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Vertical transport of anthropogenic soot aerosol into the middle atmosphere

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Abstract. Gravito-photophoresis, a sunlight-induced force acting on particles which are geometrically asymmetric and which have uneven surface distribution of thermal accommodation coefficients, explains vertical transport of fractal soot aerosol emitted by aircraft in conventional flight corridors (10-12 km altitude) into the mesosphere (>80 km altitude). While direct optical effects of this aerosol appear nonsignificant, it is conceivable that they play a role in mesospheric physics by providing nuclei for polar mesospheric cloud formation and by affecting the ionization of the mesosphere to contribute to polar mesospheric summer echoes.

1. Introduction

The levitation or even lofting in the atmosphere of certain classes of aerosol would prolong the particles atmospheric residence times and extend their associated effects, if any. The phenomenon of aerosol lofting has been documented, for example, by *Pueschel et al.* [1997] who showed that soot aerosol, the source of which is arguably airline traffic in flight corridors near 10-12 km altitude, exist at up to 20 km geopotential altitude, and by *Rietmeijer* [1993] who sampled volcanic ash aerosol at 19 km altitude. Neither dynamic nor isentropic mixing is likely to explain vertical transport of aerosol against gravity in the thermally extremely stable stratosphere. *Rietmeijer* [1993] invoked stable autorotation that generates a sufficient lift force to loft nonspherical volcanic ash particles to 17-19 km altitude at latitudes that are higher than the latitude of eruptions. While such forces may act also on asymmetric fractal soot aerosol particles, in this paper we show that another nonconventional force, namely gravito-photophoresis, can move particles from 10 km to high altitudes that previously appeared off-limits to aerosols of terrestrial origin. Possible effects of those aerosols therefore have been largely ignored. If lofted to above 80 km altitudes, for example, the particles could participate in mesospheric physics; in particular, they could nucleate noctilucent clouds (NLCs) in the mesosphere which have been observed to intensify over the past 100 years and possibly could help in the interpretation of yet unexplained radar echoes in the mesosphere known as polar mesospheric summer echoes (PMSEs).

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Photophoresis in the broadest sense means motions of freely movable solid or liquid particles during their irradiation by light under action of the surrounding gas. The motions are caused by radiometric forces resulting from normal and tangential stresses on the particle surface due to temperature gradients in the gas surrounding the surface. Such gradients can be produced in two ways, either by a difference in temperature over the (homogeneous) surface of the particle resulting in ΔT_s forces, or by a difference of thermal accommodation coefficients over the (even isothermal) surface yielding $\Delta\alpha$ forces [*Preining*, 1966; *Rohatschek* 1985,1989].

In the past three decades the question was raised repeatedly whether photophoretic forces arising from irradiation of particles by sunlight could cause a rising motion and thus prolong the atmospheric residence time of certain types of atmospheric aerosols. The best-known example of photophoresis is longitudinal photophoresis. It describes motions in the direction (positive photophoresis) or against the direction (negative photophoresis) of light [*Preining*, 1966]. This effect is caused by ΔT_s forces resulting from a temperature difference over the particle surface. If the sun is the source of irradiation, only negative photophoresis, that is, a force pointing in the direction of the sun, could pose a lifting component that opposes the force of gravity.

Theoretical treatments of photophoresis followed the standard procedure of calculating the photophoretic force from the distribution of surface temperatures due to light absorption within the particle. Application of the principles of photophoresis to stratospheric particles was pioneered by *Hidy and Brock* [1967]. However, because these researchers utilized the opaque particle model which exclusively yields positive photophoresis, their work did not contribute to solving the levitation and lofting problems in a sunlit atmosphere. Using complex refractive indices, *Kerker and Cooke* [1982] obtained positive and negative photophoretic forces. However, the resulting ratio of photophoretic to gravitational forces was never larger than 0.04. *Orr and Keng* [1964] investigated experimentally the behavior of small salt and metal particles exposed to a light beam directed vertically upward or downward. They found that some particles in the stratosphere may actually be lofted against gravity, while others were induced to fall more rapidly than they would under gravity alone. However, their test particles were atypical of most atmospheric aerosols, their light intensity exceeded that of the sun severalfold, and the spatial distribution of their light field was poorly defined. *Tong* [1973] investigated photophoresis of centimeter-sized suspended spheres and was able to produce a negative force on a transparent glass sphere that was blackened over its rear, but not its front, surface. *Lewittes et al.* [1982] and *Arnold and Lewittes* [1982] concluded from experimental results obtained from 10 μm-sized dyed glycerol droplets irradiated by IR radiation that it was conceivable that certain aerosols may be levitated in the upper atmosphere by photophoresis.

Solid particles can exhibit a great variety of motions sustained by absorbed light which extends the action of light beyond that of longitudinal photophoresis. One of those motions is gravito-photophoresis which includes helical motions around the vertical direction [*Ehrenhaft and Reeger*, 1951]. As shown by *Rohatschek* [1956], gravito-photophoresis is related to two basic requirements: a body-fixed photophoretic force and a restoring torque which orients the particle with respect to the direction of gravity. The restoring torque is induced by a different location of the center of gravity, the point of action of the gravitational force, from the center of reaction at which drag acts. The body-fixed force may result from differences in thermal accommodation coefficients α, resulting in the action of $\Delta\alpha$-forces [*Rohatschek* 1984, 1985, 1995, 1996]. These forces are independent of a temperature gradient at the surface of the particles.

In only one case has a body-fixed photophoretic force acting on liquid particles been observed. It deals with electro-photophoresis causing an analogue motion sustained by light because of $\Delta\alpha$-forces related, however, to the direction of an electric field instead of to gravity. This effect was observed on Hg droplets slightly contaminated by Sn, Zn, etc., but not with pure Hg [*Reiss*, 1932].

