3. AEROSOLS

What role do aerosols play in modulating earth's climate? Citing the IPCC, the EPA's Technical Support Document estimates the net effect of all aerosols is to produce a cooling effect, with a total direct radiative forcing of -0.5 Wm^{-2}, and an additional indirect cloud albedo forcing of -0.7 Wm^{-2}. However, there are numerous studies in the scientific literature that indicate these estimates are far too small. In fact, many indicate the radiative forcing of aerosols may be as large as, or larger than the radiative forcing due to atmospheric CO_2.

Consider, for example, the study of Vogelmann *et al*. (2003), who report that "mineral aerosols have complex, highly varied optical properties that, for equal loadings, can cause differences in the surface IR flux between 7 and 25 Wm^{-2} (Sokolik *et al*., 1998)," and that "only a few large-scale climate models currently consider aerosol IR effects (e.g., Tegen *et al*., 1996; Jacobson, 2001) despite their potentially large forcing." In an attempt to persuade climate modelers to rectify this situation, they used high-resolution spectra to obtain the IR radiative forcing at the earth's surface for aerosols encountered in the outflow from northeastern Asia, based on measurements made by the Marine-Atmospheric Emitted Radiance Interferometer from the NOAA Ship *Ronald H. Brown* during the Aerosol Characterization Experiment-Asia. As a result of this work, the five scientists determined that "daytime surface IR forcings are often a few Wm^{-2} and can reach almost 10 Wm^{-2} for large aerosol loadings." These values, in their words, "are comparable to or larger than the 1 to 2 Wm^{-2} change in the globally averaged surface IR forcing caused by greenhouse gas increases since pre-industrial times" and "highlight the importance of aerosol IR forcing which should be included in climate model simulations."

In another study that addressed the magnitude of aerosol radiative effects, Chou *et al*. (2002) analyzed aerosol optical properties retrieved from the satellite-mounted Sea-viewing Wide Field-of-view Sensor (SeaWiFS) and used them in conjunction with a radiative transfer model of the planet's atmosphere to calculate the climatic effects of aerosols over earth's major oceans. In general, this effort revealed that "aerosols reduce the annual-mean net downward solar flux by 5.4 Wm^{-2} at the top of the atmosphere, and by 5.9 Wm^{-2} at the surface." During the large Indonesian fires of September-December 1997, however, the radiative impetus for cooling at the top of the atmosphere was more than 10 Wm^{-2}, while it was more than 25 Wm^{-2} at the surface of the sea in the vicinity of Indonesia.

These latter results are similar to those obtained earlier by Wild (1999), who used a comprehensive set of collocated surface and satellite observations to calculate the amount of solar radiation absorbed in the atmosphere over equatorial Africa and compared the results with the predictions of three general circulation models of the atmosphere. This work revealed that the climate models did not properly account for spatial and temporal variations in atmospheric aerosol concentrations, leading them to predict regional and seasonal values of solar radiation absorption in the atmosphere with underestimation biases of up to 30 Wm^{-2}.

By way of comparison, as noted in the study of Vogelmann *et al*., the globally averaged surface IR forcing caused by greenhouse gas increases since pre-industrial times is only 1 to 2 Wm^{-2}. Hence, it can be appreciated that over much of the planet's surface, the radiative cooling influence of atmospheric aerosols (many of which are produced by anthropogenic activities) must prevail, suggesting a probable net anthropogenic-induced climatic signal that must be very close to zero and nowhere near capable of producing what climate alarmists refer to as the *unprecedented* warming of the 20th century. Thus, one might conclude that the air temperature record on which they rely is either grossly in error or that the warming, if real, is due to something other than anthropogenic CO_2 emissions.

Aerosol uncertainties and the problems they generate also figure prominently in the study of Anderson *et al*. (2003), who note there are two different ways by which the aerosol forcing of climate may be computed. The first of these approaches is that of *forward* calculation, which is based, in their words, on "knowledge of the pertinent aerosol physics and chemistry." The second approach is *inverse* calculation, based on "the total forcing required to match climate model simulations with observed temperature changes."

The first approach, which relies heavily on *first principles*, utilizes known physical and chemical laws and assumes nothing about the outcome of the calculation. The second approach, in considerable contrast, is based on matching *residuals*, where the aerosol forcing is computed from what is required to match the calculated change in temperature with the observed change over some period of time. Consequently, in the words of Anderson *et al*., "to the extent that climate models rely on the results of inverse calculations, the possibility of circular reasoning arises."

So which approach do climate models typically employ? "Unfortunately," according to Anderson *et al*., "virtually all climate model studies that have included anthropogenic aerosol forcing as a driver of climate change have used only aerosol forcing values that are consistent with the inverse approach."

How significant is this choice? Anderson *et al*. report that the negative forcing of anthropogenic aerosols derived by forward calculation is "considerably greater" than that derived by inverse calculation, so much so, in fact, that if forward calculation is employed, the results "differ greatly" and "even the sign of the total forcing is in question," which implies that "natural variability (that is, variability not forced by anthropogenic emissions) is much larger than climate models currently indicate." The bottom line, in the words of Anderson *et al*., is that "inferences about the causes of surface warming over the industrial period and about climate sensitivity may therefore be in error."

Schwartz (2004) also addressed the subject of uncertainty as it applies to the role of aerosols in climate models. Noting that the National Research Council (1979) concluded that "climate sensitivity [to CO_2 doubling] is likely to be in the range 1.5-4.5°C" and that "remarkably, despite some two decades of intervening work, neither the central value nor the uncertainty range has changed," he opined that this continuing uncertainty "precludes meaningful model evaluation

by comparison with observed global temperature change or empirical determination of climate sensitivity," and that it "raises questions regarding claims of having reproduced observed large-scale changes in surface temperature over the 20th century."

Schwartz thus contends that climate model predictions of CO_2-induced global warming "are limited at present by uncertainty in radiative forcing of climate change over the industrial period, which is dominated by uncertainty in forcing by aerosols," and that if this situation is not improved, "it is likely that in another 20 years it will still not be possible to specify the climate sensitivity with [an] uncertainty range appreciably narrower than it is at present." Indeed, he says "the need for reducing the uncertainty from its present estimated value by *at least a factor of 3* and perhaps *a factor of 10 or more* seems *inescapable* [all our italics] if the uncertainty in climate sensitivity is to be reduced to an extent where it becomes useful for formulating policy to deal with global change," which surely suggests that even the best climate models of the day are wholly inadequate for this purpose.

