Coupling between Land Ecosystems and the Atmospheric Hydrologic Cycle through Biogenic Aerosol Pathways

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The importance of land surface properties in controlling the exchanges of energy, water, and momentum with the atmosphere, and thus in influencing local and regional climate, is well recognized. Important land surface characteristics—including albedo, surface roughness, latent and sensible heat fluxes, and rates of evapotranspiration—affect the way in which water is transferred to the atmosphere, processed in the atmosphere, and eventually returned to the surface. The amount of water in the atmosphere and returning to Earth, in turn, affects many of the key properties of the land surface.

Terrestrial ecosystems also modulate aerosols, such as dust and biogenically derived particles. Such aerosols may play an important role (Fig. 1) in exerting control over cloud development and precipitation through their effects on the nuclei on which cloud droplets condense or ice forms. Biogenic and other aerosols, which incorporate organic material, can be effective cloud condensation nuclei (CCN) and, thus, influence microphysical and optical properties of clouds.

Likewise, cloud properties affect the quantity and type of biogenic aerosols in the atmosphere. Changes in cloudiness, cloud optical properties, precipitation, and other meteorological variables directly control the emission and formation of biogenic aerosols by altering the availability of radiation for photosynthesis and by altering temperature. By affecting vapor pressure, temperature also determines the phase distribution of some biogenic aerosols.

Conceptually, the potential feedbacks between aerosols originating in the biosphere and the hydrologic cycle are clear. However, the processes controlling each step in this coupled system are highly uncertain, and the relative importance of these processes to the atmospheric hydrologic cycle is unknown. Thus, these processes have not yet been incorporated into quantitative numerical models describing the interactions between the terrestrial biogeochemical aerosol cycle and the hydrologic cycle.

We hypothesize that feedbacks between the terrestrial biosphere and the atmosphere by way of biogenic aerosol pathways exist, and that these feedbacks can be important in both direct and indirect radiative processes. An investigation into this coupled cycle is necessary for a better understanding of the Earth system, including climate change, regional and global atmospheric chemistry, haze and visibility, weather, and changes in land cover, including biodiversity and ecological changes. Our discussions about these feedback mechanisms led to recommendations for future research.

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Energy and water exchange. Land-surface characteristics, such as albedo, land cover, and surface roughness, control the transfer of water and energy to the atmosphere. Evapotranspiration is controlled not only by land-surface hydrology, but also by vegetation structure, plant physiology, and atmospheric variables such as solar radiation, air temperature, and humidity. These atmospheric state parameters are modulated by the absence or presence of clouds, the cloud lifetime, the cloud's optical properties, and precipitation. One example of this modulation is the gradual change of vegetation patterns as soil dries and vegetation returns after heavy rainfall. The importance of aerosols linking the biogeochemical and atmospheric hydrological cycles should be assessed in the context of energy and water exchanges between ecosystems and the atmosphere.

Aerosols, CCN, and cloud properties. Clouds and perturbations of clouds affect the CCN concentrations and chemistry (of both aerosols and precursors) by modifying the structure and radiation in the boundary layer. Additionally, biogenic aerosols are linked with cloud formation and rainfall through aerosol growth and cloud drop formation. The growth of aerosols through condensation of semi-volatile and low volatility species and coagulation of aerosols to the size range of CCN has an important effect on the CCN properties that modify cloud drop formation. In traditional theory the droplet formation potential of aerosols is controlled by the hygroscopicity (or, the effective moles of solute released into the droplet solution) and the surface tension of the droplet. Both of these properties vary significantly with dry size, density, and chemical composition. Water soluble organic compounds tend to increase the solution density and decrease the surface tension, therefore increasing CCN number concentrations.

The number of CCN is a critical link between aerosols, clouds, and precipitation. When CCN number concentrations are higher, more cloud drops are formed for the same liquid water content. Higher numbers of cloud drops increase the albedo of the cloud, prolong the lifetime of the cloud, reduce precipitation, and affect the cloud thickness. Ice nuclei may be equally important in modulating mixed phase clouds and the resulting precipitation, but are less well understood. These effects on clouds then feed back on ecosystems by modifying the light, temperature, and humidity within plant canopies.

Cloud processing also alters CCN properties. CCN may condense and release water repeatedly before they are rained out and removed from the atmosphere. Each time a cloud forms, aqueous-phase chemistry modifies the CCN properties. Volatile organic compounds (VOCs) will dissolve into the liquid phase while some compounds will partition onto the particle. As the drop evaporates and the solid residue remains, unique organic species may be formed from the ionic species that were in the aqueous phase. The resulting CCN may have very different characteristics than the original CCN, thus altering future cloud formation.