In laboratory experiments at air densities typical of the middle atmosphere from the stratosphere to the mesosphere, the lofting of micrometer-sized particles of graphite powder and carbonized sunflower marrow powder was demonstrated [*Rohatschek* 1984, 1989]. Experiments with agglomerates of chimney soot (unpublished) demonstrated the lofting under solar-constant illumination also of this aerosol. Theoretical treatises [*Rohatschek* 1995, 1996] documented the possibility of levitation and lofting of atmospheric aerosol over a wide altitude range up to 85 km into the mesosphere. The mesosphere has been subject to intensive research since the discovery of NLCs, which today are also known as polar mesospheric clouds (PMCs), more than 100 years ago [*Blackhouse*, 1885; *Jesse*, 1885], and has also received attention in the popular press in recent years [*Stone*, 1991; *Gore*, 1992] because of the proposition that PMCs are a harbinger of global change. *Thomas et al.* [1989] argue that PMCs actually did not exist before the discovery of NLCs in 1885 and that they have been steadily increasing due to the increase in anthropogenic methane which, after upward transport in the tropics and subsequent photodissociation, contributes about half to the water content in the mesosphere. Alternatively, a PMC increase could also be caused by a drop in mesopause temperature [*Gadsden*, 1990] due to the increase in anthropogenic CO_2.

In this paper we document that soot particles emitted by aircraft in flight corridors at 10-12 km altitude are subject to transport into the mesosphere by gravito-photophoresis. This is shown by applying *Rohatschek's* [1996] concepts of gravito-photophoresis to atmospheric soot aerosol that was sampled in commercial airline flight corridors over the eastern Atlantic Ocean. The largest particles out of an actually measured soot aerosol size distribution can be lofted against gravity from the upper troposphere/lower stratosphere near 10 km altitude into the mesosphere. There they could provide the freezing nuclei around which mesospheric ice particles form. Thus the possibility exists that anthropogenic soot aerosol emitted from commercial aircraft operated in conventional flight corridors also could affect the microphysics and optical properties of the middle atmosphere.

2. Gravito-Photophoresis of a Model Particle

Adopting the example originally presented by *Rohatschek* [1996], consider a spherical model-particle (Figure 1) that possesses rotational symmetry with respect to the distribution of

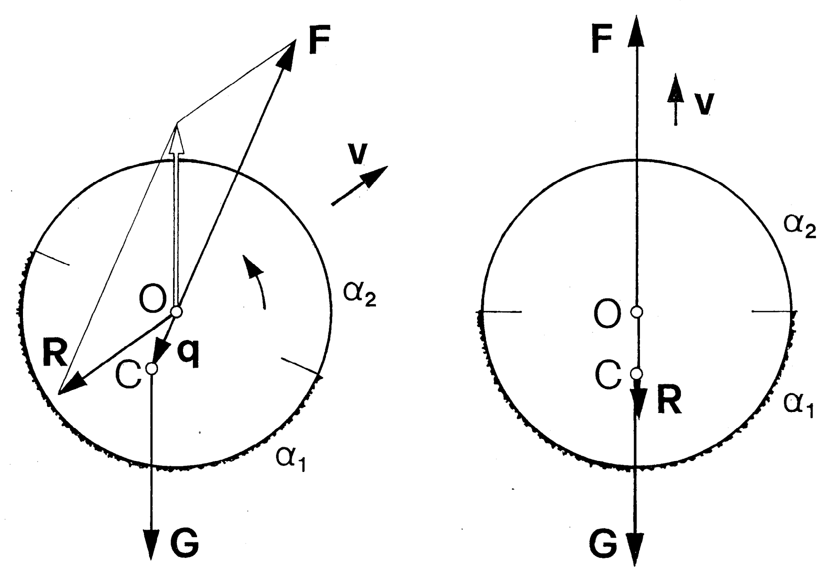

Figure 1. Forces acting on the model particle with gravito-photophoresis: **F**, photophoretic $\Delta\alpha$-force; **G**, force of gravity (weight); **R**, resistance (drag) force. (left) Nonequilibrium state; the forces add up to a restoring torque by a couple of forces: **G** and **F+R=-G** and (right) Stable equlibrium is shown. (Reprinted from Journal of Aerosol Science, Volume 27, H. Rohatschek, Levitation of stratospheric and mesospheric aerosols by gravito-photophoresis, pp.467-469, Copyright (1996), with permission from Elsevier Science).

thermal accomodation coefficients α and the mass density as follows: Let the surface be divided into two hemispheres with different accommodation coefficients α_1 and α_2. The difference in accommodation coefficients could be caused, besides an uneven distribution of ruggedness or gaps (see Figure 3), by adsorption of H_2SO_4, formed in aircraft exhaust coincidentally with the production of soot, unevenly across the particle surface [*Kärcher*, 1996]. If the temperature of that particle is elevated, for example, because of irradiation from the sun, then a body-fixed force **F** pointing from the side of higher to the side of lower α arises. It has been shown [*Rohatschek* 1995, 1996] that this gravito-photophoretic force acting on a particle is

$$F = \left(\frac{1}{12\bar{c}}\right)\left(\frac{1}{1+\left(\frac{p}{p^*}\right)^2}\right)\left(\frac{\Delta\alpha}{\bar{\alpha}}\right)H \tag{1}$$

where the factor 1/12 holds for diatomic gases and hemispheric distribution of α, $\bar{c}$ is the mean speed of the air molecules, p is atmospheric pressure, p^* is a characteristic pressure inversely proportional to the radius r of the particle (the corresponding Knudsen number has a value of approximately 1/2), $\Delta\alpha=\alpha_1-\alpha_2$, $\bar{\alpha}=(\alpha_1+\alpha_2)/2$, and H denotes the net energy flux transferred by the molecules.

The material is assumed to conduct heat so well that any photophoretic force due to inhomogeneous surface heating (ΔT_s force) can be neglected. For a $\Delta\alpha$ force, the direction of incidence of light, its direct or diffuse character, and the distribution of heat sources over the volume are irrelevant. This force is sustained not by a temperature difference over the surface but by the temperature difference between the particle and the surrounding gas, whatever its cause may be. A $\Delta\alpha$ force is independent of the heat conductivity of the particle.

Let the center of gravity C be displaced from the geometric center O by a distance **q** along the axis into the hemisphere of higher α (Figure 1). The photophoretic force **F** (acting along the axis which passes through both O and C), the drag force **R** (acting through O which is identical to the center of reaction), and the force of gravity **G** (acting through C) yield zero vector sum, provided that inertial reactions can be neglected. These forces produce a directional torque (Figure 1, left) which results in stable equilibrium (Figure 1, right). There the axis is vertical, C is below O, and the force **F** points upward.

