Appendix

A. Definition of exposure time and electrical time

The time interval during which particles stay close enough to risk collision mainly depends on the difference v_R between their relative velocities, we define this as the exposure time as in eq. A1

As shown by the sketch in figure A1 the exposure time t_{exp} represents the time interval during which there is at least one point belonging to one sphere which has the same altitude of at least one point belonging to the other sphere.

$$t_{exp} = \frac{D_1+D_2}{v_{RY}} \qquad \textit{Eq. A1}$$

The exposure time t_{exp} can be alternatively defined as the time during which the distance between the centres of two neutral spherical particles travelling at their terminal velocity along parallel trajectories stays lower than $\sqrt{L^2 + (R_1 + R_1)^2}$. It follows that, in the context of critical distance estimation, definition A1 implies that electrical interaction between the objects is neglected until the vertical offset gets down to the sum of their radii. Regarding the time that electrical forces need to bring the objects to collision, we can note that as long as the horizontal offset is high compared to the diameter, particles will move towards each other, and the vertical component of the electrical force is negligible with respect to the vertical component. Therefore, as a first approximation, we estimate the electrical time as the time required by two objects to approach each other horizontally from an initial horizontal offset h_0, under the absence of vertical forces. With respect to figure A1, we can define the electrical time as:

$$\Delta t_{el} = t_2 - t_1 \qquad \textit{Eq. A2}$$

Where t_1 and t_2 represent respectively the instants at which particles are released with a distance d0, and the distance at which they reach each other (see figure A1-B).

In Figure A1-B the two falling objects are shown with respect to an external reference system. The free body diagram shows the forces acting on both objects.

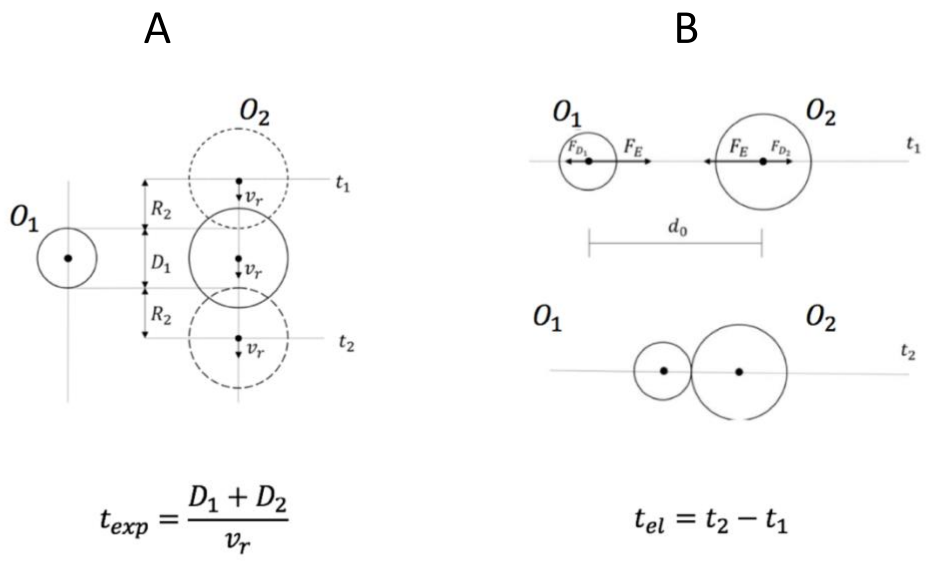

Figure A1

A Sketch of two falling spherical particles in the reference frame joined to the particle on the left O_1. vr represents the relative velocity between the particles; D_1 and D_2 denote the particle diameters, t_1 and t_2 represent the time instants that are used to compute the electrical time.

B - *Oppositely charged spheres approaching each other under the effect of Coulomb force FE and drag forces FD1 and FD2. It is worth to note that while the intensity of Coulomb forces acting on both the particles is the same, the drag forces are different for the two particles as they depend on both their sizes and their velocities*

B. Computation of relative velocity, exposure time and electrical time

The first step of the procedure is to evaluate the relative velocity between objects, since it governs the swept volume. In our model, the vertical component of the relative velocity is given by the difference between the object terminal velocities, and its horizontal component is due to electrical forces. Therefore, for neutral particles, the horizontal component is zero, and the relative velocity is given by the difference between the terminal velocities. The object terminal velocities were computed solving numerically the equation of motion for a sphere settling in air under its weight until an acceleration lower than 10^{-5} m/s^2 was reached. Figure B1 shows the terminal velocities reached at different altitudes by objects of different size and density. Blue lines represent lower density objects and red lines represent higher density objects. For each object density, four different altitudes were considered: sea level; 5 km, 10 km, 15 km.

The trends in figure B1 show that besides increasing with object diameter, the terminal velocity is higher at higher altitudes due to the lower viscosity of air. Moreover, it is worth noticing that high density objects reach higher terminal velocity than low density objects.

For objects settling in the Stokes regime, it can be of great benefit to evaluate the terminal velocity with the formula:

$$v_R = \frac{1}{18}\frac{g}{\mu}\left(\rho_1 {d_1}^2 - \rho_2 {d_2}^2\right) \qquad \textit{Eq. B1}$$

However, it is important to constrain for which combinations of size, object density and air density this simplified expression for the relative velocity holds, if compared to the complete solution obtained with the numerical integration. In order to show the range of applicability of the formula, in figure B1 we plot circle markers in correspondence of the threshold diameters below which the Stokes terminal velocity overestimates the terminal velocity by less than 1%; and star markers in correspondence of a 10% overestimation. It is worth stressing how the Stokes formula tends to always overestimate the terminal velocity due the lack of the contribution of pressure drag in the equation.