The organic compounds in the aerosols strongly influence the radiative properties of the aerosol. However, the optical effects of water soluble organic compounds in aerosols are not well characterized. In general, organic aerosols are thought to scatter radiation (although some absorption occurs) in the atmosphere, but to a lesser extent than sulfate aerosols. Thus, cooling is expected below regions where

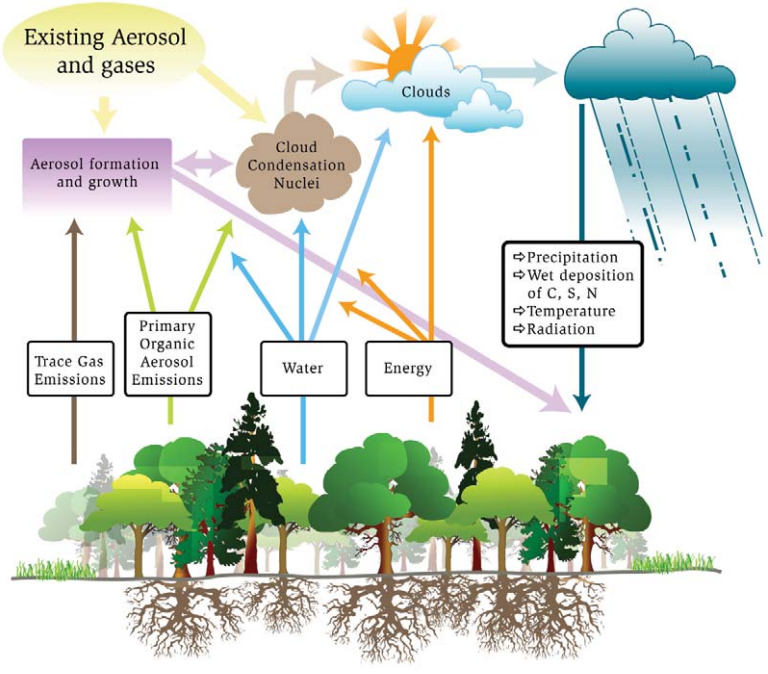

Fig. 1. Schematic of the coupling of terrestrial ecosystems and the hydrologic cycle via energy and water exchange and aerosol processing.

the aerosols composed of sulfate and/or organic compounds reside during daytime. Additionally, organic compounds often contribute to aerosol heterogeneity, modifying both the bulk radiative effects of these aerosols as well as photochemical processes. Often these radiative effects also depend on the wavelength of the incident radiation.

Aerosols affect terrestrial ecosystems. For example, after the eruption of Mt. Pinatubo, measurements in the Harvard forest indicated that the photosynthetic rate was enhanced. The increase was attributed to decreased direct radiation and increased diffuse radiation brought about by stratospheric aerosols. Sedimentation of aerosols onto leaf surfaces can also decrease photosynthetically available radiation (PAR).

Biogenic aerosols. Research on biogenic volatile organic compounds (BVOCs, especially isoprene and monoterpenes) has focused largely on their direct effects on photochemistry and oxidant production, while recent effort has been directed toward understanding their aerosol-forming potential. There is growing evidence that other BVOCs, including alkyl halides and oxidation products of sesquiterpenes, may produce aerosols with high efficiency. However, an evaluation of their significance awaits new collection and analytical techniques, as well as an improved understanding of the emission of sesquiterpenes and their oxidation. Biogenic nitrogen (ammonia, oxides of nitrogen, amines) and sulfur (dimethyl sulfide) gases are also likely to be important contributors to CCN in pristine regions, but the magnitude of these emissions and the processes controlling their release are not well known, particularly in tropical ecosystems. In arid regions, vegetation and soil moisture modulate the entrainment of mineral soil particles into the atmosphere. (We consider minerals, one of the largest aerosol emissions, in terms of mass, to be "biogenic aerosols.")

