., 2025 (as referenced by the authors) states that contact freezing is thought to be most effective heterogeneous mode of ice nucleation at the highest temperatures, depending on the nucleating particle. So why are the contact mode freezing temperatures in this study lower than immersion mode? An explanation of difference in results for the different modes should be added. This would also be improved by comparing the freezing temperatures to other literature values in lines 261 and 266.

As detailed in our response to Comment R2-10, a thorough re-examination of our initial comparative dataset revealed that both freezing modes consistently demonstrated the same core finding: oleamide particles possess very weak ice nucleation ability, exhibiting freezing behavior close to that of background ultrapure water, in sharp contrast to highly active

mineral dust controls such as K-feldspar. Because fully addressing the anomalous temperature offset in contact freezing without speculative microphysical assumptions proved difficult and risked distracting from the central focus of the study, we will omit the contact freezing dataset in the revised manuscript and restrict our analysis solely to immersion freezing.

In droplet freezing experiments, absolute freezing temperatures are inherently sensitive to setup-specific parameters, including droplet volume, cooling rate, substrate surface properties, and the baseline purity of the ultrapure water used. Because the background freezing temperature of ultrapure water blanks varies across different cold-stage systems reported in the literature, a direct numerical comparison of raw freezing temperatures between different experimental setups can be scientifically misleading (Hartmann et al., 2019; Schrod et al., 2019; Tobo et al., 2016). To maintain analytical rigor, we will evaluate the ice-nucleating efficiency of oleamide strictly relative to our internal ultrapure water blank and positive reference standard (K-feldspar) measured under identical experimental conditions within the exact same setup.

In Section 4 (Discussion) of the revised manuscript, we will clarify this relative evaluation framework. We will explicitly highlight that because the freezing temperature spectrum of oleamide-containing droplets is indistinguishable from our setup's ultrapure water background control, oleamide does not exhibit significant heterogeneous ice-nucleating activity under mixed-phase cloud conditions.

Hartmann, M., Blunier, T., Brügger, S. O., Schmale, J., Schwikowski, M., Vogel, A., Wex, H., and Stratmann, F.: Variation of Ice Nucleating Particles in the European Arctic Over the Last Centuries, *Geophysical Research Letters*, 46, 4007–4016, <https://doi.org/10.1029/2019GL082311>, 2019.

Schrod, J., Kleinhenz, D., Hörhold, M., Erhardt, T., Richter, S., Wilhelms, F., Fischer, H., Ebert, M., Twarloh, B., Della Lunga, D., Jensen, C. M., Curtius, J., and Bingemer, H. G.: Ice-nucleating particle concentrations of the past: insights from a 600-year-old Greenland ice core, *Atmos. Chem. Phys.*, 20, 12459–12482, <https://doi.org/10.5194/acp-20-12459-2020>, 2020.

Tobo, Y.: An improved approach for measuring immersion freezing in large droplets over a wide temperature range, *Sci Rep*, 6, 32930, <https://doi.org/10.1038/srep32930>, 2016.

Comment R2-13

Other issues with the ice nucleation discussion arise in lines 271 & 275, which contradict each other but reference the same paper. There is a misunderstanding of the conclusions of DeMott et. al., 2018, which discusses the difference in fatty acid vs long chain alcohol monolayers and the requirement for crystalline fatty acid particles to promote heterogeneous ice nucleation. The discussion in lines 269 – 280 needs re-writing once a better evaluation of the literature has been conducted. DeMott et. al., 2018 is also not included in the reference list - this needs to be added. I have referenced below the paper I believe you intended to reference but DeMott has several publications from this year so this will have to be double checked.

We fully acknowledge that our original interpretation of DeMott et al. (2018) was inaccurate regarding the microphysical conditions required for organic monolayers versus crystalline particles to act as ice-nucleating agents. In response, we will completely rewrite the discussion in lines 269–280 of Section 4 (Discussion) to ensure an accurate, rigorous evaluation of the literature. We will clarify the fundamental distinction between different long-chain organic structures. While long-chain alcohols can organize water molecules into ice-like structures via ordered, compressed monolayers, fatty acids and long-chain primary amides typically require a crystalline solid state with high surface packing density to promote heterogeneous ice nucleation. We will explain that in aqueous environments, the *cis*-double bond in oleamide induces a structural "kink" that prevents tight intermolecular packing, favoring a liquid-expanded monolayer phase over a rigid crystalline structure. This disordered state lacks the surface pressure and crystallographic template needed to lower the thermodynamic barrier for ice crystallization, explaining its observed IN-inactivity. We will add the complete, verified citation for DeMott et al. (2018) to the Reference list and audit all

associated citations across the manuscript to ensure complete bibliographical accuracy

Comment R2-14

Finally, discussion of the ice nucleation results should include how the deduced low ice nucleation ability informs our understanding of oleamide and its use as an ice core proxy and how these results might influence future research.

