

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.

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.

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.

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).

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.

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.

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.

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.

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.

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.

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.

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.² 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.

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.³ 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.³ 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.

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.

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.

Missing reference(s):

There are also 2 references I feel are missing from this paper. Primarily, Mazur et. al., 2020 who identified oleamide as a major biomass burning marker in Arctic snow and discuss a range of potential sources.⁴ Also, Wang et. al. 2017 who identified oleamide in sediments and highlight its potential for paleoreconstruction.⁵ These studies are very relevant to the work presented here and have to be included in the introduction and / or discussion.

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.

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

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

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

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

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

64 – enables = enabled

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

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

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.

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

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?)

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

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

References:

- 1) 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.
- 2) 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.
- 3) 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.
- 4) 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.
- 5) J. Wang, B. R. T. Simoneit, G. Sheng, L. Chen, L. Xu, X. Wang, Y. Wang and L. Sun, *Sci Rep*, 2017, **7**, 14667.
- 6) 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.