Molecular perturbation of the orientation of the particle (Brownian rotation) causes the body-fixed force to deviate from the upward direction in a random fashion. As a consequence, merely the average component of F upon the vertical direction becomes effective for lifting

$$F_g = F\mathfrak{L}(x), \tag{2}$$

where $\mathfrak{L}(x) = \coth x - 1/x$ denotes Langevin's function with the argument

$$x=qG/kT, \tag{3}$$

where k is Boltzmann's constant and T is the gas temperature far away from the particle.

Depending on the ratio F_g/G, the particle settles more slowly than under the action of G alone, it remains suspended, or it moves upward against gravity owing to the symmetry assumed. Any ratio $F_g/G>0$ for a particular aerosol would separate this aerosol from any other with $F_g/G=0$.

The atmospheric variables that affect gravito-photophoresis most strongly (equations (1) and (2)) are pressure and temperature. The ratio p/p^* accounts for the fact that the radiometric $\Delta\alpha$ force F for given H is constant at extremely low pressures but inversely proportional to the square of p at extremely high pressures. As a consequence, on a plot of F versus p on logarithmic scales the asymptotes of both branches intersect at $p=p^*$. The dependence of F_g on temperature results from its relationship to the mean speed of the molecules, $\bar{c}=(8kT/\pi m)^{1/2}$, and of $\mathfrak{L}$ to its argument qG/kT.

The molecular energy (heat) flux H appearing in equation (1) can be determined from the energy balance

$$H+E_{emi}=AI+E_{abs} \tag{4}$$

where AI is the energy flux absorbed from the sun and E_{abs} and E_{emi} are the absorbed and emitted energy fluxes due to thermal radiation, respectively. For sufficiently high pressures, the expression E_{emi}-E_{abs} can be neglected in relation to AI and

$$H=AI \tag{5}$$

This relationship implies that all energy absorbed, AI, is removed from the particle by molecular heat transfer, H. This is a useful approximation for pressures down to typically 1 hPa or up to about 50 km pressure altitude. As the pressure decreases further, however, the particle becomes increasingly hotter, and the energy emitted by radiation, E_{emi}, increases at the expense of H such that gravito-photophoresis eventually ceases to play a role.

In the general case therefore, all terms of equation (4) must be taken into account. H and E_{emi} can be expressed by the mean temperature T_s of the particle surface. E_{abs} is determined by the temperature of the radiating surroundings. For calculating H, an interpolation formula [*Rohatschek*, 1995]

$$H=4\pi r^2 (3/4)\ \bar{\alpha}\ (p\,\bar{c}\,/\,T)\ (1/[1+\mu p/p^*])(T_s-T) \tag{6}$$

where r is particle radius and μ is a coefficient of the order of ½ and can be used.

Figure 2 shows the ratio F_g/G for the lower and middle atmosphere with data from the U. S. Extension to International Civil Aviation Organization Standard Atmosphere [*Weast*, 1976]. The model particle is assumed to be a perfectly absorbing sphere with radius r=1µm, $\Delta\alpha\,/\,\bar{\alpha}\,=0.15$, ρ_p=1 g cm^{-3}, q=0.4 µm, $A=\pi r^2$, and n=1.95-0.66i. The energy balance (4) is taken fully into account. The assumption is made that particles at altitudes above 6 km receive planetary thermal radiation corresponding to T=255 K (240 W m^{-2}).

With increasing pressure toward the Earth's surface, the lifting force falls off quickly (equation 1). The result is $F_g/G<1$, and particles will settle. Thus only the particles emitted at $H\geq10$ km by aircraft, or surface-emitted particles raised to this altitude by convection, have a chance to be lofted by gravito-photophoresis ($F_g/G>1$). Above that altitude, throughout the middle atmosphere, particles can get lofted against gravity. When $F_g/G>2$, average suspension can take place even at a 12-12 hour ratio of daylight and night. Both maxima in the stratosphere and the mesosphere are due to low atmospheric temperatures. The minimum in be-

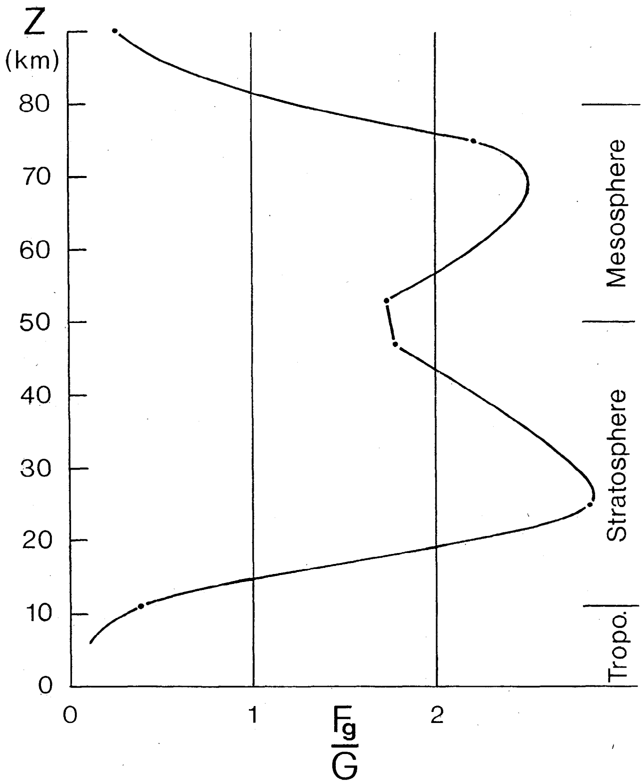

Figure 2. Average lifting component F_g in relation to weight G in a standard atmosphere at solar-constant irradiation. (Reprinted from Journal of Aerosol Science, Volume 27, H.Rohatschek, Levitation of stratospheric and mesospheric aerosols by gravito-photophoresis, pp. 467-475, Copyright (1996), with permission from Elsevier Science).

tween is due to the high temperature at the stratopause. In the mesosphere, below pressures of the order of 1 hPa, F_g/G becomes smaller because the dominating energy loss, E_{emi}, is due to thermal radiation of the particles.