Regardless of all the limitations related to the applicability range of the Stokes regime figure B1 shows that the Stokes formula is accurate for objects whose diameter is lower than 15µm, regardless their density and the settling altitude.

After computing terminal velocities, exposure times between couples of objects were calculated with eq. A1. Figure B2 shows the dependency of exposure time on the size, altitude and density of the considered objects.

To limit the number of variables, only interactions between couples of objects of the same density were considered throughout the paper. Figure B2 shows that the exposure time between objects of the same size goes to infinite. In fact, the denominator in eq. A1 goes to zero if the particles have the same settling velocity. Moreover, low density objects are always characterized by higher exposure times with respect to higher density particles.

In figure B3, we show the electrical time computed for couples of objects released by a centre to centre distance equal to 100 times the sum of their radii. Figures B3-A; B3-B and B3-C are relative to surface charges of 25%; 50% and 100% of the ionization limit value.

The trends shown in figure B3 are the result of the interplay between several effects:

large particles have a higher initial release distance than small ones, which contributes to increasing the electrical time;

large particles have higher inertia than small ones, which also contributes to increasing the electrical time;

large particles are more charged than small particles (due to their larger surface), which contributes to decreasing the electrical time.

The minimum comes as a result of a switch in the dominant effect: for small objects, the electrical time has a decreasing trend due to the fact that the net charge is increasing; for large objects, the curve has an increasing trend due to the increase in their inertia.

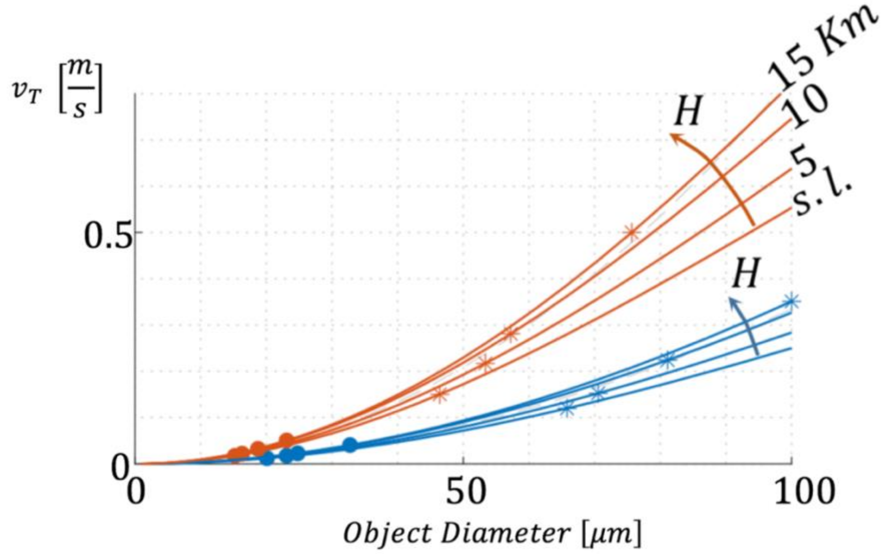

Figure B1 - Terminal velocity against object size for 4 altitudes and two object densities. Red lines represent 2500 kg/m³ objects, and blue lines represent 1000 kg/m³ objects. Different lines represent different values of altitude at which the object is settling. Circles on the graph represent the maximum diameter for which terminal velocity can be calculated with the Stokes formula, with an overestimation lower than 1% Star markers are relative to a 10% overestimation obtained with the Stokes formula.

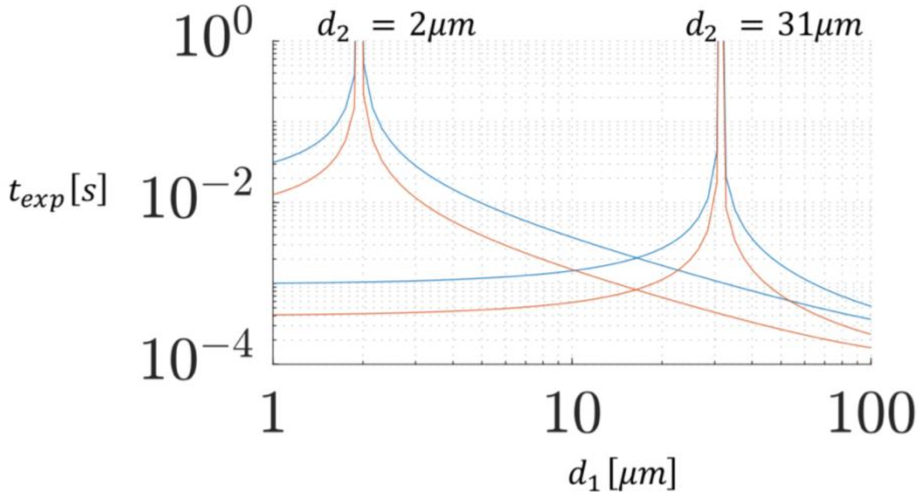

Figure B2 - Exposure time against size for different size combinations and different densities . d1 and d2 represent the diameters of the interacting objects. The diameter d1 of one interacting object is given on the x-axis. Two diameters of the other object (d1= 2μm; d2= 31μm) are represented by different lines. Red lines represent 2500 kg/m³ objects, and blue lines represent 1000 kg/m³ objects.

