Anwers to anonymous Referee #1

General Authors answer: We would like to thank the anonymous reviewer (Reviewer #1) for taking the time to carefully examine our paper. We have moved parts from the 'Discussion and Conclusions' section to the 'Results' section, as suggested. Three paragraphs were moved from the former 'Discussion and Conclusions' section to the 'Results' section, where they belong. The former 'Discussion and Conclusions' section is now just the Conclusion section. A few sentences in the conclusion had to be modified slightly to ensure it could stand alone. Modifications are highlighted in the document (bold blue). Some typos and minor syntax errors have also been corrected in the text and appear in bold blue in the revised version of the document.

Minor issues:
Introduction: I suggest providing a supporting discussion why the 1 and the 2 mm drop sizes have been chosen.

Authors answer: In the case of deformable drops, this paper considered drops of 1.39 mm and 2 mm in freefall in ground-based atmospheric conditions (Reynolds numbers of 500 and 876, respectively). These drop sizes were chosen to enable comparison with existing literature data on capture efficiency (Lai et al., 1978; Querel et al., 2014) and drop dynamics (terminal velocity, mean axis ratio, and oscillation frequency: Beard, 1976; Szakall et al., 2010). Obviously, in the future, results would benefit from being enriched with larger drop results (up to 4-6 mm).

- We propose to add the following justification to the introduction of the paper : **Initial text** L47: “Secondly, simulations are performed for flow regimes with drop oscillation and deformation (drop Reynolds of 500 and 876), for which -- to our knowledge -- no simulation results of aerosol particle collection efficiency are available in the literature. For these regimes, the simulation results for the continuous phases are validated (…) “

  New text proposal :"Secondly, simulations are performed for flow regimes with drop oscillation and deformation, for which -- to our knowledge -- no simulation results of aerosol particle collection efficiency are available in the literature. We chose the case of 1.39 mm and 2 mm diameter water drops falling freely in ground-based atmospheric conditions (drop Reynolds numbers of 500 and 876),for which experimental validation data is available. For these situations, the simulation results for the continuous phases are hence validated (…)"

Line 194: Did the authors observe multiple oscillation modes, or only the axisymmetric one? Is that at all possible to observe the different oscillation modes in the numerical model?
Authors answer: It is a limitation, but in this work, we did not analyze in detail the oscillations modes of the drop, since the primary objective focused on aerosol capture efficiencies. However, we would firstly like to thank the reviewer for suggesting this idea, which we had not previously considered. It offers the potential for more advanced physical validation of droplet dynamics and has very real applications in atmospheric science. Clearly, the simulation data

enables all oscillation modes to be observed and quantified, provided that the simulated physical time is long enough to capture several periods of the slowest oscillation mode once the drop has reached its terminal pseudo-stationary regime. Indeed, simulations predict the time-resolved 3D surface of the drop, the dynamics of which can be analysed in detail to highlight the existence of multimode oscillations. However, this data was not saved in the present study, so simulations must be run again to extract suitable (and very large) 3D data. Nevertheless, a close examination of the variation in the axis ratio in Figure 10 when the drop has reached its mean terminal velocity reveals that the amplitude of the axis ratio oscillations appears to be periodically modulated. This suggests the presence of multiple oscillation modes, as observed by Szakall et al. (2009, DOI: 10.1175/2008JAS2777.1, Figure 3). For this reason, we believe that the simulation probably predicts multiple oscillation modes, but we did not quantify them.

Line 206: Can the aerosol of 2 mm size be considered as one with negligible fall speed wrt the drop?
Authors answer: Here, we assume that the reviewer is referring to aerosols with the largest considered aerodynamic diameter, i.e. 20 micrometres (not 2mm). Indeed, aerosols of this size have a negligible sedimentation velocity compared to that of the drop (typically 100 to 500 times less for the drop diameters considered in the present paper). However, note that even if the computations are performed in the assumed Galilean coordinate system of the moving drop, changing the coordinate system of the Navier–Stokes equation does not affect particle acceleration due to gravity. Hence, the model fully accounts for the difference in fall velocity between the aerosols and the drop.