3. Gravito-Photophoresis of Soot Particles in the Atmosphere

Figure 3 is a collage of soot particles as they appear in a scanning electron microscope (SEM) at 50,000 times magnification. The images resulted from a sample that was collected on October 23, 1997, near 10 km altitude over the northeastern Atlantic Ocean (latitude 57.0° N; longitude 9.0° W) during the Subsonic Assessment Ozone and Nitrogen Oxide Experiment (SONEX). It is a subset of seven particles out of a total population of 54 for which the lofting ratios of gravito-photophoretic force to gravity, F_g/G, have been computed. The indicated longest dimensions D_{ge} of the fractals and the diameter d_0 of individual spherules making up the fractals can easily be measured from the photographs. The fractal dimension typical for soot is $f\approx2$ [*Nyeki and Colbeck*, 1994], and the density of individual spherules ρ_0=2 g cm^{-3} is typically that of graphite. Thus the fractal characteristics [*Magill*, 1991] of those particles that determine their gravito-photophoretic behavior can be computed with the information in Figure 3: $N=(D_{ge}/d_0)^f$ is the number of individual spherules within a particle; $M=Nm_0$ (where $m_0=\rho_0\pi d_0^3/6$ is the mass of a spherule) is the mass of a fractal particle; $G=Mg$ (where g=981 cm s^{-2} is the acceleration due to gravity) is the gravitational force acting on a fractal particle. For the computations of F_g we assumed thermal accommodation coefficients $\alpha_1=2\alpha_2$ and the distance between the center of gravity C and the geometric center O (Figure 2) to be $q = D_{ge}/4$.

There is extensive literature on the optical properties of carbon soot reporting some widely varying values [see, e.g., *Colbeck et al.*, 1989]. *Lee and Tien* [1981] established a model for the optical constants of soot including a rigorous consideration of the electronic band structure and concluded that n=2.0-1.0i was the maximum refractive index for visible wavelengths. For our calculations we adopted the refractive index n=1.95-0.66i established by *Senftleben and Benedict* [1919] which also was used by *Rohatschek* [1996].

The variation with wavelength and refractive index of specific absorption (absorption per unit mass) follows from the Rayleigh-Debye theory [*Berry and Percival*, 1986] as

$$B_a \approx \left(\frac{6\pi}{\rho\lambda}\right) \mathrm{Im}\frac{\left(n^2-1\right)}{\left(n^2+2\right)} \tag{7}$$

where Im denotes the imaginary part of the ratio $(n^2-1)/(n^2+2)$ and the fractal particle density is $\rho=\rho_0(d_0/D_{ge})^{3-f}$ [*Magill*, 1991]. The molecular heat flux then results as $H=B_aMI$ where $I=1.36\times10^6$ ergs s^{-1} cm^{-2} is the solar constant.

With these values the forces G and F_g were computed for temperatures and pressures corresponding to a standard atmosphere [*Valley*, 1965]. Table 1 shows relevant characteristics for the particles illustrated in Figure 3 at 10 km altitude

It follows from Table 1 that only one out of the seven particles illustrated in Figure 3 had a chance to get lofted against gravity. Because it is the largest particle out of this ensemble, however, it would have carried 75% of the mass of the selected particles. The overall size distribution consisted of a total of 54 particles. Sixteen percent of those particles, comprising 51% of the mass of the particle size distribution, were determined to be lofted against gravity. The results are summarized in Figure 4 where the ratio between the vertical component of the gravito-photophoretic force, F_g, computed according to (1)-(3), and gravity, G, is plotted against altitude. A ratio F_g/G=1 results in levitation. A ratio F_g/G>1 results in lofting. A ratio $F_g/G\geq2$ means continuous lofting over 24 hour days consisting of 12 hours night and 12 hours sunshine.

The six thin curves in Figure 4 were derived by binning the 54 total particles into 6 classes, each consisting of 9 particles. Table 2 shows the average physical characteristics of the fractals in each particle class.

The seventh heavy curve in Figure 4 is the lower portion (5-50 km altitude range) of the curve in Figure 2. Thus it follows that Rohatschek's model sphere of 1 µm radius is equivalent to atmospheric fractal soot particles of longest dimension $0.7<D_{ge}<1.0$ µm.

It follows from Figure 4 that at altitudes at or below 5 km none of the particles will get levitated. At altitudes that low in the atmosphere, the particles unambiguously will settle, albeit at a smaller rate than those particles which are not subject to gravito-photophoresis, i.e., spherical and nonabsorbing particles. At 10 km altitude and above, however, a significant fraction of atmospheric soot particles can be lofted against gravity up to 50 km and beyond. Even though the particles that have a chance to get lofted are only 16% by number, they carry 51% of the mass of the soot aerosol size distribution that we investigate here. Once lofted to 50 km, further transport to above 80 km is easily accomplished (see Figure 2).