We thank the reviewer for this excellent suggestion to explicitly connect our ice nucleation findings to atmospheric transport dynamics, paleoclimate proxy fidelity, and future research directions. In the revised manuscript, we will add a dedicated paragraph in Section 4 (Discussion) to thoroughly discuss how oleamide's low ice-nucleating efficiency enhances its utility as a long-range paleoclimate tracer and influences Arctic climate feedback. Specifically, we will incorporate the following revised text into Section 4:

"The finding that oleamide is water-insoluble and possesses exceptionally low ice-nucleating activity (INA) has profound implications for both its atmospheric residence time and its preservation within ice sheet archives. Because oleamide particles do not act as efficient heterogeneous ice nuclei (and lack hygroscopic growth without extensive photochemical aging), they are largely resistant to cloud-mediated wet scavenging processes such as immersion freezing, rainout, and in-cloud nucleation. This low scavenging efficiency minimizes in-transit removal, enabling insoluble oleamide microparticles to undergo efficient, long-range atmospheric transport from regional biomass burning source regions to the Greenland Ice Sheet. Furthermore, from an atmospheric radiative standpoint, the inability of oleamide particles to seed mixed-phase clouds implies that they do not induce cloud-mediated cooling effects during transport, maintaining direct aerosol–radiation interactions consistent with biomass burning aerosol dynamics observed in other regimes (e.g., Zhang et al., 2023). Consequently, oleamide represents a uniquely resilient, insoluble organic proxy whose presence in ice cores reflects long-range atmospheric transport efficiency rather than localized precipitation scavenging artifacts."

Zhang, A., Wang, Y., and Zou, Y.: Positive feedback to regional climate enhances African wildfires, *iScience*, 26, 108533, <https://doi.org/10.1016/j.isci.2023.108533>, 2023.

Comment R2-15

Additional Figure(s):

I suggest the inclusion of a map in the methods section which identifies the location of the ice core in Greenland and the regions affected by the “*major North American forest fire events*” which are mentioned several times in the paper. The wildfire events should be labelled with the year they occurred and thus the ice core sample they influence. This would provide useful context and expand on the currently vague description of these wildfires which are key to this study.

In the revised manuscript, we will incorporate a new multi-panel map figure. To establish the scientific foundation for this map, we have synthesized prior Greenland ice core wildfire reconstruction literature in the table below. In the revised manuscript, we will compile these records and project these regional fire event horizons.

Summary of Forest Fire Event Years Recorded in Greenland Ice Cores

Ice Core Site	Region in Greenland	Reference Paper	Detected Forest Fire Event Years	Prominent / Major Fire Event Years
Tunu	Northeast Greenland	Grieman et al. (2018)	1863, 1888, 1908, 1994–1995, 2012	1863 (largest signal), 1888, 1908, 2012
NEEM	North Greenland	Legrand et al. (2016)	1863, 1921, 1923, 1929, 1937, 1941,	1863, 1950, 1980–1981

Ice Core Site	Region in Greenland	Reference Paper	Detected Forest Fire Event Years	Prominent / Major Fire Event Years
			1950, 1961, 1973, 1980–1981	<i>(strongest peak in NEEM-2011-S1)</i>
Summit	Central Greenland	Legrand et al. (2016)	Ice core records: 1839, 1863, 1888, 1908, 1923, 1929, 1937/1938, 1941, 1950, 1961, 1980/1981 Direct observations: 1993, 1994	1863, 1908, 1961, 1980/1981, 1994
D4	West-Central Greenland	McConnell et al. (2007)	1806–1808, 1854, 1863–1868, 1888–1894, 1908, mid-1930s–1940s, 1980s–early 1990s	1854 <i>(highest monthly BC peak)</i> , 1863–1868, 1908 <i>(highest annual BC peak)</i>
Site A / Eurocore	Central Greenland	Savarino and Legrand (1998)	1853, 1896, 1908, 1961 <i>(Active period: 1830–1930)</i>	1853, 1896, 1908
SE-Dome	Southeast Greenland	Parvin et al. (2019)	1961, 1964, 1994, 1998, 2003, 2010, 2013, 2014 <i>(Reconstruction period: 1958–2014)</i>	1964 <i>(largest Levoglucosan peak)</i> , 1961, 1998, 2013, 2014 <i>(highest DA peak)</i>