Line 244: Are there any old literature results of particles collection efficiencies? Analytical or experimental?
Authors answer: Indeed, the connection with other available literature is probably not obvious. An overview of the available literature results for these drop sizes (Re = 30, 70) and similar flow regimes (i.e. laminar axisymmetric) would be as follows:

1) Analytical :
   - so-called “model II” or “flux model” by H.R. Pruppacher team (P. K. Wang, S. N. Grover and H. R. Pruppacher 1978) : Brownian motion only, no inertia, “moderate” Reynolds number in the axisymmetric laminar flow regime ;
   - Slinn 1977 : largely inaccurate semi-empirical model (does not fit well the other numerical or experimental work listed here).
2) Simulations:
   - Grover et al. work, beginning with Beard and Grover 1974 and successive papers (so-called “model I” by H.R. Pruppacher team) : Lagrangian modelling without Brownian motion ;
   - Wang et al. 2016 (Study on inertial capture of particles by a droplet in a wide Reynolds number range) : inertial aerosol only, no Brownian motion ;
   - Yu et al. 2022 (Effects of rotation on collection characteristics of fine particles by droplets) : only one inertial point at Re=72, aerodynamic diameter 3.8 micrometer ;
   - Cherrier et. al. 2016 : unify both experimental and numerical results with fair accuracy for Reg<100 and Brownian/inertial particles : for this reason, data of Cherrier et al. 2016 were chosen as reference for the validation of the current model, without repeating other available results that are similar.
3) Experimental:
   - Starr and Mason 1966 : only inertial particles and pendant drop technique (2 aerodynamic diameter > 4 micrometers)

> - Horn et al 1988 (Collection efficiency of aerosol particles by raindrops) : only 2 points, aerodyn. diam. of 0.7 and 3.3 micrometers at Reg=66.
> - Yu et al. 2022 (Effects of rotation on collection characteristics of fine particles by droplets) : only one experimental point at Re=72, aerodynamic diameter 3.8 micrometer (inertial particle, no influence of Brownian motion).
>
> For the laminar, axisymmetric flow regime past a drop, the results of Cherrier et al. (2016) thus cover the widest range of Reynolds numbers and aerodynamic diameters. They link the purely Brownian and the purely inertial behaviours of the aerosol particles and retrieve data from other authors (experimental, theoretical, or numerical) (Cherrier et al., 2016; Depée et al., 2021). For this reason, it was chosen as a reference in the present paper. As this justification is not clearly evident in the paper, we propose adding the following sentence after line 243:
>
> "As stated in the introduction, the simulation data of Cherrier et al. (2016) were chosen as the reference, as they cover the widest range of Reynolds numbers and aerosol aerodynamic diameters. This data links the purely Brownian and purely inertial behaviours of the aerosol particles, and retrieves data from other authors, either analytical, numerical of experimental (Cherrier et al., 2016; Depée et al., 2021)"

Line 287: In my opinion, the deformation of a 2 mm drop is still not really substantial
Authors answer: Indeed, we should remain objective. We therefore propose reformulating the sentence "In this flow regime, the droplets experience substantial deformations, oscillations, and vortex releases" as "In this flow regime, the droplets experience noticeable deformations, oscillations, and vortex releases (see Table 2)".

Figure 10, axis ratio: in Table 1 the axis ratio values are referred to data from Szakall et al. 2010. In Fig. 10 they are from Beard et al. 1991. Is there any difference?

Authors answer: The information in Table 1 is actually imprecise. Szakall et al. (2010) collected literature information and incorporated the values of Beard et al. (1991) (drop of 1.39 mm, used in Fig. 10) and Thurai et al. (2009) (drop of 2 mm). We propose modifying the headers in Table 2 for clarity (see revised paper).

