Supplementary Information

Surface Charge of Environmental and Radioactive Airborne Particles

Gyoung G. Jang[1], Alexander I. Wiechert[2], Austin P. Ladshaw[1], Tyler Spano[1], Joanna McFarlane[1], Kristian Myhre[1], Sotira Yiacoumi[2], Costas Tsouris[1]

[1] Oak Ridge National Laboratory (ORNL), Oak Ridge, TN 37831-6181
[2] Georgia Institute of Technology, Atlanta, GA 30332-0373, United States

Correspondence to: Costas Tsouris (tsourisc@ornl.gov)

[*]Corresponding author: tsourisc@ornl.gov

Submitted to:
(Atmospheric Chemistry and Physics)

2021

1. Theoretical Method of charge measurement

Figure S1 shows the forces acting on a particle freely falling through the air as it approaches its terminal velocity (v_t) in the absence of an electric field. Only two forces are acting on the particle in this system: the viscous force ($\boldsymbol{F_v}$) that arises from the friction applied to the particle as it moves through the air and the force of gravity ($\boldsymbol{F_g}$). When the particle reaches its terminal velocity, the net force acting on the particle is zero since the viscous force is equal and opposite to the force of gravity:

$$mg = kv_f \tag{S1}$$

where, $\boldsymbol{v_f}$ is the free fall velocity, $\boldsymbol{k}$ is the coefficient of friction between the air and the particle, $\boldsymbol{m}$ is the mass of the particle, and $\boldsymbol{g}$ is gravitational acceleration (m/s^2).

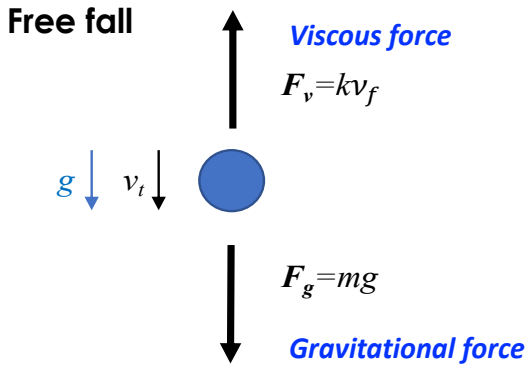

Figure S1: Free-body diagram of a particle in free fall when no electric field is present.

By adjusting the magnitude of the electric field E between two plates in the electrodynamic balance apparatus that are separated by a distance of d, we may alter the magnitude of the electric force acting on a particle of charge q (F_E=qE). These adjustments can cause the particle to rise, fall faster, or even stop moving entirely. If the magnitude of the field is properly tuned such that F_E = F_g, the particle will levitate as illustrated by Fig. S2. In this state, the charge of an individual particle can be easily determined since the force of gravity acting on the particle, the distance between the two plates, and the potential applied by the electric field are all known quantities.

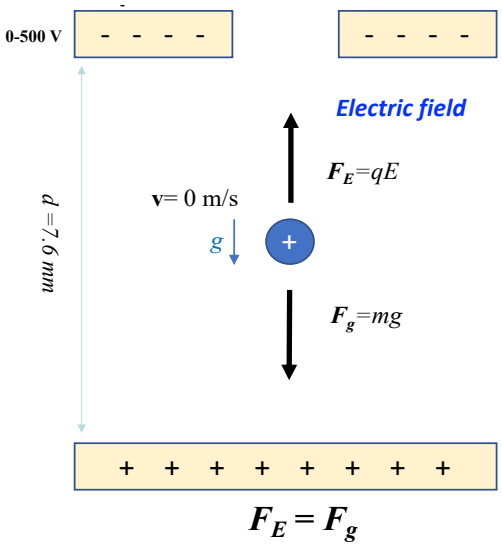

Figure S2: Free-body diagram for a particle suspended in an electric field.

The particle charge q can also be determined by observing the particle's velocity when it is rising under the influence of an electric field. Figure S3 shows the force balance acting a particle rising in an applied electric field. Adding the viscous force (kv_r) to the force balance yields:

$$qE = mg + kv_r \tag{S2}$$

where E is the electric field strength ($E=V/d$), d is the distance separating the plates in the electrodynamic balance apparatus, m is the droplet mass, V is the potential difference across the plates (volts), q is the charge carried by the particle (Coulombs), and v_r is the rise velocity.

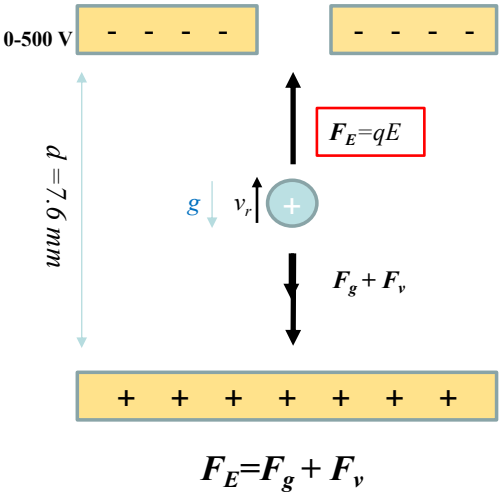

Figure S3: Free-body diagram of a particle rising in an electric field.

