1. There are no references in the first two paragraphs; however, it would be desirable to support those introductory summaries. Furthermore, what are the “radioactive releases from a nuclear accident”? Are they of particulate matter? From the point of view of the paper it should be clarified.

Radioactive releases from a nuclear accident are composed by particulate matter and gases. On this paper, we are only interested by particulate matter (Baklanov and Sørensen, 2001). At the line 15, It is added “Radioactive releases consecutive from a nuclear accident are compounded by gases and aerosols. This paper is only focused the second component.”

In order to take into account this remark

Radioactive releases consecutive from a nuclear accident are compounded of both gases and aerosol particles (Baklanov and Sørensen, 2001). This paper is only focused this second component.

2. p. 512, line 3: Inertia is not a phenomenon.

This sentence:
“For the small ones, Brownian motion allows them to leave the streamlines and, for large particles, inertia or interception are phenomena that increase the collection efficiency.”
Is replaced by
“For the small ones, Brownian motion allows them to leave the streamlines and, for large particles, their inertia induces their impaction or interception by the drop.”

3. I have some reservations about the reliability of the experiments. First, it is not described how the sizes of the drops were determined, and how the uncertainties given in Table 1 were calculated.

The sizes of the drops are determined by the shadowgraphy technique. This technique is applied inside the aerosol chamber (figure 2 is modified to show the drop size measurement location).The uncertainties come from the numerical determination of the edge determination of the drop image, the drops oscillations, and from the size distribution of the drops.
A paragraph is added at the end of part 2.1 to clarify the article:

“Both drop diameters and axis ratios of each drop crossing the BERGAME aerosol chamber are directly measured inside of it by processing the shadow images of the drops.

The velocity of each drop is also directly measured inside the aerosol chamber. This measurement is performed by taking a second picture of the shadow of the same drop with a controlled time between these two images. The drop velocity is finally deduced from the ratio between the displacement of the center drop on the two images and the time between these two images. Unfortunately the experimental precision of this method is low."

4. The drop generator is stated to be able to generate monodispersedly distributed drops, but without giving any uncertainty. In Section 2.3 the shadowgraphy technique is mentioned; it is how the drop sizes were determined? If the drops are oscillating, it can lead to some uncertainties in the size determination.

The stability of the drop generator is checked with the help of the shadowgrapy technique applied inside the aerosol chamber, during each experiment (see small paragraph added consecutively to previous question) on each drop passing through the chamber. The equivalent spherical diameter of the drop is deduced from the surface of the shadow, and the axis ratio from the ratio between the smallest axis to the longest one on the shadow. The uncertainty induced by the 2D projection on the camera of the 3D drop is included inside the uncertainties on the drop size presented in table 1 (± 100 µm). The contribution of this projection uncertainty is assumed very small and confirmed by the fact that the drops equivalent spherical diameter measured is very stable from one drop to the others. This is due to the fact that the drops, even non spherical, are axisymmetric (Szakáll et al., 2009, for same range of drop).

5. This theory regarding the incorrect size determination can probably supported by the shifted mode and the one-side skewness of the axis ratio distribution shown in Figure 3.

For the axis ratio determination, the skewness of the drops is resolved with the determination of the drop center, and the determination of the longer and shorter axis throwing this point. The method has been compared to the one used by Szakáll et al., 2009 and Szakáll et al., 2010 and give equivalent results.

6. Furthermore, how had been the terminal velocities determined? Why are the measurement errors so high (+/- 1 m/s)? If one calculates the drop sizes from the drop velocities in Table 1, they found to be between 1.44 and 2.6 mm (for the 2 mm diameter drops), and 2.1 to 3.58 mm for the 2.6 mm drops. Thus, the size uncertainty is very high. Please comment and clarify it by giving also some experimental details.

The drop velocity is determined through the measurement of the displacement of the drop on two consecutives images. In this particular condition, we preferred to focus the precision of the device on the drop size (precision of ± 100 µm), however as the field of view of the camera is relatively small, the drop displacement between the two images is small as well. That induces a relatively small resolution on the velocity measured.

7. Another comment on the experimental setup: The height of the fall shaft is enough for experiments with drops of 2 mm diameter. But it seems to be a little bit too short for the 2.6 mm drops. (see Andsager et al, J. Atmos. Sci., 1999)

The shaft have been considering enough high for the 2.6mm drops due the observation of the good velocity (according Beard, 1976) and relative good axis ratio. But, yes, we certainly are at the limit of this experiment.

8. p. 516, Eq. 2: It is not given, what v(air) in the equation means. I cannot follow the comment right after the equation, namely that the setup is able to simulate raindrops of diameters up to 2.7 mm. Please clarify.

ν_{air} is the kinematic viscosity of the gas around the drop (m²/s). The sentence concerning the Reynolds number has been removed to keep the development continuity.