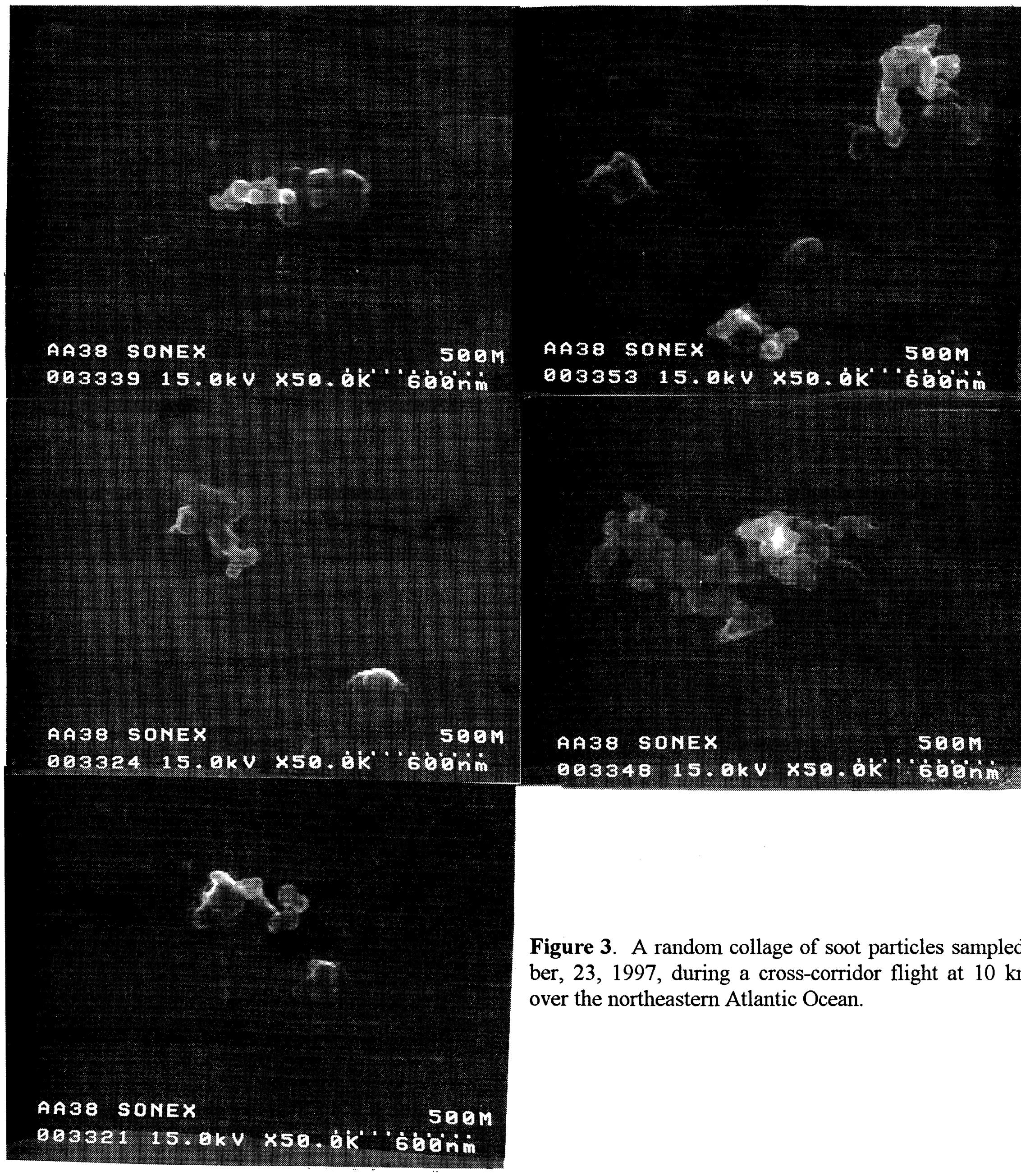

Figure 3. A random collage of soot particles sampled on October, 23, 1997, during a cross-corridor flight at 10 km altitude over the northeastern Atlantic Ocean.

The vertical velocity v_g is found from

$$v_g = v_s\left(1-\frac{F_g}{G}\right) \tag{8}$$

where

$$v_s = \left(\frac{\rho_0 g d_0^2}{18\eta_f}\right)\left(\frac{D_{ge}}{d_0}\right)^{f-1} \tag{9}$$

is the settling velocity of the fractal soot particles [*Magill*, 1991], where η=1.5×10^{-4} g cm^{-1} s^{-1} is the viscosity of air. The fractal dimension of a sphere is f=3. From (9) it follows that $v_{S,SPHERE}=(D_{ge}/d_0)v_{S,FRACTAL}$ with $3.2 \leq D_{ge}/d_0 \leq 15.2$ (Table 2). Thus it follows that a sphere settles up to 15 times faster than does a fractal. In terms of atmospheric residence times, this means that a fractal particle can last up to 15 times longer than a corresponding spherical particle.

Taking d_0 = 65 nm as the average diameter of individual spherules and $\overline{D}_{ge}$ = 1000 nm as the average diameter of a sphere just sufficient to enclose the particle in the largest-size class (Figure 3 and Table 2), and assuming f=2.0 as the fractal dimension characteristic of soot, the average settling velocity for the largest-size class of soot particles is v_s=4.7×10^{-4} cm s^{-1}. Hence, the average vertical ascent velocity for the largest-size class is v_g=9.5×10^{-4} cm s^{-1} at 10 km and v_g= 9.0×10^{-3} cm s^{-1} at 20 km altitude, respectively. Thus it takes approximately 30 years for transport from 10 to 20 km altitude, and 20 years from 20 to 80 km altitude.

Table 1. Fractal Characteristics as Function of Measured D_{ge} and Measured d_0, and Gravity $G=Mg$ and F_g/G of the Particles illustrated in Figure 3

Particle Number	D_{ge}, cm	d_0, cm	N	ρ, g cm^{-3}	M, g	G, dynes	F_g/G
1	7.1E-5	8.0E-6	79	0.23	4.2E-14	4.2E-11	0.11
2	3.0E-5	6.4E-6	22	0.42	6.0E-15	5.9E-12	0.03
3	5.8E-5	8.0E-6	53	0.28	2.8E-14	2.8E-11	0.33
4	4.2E-5	6.6E-6	41	0.31	1.2E-14	1.2E-11	0.10
5	5.0E-5	6.0E-6	69	0.24	1.6E-14	1.5E-11	0.19
6	1.4E-4	4.0E-6	1173	0.06	3.7E-13	3.6E-10	10.7
7	5.6E-5	8.0E-6	49	0.29	2.6E-14	2.6E-11	0.30

Read 7.1E-5 as 7.1×10^{-5}. Fractal characteristics are $N=(D_{ge}/d_0)^f$ = number of monomers per fractal; $\rho=\rho_0(d_0/D_{ge})^{3-f}$ = density of fractal; $M=Nm_0$ = mass of fractal where D_{ge} is the diameter of a sphere surrounding the fractal and d_0 is the diameter of a monomer. Fg/G is the ratio of gravito-photophoretic force to gravity.