In this case, there is also a small buoyant force exerted by the air on the particle; however, since the density of air is much smaller than that of the particle (e.g., only about one-thousandth), this force may be neglected. Eliminating k from Eg. S1 and S2 and solving for q yields:

$$q = \frac{mg(v_f + v_r)}{Ev_f} \quad \text{(S3)}$$

To eliminate m from Eg. S3, the expressions for the volume of a sphere ($4/3\pi a^3$) and particle density are used:

$$mg = \frac{4}{3}\pi a^3 \rho g \quad \text{(S4)}$$

where, a is the radius of the particle in meters and ρ is the density of the particle in kg/m^3. Substituting Eg. S4 into Eq. S3 yields:

$$q = \frac{4\pi a^3 \rho g(v_f + v_r)}{3(Ev_f)} \quad \text{(S5)}$$

To determine a, the formula for Stokes' Law ($F_v = 6\pi\eta a v_t$) can be applied. This formula associates the radius of a spherical body to its fall velocity (v_f) in a medium with a viscosity of η. Setting the expression for Stoke's Law (i.e., viscous force, F_v) equal to the right-hand term (i.e., F_g) in Eg. S4 and solving for a yields:

$$a^2 = \frac{9\eta v_f}{2\rho g} \quad \text{(S6)}$$

where, η is the viscosity of air (N • s/m^2). Stokes' Law, however, is inaccurate when the fall velocity of the particle is less than 0.1 cm/s. Since the particles used in this research (e.g., radioactive aerosols with an average particle size of ~0.5 μm) may have a fall velocity of 0.01 to 0.001 cm/s (10^{-4} to 10^{-6} m/s), the viscosity must be multiplied by a correction factor. The resulting effective viscosity is:

$$\eta_{eff} = \eta\left(\frac{1}{1+\frac{b}{pa}}\right) \quad \text{(S7)}$$

where b is a constant equal to 8.2×10^{-3} Pa • m, p is the atmospheric pressure (barometric pressure, Pascals), and a is the radius of the particle as calculated by the uncorrected form of Stokes' Law, Eq. S6. Substituting η_{eff} from Eq. S7 into Eg. S6 for η gives:

$$a^2 = \frac{9\eta v_f}{2\rho g}\left(\frac{1}{1+\frac{b}{pa}}\right) \quad \text{(S8)}$$

Substituting Eq. S8 into Eq. S5 for a yields:

$$q = \frac{4\pi\rho g(v_f+v_r)}{3(Ev_f)}\left[\frac{9\eta v_f}{2\rho g}\left(\frac{1}{1+\frac{b}{pa}}\right)\right]^{3/2} \quad \text{(S9)}$$

Substituting the equation for electric intensity (E) into Eq. S9 for E and rearranging yields:

$$q = \frac{4\pi d\rho g(v_f+v_r)}{3(Vv_f)}\left[\frac{9\eta v_f}{2\rho g}\left(\frac{1}{1+\frac{b}{pa}}\right)\right]^{3/2} \quad \text{(S10)}$$

Thus, the electric charge (q) carried by the particle can be determined using Eq. S10. There is an alternative method to calculate the charge on a particle. Briefly, k can be determined from Eq. S1:

$$k = -\frac{mg}{v_0} \quad \text{(S11)}$$

where v_0 is the terminal falling velocity at $V=0$. Combining Eq. S11 with Eq. S2 yields:

$$qE = -\frac{mg}{v_0}v + mg \quad \text{(S12)}$$

Rearranging Eq. S12 to solve for v gives:

$$v = -\frac{qv_0}{mg}E + v_0 \quad \text{(S13)}$$

A plot of v versus E yields a slope (s):

$$s = -\frac{qv_0}{mg} \quad \text{(S14)}$$

Rearranging for the value of the charge (q) gives:

$$q = -\frac{smg}{v_0} \quad \text{(S15)}$$

2. Particle characterization

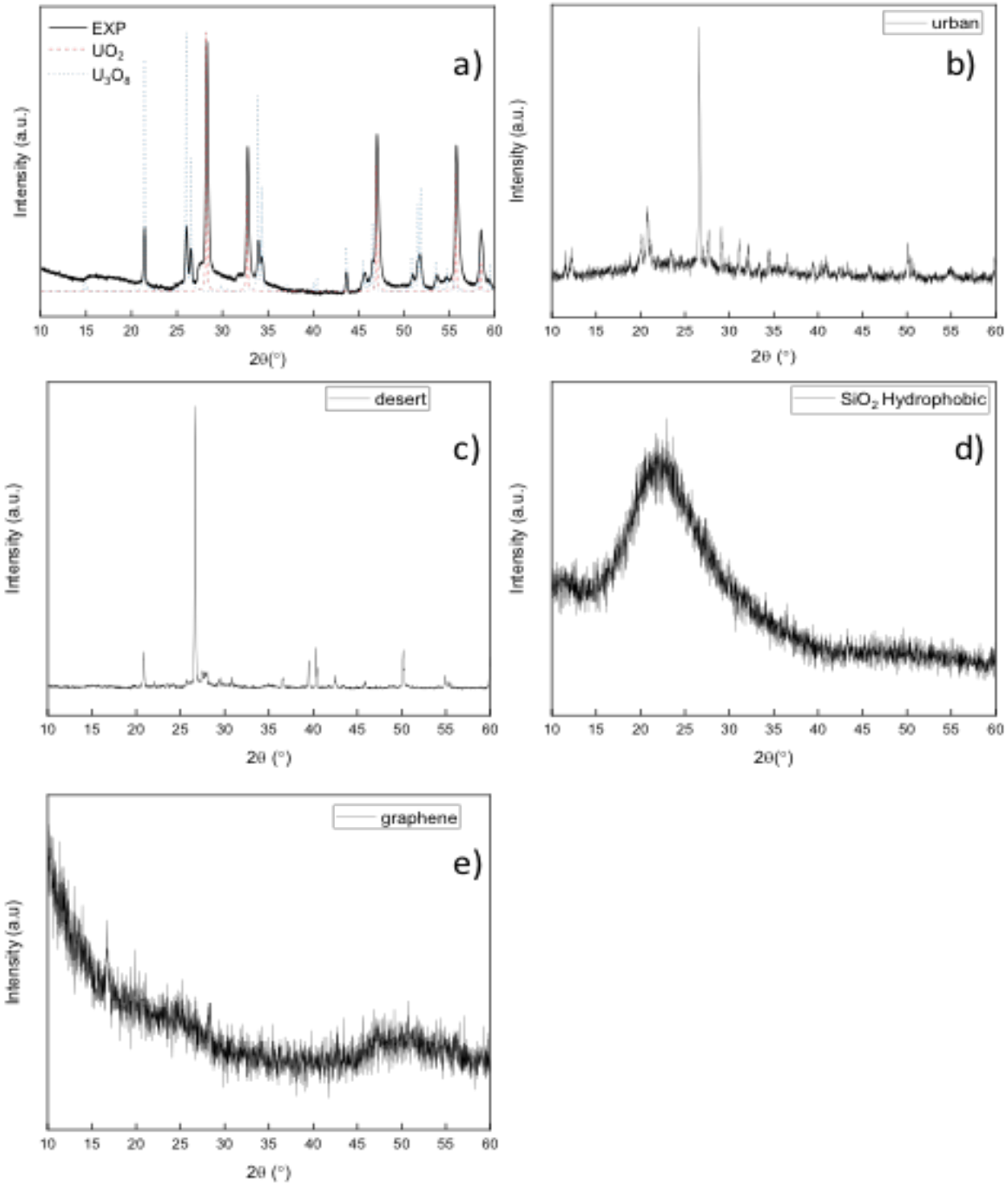

Figure S4. Powder X-ray diffractograms collected for a) UO_2 compared with reference diffraction patterns for UO_2 and U_3O_8, b) urban dust, c) desert dust, d) hydrophobic SiO_2, and e) graphene. PXRD data were not collected for hydrophilic SiO_2 due to high humidity in the laboratory

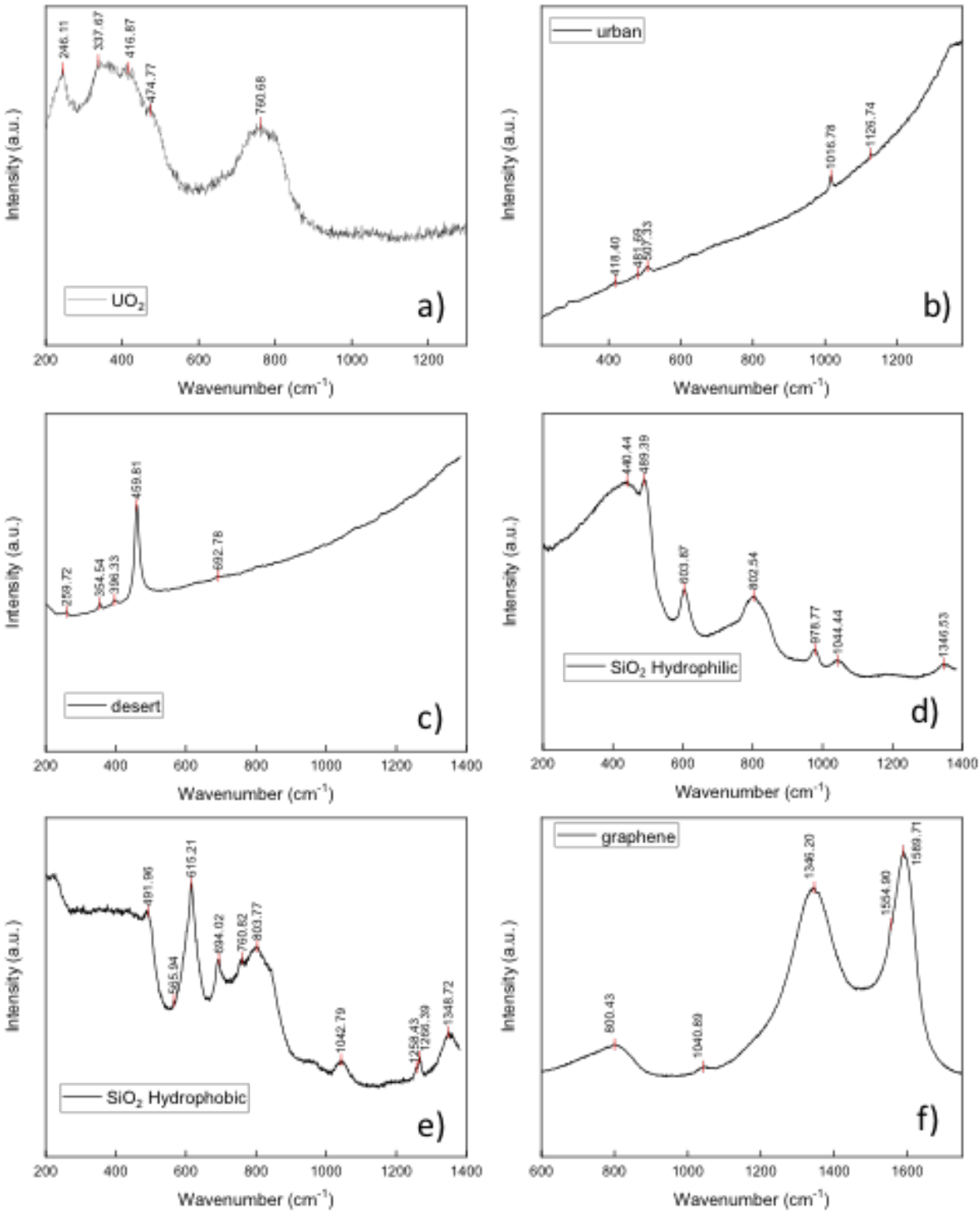

Figure S5. Raman spectra with peak centers labeled for samples of a) UO_2, b) urban dust, c) desert dust, d) hydrophilic SiO_2, e) hydrophobic SiO_2, and f) graphene