Line 303: Are the fluctuations seen in the model at the beginning of the simulations of physical or of numerical origin?
Author's answer: We could say that they would be physical if the initial conditions given in the simulation were replicated in an experiment; in other words, these fluctuations are not the result of a numerical error. However, in the natural process of a raindrop falling, this initial condition does not reflect the reality of raindrop formation. Nevertheless, we demonstrate that the steady-state free-fall situation is independent of this initial condition, whether it is physical or not, and that the predicted free-fall dynamics of the drop is consistent with the literature - therefore rendering the fluctuation event inconsequential.

Line 400: Is it possible to provide an estimation on the magnitude of the phoretic forces (probably it would be just enough to move the corresponding discussion from the Conclusions section to this line).

Authors answer: We propose to do as suggested: in line also with your initial general suggestion, we have moved the discussion paragraphs from the former "discussion and conclusion" section to the results section.

Anwers to anonymous Referee #2

General Authors answer: We would like to thank Reviewer 2 for reviewing our article proposal and providing comments. Below, we have summarized their feedback and addressed each point individually. As a general comment, we would like to point out that this paper aims to present an initial numerical study of the capture of aerosols by deformable droplets. To the best of our knowledge, this is the first study of its kind in the literature. While it is certainly imperfect, it will be further developed by either us or the research community, possibly based on the areas for improvement identified in the paper. Since the initial draft was submitted, the article has been revised based on the comments provided by Reviewer 1. We believe that this version already addresses some of the concerns raised by Reviewer 2.

Bullet points identified by Reviewer 2 :

- ***1)*** *discrepancy observed in the predicted collection efficiency : the deviations from available experimental data are not sufficiently analyzed or explained (particularly in the deformable droplet regime).*
    - *the sensitivity of the results to numerical choices, particularly interface treatment, interpolation schemes near the liquid–gas boundary, and grid resolution, has not been adequately assessed. The discussion of these aspects remains too limited for a DNS-based study of this complexity.*
    - *A more systematic error and convergence analysis would significantly strengthen confidence in the reported results.*

Authors' response :

Our impression is that the considerable effort made in the article to explain the discrepancies between simulations and experiments was likely not sufficiently clear in the initial version of the submitted manuscript. Indeed, most of the explanations were not included immediately following the results, but rather in the discussion and conclusion sections. Following the suggestions of Reviewer #1, we believe that the new structure of the article highlights much more clearly the reasoning behind the discrepancies between simulation and measurement, as the discussions are now integrated into the results section. We hope that Reviewer #2 will find, with this new presentation, a more nuanced view of the discrepancies that exist regarding collection efficiency among the various studies on this subject, as no single dataset—whether experimental or numerical—is definitive for many regimes of this three-phase droplet/gas/aerosol flow.

From a general perspective, it is also important to remain objective regarding the significance of the discrepancies between simulation results and reference data, as well as regarding the interpretation of these discrepancies.

In the case of non-deformed drops (Re 30.70), it seems clear to us that the sensitivity of the results to numerical choices is already extensively assessed and that no more verifications are needed. The results are consistent with the state of the art, and convergence tests were conducted with respect to the velocity interpolation scheme and the DNS grid discretization, up to the finest resolution technically feasible for us. These tests reveal a residual uncertainty in E due to numerical approximations, which we estimate to be 1e-3, compared to the results of Cherrier et al. (2016). This residual uncertainty, amounting to one-thousandth of the range of variation of E, is beyond the sensitivity of the available experimental measurements, for reasons we discuss in detail in the article (formerly in the discussion section, now integrated into the results). Furthermore, whether or not this uncertainty is significant is primarily a matter of the application domain, and we believe it is up to the reader to judge whether the results are sufficiently precise for their application, provided that the residual uncertainty is clearly stated.

