The effect of electrostatic charges on the removal of radioactive aerosols in the atmosphere by raindrops

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Abstract. In this article, we report results of self-charged water drop generated by hypodermic needle over charge values comparable to those reported in the literature during stormy rainfall. We also controllably charged aerosols particles by corona discharge and evaluate how it affects their collection efficiency. Electric charges on drops and aerosols are precisely monitored by high resolution electrometers. Our preliminary results tend to accredit the impact of electric charges in collection efficiency.

1. Introduction
Measurements of soil contamination by cesium particles in the first days following the Fukushima accident have shown that higher doses were located downwind of the radioactive plumes after snow and rainfall events. In France, given the location of the nuclear plants, the probabilities of occurrence of rainfall coupled to a nuclear accident are real. To establish procedures that should protect people against exposure to radionuclides, it is essential to understand the interactions between aerosols and precipitation that determine their removal from the atmosphere. The capture of aerosol particles by raindrops (washout) has been studied exhaustively in laboratory under simulated storm conditions, and by modeling. However, to date there are discrepancies on the estimation of the aerosol capture efficiency between experimental studies and modeling results that tend to underestimate it. We believe that this discrepancy is partly related to the non-inclusion of electrostatic interactions in the calculations. More importantly, it is well recognized that raindrops as well as aerosol particles naturally and systematically carry electric charges through various mechanisms in the atmosphere [1], which could lead to strong electrostatic interactions. For electrically charged drops, Barlow and Latham [2] in their laboratory experiments on sub-micron aerosols exhibit significant increase in the washout efficiency. More recently, Tinsley et al. [3] model calculations in the same size range pointed out that image charge effect due to aerosol particles charging can significantly enhance their collision efficiency with raindrops compared to uncharged particles, even if the particles and drops carry electric charges of same polarity. Indeed, radioactive aerosols positively self-charge because of emission of electrons during decay [4] and non-radioactive aerosol get electric charges by adsorption of atmospheric ions resulting from cosmic radiations [5].

We are currently investigating the influence of electric charges on both water drops simulating rainfalls and aerosol particles of micron size range on the washout efficiency in BERGAME facility –
French acronym for Facility to study aerosol washout and measure collection efficiency – developed at IRSN (figure 1).

This article focuses on our first results on the evolution of washout efficiency when aerosol particles are controllably charged by a bipolar corona charger. The initial charge state of the drops is monitored with a Faraday pail but it is not intentionally charged. Other experiments on the influence of drops charge on washout efficiency for neutralized aerosols are presently explored but not reported here.

2. Experimental arrangement

BERGAME has been designed specifically to assess the washout efficiency of aerosol particles by raindrops. It consists of a shaft of ten meters high with a square cross section of 0.45 × 0.45 m² through which millimeter drops free fall toward their terminal velocity before entering in a seeded aerosol atmosphere, and collected beneath.

Drops are generated from a hypodermic needle filled with water in a constant level that shapes them uniformly. Drop diameter and velocity are measured inside the aerosol chamber by shadow imaging and image velocimetry (PIV) respectively. The aerosol chamber is a stainless steel rectangular cube of 1 m height with a cross section of 0.8 × 0.8 m². It is equipped with two apertures on the top and the bottom that allows drops to enter and exit the chamber; dynamic containment systems prevent aerosol particles from reaching the shaft.

The aerosols used in this study are soda fluorescein particles that are easily detectable by fluorescence spectroscopy. Aerosols are generated with ultrasonic nebulizers (Sinaptec GA 500) that produce particles of micron size nearly mono-dispersed. Particle size in the aerosol chamber is measured with both APS (Aerodynamic Particle Sizer) and ELPI (Electrical Low Pressure Impactor). To evaluate the mass density of aerosol particles inside the chamber, a known volume is pumped
through a particle air filter. The mass of soda fluorescein collected on the filter and by the drops are
together measured with fluorescence spectroscopy to determine respectively the mass concentration of
soda fluorescein particles in suspension inside the aerosol chamber, and the mass concentration of
soda fluorescein in the drops. The washout efficiency (E) is simply defined as the ratio of the mass of
aerosol particles (AP) collected by the drop during its fall to the mass of aerosol particles in the
volume swept out by a drop of same equivalent spherical diameter ($E = \frac{m_{AP, collected}}{m_{AP, swept}}$).

The aerosol charges are measured with an impactor ELPI. This instrument consists of 13-stage
cascade low pressure impactor for a size range from 30 nm to 10 µm. Electrometers are used to
measure the current signals on the 12 impactor stages (the first stage is a pre-selector). They can read
current signals in the range of 10 000 to 400 000 fA. In usual operation, aerosols are preloaded by a
corona charger with a known charge efficiency function from which a computer program deducts the
number of particles impacting each stage of the impactor. To assess the aerosol charges, the ELPI is
operated with the charger off, so the measured current is only due to aerosol charges on each stage.

Our aerosol particles are subject to contact electrification and are usually highly charged when they
exit from the generator. To pilot the charging, particles are allowed to pass through a neutralizer
(Topas - Electrostatic Aerosol Neutralizer Series EAN) which is equipped with two corona chargers
that generate positive and negative ions in large amount that can either neutralize them or impose a
controlled charge level and polarity.

3. Results on aerosol washout efficiency

We calculated washout efficiency for non-charged drop sizes of 2.5 mm, of 3 different aerosol mean
diameter diameters and varied the number of elementary charges on the particles (figure 2). For nearly
neutral particles, the washout efficiencies increase with diameter (2.10⁻³, 4.10⁻³, 38.10⁻³ for 1.4 µm,
2.34 µm, 3.5 µm respectively). These sizes are in the range where the main mechanisms responsible of
the washout are impaction and interception of the particles by the drop due to gravity. So it is not
surprising that washout efficiency increases as the size becomes larger. If the charge level is raised in
small steps, the washout efficiency increase very quickly and starts decaying for less than 10
elementary charges and stabilizes after around 20 elementary charges. We can also observe that the
influence of the aerosol charge level seems to diminish as the particle size enlarges. This can easily be
interpreted by smaller particles having higher charge density compared to the bigger particles for the
same number of charges. This trend agrees partly with Tinsley et al. [3] predictions. Indeed these
authors showed a rapid increase of the washout efficiency when the drop and the aerosols particles
have electric charges of comparable magnitudes due to short range image force interactions, this, even
if the drop carries a net charge that is of same sign as the particles. If the charges on the particles are
too high, the Coulombic long range repulsion becomes strong enough to prevent particles from being
captured by the drop. However, the washout efficiency in these calculations keeps decreasing with
increasing charges on the particles until a zero value, while our results show an asymptotic behavior.

These preliminary results seem to confirm that electric charges need to be considered when
investigating aerosol atmospheric washout by rainfall. For aerosol particles of around 1 µm in
diameter, the washout efficiency has almost tripled when they carry few elementary charges.

Further experiments are currently performed for more aerosols sizes, including the sub-micron
range to consolidate these observations.
Figure 2 Measured washout efficiency vs number of elementary charges on the aerosols particles for 3 mean size diameters, for a drop diameter of 2.5 mm. For each datapoint 200 drops were collected and analyzed.

References

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