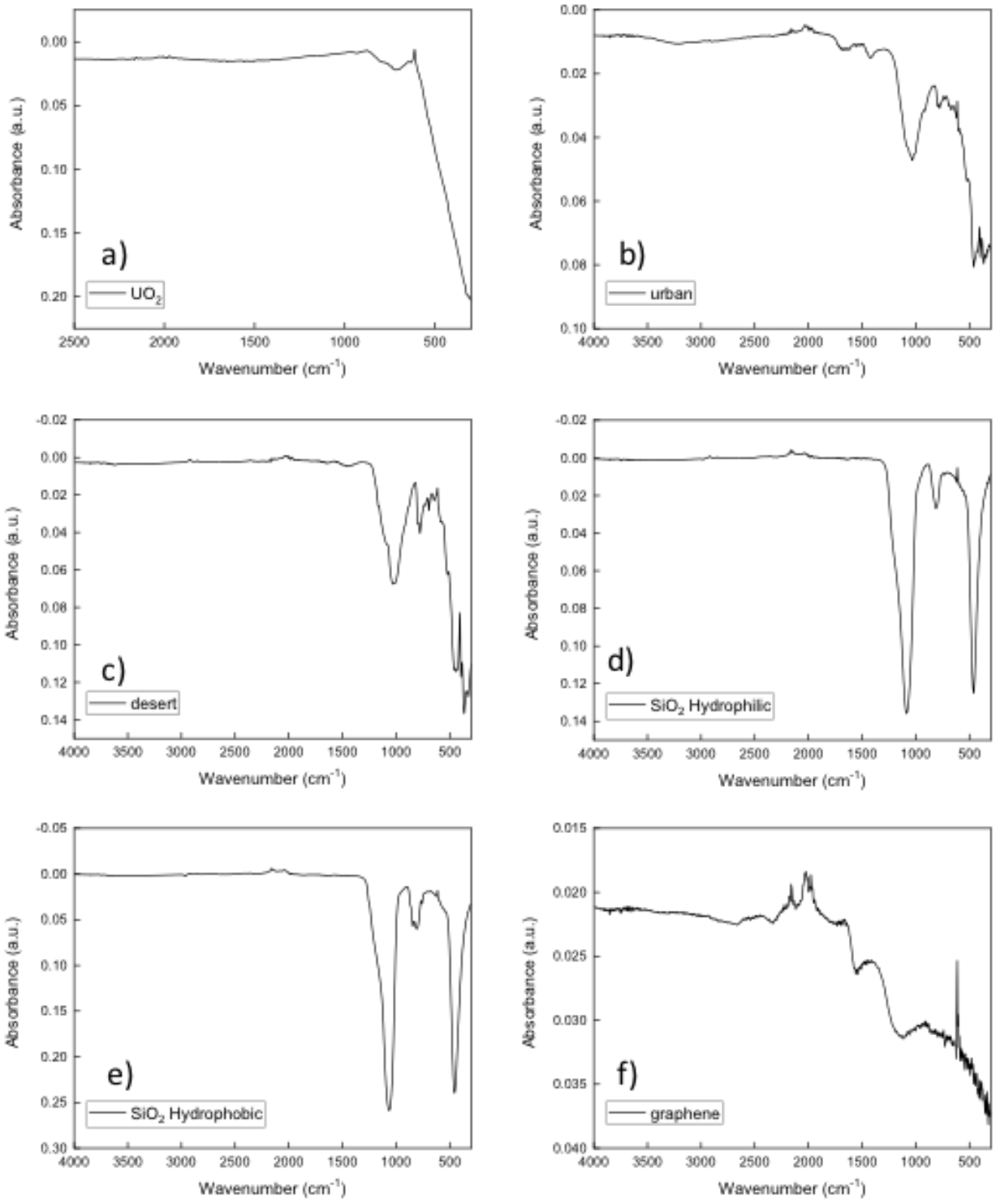

Figure S6. FTIR spectra collected for samples of a) UO_2, b) urban dust, c) desert dust, d) hydrophilic SiO_2, e) hydrophobic SiO_2, and f) graphene