Coming to much the same conclusion was the study of Jaenicke *et al*. (2007), who reviewed the status of research being conducted on biological materials in the atmosphere, which they denominate *primary biological atmospheric particles* or PBAPs. Originally, these particles were restricted to *culture forming units*, including pollen, bacteria, mold and viruses, but they also include fragments of living and dead organisms and plant debris, human and animal epithelial cells, broken hair filaments, parts of insects, shed feather fractions, etc., which they lump together under the category of *dead biological matter*.

With respect to the meteorological and climatic relevance of these particles, they note that many PBAPs, including "decaying vegetation, marine plankton and bacteria are excellent ice nuclei," and they say that "one can easily imagine the[ir] influence on cloud cover, climate forcing and feedback and global precipitation distribution."

In describing their own measurements and those of others, which they say "have now been carried out at several geographical locations covering all seasons of the year and many characteristic environments," Jaenicke *et al*. report that "by number and volume, the PBAP fraction is ~20% of the total aerosol, and appears rather constant during the year." In addition, they write that "the impression prevails that the biological material, whether produced directly or shed during the seasons, sits on surfaces, ready to be lifted again in resuspension."

In a brief summation of their findings, the German researchers say "the overall conclusion can only be that PBAPs are a major fraction of atmospheric aerosols, and are comparable to sea salt over the oceans and mineral particles over the continents," and, consequently, that "the biosphere must be a major source for directly injected biological particles, and those particles should be taken into account in understanding and modeling atmospheric processes." *However*, they note that "the IPCC-Report of 2007 *does not even* ***mention*** *these particles* [our italics and boldface]," and that "this disregard of the biological particles requires a new attitude."

We agree. And we hope that that attitude includes a willingness to acknowledge that, as comprehensive as current state-of-the-art climate models might be, there is much of significance that is not included in them, and that when that which is missing is finally factored in and properly modeled, it could well result in radically different conclusions being reached from those that are currently held by the IPCC.

Our review of important aerosol studies continues below with a separate discussion of four important aerosol categories: (1) Biological (Aquatic), (2) Biological (Terrestrial), (3) Non-Biological (Anthropogenic) and (4) Non-Biological (Natural). Additional information on this topic, including reviews of aerosols not discussed here, can be found at http://www.co2science.org/subject/a/subject_a.php under the heading Aerosols.

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3.1. Biological (Aquatic)

Charlson *et al*. (1987) described a multi-stage negative feedback phenomenon - several components of which have been verified by subsequent scientific studies - that links biology with climate change. The process begins with an initial impetus for warming that stimulates primary production in marine phytoplankton. This enhanced process leads to the production of more copious quantities of dimethylsulphoniopropionate, which leads in turn to the evolution of greater amounts of dimethyl sulphide, or DMS, in the surface waters of the world's oceans. Larger quantities of DMS thus diffuse into the atmosphere, where the gas is oxidized, leading to the creation of greater amounts of acidic aerosols that function as cloud condensation nuclei. This phenomenon then leads to the creation of more and brighter clouds that reflect more incoming solar radiation back to space, thereby providing a cooling influence that counters the initial impetus for warming.

Several recent studies have shed additional light on this complex hypothesis. Simo and Pedros-Alio (1999), for example, used satellite imagery and *in situ* experiments to study the production of DMS by enzymatic cleavage of dimethylsulphoniopropionate in the North Atlantic Ocean about 400 km south of Iceland, finding that the depth of the surface mixing-layer has a substantial influence on DMS yield in the short term, as do seasonal variations in vertical mixing in the longer term, which observations led them to conclude that "climate-controlled mixing controls DMS production over vast regions of the ocean."

Amplifying the significance of this finding, Hopke *et al*. (1999) analyzed weekly concentrations of 24 different airborne particulates measured at the northernmost manned site in the world - Alert, Northwest Territories, Canada - from 1980 to 1991. One of their more interesting discoveries was the finding that concentrations of biogenic sulfur, including sulfate and methane sulfonate, were low in winter but high in summer, and that the year-to-year variability in the strength of the biogenic sulfur signal was strongly correlated with the mean temperature of the Northern Hemisphere. "This result," as the authors say, "suggests that as the temperature rises, there is increased biogenic production of the reduced sulfur precursor compounds that are oxidized in the atmosphere to sulfate and methane sulfonate and could be evidence of a negative feedback mechanism in the global climate system."

Further support for this aquatic life-climate linkage was provided by Ayers and Gillett (2000), who summarized relevant empirical evidence collected at Cape Grim, Tasmania, along with pertinent evidence reported in many peer-reviewed scientific papers on the subject. This exercise led them to conclude that "major links in the feedback chain proposed by Charlson *et al*. (1987) have a sound physical basis." More specifically, they noted there is "compelling observational evidence to suggest that DMS and its atmospheric products participate significantly in processes of climate regulation and reactive atmospheric chemistry in the remote marine boundary layer of the Southern Hemisphere."

In another test of the Charlson *et al*. hypothesis, Sciare *et al*. (2000) made continuous measurements of atmospheric DMS concentration over the 10-year period 1990-1999 at Amsterdam Island in the southern Indian Ocean. Their study revealed "a clear seasonal

variation with a factor of 20 in amplitude between its maximum in January (austral summer) and minimum in July-August (austral winter)." In addition, they found DMS anomalies to be "closely related to sea surface temperature anomalies, clearly indicating a link between DMS and climate changes." In fact, they found that a temperature increase of only 1°C was sufficient to increase the atmospheric DMS concentration by as much as 50% on a monthly basis, noting that "this is the first time that a direct link between SSTs [sea surface temperatures] and atmospheric DMS is established for a large oceanic area."

In a related study, Baboukas *et al*. (2002) report the results of nine years of measurements of methanesulfonate (MS^-), an exclusive oxidation product of DMS, in *rainwater* at Amsterdam Island. Their data, too, reveal "a well distinguished seasonal variation with higher values in summer, in line with the seasonal variation of its gaseous precursor (DMS)," which, as they say, "further confirms the findings of Sciare *et al*. (2000)." In addition, the MS^- anomalies in the rainwater were found to be closely related to sea surface temperature anomalies; and the authors say this observation provides even more support for "the existence of a positive ocean-atmosphere feedback on the biogenic sulfur cycle above the Austral Ocean, one of the most important DMS sources of the world."