In the case of deformed droplets (Re=500, 876), while we agree that more effort could be made to highlight the effect of numerical choices on the results, but we do not feel it is objective to say that the deviations from available experimental data are not sufficiently analyzed or explained. The article honestly acknowledges that the simulations do not reproduce the few available experimental results with a discrepancy of less than one order of magnitude for E. The numerous reasons for this are explained in great detail (formerly in the discussion section, now integrated into the results for non-spherical droplets), but are not necessarily quantifiable, particularly with regard to the experiments. One should not hastily assume that the experimental results are reliable, as we explain in detail in the article, which suggests numerous ways to increase the reliability of the reference data for E, both numerically and experimentally. Making these proposals is, moreover, one of the article's objectives and a useful conclusion for the scientific community, as stated in the abstract.
We should mention that two of us participated in former experimental and numerical investigations on this topic in several papers in the literature. As such, we are particularly aware of how difficult it is to obtain reliable E results, particularly in the Greenfield gap, and of the residual sources of error that remain, however carefully the experimental protocol is designed and conducted. The role of humidity control is particularly crucial (for instance Lemaitre et al. (2020) highlight a 100% increase in the collection efficiency for each percentage decrease in the atmospheric relative humidity).

It therefore seems highly unlikely that experimental results without phoretic forces could be obtained that would exactly match the simulated cases. The ideal approach would, of course, be to simulate the entire non-isothermal process, including droplet phase changes and the resulting phoretic forces on the particles. These physical phenomena can be incorporated into the simulations, but they introduce multiple new steps for verifying and validating the results. Since the approach excluding these phenomena is an essential prerequisite, this article should therefore be viewed as a building block—necessary but not final.

In any case, it seems relatively clear to us, based on the numerical convergence tests performed, that minimizing the numerical uncertainties associated with the spatial discretization of the fluid field and the velocity interpolation scheme will not be sufficient to bridge the gap between simulation and experiment for E. These discrepancies must stem primarily from excessive differences between the simulated and experimental situations, as discussed in the article (phoretic and electric forces, uncertainties regarding particle size distributions, and hygroscopic growth of aerosols in the drop boundary layer).

Lemaitre, P., Sow, M., Quérel, A., Dépée, A., Monier, M., Menard, T., & Flossmann, A. (2020). Contribution of phoretic and electrostatic effects to the collection efficiency of submicron aerosol particles by raindrops. Atmosphere, 11(10), 1028.

- ***2)*** *The manuscript overlooks several relevant and recent studies*

Authors' response :

We would like to thank Reviewer #2 for bringing these interesting references to our attention, which we had overlooked. These references deal exclusively with the hydrodynamics of deformable droplets; therefore, the connection to the capture of aerosols by free-falling droplets is not direct, but some of these studies nevertheless provide useful insights into the paper under review. We propose evaluating their suitability as bibliographic references on a case-by-case basis, as outlined below. In summary, our analysis has led us to select one of these four publications for inclusion in our bibliography.

a) Journal of Fluid Mechanics (2026, 1031, A21) (Evaporation of a freely floating droplet in an airstream: effects of temperature, humidity and shape oscillations)
This paper is very recent (March 2026), and we were, of course, unaware of it at the time of our article's submission. This paper focuses on the analysis of the evaporation dynamics of highly deformed droplets, based on experimental measurements, and does not contain information on aerosol capture by droplets in the absence of evaporation, which is the subject of our article. It deals with droplets initially 3 mm to 4.5 mm in diameter (and thus of much greater volume than those of interest here), with very substantial deformations, at the limit of aerodynamic breakup (Re= [1700 2800] We=[3.4 6.2] compared to Re=[30 876] We=[1e-3 1.39] in our case, where We is the Weber number reflecting the relative importance of the incident aerodynamic pressure on the surface tension). In summary, this article, although very interesting, does not contain information that we could use as a suitable bibliographic reference for our purposes. It does, however, report measured oscillation frequencies for free-falling droplets, which eventually reach the diameters of interest to us after evaporation. Nevertheless, these frequencies were already known in the literature and with greater certainty (The paper in the "Journal of Fluid Mechanics" (2026, 1031, A21) does not compare its results with previous measurements - Szakall et al. 2010, Thurai et al. (2009) - and reports substantial discrepancies with respect to theory : see figure 5,6,7 and equation 3.2 of the paper, and Figure 8 that shows that the standard deviation of the droplet oscillation period measurements is typically 25% of the theoretical value - one standard deviation being a low estimate of the discrepancy between theory and experiment.). Thus, we do not believe it is relevant to include it in the bibliography