The vertical mass flux of ascending soot becomes

$$\text{Flux} = v_g\, m \tag{10}$$

where m is the mass density of soot particles in the stratosphere. With an average stratospheric soot mass loading m=0.5 ng m^{-3} [*Pueschel et al.*, 1992] in the northern hemisphere, the vertical flux of soot at 20 km amounts to $5.\times10^{-18}$ g cm^{-2} s^{-1} which is within 1 order of magnitude of the mass influx of meteoritic dust [*Hunten et al.*, 1980].

The strong influence of fractal particle characteristics on its physical properties (Tables 1 and 2) strongly suggests that we re-evaluate soot data that we published previously [*Pueschel et al.*, 1992; 1997; IPCC Report 1999]. In the past we used the arithmetic mean of long and short dimensions of SEM images of the particles to determine the diameter of an "equivalent sphere," defined as a sphere whose volume is equal to that of the fractal particle, and we adjusted the particle density to unity to account for the fact that the sphere thus designated was not completely filled by soot monomers. This analysis has been repeated for the seven particles shown in Figure 3 and the resulting surface area and particle mass (subscript *ES*) is compared with the corresponding fractal properties (subscript *FR*) in Table 3.

It follows from Table 3 that our "equivalent sphere" method underestimates the soot particle surface area by 80% but overestimates particle mass by up to 90%. The discrepancy in mass is, furthermore, strongly size-dependent owing to the strong size-dependence of fractal particle density (column 5 in Table 1).

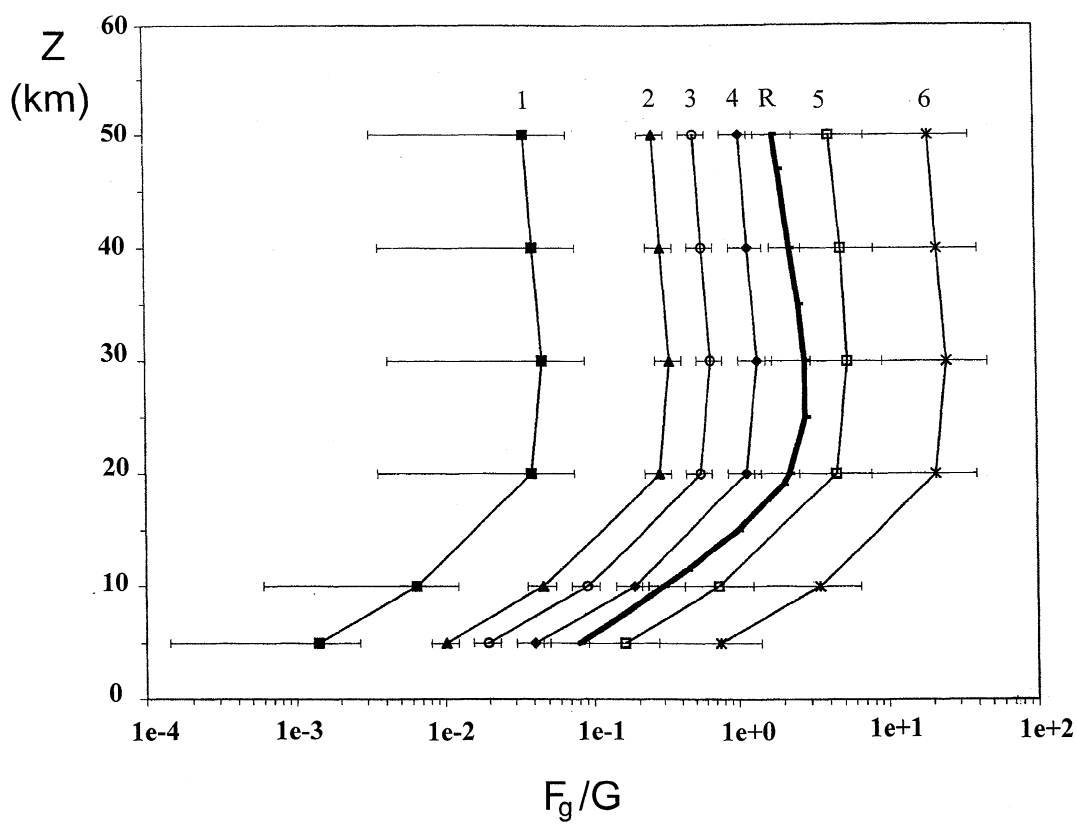

Figure 4. Ratio of gravito-photophoresis to gravity, F_g/G, versus altitude for six particle classes defined in Table 2. The heavy curve, R, is a section of the plot in Figure 2 for the 5-50 km altitude range.

Table 2. Average Physical Characteristicsas Function of Mean $\overline{D}_{ge}$ and Mean $\overline{d}_0$ for Six Size Classes.

Particle Class	$\overline{D}_{ge}$, cm	$\overline{d}_0$, cm	$\overline{N}=(\overline{D}_{ge}/\overline{d}_0)^f$	$\overline{\rho}=\rho_0(\overline{d}_0/\overline{D}_{ge})^{3-f}$, g cm^{-3}	$\overline{M}=\overline{N}m_0$, g
1	(2.0±0.4)E5	(6.3±1.6)E-6	10	0.63	2.6E15
2	(3.5±0.2)E-5	(7.0±1.1)E-6	25	0.40	9.0E-15
3	(4.1±0.2)E-5	(6.2±1.0)E-6	44	0.30	1.1E-14
4	(5.0±0.3)E-5	(6.1±0.6)E-6	67	0.24	1.6E-14
5	(6.7±1.1)E-5	(6.8±0.6)E-6	97	0.20	3.2E-14
6	(9.9±1.6)E-5	(6.5±1.5)E-6	232	0.13	6.6E-14

Read 2.0E-5 as 2.0×10^{-5}. $\overline{N}$ is the average number of monomers, $\overline{M}$ the average mass, $\overline{\rho}$ is the average density, $\overline{D}_{ge}$ is the average diameter of the smallest spheres surrounding a fractal, and $\overline{d}_0$ the average diameter of the monomers per fractal class.