Another pertinent study was conducted by Kouvarakis and Mihalopoulos (2002), who investigated the seasonal variations of gaseous dimethylsulfide (DMS) and its oxidation products - non-sea-salt sulfate (nss-SO_4^{2-}) and methanesulfonic acid (MSA) - at a remote coastal location in the Eastern Mediterranean Sea from May 1997 through October 1999, as well as the diurnal variation of DMS during two intensive measurement campaigns in September 1997. In the seasonal investigation, DMS concentrations tracked sea surface temperature (SST) almost perfectly, going from a low of 0.87 nmol m^{-3} in the winter to a high of 3.74 nmol m^{-3} in the summer. Such was also the case in the diurnal study: DMS concentrations were lowest just before sunrise, rose rapidly thereafter to about 1100, were followed by a little dip and then a further rise to 2000, whereupon a decline set in that continued until just before sunrise. MSA concentrations exhibited a similar seasonal variation to that displayed by DMS, ranging from a wintertime low of 0.04 nmol m^{-3} to a summertime high of 0.99 nmol m^{-3}. The same was also true of aerosol nss-SO_4^{2-}, which varied from 0.6 to 123.9 nmol m^{-3} in going from winter to summer.

As time marches on, therefore, and as ever more studies of the Charlson *et al*. hypothesis are conducted (O'Dowd *et al*., 2004; Toole and Siegel, 2004), it is becoming ever more clear that the normal hour-to hour, day-to-day and season-to-season behaviors of the phytoplanktonic inhabitants of earth's marine ecosystems effectively combat extreme environmental temperature changes; and in view of their efficacy, it would seem we have little to worry about in the way of catastrophic CO_2-induced global warming.

Additional information on this topic, including reviews of newer publications as they become available, can be found at http://www.co2science.org/subject/a/aerosolsbioaqua.php.

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3.2. Biological (Terrestrial)

Just as marine phytoplankton that are exposed to rising temperatures give off greater quantities of gases that lead to the production of greater quantities of cloud condensation nuclei, which create more and brighter clouds that reflect more incoming solar radiation back to space and thereby either reverse, stop or slow the warming that initiated this negative feedback phenomenon, so too do terrestrial plants respond in like manner, thereby enhancing their ability to survive in a changing global environment. What is more, earth's terrestrial plants have a tendency to operate in this manner ever more effectively as the air's CO_2 content rises ever higher.

A good introduction to this subject is provided by the review paper of Peñuelas and Llusia (2003), who say that *biogenic volatile organic compounds* -- or BVOCs -- constitute "one of nature's biodiversity treasures." Comprised of isoprene, terpenes, alkanes, alkenes, alcohols, esters, carbonyls and acids, this diverse group of substances is produced by a variety of processes occurring in many plant tissues. Some of the functions of these substances, according to the two scientists, include acting as "deterrents against pathogens and herbivores, or to aid wound sealing after damage (Pichersky and Gershenzon, 2002)." They also say that BVOCs provide a means "to attract pollinators and herbivore predators, and to communicate with other plants and organisms (Peñuelas *et al*., 1995; Shulaev *et al*., 1997)."

Of particular importance within the context of global climate change, in the opinion of Peñuelas and Llusia, is the growing realization that "isoprene and monoterpenes, which constitute a major fraction of BVOCs, might confer protection against high temperatures" by acting "as scavengers of reactive oxygen species produced [within plants] under high temperatures." If this is indeed the case, it can be appreciated that with respect to the claimed ill effects of CO_2-induced global warming on earth's vegetation, there are likely to be two strong ameliorative phenomena that act to protect the planet's plants: (1) the aerial fertilization effect of atmospheric CO_2 enrichment, which is typically more strongly expressed at higher temperatures, and (2) the tendency for rising air temperatures and CO_2 concentrations to spur the production of higher concentrations of heat-stress-reducing BVOCs. With respect to temperature, for example, Peñuelas and Llusia calculate that "global warming over the past 30 years could have increased the BVOC global emissions by approximately 10%, and a further 2-3°C rise in the mean global temperature ... could increase BVOC global emissions by an additional 30-45%."

There may also be a couple of other phenomena that favor earth's plants within this context. Peñuelas and Llusia note, for example, that "the increased release of nitrogen into the biosphere by man probably also enhances BVOC emissions by increasing the level of carbon fixation and the activity of the responsible enzymes (Litvak *et al*., 1996)." In addition, they indicate that the conversion of abandoned agricultural lands to forests and the implementation of planned reforestation projects should help the rest of the biosphere too, reporting that additional numbers of "*Populus*, *Eucalyptus* or *Pinus*, which are major emitters, might greatly increase BVOC emissions."

Most intriguing of all, perhaps, is how increased BVOC emissions might impact climate change. Peñuelas and Llusia say that "BVOCs generate large quantities of organic aerosols that could affect climate significantly by forming cloud condensation nuclei." As a result, they say "there should be a net cooling of the Earth's surface during the day because of radiation interception," noting that Shallcross and Monks (2000) "have suggested that one of the reasons plants emit the aerosol isoprene might be to cool the surroundings in addition to any physiological or evaporative effects that might cool the plant directly."

However, not all experiments have reported increases in plant BVOC emissions with increasing atmospheric CO_2 concentrations, one example being that of Constable *et al*. (1999), who found

no effect of elevated CO_2 on monoterpene emissions from Ponderosa pine and Douglas fir trees. Some studies, in fact, have even reported *decreases* in BVOC emissions, such as those of Vuorinen *et al*. (2004), who worked with cabbage plants, and Loreto *et al*. (2001), who studied monoterpene emissions from oak seedlings. On the other hand, Staudt *et al*. (2001) observed CO_2-induced *increases* in BVOC emissions in *the identical species of oak*. An explanation for this wide range of results comes from Baraldi *et al*. (2004), who -- after exposing sections of a southern California chaparral ecosystem to atmospheric CO_2 concentrations ranging from 250 to 750 ppm in 100-ppm increments for a period of four years -- concluded that "BVOC emission can remain nearly constant as rising CO_2 reduces emission per unit leaf area while stimulating biomass growth and leaf area per unit ground area." In most of the cases investigated, however, BVOC emissions tend to increase with atmospheric CO_2 enrichment; and the increases are often huge.

