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Reply to Gañán-Calvo: Aerosol production from the bursting of submillimeter bubbles

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Our recent paper (1) demonstrates that the major pathway to produce submicron drops from bursting submillimeter bubbles depends crucially on the gas density, and justifies how the flapping mechanism of the bubbles' cap as they burst might be responsible for it. Gañán-Calvo's (2) comment raises concerns about this paper. His arguments are incorrect.

First, Gañán-Calvo (2) argues that ref. 1 does not rule out jet drop formation from a cascade of daughter bubbles. On the contrary, ref. 1 has precisely addressed this issue. Bird et al. (3) report that the average daughter bubble size is roughly 0.08 times the parent bubble size. If the bubbles with cap radii of curvature $R = 73$, 137, 199, 870, and 1,080 μm do produce daughter bubbles, based on the relation determined by Berny et al. (4), the theoretical dried sizes of jet drops from these daughter bubbles would be 24, 52, 83, 532, and 700 nm. However, the size distributions in ref. 1's figure 2 peak at 35, 50, 76, 66, and 71 nm, respectively. Gañán-Calvo argues that the first three values are similar between these two pairs. However, the differences between the last two values (532 nm vs. 66 nm, 700 nm vs. 71 nm) are very substantial and cannot be explained by jetting from a cascade of daughter bubbles. Moreover, there is no reason to believe that the daughter bubble mechanism would only work for bubbles with $R = 73$, 137, and 199 μm, and not for bubbles with $R = 870$ and 1,080 μm. The truth is that the droplets are formed by another mechanism, which involves the gaseous atmosphere, as ref. 1's observations demonstrate.

Second, Gañán-Calvo (2) argues that the number of aerosols produced from single bubbles with $R = 73$, 137, and 199 nm is much lower than the theoretical number of jet drops. To be clear, these measured numbers were obtained using a condensational particle counter (CPC), not scanning mobility particle sizer or aerodynamic particle sizer as Gañán-Calvo claims. Ref. 1 is concerned with submicron drops. Thus, we intentionally put an impactor before CPC to remove the micrometer-sized drops. Based on Berny et al. (4), the jet drop diameters produced from bubbles with $R = 73$, 137, and 199 nm would be 2.3, 5.0, and 8.0 μm, these sizes being readily removed by the impactor. Therefore, these micrometer-sized jet drops were not measured at all. There is no artifact.

We understand Gañán-Calvo (2)'s interest in jet drops but would like to recall down-to-earth facts: Air drag exerted on these ejected micrometer-sized drops in this way prevents them from rising appreciably high above the surface, before they settle back down(5). How much a fraction of these drops can penetrate the free air to contribute to the spray aerosol is still an open question, in need of a precise investigation (6).

Finally, let us mention that the admitted value of the mean free path in the air at ambient conditions is ∼65 nm rather than 143 nm. But that is, admittedly, a detail.

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The authors declare no competing interest.

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1. X. Jiang, L. Rotily, E. Villermaux, X. Wang, Submicron drops from flapping bursting bubbles. *Proc. Natl. Acad. Sci. U.S.A.* **119**, e2112924119 (2022).
2. A. M. Gañán-Calvo, The complexity of aerosol production from bubble bursting. *Proc. Natl. Acad. Sci. U.S.A.*, 10.1073/pnas.2208770119 (2022).
3. J. C. Bird, R. de Ruiter, L. Courbin, H. A. Stone, Daughter bubble cascades produced by folding of ruptured thin films. *Nature* **465**, 759–762 (2010).
4. A. Berny, S. Popinet, T. Séon, L. Deike, Statistics of jet drop production. *Geophys. Res. Lett.* **48**, e2021GL092919 (2021).
5. D. C. Blanchard,, The electrification of the atmosphere by particles from bubbles in the sea. *Prog. Oceanogr.* **1**, 73–202 (1963).
6. X. Wang *et al.*, The role of jet and film drops in controlling the mixing state of submicron sea spray aerosol particles. *Proc. Natl. Acad. Sci. U.S.A.* **114**, 6978–6983 (2017).