Taking into account the difference in aerodynamic characteristics of fractals in relation to spheres and accounting for the fact that soot particles are solid and therefore subject to bounce, *Strawa et al.* [1999] showed that the collection efficiency of our impactors for soot is only 25% of that for equivalent spherical droplets. Thus, fortuitously, the smaller mass due to a lower fractal density is compensated, at least in part, by the gain due to an adjusted collection efficiency for soot aerosol.

4. Discussion

We have documented that approximately 50%, by mass, of a soot particle size distribution measured in flight corridors over the northeast Atlantic Ocean can be lofted into the middle atmosphere by gravito-photophoretic forces. Possible significance of this phenomenon are potential effects on atmospheric optics (direct effect) and on mesospheric cloud physics (indirect effect).

Mesospheric ice particles at 80-100 km geopotential altitude are designated harbingers of global change [*Thomas*, 1991; *Thomas et al.*, 1989]. The argument is that the first observation of noctilucent (night-luminous) clouds (NLCs), a little more than a century ago [*Blackhouse*, 1885; *Jesse*, 1885], was not an accidental discovery of an existing phenomenon but an emergence of a previously undetectable cloud into sudden visibility. Previous to that time, NLCs would not have been routinely visible because the pre-1885 water vapor (H_2O) concentration in the mesosphere was insufficient to produce visible clouds. The microphysical model of *Jensen* [1989] shows a century-long increase in NLC brightness by nearly a factor of 10. There is also evidence for an increase in frequency of NLC occurrence in the last 20-30 years [*Gadsden*, 1990]. The cause of this increase in mesospheric cloudiness has been attributed to an increase in mesospheric water vapor, or a decrease of mesospheric temperatures, or both.

Temperature and water vapor are important for the brightness of PMCs as they influence nucleation, growth, and sedimentation of the ice particles. At the cold mesopause at high latitudes, IR effects due to increasing CO_2 could warm the region without dynamical feedbacks, causing a net cooling of the atmosphere below at all latitudes. *Thomas* [1996] argues that the mesospheric cloud existence region, defined in terms of water-ice saturation, may have advanced from near the pole to its current location inside the 50°-90° latitude zone because temperature lowered owing to an increase in CO_2 since the industrial revolution. However, *Lübken* et al. [1996] report a remarkable repeatability of the mean temperature (150±2 K) below the mesopause ever since the first measurement 30 years ago and a persistent height at which NLCs occur (83.1 km) since the very first measurements more than 100 years ago.

This still leaves water vapor increase as a plausible explanation of the increase in frequency of occurrence and extent of PMCs. Both methane (CH_4) (currently 1.6 ppmv) and tropospheric water vapor (H_2O) enter the stratosphere within air rising mainly through the cold tropical tropopause in the so-called Hadley cell circulation. All the CH_4 survives the passage, but most of the H_2O is precipitated out at the cold tropopause, leav-

Table 3: Particle Surface Area and Mass for fractal Soot Particles shown in Figure 3 and corresponding Values of an equivalent Sphere.

Particle No.	Fractal Particle Characteristics		Equivalent Sphere Particle Characteristics			
	A_{FR} (cm^2)	M_{FR} (grams)	A_{ES} (cm^2)	M_{ES} (grams)	A_{FR}/A_{ES}	M_{FR}/M_{ES}
1	1.6E-8	2.1E-14	8.9E-9	7.9E-14	1.8	0.3
2	2.8E-9	3.0E-14	1.6E-9	6.1E-15	1.8	0.5
3	1.1E-8	1.4E-14	5.9E-9	4.4E-14	1.9	0.1
4	5.6E-9	6.2E-15	3.1E-9	1.7E-14	1.8	0.4
5	7.8E-9	7.8E-15	4.4E-9	2.8E-14	1.7	0.3
6	6.2E-8	4.1E-14	3.5E-8	6.3E-13	1.8	0.1
7	9.8E-9	6.6E-15	5.5E-9	3.9E-14	1.8	0.2

Read 1.6E-8 as 1.6×10^{-8}.

ing only 3 ppmv [*Jones et al.*, 1986] to enter the stratosphere. In the upper stratosphere, CH_4 is both photodissociated and chemically oxidized to H_2O in a complex chain of reactions, yielding an average of about 2 water molecules per methane molecule. Thus CH_4 oxidation accounts for roughly half of the H_2O content of the air reaching the mesosphere. CH_4 has been increasing at a rate of 1% per annum in the atmosphere since the industrial revolution [*Harriss*, 1989]. This increase is attributed to increases in biological sources in rice paddies, landfills, domesticated animals, etc. and in nonbiological sources from mining and industrial activities. Thus CH_4 has doubled since 1800, and most of this increase occurred since 1900 [*Khalil and Rasmussen*, 1987; *Pearman and Fraser*, 1988]. A CH_4 doubling corresponds to a 30% increase in upper level H_2O [*Ehhalt*, 1986; *Blake and Rowland*, 1988]. Actually, the H_2O has increased from 4.3 ppmv (the prevailing level in 1885) to the present level of 6 ppmv.

This 30% increase in H_2O has to be weighted against a tenfold increase in NLC brightness [*Jensen*, 1989]. An exponential dependence of the ice particle nucleation rate on the water saturation ratio is not sufficient to explain this discrepancy. We therefore would like to propose a different (additional) explanation, namely a simultaneous increase over the past century with H_2O of soot aerosols in the mesosphere which could serve as more efficient freezing nuclei than ablated meteoritic dust and/or ions, both of which are generally accepted to serve as nuclei for ice formation in the mesosphere [*Hunten et al.*, 1980]. Indeed, *Havnes et al.* [1996] report that large amounts of "dust" with average grain sizes of 0.1 μm or less at concentrations of several thousand particles per cubic centimeter were present during both PMSE and NLC conditions.

It is believed that PMCs consist of ice particles which start to nucleate around the mesopause, settle to lower altitudes while growing, become observable by lidar and/or by the naked eye, and finally evaporate once they approach the higher temperature near 82 km. Soot particles as freezing nuclei fit that pattern in that they get lofted gravito-photophoretically up to the mesopause, where nucleation of ice at the coldest temperature would destroy their lofting capability, whereupon they would settle to altitudes where temperatures are too high to sustain the existence of ice. After the evaporation of water, the soot nuclei would have regained their lofting capabilities to be raised back to the mesopause where the cycle would repeat itself.