A case in point is provided by the study of Jasoni *et al*. (2003), who grew onions from seed for 30 days in individual cylindrical flow-through growth chambers under controlled environmental conditions at atmospheric CO_2 concentrations of either 400 or 1000 ppm. At the end of the study, the plants in the CO_2-enriched chambers had 40% more biomass than the plants grown in ambient air, and their photosynthetic rates were 22% greater. In addition, the CO_2-enriched plants exhibited *17-fold* and *38-fold* increases in emissions of the BVOC hydrocarbons 2-undecanone and 2-tridecanone, respectively, which Jasoni *et al*. make a point of noting "confer insect resistance against a major agricultural pest, spider mites." More generally, they conclude that "plants grown under elevated CO_2 will accumulate excess carbon and that at least a portion of this excess carbon is funneled into an increased production of BVOCs," which have many positive implications in the realms of both biology and climate, as noted above.

Another example is seen in the study of Raisanen *et al*. (2008), who developed an experiment designed to see to what extent a doubling of the air's CO_2 content and a 2-6°C increase in air temperature might impact the emission of monoterpenes from 20-year-old Scots pine (*Pinus sylvestris* L.) seedlings, Raisanen *et al*. (2008) studied the two phenomena (and their interaction) within closed-top chambers built on a naturally-seeded stand of the trees in eastern Finland that had been exposed to the four treatments -- ambient CO_2 and ambient temperature, ambient temperature and elevated CO_2, ambient CO_2 and elevated temperature, elevated temperature and elevated CO_2 -- for the prior five years.

Over the five-month growing season of May-September, the three Finnish researchers found that total monoterpene emissions in the elevated-CO_2-only treatment were 5% greater than those in the ambient CO_2, ambient temperature treatment, and that emissions in the elevated-temperature-only treatment were *9% less* than those in ambient air. In the presence of both elevated CO_2 *and* elevated temperature, however, there was an increase of fully 126% in the total amount of monoterpenes emitted over the growing season, which led the authors to conclude that "the amount of monoterpenes released by Scots pines into the atmosphere during a growing season will increase substantially in the predicted future climate."

A number of studies suggest that the phenomena discussed in the preceding paragraphs do indeed operate in the real world. Kavouras *et al*. (1998), for example, measured a number of atmospheric gases and particles in a eucalyptus forest in Portugal and analyzed their observations to see if there was any evidence of biologically-produced gases being converted to particles that could function as cloud condensation nuclei. Their work demonstrated that certain hydrocarbons emitted by vegetation (isoprene and terpenes, in particular) do indeed experience gas-to-particle transformations. In fact, aerosols (or *bio*sols) produced from two of these organic acids (*cis*- and *trans*-pinonic acid) comprised as much as 40% of the fine particle atmospheric mass during daytime hours.

These findings clearly demonstrate that the *biology* of the earth can indeed influence the *climate* of the earth. Specifically, they reveal a direct connection between the metabolic activity of trees and the propensity for the atmosphere to produce clouds. What is more, the relationship is one that is self-protecting of the biosphere: as the air's CO_2 content rises, plant productivity rises, which leads to an enhanced evolution of biogenic gases, which leads to the production of more cloud condensation nuclei, which leads to the creation of more clouds that reflect more solar radiation back to space, which tends to counter any increase in the strength of the atmosphere's greenhouse effect that may have been produced by the initial rise in the air's CO_2 content.

A similar study was conducted by O'Dowd *et al*. (2002), who measured aerosol electrical-mobility size-distributions before and during the initial stage of an atmospheric nucleation event over a boreal forest in Finland. Simultaneously, organic vapor growth rate measurements were made of particles that nucleated into organic cloud-droplets in the flow-tube cloud chamber of a modified condensation-particle counter. This work demonstrated, in their words, that newly-formed aerosol particles over forested areas "are composed primarily of organic species, such as cis-pinonic acid and pinonic acid, produced by oxidation of terpenes in organic vapours released from the canopy."

Commenting on this finding, O'Dowd *et al*. note that "aerosol particles produced over forested areas may affect climate by acting as nuclei for cloud condensation," but they say there remain numerous uncertainties involving complex feedback processes "that must be determined if we are to predict future changes in global climate." This being the case, we wonder how anyone can presume to decide what should or should not be done about anthropogenic CO_2 emissions; for if we can't predict future changes in global climate without the knowledge just specified, how do we know if we even need to be worried about the matter?

Shifting from trees to a much tinier plant, Kuhn and Kesselmeier (2000) collected lichens from an open oak woodland in central California, USA, and studied their uptake of carbonyl sulfide or COS in a dynamic cuvette system under controlled conditions in the laboratory. When optimally hydrated, COS was absorbed from the atmosphere by the lichens at a rate that gradually doubled as air temperature rose from approximately 3 to 25°C, whereupon the rate of COS absorption dropped precipitously, reaching a value of zero at 35°C. Why is this significant?

COS is the most stable and abundant reduced sulfur gas in the atmosphere and is thus a major player in determining earth's radiation budget. After making its way into the stratosphere, it can be photo-dissociated, as well as oxidized, to form SO_2, which is typically converted to sulfate aerosol particles that are highly reflective of incoming solar radiation and, therefore, have the capacity to significantly cool the earth as more and more of them collect above the tropopause. This being the case, it is only natural to suspect that biologically-modulated COS concentrations may play a role in keeping earth's surface air temperature within bounds conducive to the continued existence of life; and that is exactly what is implied by the observations of Kuhn and Kesselmeier. Once air temperature rises above 25°C, the rate of removal of COS from the air by this particular species of lichen declines dramatically; and when this happens, more COS remains in the air, which increases the potential for more COS to make its way into the stratosphere, where it can be converted into sulfate aerosol particles that can reflect more incoming solar radiation back to space and thereby cool the earth. And since the consumption of COS by lichens is under the physiological control of carbonic anhydrase -- which is the key enzyme for COS uptake in all higher plants, algae and soil organisms -- we could expect this phenomenon to be generally operative throughout much of the plant kingdom. Hence, this biological "thermostat" may well be powerful enough to define an upper limit above which the surface air temperature of the planet may be restricted from rising, even when changes in other forcing factors, such as greenhouse gases, produce an impetus for it to do so.