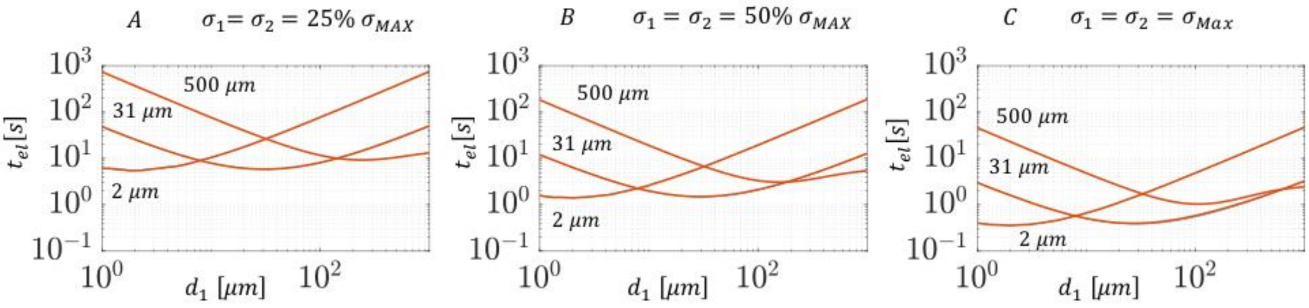

Figure B3 - Electrical time variation with object diametersnd surface charge. Object density is set to 2500 kg/m^3 for both the colliding objects, and the collision altitude at H=10000 m. A release distance of 100 y_G was considered. The diameter of one object is considered on the abscissa. Different lines represents three different diameters of the second object. Figures A, B and C are relative to surface charges of 25% σ_{max}, 50% σ_{max}, 100% σ_{max} respectively. Where σ_{max} is maximum charge that a spherical particle can carry in air, considering a dielectric strength of 3 MV/m.

C. Sticking maps for different values of particle density, collision altitude and surface energy.

In general, the sticking map changes its shape when one of these parameters varies. To show how variations of object density, collision altitude and object surface energy affect the collision maps, we fixed two parameters and let the third one vary. The results are shown in figures C1, C2 and C3.

Figure C1-A shows how the sticking map is affected when the density of the colliding object is varied between 500 kg/m^3 and 2500 kg/m^3.
As we can see, the variation of the sticking map strongly depends on whether the colliding objects are charged or not. When the colliding objects are neutral (see the blue lines), an increase in the object density has the effect of significantly shrinking the sticking region without changing its shape. Conversely, when the colliding objects are charged (see orange lines), an increase in the density has the effect of slightly expanding the sticking region. This expansion is accompanied by a change in the shape of the boundary lines. In fact, the sticking area expands close to the bisector, where the colliding particles have similar sizes, and contract far from the bisector, where particle sizes are more different. In order to appreciate how the whole sticking region changes with object density, in figure C1-B we show the square root of the area under the curves. Figure C1-B shows that an increase in particle density results in a sharp decrease of the sticking Area if particles are neutral, and a gentle increase in the sticking area if particles are charged. The x and y intercept on figure C1-A (see the dashed ellipses) are relative to the maximum diameter with which a 1µm particle can stick. These values are plotted against the object density in figure C1 where it is displayed the maximum diameter that a collector can have in order to capture a 1µm object of the same density. When the object density increases, the maximum size that a collector can have gets lower for both charged and neutral objects.

The influence of the altitude at which the collision occurs on the final outcome of the impact in figure C2-A. When a collision happens at higher altitudes, the sticking region gets smaller for both neutral and charged particles. This is also shown in the decreasing trend for the square root of the area under the curves shown in figure C2-B. The spreading of the curve towards the coordinate axes shows that the effect of altitude on sticking is more important when the colliding objects have different sizes (when they are far from the bisector). In fact, the maximum effect of altitude is shown in figure C2-C, which shows the maximum diameter with which a 1µm object can stick.

Figure C3-A shows the effect of surface energy on the sticking maps. As could be expected, an increase on the object surface energy is reflected in an expansion of the sticking region. Figures C3-B and C3-C reveal that such an expansion is more important for charged object than for neutral ones.