"In Table 1 the drop Reynolds number is calculated using equation (2).

$$Re = \frac{v_d D_d}{\nu_{air}} \qquad (2)$$

The agreement of these measurements with the literature models ensures the representativeness of the BERGAME set-up to simulate raindrops with a diameter up to 2.7 mm"

<u>**Is replace by :**</u>

"In Table 1 the drop Reynolds number is calculated using equation (2) in which ν_{air} is the kinematic viscosity of the gas around the drop (m²/s).

$$Re = \frac{v_d D_d}{\nu_{air}} \qquad (2)$$

The agreement of these measurements with the literature models ensures the representativeness of the BERGAME set-up, in the drop size range investigated. However beyond the drop size of 2.7 mm, the velocity allowed by this shaft is no more satisfying."

9. Section 2.4.: It is not clear here why the fluorescence properties of the particles are important. Later it will be obvious: because of the applied spectroscopy technique. It would be desirable to mention this here.

The sentence “(which is essential for the spectroscopy technique used latter to measure their concentrations insides the drops)” is added after the sentence pointed by the reviewer.

Remplacer ligne 14 “These aerosol particles are selected because of their very important fluorescence properties”

Par “These aerosol particles are selected because of their very important fluorescence properties (which is essential for the spectroscopy technique used latter to measured their concentrations insides the drops)”

10. p. 518, line 3: I suppose the ratio of the c-s is considered equal to 1 and not equal to Eq. 1.

That’s right. Thanks.

Replace by :

The ratio $\frac{C_{c,\ d_{ae}}}{C_{c,\ d_{ap}}}$ is considered equal to 1

11. p. 518, Eq. 4: What are the Cs here mean?

We don’t find any Cs in the article.

12. In Section 2.5 C is used for the concentration, earlier for the slip correction factor. It is a little bit confusing for the reader.

To avoid any confusion between slip correction factor and concentrations we modified the concentrations notations as follow :

$C_{C_{10}H_{10}Na_2O_5}$ **replaced by** $[C_{10}H_{10}Na_2O_5]$

C_{fluoAC} **replaced by** $[fluoAC]$

$C_{fluoDrop}$ **replaced by** $[fluoDrop]$

13. Section 4, Fig. 6: How were the error bars calculated?

On figure 6 the error bars represents the experimental uncertainties on the collection efficiency calculated with the propagation of the uncertainty of each term of eq 5.

It is added p 520 line 15:

“Figure 6 presents all the collection efficiencies calculated from measurements with the associated uncertainties evaluated with the propagation of the uncertainty of each term of eq 5.”

14. p. 521, line 7: I cannot understand the sentence. Next sentence: What does “difference” mean? Is it larger or smaller?

The sentence line 7 is replace by :

“If theoretical computations for the inertial terms (Eq. 6) show the same behavior at large particle diameter, the magnitude of the collection efficiency is not reproduced. The Slinn model misses this increase due to the non-validity of the potential flow hypothesis, especially in the wake of the drop.”

The sentence line 9 is replaced by:

“The experimental results are at least one order of magnitude larger than the Slinn model results.”

15. p. 521: The Slinn model is mentioned and the experiments are compared to it. It would be therefore good if the model would shortly be introduced in the paper. The Slinn model is stated to be developed for spherical drops. Is it possible to modify it for flattened drops?

We think it is feasible. Two approaches can be considered to take into account drop non-sphericity and oscillations:

- An empirical approach, with the measurement of collection efficiency for different drop sizes and an empirical re-parameterization of the Slinn modell.
- A theoretical approach inspires from Beard, (1974), and based on a Lagrangian tracking of aerosol particles, in a two phase flow precisely calculated with recent method (Tanguy et al. 2007, ref at the end of the answer)

These two approach approach are undertaken at the laboratory.

16. p. 521, line 15: What does “relaxation time of the particle” mean?

It is the time needed for a particle to reach $\frac{1}{e^1}$ of its terminal velocity in a gravitational field and exposed to the drag forces.

$$\tau = \frac{\rho_d d_d^2 C_c}{18\ \mu_g}$$

where ρ_d is the particle density, d_d is the particle diameter and μ_g is the gas dynamic viscosity

17. Figure 7 is not introduced in the text.

The figure 7 is introduced at the line 17, p521

18. Figure 8: Why theoretical curves for so small diameters are shown? They are also very far from the measured data points.

On that figure all collection efficiencies calculated from Beard (1974) model are shown

19. p. 523, line 18: The authors claim that they observed that the collection is driven by the inertial impaction without any contribution of phoretic forces. Is it not something which can be characterized by the Péclet number by chance?

The Péclet number is the ratio between advection rate and diffusion rate, it thus seems to be a good parameter to characterize diffusiophoresis. If any influence of phoretic forces is measured in future measurement (for smaller drops 1 mm) as advises, it will be correlated to Péclet number.

20. p. 524, line 1: “raindrops at a given size collect aerosol particles at a given size does not sound good.

<u>We Replaced :</u>

“This study provides 163 measurements of the efficiency with which raindrops at a given size collect aerosol particles at a given size. “

<u>by :</u>

“This study provides 163 measurements of the collection efficiency with raindrops and particles sizes controlled and measured.”

21. In general, the English of the manuscript should be revised, and the typos should be corrected.

Thanks for the review and your advices.

Ref

Tanguy, S. Ménard, T.,Berlemont, A. A Level Set Method for vaporizing two-phase flowsJournal of Computational Physics. Volume 221, Issue 2, 10 February 2007, Pages 837-853.