Although BVOCs emitted from terrestrial plants both small and large are thus important to earth's climate, trees tend to dominate in this regard; and recent research suggests yet another way in which their response to atmospheric CO_2 enrichment may provide an effective counterbalance to the greenhouse properties of CO_2. The phenomenon begins with the propensity for CO_2-induced increases in BVOCs, together with the cloud particles they spawn, to enhance the amount of diffuse solar radiation reaching the earth's surface (Suraqui *et al*., 1974; Abakumova *et al*., 1996), which is followed by the ability of enhanced diffuse lighting to reduce the volume of shade within vegetative canopies (Roderick *et al*., 2001), which is followed by the tendency for less internal canopy shading to enhance whole-canopy photosynthesis (Healey *et al*., 1998), which finally produces the end result: a greater photosynthetic extraction of CO_2 from the air and the subsequent reduction of the strength of the atmosphere's greenhouse effect.

How significant is this process? Roderick *et al*. provide a good estimate based on the utilization of a unique "natural experiment," a technique that has been used extensively by Idso (1998) to evaluate the overall climatic sensitivity of the planet. Specifically, Roderick and his colleagues consider the volcanic eruption of Mt. Pinatubo in June of 1991. This event ejected enough gases and fine materials into the atmosphere that it produced sufficient aerosol particles to greatly increase the diffuse component of the solar radiation reaching the surface of the earth from that point in time through much of 1993, while only slightly reducing the receipt of total solar radiation.

Based on a set of lengthy calculations, Roderick *et al*. concluded that the Mt. Pinatubo eruption may well have resulted in the removal of an extra 2.5 Gt of carbon from the atmosphere due to its diffuse-light-enhancing stimulation of terrestrial photosynthesis in the year following the eruption, which would have reduced the ongoing rise in the air's CO_2 concentration that year by about 1.2 ppm. Interestingly, this reduction is about the size of the real-world reduction that was measured that year (Sarmiento, 1993). What makes this observation even more impressive is the fact that the CO_2 reduction was coincident with an El Niño event; because, in the words of Roderick *et al*., "previous and subsequent such events have been associated with *increases* in atmospheric CO_2." In addition, the observed reduction in total solar radiation received at the earth's surface during this period would have had a tendency to reduce the amount of photosynthetically active radiation incident upon earth's plants, which would also have had a tendency to cause the air's CO_2 content to rise, as it would tend to lessen global photosynthetic activity.

Additional real-world evidence for the existence of this phenomenon was provided by Gu *et al*. (2003), who reported using "two independent and direct methods to examine the photosynthetic response of a northern hardwood forest (Harvard Forest, 42.5°N, 72.2°W) to changes in diffuse radiation caused by Mount Pinatubo's volcanic aerosols." They found that "around noontime in the midgrowing season, the gross photosynthetic rate under the perturbed cloudless solar radiation regime was 23, 8, and 4% higher than that under the normal cloudless solar radiation regime in 1992, 1993, and 1994, respectively," and that "integrated over a day, the enhancement for canopy gross photosynthesis by the volcanic aerosols was 21% in 1992, 6% in 1993 and 3% in 1994." In reflecting on the significance of these observations, Gu *et al*. stated that "because of substantial increases in diffuse radiation world-wide after the eruption and strong positive effects of diffuse radiation for a variety of vegetation types, it is likely that our findings at Harvard Forest represent a global phenomenon."

As impressive as these findings are, Gu *et al*. did not stop there. They went on to document a powerful propensity for the extra diffuse light created by increased *cloud cover* to *further* enhance forest photosynthesis, and to do so even though the total flux of solar radiation received at the earth's surface is typically significantly *reduced* under such conditions (Stanhill and Cohen, 2001). Based on still more real-world data, for example, they reported finding that "Harvard Forest photosynthesis also increases with cloud cover, with a peak at about 50% cloud cover."

In all of the original investigations of this phenomenon, which also include the studies of Law *et al*. (2002), Farquhar and Roderick (2003) and Reichenau and Esser (2003), the source of the enhanced aerosol concentration, i.e., the key *natural experiment*, was a massive volcanic eruption. So what happens under more normal conditions? This is the question that was asked by Niyogi *et al*. (2004): "can we detect the effect of relatively routine aerosol variability on field measurements of CO_2 fluxes, and if so, how does the variability in aerosol loading affect CO_2 fluxes over different landscapes?"

To answer this question, the group of sixteen researchers used CO_2 flux data from the AmeriFlux network (Baldocchi *et al*., 2001) together with cloud-free aerosol optical depth data from the NASA Robotic Network (AERONET; Holben *et al*., 2001) to assess the effect of aerosol loading on the net assimilation of CO_2 by three types of vegetation: trees (broadleaf deciduous forest and mixed forest), crops (winter wheat, soybeans and corn) and grasslands. Their work revealed that an aerosol-induced increase in *diffuse radiative-flux fraction* [DRF = ratio of diffuse (Rd) to total or global (Rg) solar irradiance] increased the net CO_2 assimilation of trees and crops, making them larger carbon sinks, but that it decreased the net CO_2 assimilation of grasslands, making them smaller carbon sinks.

How significant were the effects observed by Niyogi *et al*.? For a summer mid-range Rg flux of 500 W m^{-2}, going from the set of all DRF values between 0.0 and 0.4 to the set of all DRF values between 0.6 and 1.0 resulted in an approximate 50% increase in net CO_2 assimilation by a broadleaf deciduous forest located in Tennessee, USA. Averaged over the entire daylight period, they further determined that the shift from the lower to the higher set of DRF values "enhances photosynthetic fluxes by about 30% at this study site." Similar results were obtained for the mixed forest and the conglomerate of crops studied. Hence, they concluded that natural variability among commonly-present aerosols can "routinely influence surface irradiance and hence the terrestrial CO_2 flux and regional carbon cycle." For these types of land-cover (forests and agricultural crops), that influence is to significantly *increase* the assimilation of CO_2 from the atmosphere; and this effect greatly overpowers the opposite effect that occurs over grasslands, primarily because earth's trees and shrubs are responsible for fully two thirds of the planet's net primary production.

What is especially exciting about these real-world observations is that much of the commonly-present aerosol burden of the atmosphere is plant-derived. Hence, it can be appreciated that earth's woody plants are themselves responsible for emitting to the air that which ultimately enhances their photosynthetic prowess. In other words, earth's trees significantly control their own destiny, i.e., they alter the atmospheric environment in a way that directly enhances their opportunities for greater growth.