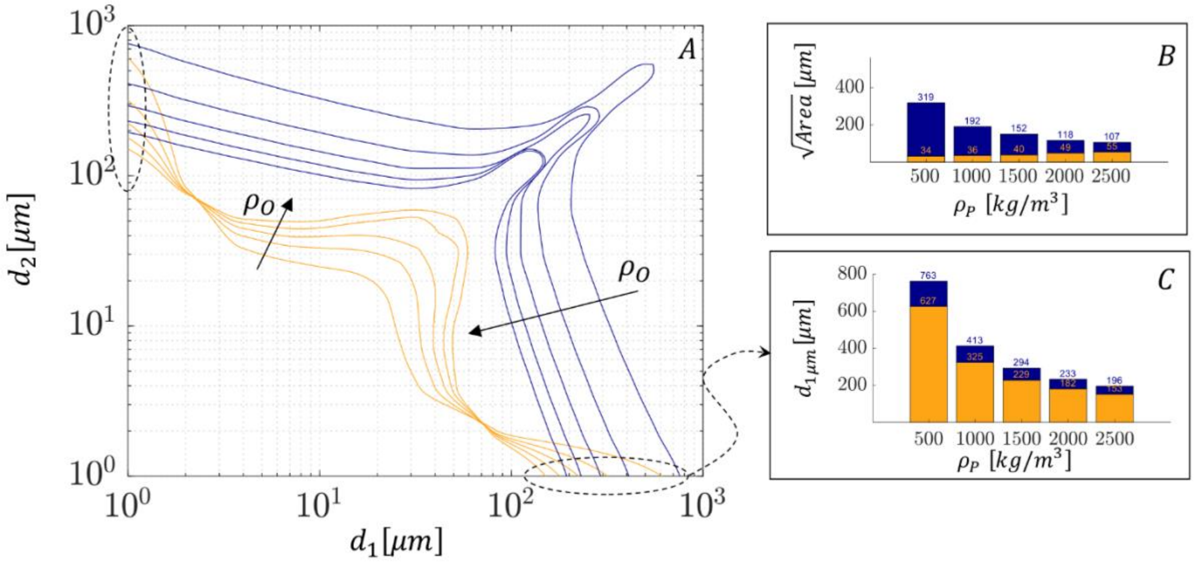

Figure C1 - Plot A represents the Sticking maps for neutral and charged objects for different values of object density. The colliding objects are characterized by a surface energy of 20 mJ/m^2 and a collision altitude of 10 km. Continuous lines separate the sticking region (internal) and the rebound region (external). Blue lines represent the boundary lines for neutral objects; yellow lines represent boundary lines for objects charged at the ionization limit. Different lines correspond to different object densities ranging from 500 kg/m^3 to 2500 kg/m^3. The black arrows over the boundary lines point towards the direction of increasing object density, e.g. the closest line to the arrowhead is relative to an object density of 2500 kg/m^3). In (B) it is displayed the square root of the area under the boundary curves showed in (A). The values of object densities are shown on the x-axis. Blue bars are relative to neutral objects, and orange bars are relative to objects charged at the ionization limit.

Fig. C shows the value of the maximum diameter that can stick with a 1µm particle. Similarly to plot B, blue bars are relative to neutral objects, and orange bars are relative to objects charged at the ionization limit.

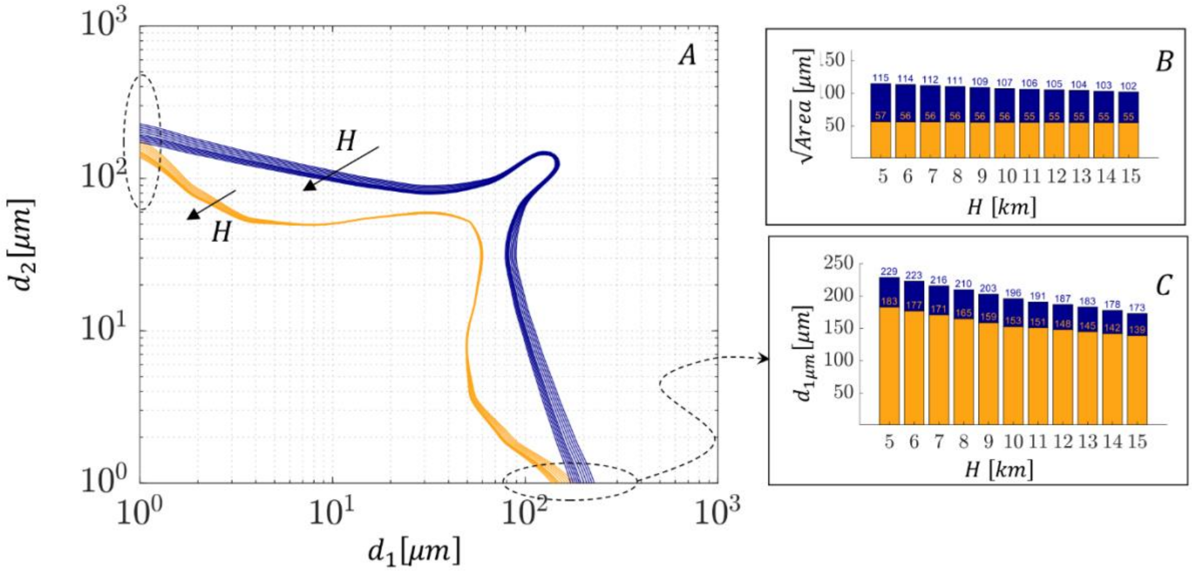

Figure C2 - The Sticking maps for neutral and charged objects for different values of collision altitude (A). The colliding objects are characterized by a surface energy of 20 mJ/m² and an object density of 2500 kg/m³. Continuous lines separate the sticking region (internal) and the rebound region (external). Blue lines represent the boundary lines for neutral objects; yellow lines represent boundary lines for objects charged at the ionization limit. Different lines correspond to different collision altitudes ranging from 5km to 15km. The black arrows over the boundary lines point towards the direction of increasing collision altitudes, e.g. the closest line to the arrowhead is relative to a collision altitude of 15km)

In (B) the square root of the area under the boundary curves displayed in (A). In Fig. C the value of the maximum diameter that can stick with a 1µm particle. Similarly to plot B, blue bars are relative to neutral objects, and orange bars are relative to objects charged at the ionization limit.