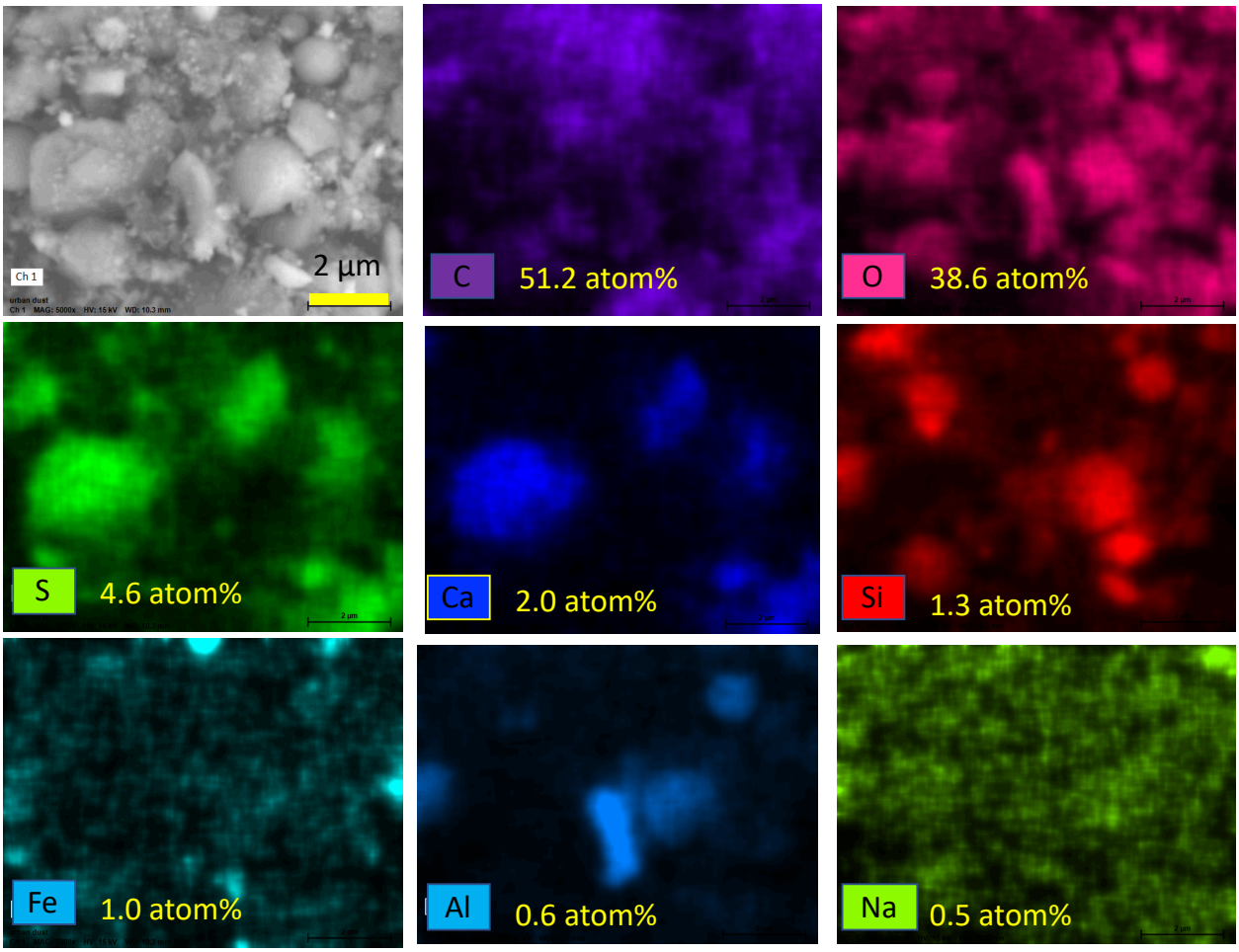

Figure S7. SEM-EDX mapping of urban dust

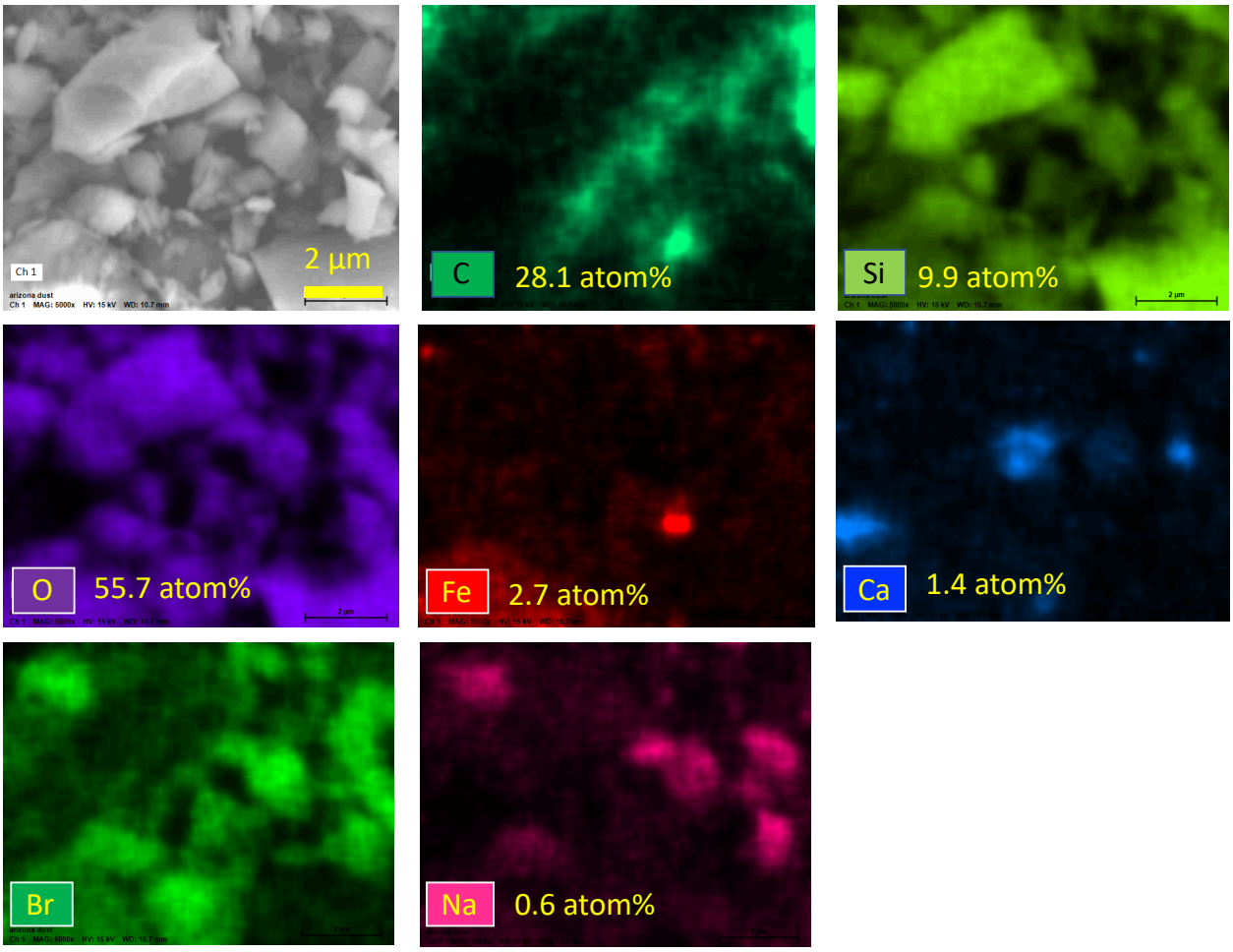

Figure S8. SEM-EDX mapping of Arizona dust

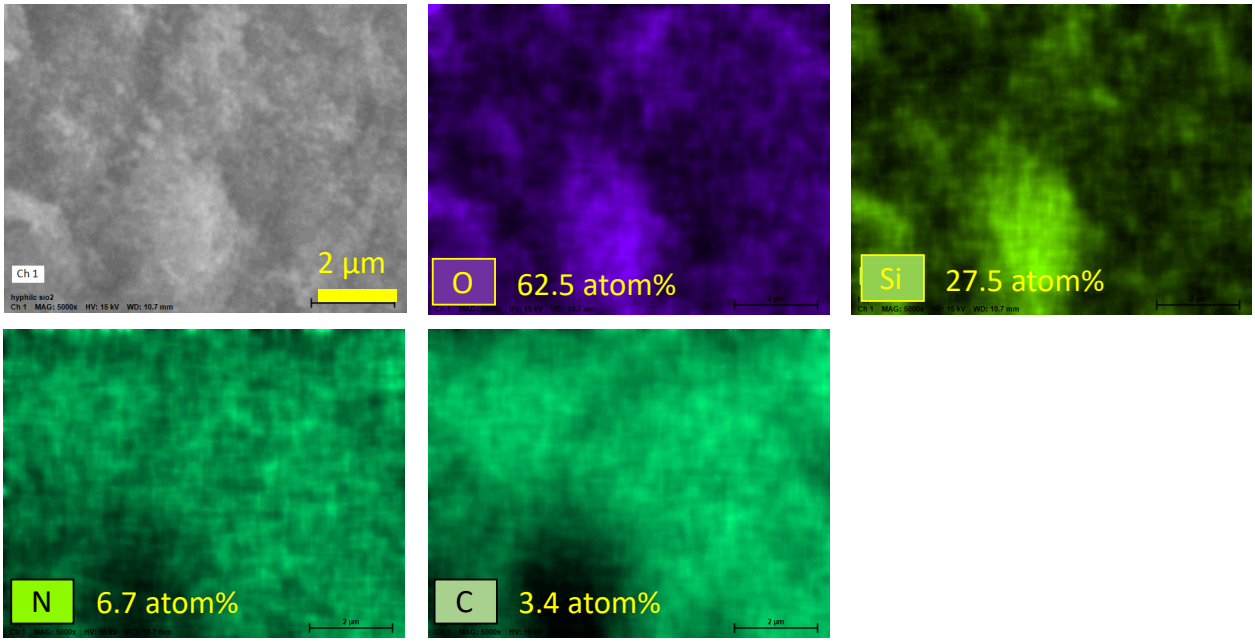

