Supporting Information

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SI Text

Photophoretic Levitation of Aerosols for Geoengineering. ***Photopheresis.*** In the free molecular regime, the photophoretic force (1) on a thin plate is

$$F = \frac{1}{4}\Delta\alpha\frac{\Delta T}{T}p,$$

where F is the force per unit area of the plate, ΔT is the temperature difference between the plate and the ambient gas, which has temperature T and pressure p, and $\Delta\alpha$ is the difference between the molecular accommodation coefficients (α) on the two sides of the plate.

The temperature difference, ΔT, is determined by solving an energy balance equation,

$$\frac{S\varepsilon_S}{4} + \sigma\varepsilon_T T_E^4 - 2\sigma\varepsilon_T(T+\Delta T)^4 = \frac{3}{2}V\bar{\alpha}\frac{\Delta T}{T}p,$$

where S is the solar constant, ε_S is the solar band absorbance, ε_T the thermal band emissivity, and T_E is the mean radiative temperature of upwelling infrared. The term on the right is the heat loss for a diatomic gas, where $\bar{\alpha}$ is the average of α's on the two sides of the plate and V is the mean molecular speed (1).

The photophoretic force is roughly independent of altitude from 10 to 100 km despite the fact that pressure declines by $\sim 10^6$ over the same range, because conductive heat loss and thus ΔT are proportional to $1/p$, canceling out the pressure dependence in the force equation. The force declines as $\sim 1/p$ above $\sim$100 km as radiation begins to dominate conduction in determining particle temperature. The force declines in proportion to increasing p below about 10 km (for a 1-μ particle) as the mean free path becomes smaller than the particle size.

Electro- and magnetophotophoresis compared with gravito-photophoresis. Brownian motion randomizes particle orientation as the orienting torque becomes small compared to the thermal energy. The mean force, F_{mean}, in the direction of the aligning torque, τ, is

$$F_{\mathrm{mean}} = F_{\mathrm{BF}}L\left(\frac{\tau}{kT}\right),$$

where F_{BF} is the body fixed force and L is Langevin's function, $L(x) = \coth(x) - 1/x$.

For gravitationally oriented particles, the net upward gravito-photophoretic force declines as r^4 for particle radii below $\sim$1 μm because $L(x)$ is linear as $x \to 0$ and, assuming the particle has a fixed aspect ratio, the gravitational aligning torque is proportional to r^4.

Magnetic or electrostatic torques can greatly exceed gravitational torques for small particles in the upper atmosphere. Consider the 1-μm radius sphere analyzed by Rohatschek (2), in which the center of mass is displaced 0.1 μm from the particle's geometric center. A similar magnetite sphere with magnetization of 10^5 J T^{-1} m^{-3} would feel magnetic torques that exceeded gravitational torque by a factor of $\sim$8,000 at the typical terrestrial magnetic field strength of 0.5×10^{-4} T. Similarly, a sphere of barium titanate (3), a common ferroelectric, with residual charge of 2×10^{-3} C m^{-2} would experience a torque 750 times the gravitational torque in the typical atmospheric electric field of 100 V/m.

The larger mass-specific torques from electric or magnetic fields enable smaller particles to remain oriented in the face of Brownian torques, increasing the size range over which levitation of particles is possible and thus greatly increasing the peak mass-specific levitating force. Either electrostatic or magnetic forces provide sufficient torque to enable particles smaller than $\sim$0.5 μm to be levitated at a pressure of one atmosphere, which is not possible for gravitationally oriented particles with reasonable shapes (2). For electric or magnetic torques that are proportional to particle volume, the mass-specific $\Delta\alpha$ force decreases with particle radius as r^{-1}; the upward force per unit mass can exceed $10 \times g$ for particles of order 0.2 μm (where g is the acceleration due to gravity, $\sim$9.8 m/s^2).

Settling velocity and horizontal motion. Neglecting photophoretic forces, the fall speed of a plate with thickness h and density ρ is $gh\rho V/4p$. For the plate described in the paper with mass of 0.15 g/m^2, the fall speed would be 0.18 km/day at the stratopause. It would take tens of years to sink through the stratosphere to the tropopause. This implies that (*i*) advection rather than settling would be the dominant transport process in the lower stratosphere and that (*ii*) particles need not settle out during winter when they are not illuminated and thus experience no solar drive for photophoretic levitation.

At the mesopause settling (and levitation) velocities are much faster. Passive settling for a 0.15 g/m^2 plate is $\sim$30 km/day at 85 km. For a particle that experienced strong photophoretic forces, like the disk design described in this paper, the mesospheric pressure and particle velocities would both experience a strong diurnal cycle.

The horizontal drift speed of a thin plate with a small inclination from the horizontal plane is the same as the settling velocity assuming the vertical photophoretic and gravitational forces are in balance as would be the case for a disk that had drifted upward until confined below the stratopause or the mesopause. The drift speed is independent of tip angle because both the horizontal drag and the horizontal component of the upward force are proportional to inclination.

For example, while ΔT forces are typically unimportant for terrestrial aerosols, a thin-walled sphere would experience significant ΔT forces arising from radiatively driven temperature inhomogeneities. If the hemispheres were coated with dissimilar materials to produce the α contrast and the required gravitational torque, the sphere might be levitated by $\Delta\alpha$ forces while it was driven poleward, away from the mean solar flux, by ΔT forces.

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3. Fong DD, et al. (2004) Ferroelectricity in ultrathin perovskite films *Science* 304:1650–1653.