Because the soot particles lofted by gravito-photophoresis are larger (0.7 μm < D_{ge} < 1.5 μm) than PMC particles (< 80 nm diameter), it is not immediately apparent that they contribute significantly to the PMC particle population, unless they acquire unipolar charges that break up the fractals into their individual spherules. In that case, each soot fractal, of dimension similar to that of PMC particles, could contribute up to thousands of individual spherules (see Tables 1 and 2).

Further and in addition to the NLC phenomenon, peculiar atmospheric radar echoes from the high-latitude summer mesosphere have spurred much research in recent years. They occur between June and August, most frequently at noon and near midnight at a height of 85±2 km [*Palmer et al.*, 1996]. Various fundamentally different theories have proliferated which all share the feature of these polar mesospheric summer echoes being dependent on the existence of electrically charged aerosols. *Cho and Röttger* [1997] critically examine both the data available and the theories proposed, with a special focus on the relationship between PMSEs and NLCs. Since the summer mesopause is characterized by very low temperatures, it has been proposed that PMSEs are related to the presence of subvisible ice particles. Concurrent measurements of radar reflectance and temperatures, however, have shown that generally the ambient temperatures in the summer mesosphere at midlatitudes are above that required to nucleate ice crystals on ions [*Chilson et al.*, 1997]. While gravity waves that can reduce temperatures to below the saturation temperature have been invoked as a possible explanation, we would like to repeat that ice formation could also have been facilitated by the presence of different nuclei, for example, soot particles, that are more effective in forming ice than are ions.

Finally, while PMCs should also occur in Antarctica, there is a large hemispheric difference in the occurrence of PMSEs, since they are almost absent (reported to be at least 30 db weaker during the first 2 years of observations) at 62°S over King George Island, Antarctica, than in the Arctic. This phenomenon has been explained by a warmer summer mesopause in the Southern Hemisphere [*Balsley et al.*, 1993; 1995]. *Olivero and Thomas* [1986] noticed that Southern Hemisphere PMCs were dimmer than Northern Hemisphere PMCs, which could be explained by either less water vapor or warmer temperatures in the south. If water vapor is assumed to be the same, then the southern summer mesopause must be 3-4 K warmer than the northern one [*Thomas*, 1996]. In this context we want to point out the strong hemispheric gradient in stratospheric soot particle concentration [*Pueschel et al.*, 1997] which, by the gravito-photophoretic transport mechanism treated above, should also be reflected in the mesosphere. This soot gradient could explain the observed PMSE gradient, if soot aerosol play a role in PMC formation.

In contrast to those conceivable indirect effects of anthropogenic soot in the mesosphere, direct effects are not soon to be expected from soot accumulation in the mesosphere. The 1990 fuel consumption by the world's commercial airline fleet amounted to 1.3×10^{11} kg [*Baughcum et al.*, 1993]. Applying a soot emission index E_{SOOT}=7.5×10^{-4} g kg^{-1} fuel burned [*Pueschel et al.*, 1998] to this consumption rate, the annual emission of soot into the atmosphere amounted to 9.8×10^{7} g. If 50% of this mass is distributed evenly into an atmospheric shell between 10 and 80 km altitude, equivalent to a volume of 3.6×10^{10} km^3, the average global concentration would amount to 1.4×10^{-3} g km^{-3}, or 1.4×10^{-12} g m^{-3}. With a specific absorption index of soot of 10 m^2 g^{-1}, the mid-visible light extinction is 1.4×10^{-11} m^{-1} which corresponds to an average optical depth between 10 km and 80 km of τ=9.7×10^{-7}. This is negligibly small compared to optical depths of 0.01 that are typical for the background atmosphere, and to optical depths of PMCs of $(1.2\text{-}1.8)\times10^{-3}$ [*Debrestian et al.*, 1997].

5. Summary

The observed existence of soot aerosol at 20 km altitude which arguably is generated by aircraft flying in corridors at 10-12 km requires a transport mechanism in a thermally stable stratosphere that is different from isentropic and/or dynamic mixing. Such a mechanism could be provided by gravito-photophoresis induced by the incidence of sunlight on strongly absorbing fractal soot particles. The particles' absorptivity, in conjunction with uneven surface coating with sulfuric acid, and their fractal nature make soot particles with maximum dimensions approaching one micrometer particularly conducive to gravito-photophoresis, because the requirement of a restoring torque that orients the particle with respect to gravity that this force requires is provided by the fractal characteristics of soot, and a body-fixed photophoretic force is given by asymmetric thermal accommotation coefficients across the particles' uneven surface.

During the SONEX field campaign in 1997, we sampled soot aerosol in commercial airline flight corridors and computed the gravitational and gravito-photophoretic forces acting on those soot particles. The result is that 16% by number, corresponding to

51% by mass, of a soot particle size distribution could be lofted against gravity by gravito-photophoresis. The calculated vertical velocities, exceeding settling velocities by up to a factor of 30, suggest that it takes approximately 30 and 20 years respectively to transport soot aerosol from 10 to 20 km and from 20 to 80 km. On the basis of stratospheric soot particle loading, the resulting soot mass flux at 20 km altitude in the northern hemisphere amounts to $5.\times10^{-18}$ g cm^{-2} s^{-1} which is within 1 order of magnitude to the influx of meteoritic dust into the mesosphere from outer space.

The effect of gravito-photophoresis is strongly altitude dependent. With increasing pressures near the Earth' surface, the lifting force falls off quickly. Above the mesopause, the lifting force becomes smaller because of a dominating energy loss by radiation rather than by molecular heat transfer. Thus gravito-photophoretic lifting forces are most effective within the altitude range 10 km $< Z <$ 85 km, making aircraft soot emitted in conventional flight corridors subject to lofting up to the mesopause.

The current mass loading of soot in the middle atmosphere is too small to cause a direct absorption effect. However, it is conceivable that soot in the mesosphere causes indirect effects by providing freezing nuclei for mesospheric ice particles. In addition, soot might affect the ionization of the mesosphere to contribute to polar mesospheric summer echoes.

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