Figure S9. SEM-EDX mapping of Hydrophilic SiO_2 nanoparticle

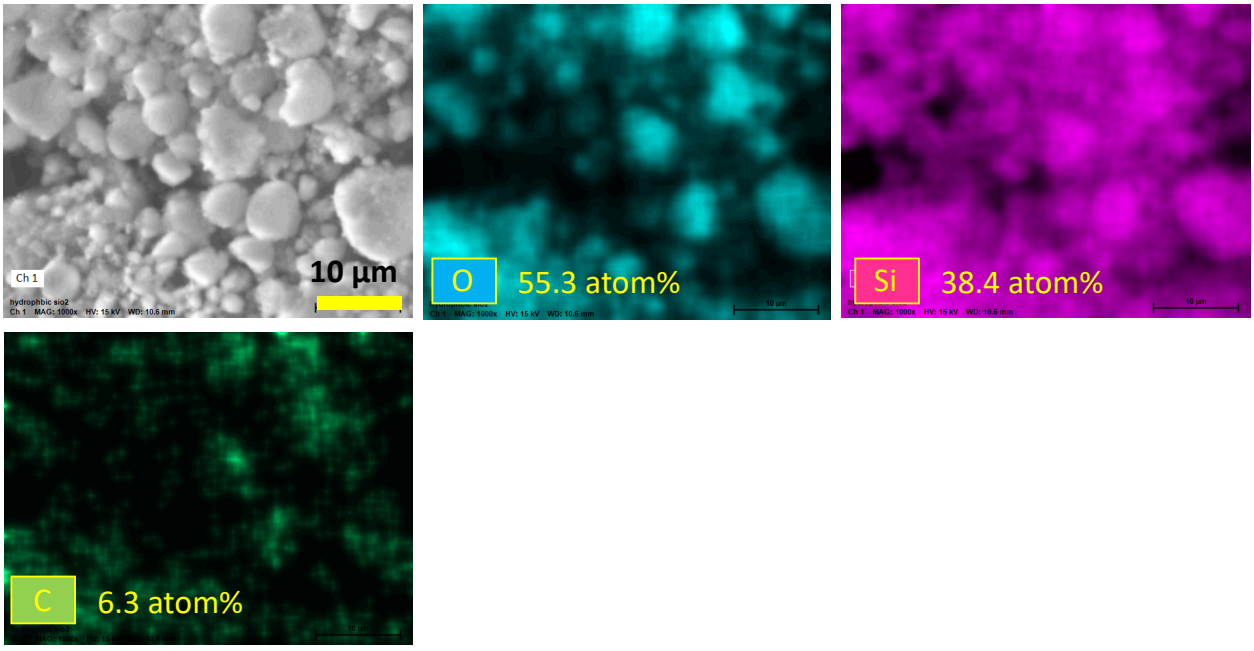

Figure S10. SEM-EDX mapping of Hydrophobic SiO_2 nanoparticle

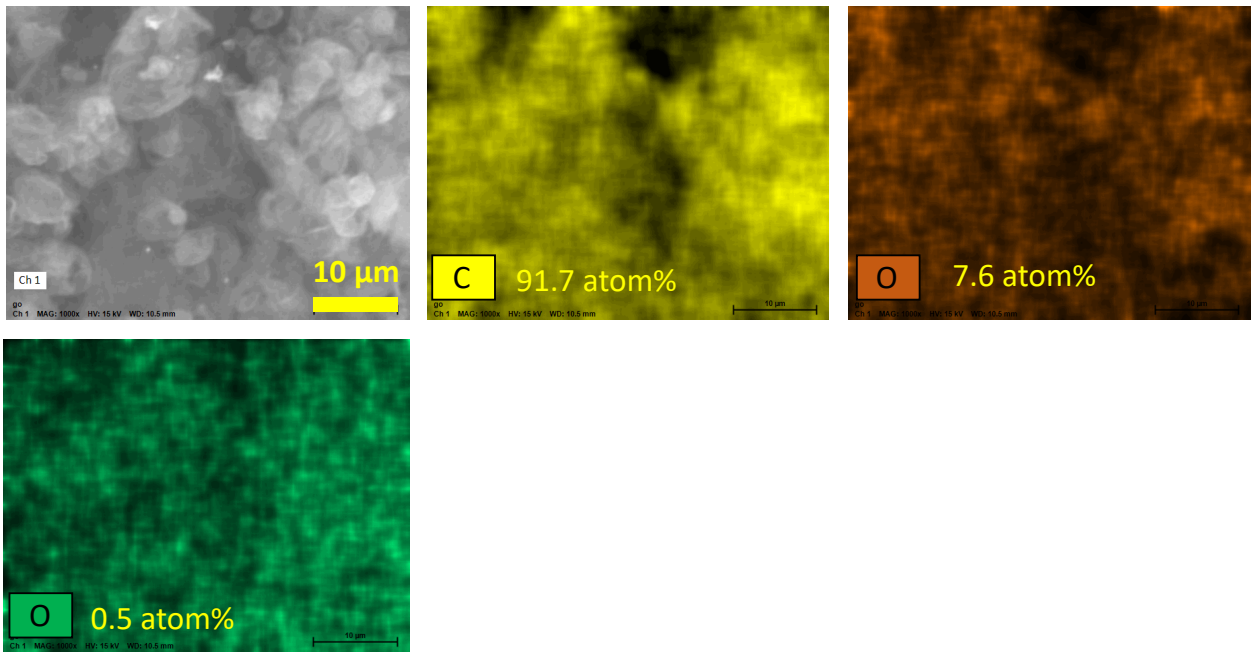