Humanity also helps in this regard; for as we pump ever more CO_2 into the atmosphere, the globe's woody plants quickly respond to its *aerial fertilization effect*, becoming ever more productive, which leads to even more plant-derived aerosols being released to the atmosphere, which stimulates this positive feedback cycle to a still greater degree. Stated another way, earth's trees use some of the CO_2 emitted to the atmosphere by man to alter the aerial environment so as to enable them to remove even more CO_2 from the air. The end result is that earth's trees and humanity are working *hand-in-hand* to significantly increase the productivity of the biosphere; and it is happening in spite of all of the *true* anthropogenic insults to the environment that work in opposition to enhanced biological activity.

In light of these several observations, it should be painfully obvious that the historical and still-ongoing CO_2-induced increase in atmospheric BVOCs should have had, and should be continuing to have, a significant cooling effect on the planet that exerts itself by both slowing

the rate of rise of the air's CO_2 content and reducing the receipt of solar radiation at the earth's surface, neither of which effects is included in any general circulation model of the atmosphere of which we are aware.

One final beneficial effect of CO_2-induced increases in BVOC emissions is described by Goldstein *et al*. (2004), and that is the propensity of BVOCs to destroy tropospheric ozone.

As a bit of a background, earth's vegetation is responsible for the production of vast amounts of ozone (O_3; Chameides *et al*., 1988; Harley *et al*., 1999), but it is also responsible for *destroying* a lot of O_3. With respect to the latter phenomenon, Goldstein *et al*. mention three major routes by which O_3 exits the air near the earth's surface: leaf stomatal uptake, surface deposition, and within-canopy gas-phase chemical reactions with BVOCs. The first of these exit routes, according to them, accounts for 30-90% of total ecosystem O_3 uptake from the atmosphere (= O_3 destruction), while the remainder has typically been attributed to deposition on non-stomatal surfaces. However, they note that "Kurpius and Goldstein (2003) recently showed that the non-stomatal flux [from the atmosphere to oblivion] increased exponentially as a function of temperature at a coniferous forest site," and that "the exponential increase with temperature was consistent with the temperature dependence of monoterpene emissions from the same ecosystem, suggesting O_3 was lost via gas phase reactions with biogenically emitted terpenes before they could escape the forest canopy."

In a study designed to take the next step towards turning the implication of this observation into something stronger than a mere suggestion, Schade and Goldstein (2003) demonstrated that forest thinning dramatically enhances monoterpene emissions. In the current study, Goldstein *et al*. take another important step towards clarifying the issue by measuring the effect of forest thinning on O_3 destruction in an attempt to see if it is enhanced in parallel fashion to the thinning-induced increase in monoterpene emissions.

In a ponderosa pine plantation in the Sierra Nevada Mountains of California, USA, a management procedure to improve forest health and optimize tree growth was initiated on 11 May and continued through 15 June 2000. This procedure involved the use of a *masticator* to mechanically "chew up" smaller unwanted trees and leave their debris on site, which operation reduced plantation green leaf biomass by just over half. Simultaneously, monoterpene mixing ratios and fluxes were measured hourly within the plantation canopy, while total ecosystem O_3 destruction was "partitioned to differentiate loss due to gas-phase chemistry from stomatal uptake and deposition."

Goldstein *et al*. report that both the destruction of ozone due to gas-phase chemistry and emissions of monoterpenes increased dramatically with the onset of thinning, and that these phenomena continued in phase with each other thereafter. Hence, they "infer that the massive increase of O_3 flux [from the atmosphere to oblivion] during and following mastication is driven by loss of O_3 through chemical reactions with unmeasured terpenes or closely related BVOCs whose emissions were enhanced due to wounding [by the masticator]." Indeed, they say that "considered together, these observations provide a conclusive picture that the chemical loss of

O_3 is due to reactions with BVOCs emitted in a similar manner as terpenes," and that "we can conceive no other possible explanation for this behavior other than chemical O_3 destruction in and above the forest canopy by reactions with BVOCs."

Goldstein *et al*. say their results "suggest that total reactive terpene emissions might be roughly a factor of 10 higher than the typically measured and modeled monoterpene emissions, making them larger than isoprene emissions on a global scale." If this proves to be the case, it will be a most important finding, for it would mean that vegetative emissions of terpenes, which lead to the *destruction* of ozone, are significantly greater than vegetative emissions of isoprene, which lead to the *creation* of ozone (Poisson *et al*., 2000). In addition, there is substantial evidence to suggest that the ongoing rise in the air's CO_2 content may well lead to an overall *reduction* in vegetative *isoprene* emissions, while at the same time enhancing vegetative productivity, which may well lead to an overall *increase* in vegetative *terpene* emissions. As a result, there is reason to believe that the ongoing rise in the air's CO_2 content will help to reduce the ongoing rise in the air's O_3 concentration, which should be a boon to the entire biosphere.

In conclusion, a wealth of real-world evidence is beginning to suggest that both rising air temperatures and CO_2 concentrations significantly increase desirable vegetative BVOC emissions, particularly from trees, which constitute the most prominent photosynthetic force on the planet, and that this phenomenon has a large number of extremely important and highly beneficial biospheric consequences.

Additional information on this topic, including reviews of newer publications as they become available, can be found at http://www.co2science.org/subject/a/aerosolsterr.php.

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3.3. Non-Biological (Anthropogenic)

There are also numerous ways in which the activities of humanity lead to the creation of aerosols that have the potential to alter earth's radiation balance and affect its climate. Contrails created in the wake of emissions from jet aircraft are one example, where Minnis *et al*. (2004) have calculated that nearly all of the surface warming observed over the United States between 1975 and 1994 (0.54°C) may well be explained by aircraft-induced increases in cirrus cloud coverage over that period. If true, this result would imply that *little to none* of the observed U.S. warming over that period could be attributed to the concomitant increase in the air's CO_2 content. *Ship tracks*, or bright streaks that form in layers of marine stratus clouds, are another example. They are created by emissions from ocean-going vessels; and these persistent and highly-reflective linear patches of low-level clouds generally always tend to cool the planet (Ferek *et al*., 1998; Schreier *et al*., 2006). Averaged over the surface of the earth both day and night and over the year, for example, Capaldo *et al*. (1999) have calculated that this phenomenon creates a mean *negative* radiative forcing of -0.16 Wm^{-2} in the Northern Hemisphere and -0.06 Wm^{-2} in the Southern Hemisphere, which values are to be compared to the much larger *positive* radiative forcing of approximately 4 Wm^{-2} due to a 300 ppm increase in the atmosphere's CO_2 concentration.