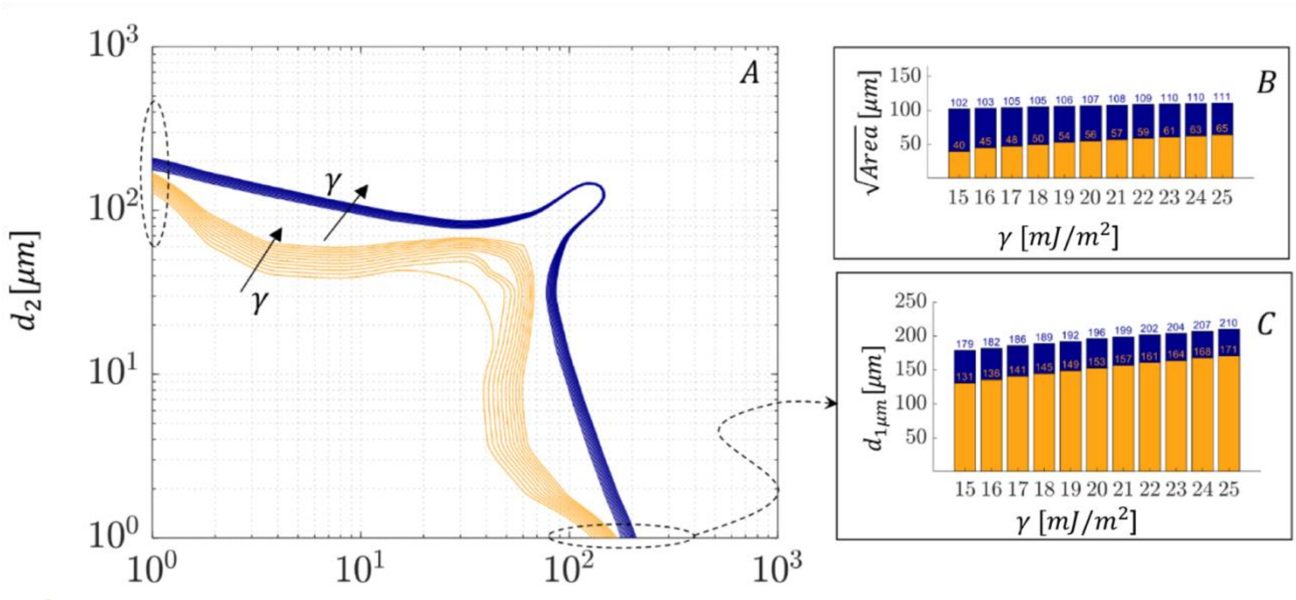

Figure C3 - Plot A represents the Sticking maps for neutral and charged objects for different values of object surface energy. The colliding objects have a density of 2500 kg/m^3 and a collision altitude of 10km. Continuous lines separate the sticking region (internal) and the rebound region (external). Blue lines represent the boundary lines for neutral objects; yellow lines represent boundary lines for objects charged at the ionization limit. Different lines correspond to different surface energies ranging from 15 mJ/m^2 to 25 mJ/m^2. The black arrows over the boundary lines point towards the direction of increasing collision altitudes, e.g. the closest line to the arrowhead is relative to a surface energy of 25 mJ/m^2.

Plot B shows the square root of the area under the boundary curves in Plot A. The values of collision altitudes are shown on the x-axis. Blue bars are relative to neutral objects, and orange bars are relative to objects charged at the ionization limit.

Plot C shows the value of the maximum diameter that can stick with a 1µm particle. Similarly to plot B, blue bars are relative to neutral objects, and orange bars are relative to objects charged at the ionization limit.

Supplementary material

Flowcharts to determine collision and sticking outcome

In this sections, we report the flowcharts that allow to apply the collision and the sticking map in figure 3 for the case of oppositely charged particles characterised by different charges.

Regarding particle collision, the flow chart in figure S1 explains how figure 3A can be used to determine whether particles will collide or not. For the sake of clarity, we will discuss here some practical examples.

Example 1: Let us consider two neutral objects O_1 and O_2, whose density is 2500 kg/m^3 settling at an altitude of about 10 km. Let their diameters be d_1 = 40μm and d_2 = 20μm. In figure 1A objects are sketched in the reference system joint to the object O_2. In this reference system, O_1 sediments at a speed given by the difference between the two terminal velocities. The values of the terminal velocities can be read in in figure 4, from which we can compute the relative velocity: $v_R \cong 0.15 - 0.03 = 0.12$. After a time interval $\Delta t = \frac{\Delta y_{(1\leftrightarrow 2)}}{v_r}$, O_1 will reach O_2. Collision will happen provided that the initial horizontal offset between the objects is lower than the geometrical distance y_g = 30μm. Therefore, with reference to figure 1A, object O_1 will collide with object O_{2A} and will not collide with O_{2B}, nor with the object O_{2C}. It is worth noticing that in a more complete description of the collision, hydrodynamic interactions may cause objects to avoid each other even when the horizontal offset is lower than y_g. However, this phenomenon is not considered in this model.

Example 2 : If the objects O_1 and O_2 are oppositely charged, the volume V_c swept by O_1 respect to O_2 is higher than the geometrical volume V_g due to the fact that objects are drawn to each other by Coulomb forces. The radius y_c of this volume is given by $y_{c(1\leftrightarrow 2)} = E_{lin(1\leftrightarrow 2)} y_{g(1\leftrightarrow 2)}$). If objects are oppositely charged at the ionization limit (see figure 1B), the value of the linear collision efficiency $E_{lin(1\leftrightarrow 2)} = \sqrt{E_{(1\leftrightarrow 2)}}$ can be read on figure 3A. For d_1 = 40μm and d_2 = 20μm, $E_{lin(1\leftrightarrow 2)} \cong 4$; and $y_{c(1\leftrightarrow 2)} \cong 120$ μm. The increase in volume respect to the neutral case, is given by $\frac{Vc}{Vg} = E \cong 16$. Collision will happen provided that the initial horizontal offset between the objects is lower than the critical distance $y_{c(1\leftrightarrow 2)} \cong 120$ μm. With reference to figure 1B, both O_{2A} and O_{2B} will collide with P_1, while P_{2C} will not collide.