Figure S11. SEM-EDX mapping of graphene oxide powder

3. Decay Chains for IBIS Simulations

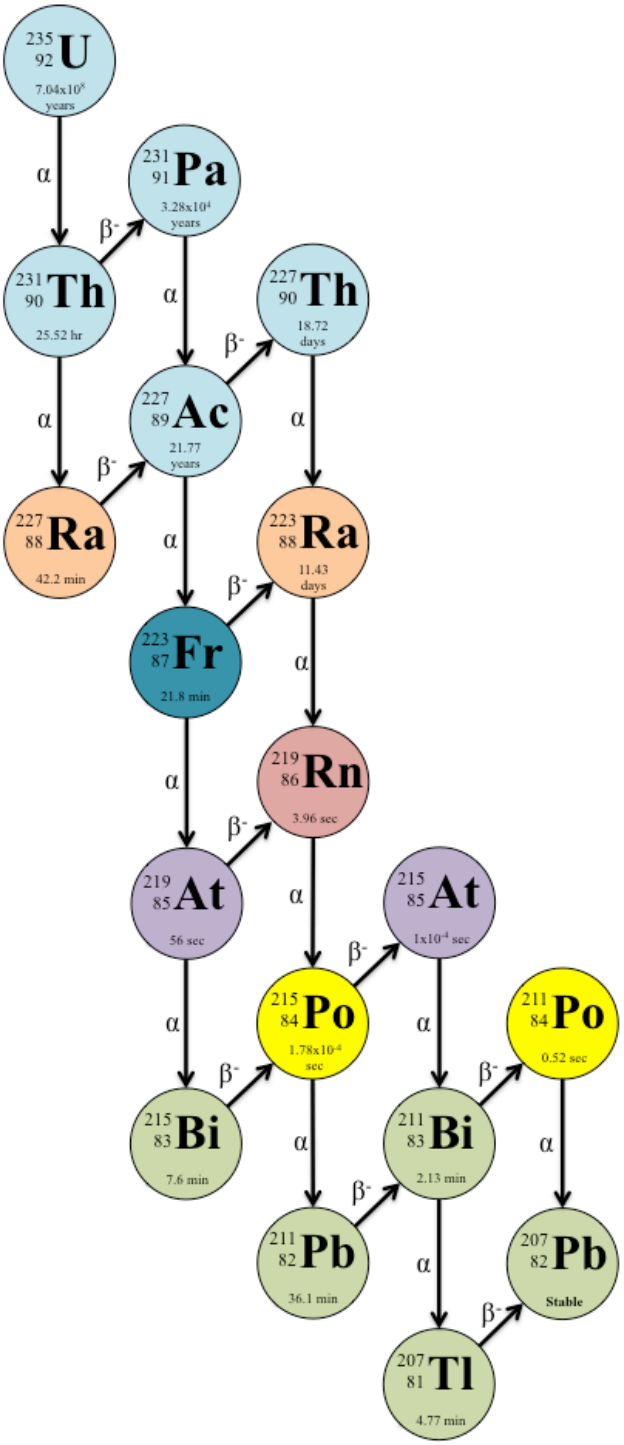

Figure S12. Decay chain used in IBIS simulations for