b) Physics of Fluids (2020, 32, 112105) (Experimental investigation of a nonspherical water droplet falling in air)
This article investigates experimentally the dynamics of initially oblate, prolate, and tilted droplets that are propelled in air using a catapult-type setup with a pneumatic piston-cylinder arrangement and a superhydrophobic coated plate. This study is extremely far removed from the case at hand, as the droplets ejected by the catapult are constantly in an accelerated state, far removed from a free-fall situation such as the one considered here. Moreover, the droplets do not travel far enough to reach terminal velocity. In addition, this study does not address the interactions of the droplet with aerosols; only the dynamic aspects of the droplet's shape are examined. In summary, it seems to us that the connection to the subject of our article is far too tenuous to justify including this article in the bibliography.

c) Theoretical and Computational Fluid Dynamics (2020, 34, 133–144) (A numerical study of a hollow water droplet falling in air)

The proposed article deals with the hydrodynamics of a hollow droplet from a purely numerical perspective; this topic is too far removed from the subject of the paper under review, which deals with the capture of aerosols by droplets. Furthermore, the Eötvös numbers considered in this article are significantly higher (0.004 to 0.01, compared to a maximum of 1.7e-4 in our work), leading to much more pronounced interface instabilities, which makes the situations incomparable. Thus, we do not believe it is relevant to include it in the bibliography

d) Physical Review E (2019, 99, 023107) (=Shape oscillations of a nonspherical water droplet)
The topic and content of this article are much more in line with the focus of our article. This article examines, from a purely numerical perspective, the shape oscillations of a drop in free fall, focusing primarily on the effect of the drop's initial aspect ratio and tilt on the evolution of its dynamics. This study is conducted over a short time scale relative to the time required for a drop to reach its terminal velocity and dynamics (for example, for a drop with a diameter of 5.76 mm discussed in Section III.A. of the article, the simulated physical time is 0.12 s — a dimensionless time of 7 — which corresponds to a dimensionless time of 190 in our dimensionless analysis, that is approximately one-quarter of the simulation time required to reach a steady-state oscillation regime in our very similar configuration. We can also interpret this as a physical interaction length of only 1 m at the fall speed, a length far too short to accurately model the atmospheric behavior of droplets of this size.

We can conclude that, in this study, the results presented are highly dependent on the initial conditions used and are not representative of the behavior of a water droplet in terminal free fall. This is, in fact, what the authors demonstrate, with different oscillation dynamics observed depending on the initialization chosen for the droplet's shape, for the same equivalent spherical diameter. Furthermore, the chosen initializations do not correspond to the situation of a raindrop in free fall, where the droplet's initial shape is not ellipsoidal.
Ultimately, we therefore conclude that none of the configurations simulated in this article corresponds to the terminal falling regime of a droplet, which is the subject of our study.

Furthermore, we can see that the authors do not reproduce the oscillatory behavior observed experimentally, for example in Figure 3, for a drop with a diameter of 5.76 mm as observed by Szakall et al. (2009), an initially spherical droplet does not enter a deformation/oscillation regime, and for the various initializations proposed, neither the average aspect ratios nor the experimental oscillation frequencies of Szakall et al. 2009 are reproduced. This contrasts with our simulations, in which an initially spherical drop spontaneously evolves toward the experimentally observed oscillatory regime, with characteristics very close to the measurements and independent of the chosen initialization.