Example 3: Let us now consider a couple of colliding objects oppositely charged at the ionization limit, whose diameters are d_1 = 70 µm and d_2 = 15 µm. From figure 3A, we see that E_{lin} = 2.5, therefore the critical distance is y_c = 106 µm. If the particle initial horizontal offset is equal to their critical distance, the outcome of the collision can be directly read on figure 3B. The corresponding point on the plot falls in the blue region. Since the point is external to the region bounded by the $\sigma = \sigma_{max}$ line, objects will rebound. This conclusion is strictly valid when initial offset between the objects is equal to the critical distance. In fact, since the critical distance is by definition the maximum distance that two colliding objects can have, when the initial horizontal offset is equal to the critical distance the relative velocity will be at its maximum. If the initial horizontal offset is lower than the critical one, particles will have less space to accelerate, and the relative velocity will be lower as a result. If the initial horizontal offset is low enough, relative velocity will be lower than the sticking velocity, and particles will stick. If the initial horizontal offset h_i is known, an auxiliary reference line can be found on the graph to predict the collision outcome with eq S6. For example, if $h_i = 50$ µm eq. S4 gives: $\sigma_{eq} = \sigma_{max}\sqrt{\frac{50\ \mu\text{m}-42.5\mu\text{m}}{106\ \mu\text{m}-42.5\mu\text{m}}} \approx 34\%\ \sigma_{max}$.

Since the point is now internal respect to the line $\sigma = 30\%\ \sigma_{max}$ in figure 3B, objects will stick when released at a horizontal offset of $h_i = 50$ µm.

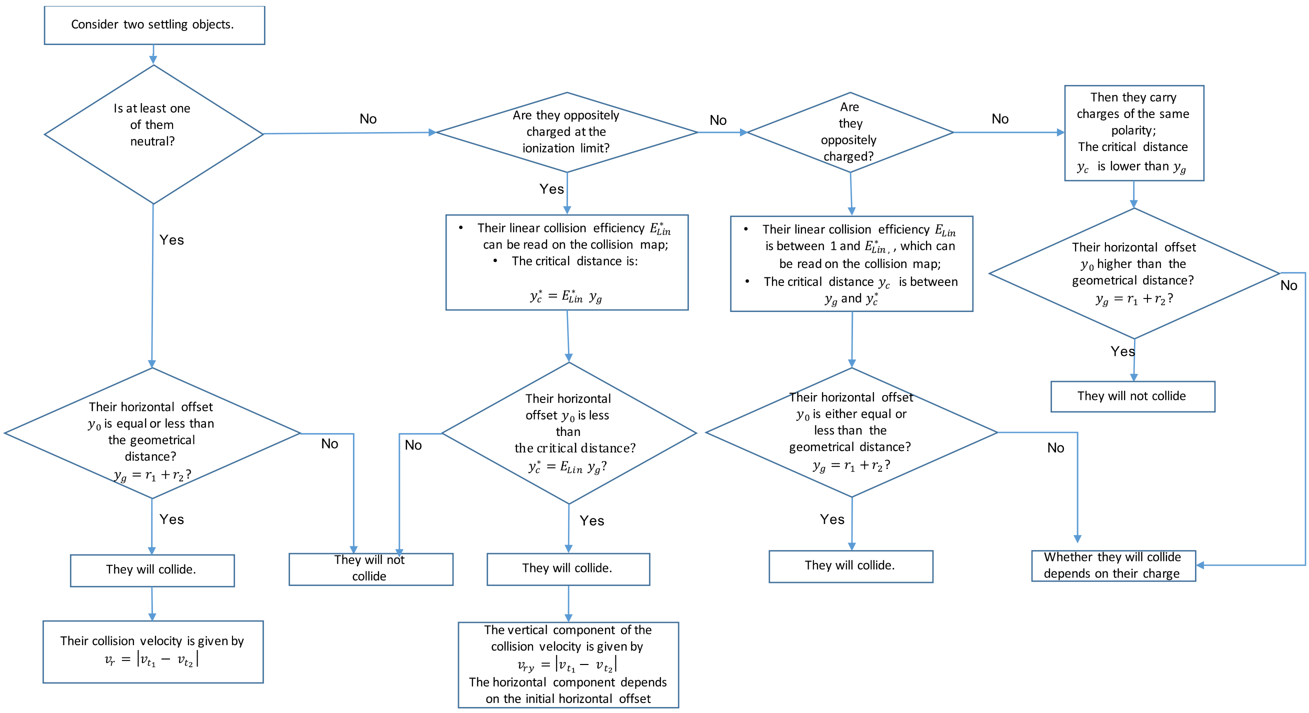

Figure S1 - The flowchart explains how to use figure 3 A to determine whether two objects will collide or not.