Grieman, M. M., Aydin, M., McConnell, J. R., and Saltzman, E. S.: Burning-derived vanillic acid in an Arctic ice core from Tunu, northeastern Greenland, *Clim. Past*, 14, 1625–1637, <https://doi.org/10.5194/cp-14-1625-2018>, 2018.

Savarino, J. and Legrand, M.: High northern latitude forest fires and vegetation emissions over the last millennium inferred from the chemistry of a central Greenland ice core, *J. Geophys. Res.*, 103, 8267–8279, <https://doi.org/10.1029/97JD03748>, 1998.

Legrand, M., McConnell, J., Fischer, H., Wolff, E. W., Preunkert, S., Arienzo, M., Chellman, N., Leuenberger, D., Maselli, O., Place, P., Sigl, M., Schüpbach, S., and Flannigan, M.: Boreal fire records in Northern Hemisphere ice cores: a review, *Clim. Past*, 12, 2033–2059, <https://doi.org/10.5194/cp-12-2033-2016>, 2016.

Parvin, F., Seki, O., Fujita, K., Iizuka, Y., Matoba, S., Ando, T., and Sawada, K.: Assessment for paleoclimatic utility of biomass burning tracers in SE-Dome ice core, Greenland, *Atmospheric Environment*, 196, 86–94, <https://doi.org/10.1016/j.atmosenv.2018.10.012>, 2019.

Comment R2-16

Missing reference(s):

There are also 2 references I feel are missing from this paper. Primarily, Mazur et. la.,

2020 who identified oleamide as a major biomass burning marker in Arctic snow and discuss a range of potential sources. Also, Wang et al. 2017 who identified oleamide in sediments and highlight its potential for paleoreconstruction. These studies are very relevant to the work presented here and have to be included in the introduction and / or discussion.

We sincerely thank the reviewer for recommending these highly relevant and valuable studies. We completely agree that incorporating these references substantially enriches the theoretical background and discussion regarding oleamide as a biomarker. In the revised manuscript, we will incorporate and cite both Mazur et al. (2020) and Wang et al. (2017) in Section 1 (Introduction) and/or Section 4 (Discussion).

Comment R2-17

Other line-by-line comments are as follows:

Title – switch out ‘the ice core’ for ‘an ice core’ as other southeastern Greenland ice cores exist.

We will correct the title to ‘an ice core’.

Comment R2-18

32 – INPS do more than just promote cloud formation. Restructure the sentence and expand on their influence on cloud microphysics.

We will restructure this sentence and will add a paragraph afterward to elaborate on the importance of INP research.

Comment R2-19

45 – how are these species more source specific? Expand.

We will correct this sentence on how specific these indicators are in the revised manuscript. We will add the sentence that “specific tracers such as vanillic acid, levoglucosan, and dehydroabietic acid are indicators specifically designed for forest fire events, e.g., Levoglucosan is formed from the pyrolysis of carbohydrates such as starch and cellulose (Zennaro et al., 2014; Legrand et al., 1992, 2016; Parvin et al., 2019).”

Legrand, M., McConnell, J., Fischer, H., Wolff, E. W., Preunkert, S., Arienzo, M., Chellman, N., Leuenberger, D., Maselli, O., Place, P., Sigl, M., Schüpbach, S., and Flannigan, M.: Boreal fire records in Northern Hemisphere ice cores: a review, *Clim. Past*, 12, 2033–2059, <https://doi.org/10.5194/cp-12-2033-2016>, 2016.

Parvin, F., Seki, O., Fujita, K., Iizuka, Y., Matoba, S., Ando, T., and Sawada, K.: Assessment for paleoclimatic utility of biomass burning tracers in SE-Dome ice core, Greenland, *Atmospheric Environment*, 196, 86–94, <https://doi.org/10.1016/j.atmosenv.2018.10.012>, 2019.