In some cases, the atmosphere over the sea also carries a considerable burden of anthropogenically-produced aerosols that come from terrestrial sites. In recent years, attention to this topic has centered on highly-polluted air from south and southeast Asia that makes its way over the northern Indian Ocean during the dry monsoon season. There has been much discussion about the impact of this phenomenon on regional climate; and within this context, Norris (2001) has looked at cloud cover as the ultimate arbiter of the various competing hypotheses, finding that daytime low-level oceanic cloud cover increased substantially over the last half of the past century in both the Northern and Southern Hemispheres at essentially all hours of the day. This finding is indicative of a pervasive net cooling effect.

Over land, aerosol-generating human activities also have a significant impact on local, as well as more wide-ranging, climatic phenomena. Most interesting in the local context is the study of Sahai (1998), who found that although suburban areas of Nagpur, India had warmed over recent decades, the central part of the city had cooled, especially during the day, because of "increasing concentrations of suspended particulate matter." Likewise, outside of, but adjacent to, industrial complexes in the Po Valley of Italy, Facchini *et al*. (1999) found that water vapor was more likely to form on aerosols that had been altered by human-produced organic solutes, and that this phenomenon led to the creation of more numerous and more-highly-reflective cloud droplets that had a tendency to cool the surface below them.

In a similar vein, Rosenfield (2000) studied terrestrial analogues of ship tracks downwind of urban/industrial complexes in Turkey, Canada and Australia, to which he gave the name *pollution tracks*. His findings indicated that the clouds comprising these pollution tracks were composed of droplets of reduced size that suppressed precipitation by inhibiting further coalescence and ice precipitation formation. In commenting on this research, Toon (2000) pointed out that when clouds are composed of smaller droplets, they will not "rain out" as quickly and will therefore last longer and cover more of the earth, both of which effects tend to cool the globe.

In reviewing these and other advances in the field of anthropogenic aerosol impacts on clouds, Charlson *et al*. (2001) note that droplet clouds "are the most important factor controlling the albedo (reflectivity) and hence the temperature of our planet." Furthermore, he and his coauthors state that man-made aerosols "have a strong influence on cloud albedo, with a global mean forcing estimated to be of the same order (but opposite in sign) as that of greenhouse gases." In fact, in reviewing the very newest advances in this field of research, which have yet to be incorporated into either the analyses or recommendations of the Intergovernmental Panel on Climate Change, Charlson *et al*. conclude that "both the forcing [of this man-induced impetus for cooling] and its magnitude may be even larger than anticipated." Hence, they rightly warn us that lack of inclusion of the consequences of these important phenomena in climate change deliberations "poses additional uncertainty beyond that already recognized by the Intergovernmental Panel on Climate Change, making the largest uncertainty in estimating climate forcing even larger."

Another assessment of the issue was provided by Ghan *et al*. (2001), who studied both the *positive* radiative forcings of *greenhouse gases* and the *negative* radiative forcings of *anthropogenic aerosols* and reported that current best estimates of "the total global mean present-day anthropogenic forcing range from 3 Wm^{-2} to -1 Wm^{-2}," which represents everything from a modest warming to a slight cooling. After performing their own analysis of the problem, they reduced the magnitude of this range somewhat; but the end result still stretched from a small cooling influence to a modest impetus for warming. "Clearly," they thus concluded, "the great uncertainty in the radiative forcing must be reduced if the observed climate record is to be reconciled with model predictions and if estimates of future climate change are to be useful in formulating emission policies."

Another pertinent observation comes from Stanhill and Cohen (2001), who reviewed numerous solar radiation measurement programs around the world to see if there had been any trend in the mean amount of solar radiation falling on the surface of the earth over the past half-century. In a finding so stunning that it stretches one's credulity, they determined there was a significant 50-year downward trend in this parameter that "has globally averaged 0.51 ± 0.05 Wm^{-2} per year, equivalent to a reduction of 2.7% per decade, [which] now totals 20 Wm^{-2}." They also concluded that the most probable explanation for this observation "is that increases in man made aerosols and other air pollutants have changed the optical properties of the atmosphere, in particular those of clouds."

Although this surface-cooling influence is huge, it falls right in the mid-range of a similar solar radiative perturbation documented by Satheesh and Ramanathan (2000) in their study of the effects of human-induced pollution over the tropical northern Indian Ocean, where they determined that "mean clear-sky solar radiative heating for the winters of 1998 and 1999 decreased at the ocean surface by 12 to 30 Wm^{-2}." Hence, the decline in solar radiation reception discovered by Stanhill and Cohen could well be real. And if it is, it represents a *tremendous* counter-influence to the enhanced greenhouse effect produced by the contemporaneous increase in atmospheric CO_2 concentration. In fact, it overwhelms it.

Finally, we turn to a more recent study by Ruckstuhl *et al*. (2008), who presented "observational evidence of a strong *decline* [our italics] in aerosol optical depth over mainland Europe during the last two decades of rapid warming" -- when air temperatures rose by about 1°C after 1980 -- via analyses of "aerosol optical depth measurements from six specific locations and surface irradiance measurements from a large number of radiation sites in Northern Germany and Switzerland."

In consequence of the observed decline in aerosol concentration of up to 60%, the authors' state there was "a statistically significant increase of solar irradiance under cloud-free skies since the 1980s." The value of the direct aerosol effect of this radiative forcing was approximately 0.84 Wm^{-2}; and when combined with the concomitant cloud-induced radiative forcing of about 0.16 Wm^{-2}, it lead to a total radiative surface climate forcing over mainland Europe of about 1 Wm^{-2} that "most probably strongly contributed to the recent rapid warming in Europe."

Cleaning up significantly polluted skies can provide an even greater impetus for climate warming than does the carbon dioxide that is concurrently emitted to them, as has apparently been the case over mainland Europe for the past quarter century. Given such findings, it may not be correct to fully ascribe the high temperatures the continent has experienced in recent years to the concomitant ongoing rise in the air's CO_2 content. The lion's share of the warming has likely been produced by the removal from the atmosphere of *true* air pollutants.