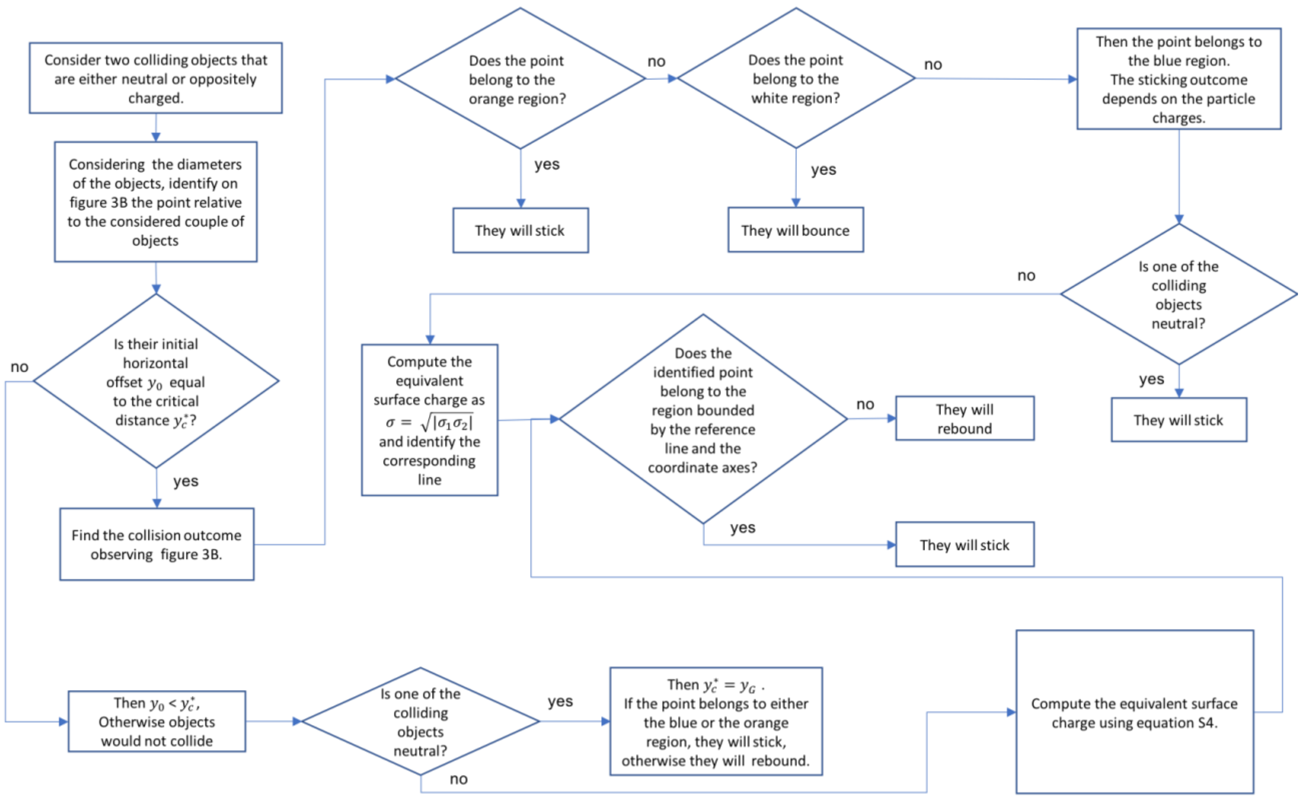

Figure S2. The flowchart explains how to use figure 3B to determine whether two objects will stick or rebound.

Sticking efficiency for an arbitrary horizontal offset

Let us consider two spherical objects characterized by an initial horizontal offset h_i approaching horizontally under the effect of Coulomb forces until they touch each other (at an horizontal offset y_g). If we neglect drag forces, both linear momentum and total energy are conserved. This leads to the following expression for the collision velocity.

$$v_{xcoll} = \sqrt{{v_{xi}}^2 - \frac{2}{4\pi\varepsilon_0}\frac{Q_1Q_2}{M^*}\left(\frac{1}{y_g} - \frac{1}{h_i}\right)} \qquad \textit{Eq. S1}$$

Where v_{xi} represents the initial relative velocity, and v_{xcoll} the relative velocity at the moment of collision. ε_0 represents the dielectric constant of vacuum, Q_1 and Q_2 the object charges, and M^* the reduced mass given by $\frac{M_1M_2}{M_1+M_2}$.

For a full derivation of *eq. S1* in the case of a gravitational field please refer to "Relative Speeds of Interacting astronomical bodies" Mungan (2006).

Assuming a null initial relative velocity, the square of collision velocity as a function of surface charges and object diameters can be rewritten as follows:

$${v_{xcoll}}^2 = \left(\frac{3}{\varepsilon_0}\frac{\rho_1{d_1}^3 + \rho_2{d_2}^3}{\rho_1d_1\rho_2d_2}\right)|\sigma_1\sigma_2|\left(\frac{1}{y_g} - \frac{1}{h_i}\right) \qquad \textit{Eq. S2}$$

Where ρ_1 and ρ_2 are the object densities; d_1 and d_2 are the object diameters; σ_1 and σ_2 the surface charges of the two objects.

Since figure 3B was computed for couples of oppositely charged objects released at the critical distance, we cannot employ it directly when the initial horizontal offset h_i is lower than the critical distance. However, since the collision outcome (sticking or rebound) depends on the collision velocity, we can determine the collision outcome of a collision by considering an equivalent collision characterised by the same collision velocity. Under the assumption that the vertical component of the collision velocity is given by the difference between the terminal velocities and that the horizontal component is given by Coulomb forces only, we can state that two collisions are equivalent if and only if they are characterised by the same component of the horizontal velocity at contact (given by eq, S2).
Therefore, if we want to determine the outcome of a collision between two objects characterised by surface charges σ_1 and σ_2 released at a distance $h_i < y_c$ we can solve the problem by considering the equivalent collision between two objects released at the critical distance that collide at the same velocity. In particular, we want to find what is the charge that these objects would need to have to reach the same collision velocity. To this purpose, we define the equivalent surface charge σ_{eq} as the geometric mean between the charges that two objects released at the critical distance y_c should have for the collision to be equivalent.