Zennaro, P., Khrwald, N., McConnell, J. R., Schüpbach, S., Maselli, O. J., Marlon, J., Vallelonga, P., Leuenberger, D., Zangrando, R., Spolaor, A., Borrotti, M., Barbaro, E., Gambaro, A., and Barbante, C.: Fire in ice: two millennia of boreal forest fire history from the Greenland NEEM ice core, *Clim. Past*, 10, 1905–1924, <https://doi.org/10.5194/cp-10-1905-2014>, 2014.

Comment R2-20

49/50 – NO₃⁻ redistribution - how does high accumulation prevent this? Clarify for the reader.

We will explain the reason for less NO₃⁻ redistribution in the revised manuscript. We will add the sentence that “because the time required for photodecomposition on snow surface is extremely short (Zatko et al., 2016)”.

Zatko, M., Geng, L., Alexander, B., Sofen, E., and Klein, K.: The impact of snow nitrate photolysis on boundary layer chemistry and the recycling and redistribution of reactive nitrogen across Antarctica and Greenland in a global chemical transport model, *Atmos. Chem. Phys.*, 16, 2819–2842, <https://doi.org/10.5194/acp-16-2819-2016>, 2016.

Comment R2-21

53 – ‘insoluble particles are hypothesised to be more stable’ – this statement should be backed up by a reference.

We will add the references of Avak et al., (2019) and Baccolo et al., (2021) in the revised manuscript.

Avak, S. E., Trachsel, J. C., Edebeli, J., Brüttsch, S., Bartels-Rausch, T., Schneebeli, M., Schwikowski, M., and Eichler, A.: Melt-Induced Fractionation of Major Ions and Trace Elements in an Alpine Snowpack, *JGR Earth Surface*, 124, 1647–1657, <https://doi.org/10.1029/2019JF005026>, 2019.

Baccolo, G., Delmonte, B., Di Stefano, E., Cibir, G., Crotti, I., Frezzotti, M., Hampai, D., Iizuka, Y., Marcelli, A., and Maggi, V.: Deep ice as a geochemical reactor: insights from iron speciation and mineralogy of dust in the Talos Dome ice core (East Antarctica), *The Cryosphere*, 15, 4807–4822, <https://doi.org/10.5194/tc-15-4807-2021>, 2021.

Comment R2-22

62 – Expand on how the age scale was derived for this ice core. Explain the methods used.

We will clearly state the source of the timescale (Kawakami et al., 2023; Hamamoto et al., 2026) in the revised text.

Hamamoto, S., Iizuka, Y., Aizawa, T., Kawakami, K., Matsumoto, M., Matoba, S., Bong, H., Yoshimura, K., Okazaki, A., Dietrich, L. J., Steen-Larsen, H. C., and Uemura, R.: Isotope-Based Age Scale with Multi-Month Precision for the SE-Dome II Ice Core from Southeast Greenland Over the Past 150 Years, <https://doi.org/10.5194/egusphere-2026-3602>, 9 July 2026.

Kawakami, K., Iizuka, Y., Sasage, M., Matsumoto, M., Saito, T., Hori, A., Ishino, S., Fujita, S., Fujita, K., Takasugi, K., Hatakeyama, T., Hamamoto, S., Watari, A., Esashi, N., Otsuka, M., Uemura, R., Horiuchi, K., Minowa, M., Hattori, S., Aoki, T., Hirabayashi, M., Kawamura, K., and Matoba, S.: SE-Dome II Ice Core Dating With Half-Year Precision: Increasing Melting Events From 1799 to 2020 in Southeastern Greenland, *JGR Atmospheres*, 128, e2023JD038874, <https://doi.org/10.1029/2023JD038874>, 2023.

Comment R2-23

64 – enables = enabled

We will correct the word in the revised manuscript.

Comment R2-24

81 - ‘reagent compounds’ needs changing to ‘reference compounds’.

We will correct the words to ‘reference compounds’.

Comment R2-25

94 – Why were these 2 dates chosen over others? Expand on this decision.

The selection of the 1908 and 1911 layers for SEM-EDS analysis was conducted as a representative initial screening to evaluate particle elemental compositions in key depth horizons, rather than an arbitrary choice.