Additional information on this topic, including reviews of newer publications as they become available, can be found at http://www.co2science.org/subject/a/aerononbioanthro.php.

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3.4. Non-Biological (Natural)

We conclude our section on aerosols with a brief discussion of a non-biological, naturally-produced aerosol -- dust. Dust is about as natural and ubiquitous a substance as there is. Hence, one would think we would have a pretty good handle on what it does to earth's climate as it is moved about by the planet's ever-active atmosphere. But, alas, such is not the case, as was made strikingly clear by Sokolik (1999), who with the help of nine colleagues summarized the sentiments of a number of scientists who have devoted their lives to studying the subject.

The Sokolik-led report notes that state-of-the-art climate models "rely heavily on oversimplified parameterizations" of many important dust-related phenomena, "while ignoring others." As a result, the group concludes that "the magnitude and even the sign of dust net direct radiative forcing of climate remains unclear." Now *that's* uncertainty.

Why is this so? According to Sokolik, there are a number of unanswered questions about airborne dust, including: (1) How does one quantify dust emission rates from both natural and anthropogenic (disturbed) sources with required levels of temporal and spatial resolution? (2) How does one accurately determine the composition, size and shape of dust particles from ground-based and aircraft measurements? (3) How does one adequately measure and model light absorption by mineral particles? (4) How does one link the ever-evolving optical, chemical and physical properties of dust to its life cycle in the air? (5) How does one model complex multi-layered aerosol stratification in the dust-laden atmosphere? (6) How does one quantify airborne dust properties from satellite observations?

In discussing these questions, Sokolik makes some interesting observations, noting that: (1) what is currently known (or believed to be known) about dust emissions "is largely from micro-scale experiments and theoretical studies," (2) new global data sets are needed to provide "missing information" on input parameters (such as soil type, surface roughness and soil moisture) required to model dust emission rates, (3) improvements in methods used to determine some of these parameters are also "sorely needed," (4) how to adequately measure

light absorption by mineral particles is still an "outstanding problem," and (5) it "remains unknown how well these measurements represent the light absorption by aerosol particles suspended in the atmosphere."

Considering these many problems, it is easy to understand why Sokolik says that "a challenge remains in relating dust climatology and the processes controlling the evolution of dust at all relevant spatial/temporal scales needed for chemistry and climate models," for until this challenge is met, we will but "see through a glass, darkly," especially when it comes to trying to discern the effects of airborne dust on earth's climate.

In consequence of - or perhaps in spite of - the murky status of the subject, the work goes on; and in tackling one of the chief challenges set forth by Sokolik, Vogelmann *et al*. (2003) reiterate the fact that "mineral aerosols have complex, highly varied optical properties that, for equal loadings, can cause differences in the surface IR flux [of] between 7 and 25 Wm^{-2} (Sokolik *et al*., 1998)," while at the same time acknowledging that "only a few large-scale climate models currently consider aerosol IR effects (e.g., Tegen *et al*., 1996; Jacobson, 2001) despite their potentially large forcing."

In an attempt to rectify this situation, Vogelmann *et al*. "use[d] high-resolution spectra to obtain the IR radiative forcing at the surface for aerosols encountered in the outflow from northeastern Asia," based on measurements made by the Marine-Atmospheric Emitted Radiance Interferometer aboard the NOAA Ship *Ronald H. Brown* during the Aerosol Characterization Experiment-Asia." This work led them to conclude that "daytime surface IR forcings are often a few Wm^{-2} and can reach almost 10 Wm^{-2} for large aerosol loadings," which values, in their words, "are comparable to or larger than the 1 to 2 Wm^{-2} change in the globally averaged surface IR forcing caused by greenhouse gas increases since pre-industrial times." And in a massive understatement of fact, Vogelmann *et al*. say that these results "highlight the importance of aerosol IR forcing which should be included in climate model simulations."

Another aspect of the dust-climate connection centers on the African Sahel, which has figured prominently in discussions of climate change ever since it began to experience extended drought conditions in the late 1960s and early 70s. Initial studies of the drought attributed it to anthropogenic factors such as overgrazing of the region's fragile grasses, which tends to increase surface albedo, which was envisioned to reduce precipitation, resulting in a further reduction in the region's vegetative cover, and so on (Otterman, 1974; Charney, 1975). This scenario, however, was challenged by Jackson and Idso (1975) and Idso (1977) on the basis of empirical observations; while Lamb (1978) and Folland *et al*. (1986) attributed the drought to large-scale atmospheric circulation changes triggered by multidecadal variations in sea surface temperature.

Building on the insights provided by these latter investigations, Giannini *et al*. (2003) presented evidence based on an ensemble of integrations with a general circulation model of the atmosphere -- *forced only by the observed record of sea surface temperature* -- which suggested that the "variability of rainfall in the Sahel results from the response of the African summer

monsoon to oceanic forcing amplified by land-atmosphere interaction." The success of this analysis led them to conclude that "the recent drying trend in the semi-arid Sahel is attributed to warmer-than-average low-latitude waters around Africa, which, by favoring the establishment of deep convection over the ocean, weaken the continental convergence associated with the monsoon and engender widespread drought from Senegal to Ethiopia." Hence, they further concluded that "the secular change in Sahel rainfall during the past century was not a direct consequence of regional environmental change, anthropogenic in nature or otherwise."

In a companion article, Prospero and Lamb (2003) report that measurements made from 1965 to 1998 in the Barbados trade winds show large interannual changes in the concentration of dust of African origin that are highly anticorrelated with the prior year's rainfall in the Soudano-Sahel. With respect to this subject, they state that the IPCC report of Houghton *et al*. (2001) "assumes that natural dust sources have been effectively constant over the past several hundred years and that all variability is attributable to human land-use impacts." Of this statement, however, they say "there is little firm evidence to support either of these assumptions," and their findings demonstrate why: the IPCC assumptions are simply wrong. Clearly, much remains to be learned about the climatic impacts of dust on both a regional and global scale before placing any high-degree of confidence in the climatic projections of the IPCC.

Additional information on this topic, including reviews of newer publications as they become available, can be found at http://www.co2science.org/subject/a/aerononbionat.php.

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