The collision velocity for the objects released at the critical distance is then given by:

$$v_{xcoll}{}^2 = \left(\frac{3}{\varepsilon_0}\ \frac{\rho_1 d_1{}^3 + \rho_2 d_2{}^3}{\rho_1 d_1 \rho_2 d_2}\right)\ \sigma_{eq}{}^2\ \left(\frac{1}{y_g} - \frac{1}{y_c}\right) \qquad \textit{Eq. S3}$$

Equating *S2* and *S3*, we get the following equation for the equivalent surface charge.

$$\sigma_{eq}{}^2 = \sigma\ \frac{h_i - y_g}{y_c - y_g} \qquad \textit{Eq. S4}$$

Where $\sigma = \sqrt{\sigma_1 \sigma_2}$

Using equation *S4* we can determine the outcome of a collision between a couple of objects characterised by the charges σ_1 and σ_2 that are released with a horizontal offset h_i. To this purpose, after reading the critical distance from figure 3A, we can use eq. *S4* to find the equivalent surface charge. The equivalent surface charge will identify the line in figure 3B that we need to use as a reference to determine whether the collision will end up in sticking or bouncing.

List of symbols

$A_{1,}A_2$	cross-sectional areas of the interacting objects.
$C_{D1,}C_{D2}$	Drag coefficients of the interacting objects.
d_1, d_2	Diameters of the interacting objects.
D_s	Dielectric strength of air. It represents the electric field above which electrical breakdown occur. Although its value can change with temperature and humidity, a constant value of 3 $\frac{MV}{m}$ in this work.
E^*	Reduced elastic modulus of two colliding objects. $$\frac{1}{E^*} = \frac{1-\nu_1}{E_1} + \frac{1-\nu_2}{E_2}$$ Where ν_1 and ν_2 represent the Poisson's ratios between the two objects. In all computations, we set: $$E_1 = E_2 = 142.2\ MPa$$ $$\nu_1 = \nu_2 = 0.21$$
E_{el}	Electrical collision efficiency. Ratio between the volume swept by two charged objects and the volume swept by two neutral objects.
$E_{lin} = \sqrt{E_{el}}$	Linear electrical collision efficiency. Represents the ratio between the critical distance and the geometrical distance.
E_{sphere}	Electric field generated by a charged sphere.
$\vec{F}_{drag}$	Drag force.
$\vec{F}_{el}$	Coulomb force.
$\vec{g}$	Acceleration of gravity
H	Collision altitude.
K_{el}	Collision Kernel. Represents the volumetric flow rate of objects characterised by a radius R_2 that collide with a particular object characterised by a radius R_1.
m_1, m_2	Masses of the interacting objects
m^*	Reduced mass of two colliding objects. $\frac{1}{m^*} = \frac{1}{m_1} + \frac{1}{m_2}$
O_1, O_2	Couple of objects that may collide to each other. They can represent either aggregates and single particles.
Q_1, Q_2	Charges carried by the interacting objects.
Q_{max}	Maximum charge carriable by a spherical object.
R^*	Reduced radius of two colliding objects. $$\frac{1}{R^*} = \frac{1}{R_1} + \frac{1}{R_2}$$
Re	Object Reynolds number
t_{el}	Electrical Time. Time spent by two oppositely charged objects to horizontally approach each other

	under the effect of coulomb interactions, and drag forces, considering an initial horizontal offset h_i.
t_{exp}	Exposure time. Time interval during which the horizontal projections of two falling objects have at least one point in common.
V_g	Volume swept by the upper object in the reference system joint to the lower object
$\overrightarrow{v_1}, \overrightarrow{v_2}$	Velocity of the approaching objects with respect to still air.
v_r	Relative velocity between two objects.
v_{Ry}	Vertical component of the relative velocity between two objects. In this work, it is assumed to be always equal to the difference between the objects terminal velocities.
V_s	Sticking velocity between particles. It represents the threshold collision velocity below which particles stick with each other.
v_T	Terminal velocity.
$\overrightarrow{x_1}, \overrightarrow{x_2}$	Horizontal coordinates of the interacting objects
y_c	Critical Distance. Maximum value for the initial horizontal offset between two falling objects that leads to collision between the objects. In other words, when the initial horizontal offset between the particles is lower than the critical distance, particles will collide.
y_g	Geometrical distance. $y_g = R_1 + R_2$
Δt	Time taken by an upper objects to reach the same altitude of a lower object, considering an initial vertical offset of Δy_i.
γ	Object surface energy.
ε_0	dielectric constant of vacuum.
ε_a	Absolute dielectric constant of air.
ε_r	relative dielectric constant of air respect to vacuum.
μ	Dynamic viscosity of air. Different values of dynamic viscosity were used for different collision altitudes. The standard atmosphere profile was considered.
ρ_f	Air density. Different values of air density were used for different collision altitudes. The standard atmosphere profile was considered.
$\overline{\sigma_g}$	Geometrical mean between the two surface charges of the interacting objects.
σ_1, σ_2	Surface charge of the interacting objects.
σ_{max}	Maximum surface charge carriable by a spherical object characterised by a uniform charge distribution, considering a dielectric strength of $3\,\frac{MV}{m}$.