Comment R2-26

136/7 – Raman spectra for all unidentified particles are described as previously unreported in ice cores. Raman spectra for black carbon (Unidentified Particle 2) in ice cores have been reported (e.g. Eichler 2019).⁶ Either provide more clarity as to how these results are novel or re-evaluate the conclusions with proper literature assessment.

Thank you for pointing that out. We have missed the previous research on black carbon (e.g. Eichler 2019); we apologize for this oversight. We will make significant revisions to the section on the new Raman spectra.

Comment R2-27

163 – Could the RRUFF database spectra used be included in the SI?

We will add the RRUFF database used in this study to SI.

Comment R2-28

183 – If you have standards for these compounds could you test the high BB years for these organics as well to confirm that these signals are from BB events (vs some non-BB years?)

We do not have equipment such as a gas or liquid chromatograph to measure the concentration of organic compounds. So, we cannot confirm that these signals are from BB events in the ice core.

Comment R2-29

213 – This is too strong of a claim given the above discussion. Edit accordingly.

We will entirely remove from the discussion claiming a statistically significant correlation between NH₄⁺ concentrations and oleamide abundance.

Comment R2-30

271 – Why would monolayers prevent ice nucleation? Provide an explanation (or a hypothesis of one).

To clarify the atmospheric and physical significance of monolayer formation in Section 4 (Discussion), we will explicitly detail the background of organic monolayer-induced ice nucleation. Specifically, while condensed organic monolayers can template ice nucleation by mimicking the hexagonal ice lattice, the structural kink introduced by oleamide's cis-double bond stabilizes an expanded, low-surface-pressure phase. This prevents ordered surface alignment and explains its negligible ice-nucleating activity in our immersion freezing experiments.

Comment R2-31

References:

C. P. Vega, V. A. Pohjola, D. Samyn, R. Pettersson, E. Isaksson, M. P. Björkman, T. Martma, A. Marca and J. Kaiser, *Journal of Geophysical Research: Atmospheres*, 2015, **120**, 313–330.

Z. R. Schiffman, K. A. McMillan, D. M. Johnson, R. V. Gough, R. D. Davis, S. D. Brooks and M. A. Tolbert, *J. Phys. Chem. A*, 2025, **129**, 7482–7495.

P. J. DeMott, R. H. Mason, C. S. McCluskey, T. C. J. Hill, R. J. Perkins, Y. Desyaterik, A. K. Bertram, J. V. Trueblood, V. H. Grassian, Y. Qiu, V. Molinero, Y. Tobo, C. M. Sultana, C. Lee and K. A. Prather, *Environ. Sci.: Processes Impacts*, 2018, **20**, 1559–1569.

D. M. Mazur, T. B. Latkin, D. S. Kosyakov, A. Y. Kozhevnikov, N. V. Ul'yanovskii, A. G. Kirilov and A. T. Lebedev, *Environmental Pollution*, 2020, **265**, 114885.

J. Wang, B. R. T. Simoneit, G. Sheng, L. Chen, L. Xu, X. Wang, Y. Wang and L. Sun, *Sci Rep*, 2017, **7**, 14667.

J. Eichler, C. Weikusat, A. Wegner, B. Twarloh, M. Behrens, H. Fischer, M. Hörhold, D. Jansen, S. Kipfstuhl, U. Ruth, F. Wilhelms and I. Weikusat, *Front. Earth Sci.*, DOI:10.3389/feart.2019.00020.

Thank you very much for recommending these great papers. All of these papers will be cited in the revised manuscript.

Finally, while we have adjusted our tone throughout the manuscript to be appropriately cautious and scientifically rigorous, we would like to respectfully highlight the foundational value and novelty of our findings. To the best of our knowledge, this study provides the first direct, single-particle identification of oleamide (at least a kind of fatty amide) within an ice core matrix, achieved through a complementary multi-spectroscopic approach (Raman, FT-IR, and SEM-EDS). By demonstrating that oleamide is prominently enriched in horizons corresponding to historically verified major wildfire events (e.g., 1961, 1980, and 1994), we provide a proof-of-concept that insoluble organic microparticles can serve as novel, complementary paleoclimate indicators. We believe that these extensive revisions resolve

the concerns raised by the reviewer and allow the genuine analytical novelty and importance of our work to stand on a solid scientific foundation.