the natural decay of Uranium-235 to stable Lead-207.

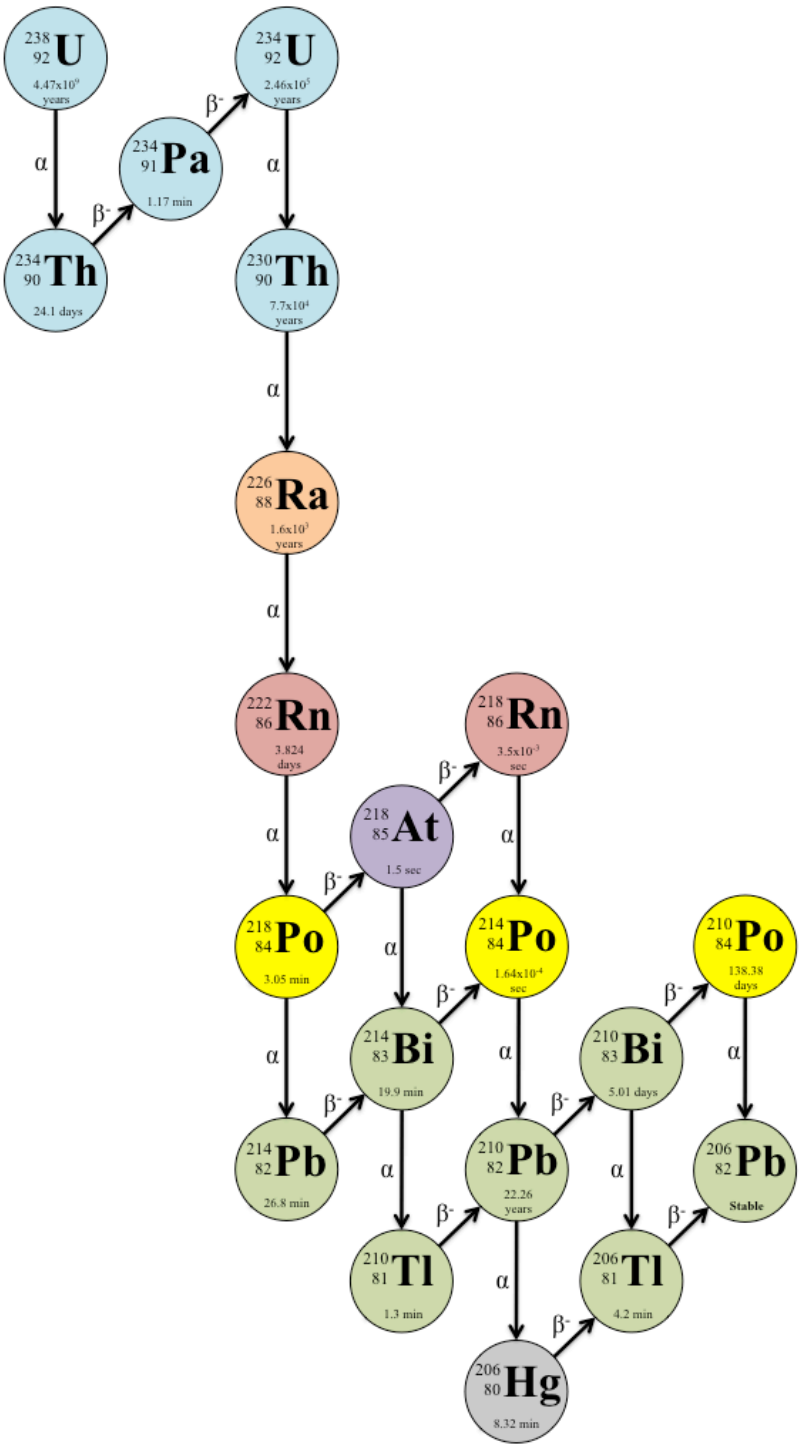

Figure S13. Decay chain used in IBIS simulations for the natural decay of Uranium-238 to stable Lead-206.