Following this review, we therefore propose adding this article to our bibliography by inserting the following sentence at the end of section 6.1.2:
"Ultimately, compared to the state of the art in simulating the dynamics of a water droplet in free fall (Balla et al. (2019)), our model successfully reproduces the spontaneous destabilization of an initially spherical water droplet into the oscillatory dynamics observed experimentally (Szakall et al. (2009, 2010))"

- ***3)** Clearer separation between methodological development and physical interpretation*

Authors' response:
We are not sure we understand what Reviewer 2 means. As Reviewer 1 did not comment on these points and the article has been reorganised slightly in response to their comments, we propose leaving this comment pending.

- ***4)** The definition of particle capture based on geometric contact with a deforming interface, require additional justification*

Authors' response:
We identify two aspects (physical and numerical) in the reviewer comment, and we try to address both in the following paragraphs

Physical aspect:
In this study, aerosol particles are considered collected upon establishing geometric contact with the droplet surface, a definition consistent with both experimental observations and theoretical models (see line 97 of the submitted manuscript, section 2.3, "the particles are considered "collected" as long as they enter into geometrical contact with the drop, which is in line with theoretical predictions Wang et al., 2015). While the implicit assumption that aerosol-droplet collision invariably results in particle collection is widely adopted, its validity has been primarily demonstrated for water-soluble aerosols, which constitute the majority of atmospheric particulate matter (Kandler et al., 2007). However, as the present work addresses aerosols of diverse physicochemical properties, a critical reassessment of this assumption is necessary. The generalization of this collection criterion was first proposed by Beard (1974), building upon Weber's (1969) seminal experiments with insoluble, hydrophobic dust particles. Weber's results indicated near-unity adhesion efficiencies under controlled conditions. Subsequent numerical investigations by Wang et al. (2015), employing Newton's laws of motion and accounting for particle wettability, demonstrated that post-impact interface oscillations and particle rebound occur only when the particle's contact velocity exceeds 4 m/s at an air-water interface.
Under atmospheric conditions, such high relative velocities—comparable to the droplet's terminal velocity—are not achieved at the moment of contact. Aerosol particles experience rapid deceleration upon entering the droplet's boundary layer due to their negligible aerodynamic response time (≤ 0.001 s for 20-µm particles). This behaviour is consistent with Weber's (1969) findings and supports the adoption of geometric contact as a robust collection criterion.

Numerical Implementation and Validation:
For simulations involving deformable droplets at elevated Reynolds numbers (Re = 500 and 818), the geometric contact criterion is evaluated using the Level Set function, ϕ, at the particle's centre. As described in Section 3.3.2 (Eq. 17), collection is confirmed if $\phi_p + r_p > 0$ at any timestep during Lagrangian tracking. The accuracy of the ϕ function is therefore paramount for reliable collection assessment.

To validate the interface tracking model (based on level set and VOF coupling - section 2.1), we benchmarked the mean axis ratio, oscillation frequency, and amplitude of deformable droplets (Re = 500 and 818) against established literature data, obtaining satisfactory agreement for all parameters. Furthermore, applying the same methodology to non-deforming droplets at lower Reynolds numbers (Re = 30 and 70) confirmed the absence of spurious deformation, as expected from experimental observations. These results collectively validate our two-step approach: (1) precise interface localization via the ϕ function, and (2) determination of geometric contact for particle collection.

Kandler, K., & Schütz, L. (2007). Climatology of the average water-soluble volume fraction of atmospheric aerosol. Atmospheric Research, 83(1), 77-92.
Weber, E. (1968). Der Einfluss der Benetzbarkeit von Stäuben bei der Naßabscheidung Staub-Reinh. Luft, 28(11), 462-467.
Wang, A., Song, Q., & Yao, Q. (2015). Behavior of hydrophobic micron particles impacting on droplet surface. Atmospheric Environment, 115, 1-8.