Terrestrial ecosystems are expected to contribute significantly to the total aerosol burden through both direct primary emission of microbial particles (e.g., bacteria, fungi, pollen, and spores), and humic matter, and through secondary aerosol production via the oxidation of BVOCs (including hemi-, mono-, and sesquiterpenes, and carbonyls) and other trace gases containing S (dimethyl sulfide), N (NO and NH_3), or halides. Several lines of evidence suggest that biogenic aerosols are potentially important to the overall aerosol burden, altering aerosol radiative and absorptive properties, and contributing to CCN formation and growth. Because the amount of some gas-phase aerosol precursors emitted from vegetation, particularly monoterpenes, is well studied, their contribution to secondary aerosol formation in the atmosphere has been calculated. Estimates of global organic aerosols formed through BVOC oxidation range from 19 to 30–270 Tg yr^{-1}. These estimates are quite uncertain because the emission rates of certain gas-phase BVOCs, such as sesquiterpenes, are less known but are expected to be substantial and to contribute to secondary aerosol formation. In addition, these estimates do not consider changes in all of the relevant environmental variables that affect secondary organic aerosol formation.

Estimates of primary aerosol emissions from biogenic sources are less well understood than many of the gaseous emissions due to the large number of potential sources and controlling processes. Primary biogenic aerosol emissions, however, could be substantial. For example, biogenic sources are the main contributors to the aerosol burden during the wet (non-biomass burning) season in Amazonia.

RESEARCH TOPICS TO ADVANCE THE HYPOTHESIS

- Biological function of BVOCs in plants
- Radiation in forest canopies
- Biological and environmental controls on emissions of aerosols from plants
- Cloud drop formation from multicomponent aerosols
- Aqueous-phase chemistry

Recent work is beginning to provide a quantitative understanding of the contribution of biogenic particles to the total aerosol number and mass over some terrestrial ecosystems. The conditions under which secondary BVOC products participate in nucleation and condensational growth have also been elucidated. For example, in coastal environments, frequent aerosol nucleation events have been attributed to the oxidation of diiodomethane emitted from marine algae. Above boreal forests, large-scale nucleation events, followed by particle growth, have been attributed to organic acids.

Atmospheric hydrologic effects on the emission of biogenic aerosols and precursors. The emissions of many BVOCs (and directly emitted biogenic aerosols, such

as waxes, spores, bacteria, and mineral dust) depend on vegetation density and species distribution, solar radiation, ambient temperature, and a variety of other environmental parameters, such as drought stress, nutrient availability, and ambient oxidant concentrations. Therefore, these emissions are closely linked to the atmospheric hydrologic cycle. For example, cloud cover determines the amount of photosynthetically active radiation that reaches terrestrial ecosystems, and the emissions of several BVOCs depend on light. Daytime convective updrafts resulting from solar radiation may also control the amount of bacteria emitted into the atmosphere. Emissions of BVOCs are altered when plants become water stressed, and thus are directly coupled with rainfall.

RESEARCH DIRECTIONS. The hypothesis proposed in this discussion is that terrestrial biogenic aerosols are important in the atmospheric hydrologic cycle, and that feedbacks between the hydrologic cycle and biogeochemical cycles control the concentration of the biogenic aerosol. Although an initial understanding of the individual processes is currently available, multiple levels of questions must be addressed in order to evaluate the feedback hypothesis.

General issues. The scales, both temporal and spatial, at which this hypothesis is tested must be established. Because aerosols and CCN may go through several cycles before being rained out of the atmosphere and water may travel as far as 1000 km before returning to the earth, evaluating this cycle at the local scale may be difficult. However, the Amazon region during its wet season may be an appropriate locale to test the coupled-cycle hypothesis. Because of the frequent rain during the wet season, the water is quickly recycled, with ~75% of the rainfall at locations such as Manaus, Brazil, being returned to the atmosphere via evapotranspiration. In addition, heterogeneous land cover over a 1000-km spatial scale may dramatically affect cloud formation. Vegetation characteristics change seasonally, and even over the course of years. These temporal changes would also influence the atmospheric hydrologic cycle. The diurnal variation in emissions and atmospheric processes will be very important to the overall understanding of the coupled cycle. Thus, understanding the impact of the diurnal cycle on the individual parts of the interactions between land ecosystems and the atmospheric hydrologic cycle should be addressed foremost.

Individual components of the coupled cycles. Three topics related to processes in the forest canopy should be addressed. First, our ability to predict how radiation is processed within canopies needs to improve. Photosynthesis is affected by the proportion of incident PAR that is received as direct or diffuse radiation. Thus, we need to quantify the distribution of PAR in a variety of canopies. Second, we need to determine the biological function of BVOCs in plants, and to improve our understanding of the biological and environmental controls on BVOC emissions. Third, not only does the flux of primary aerosols from the canopy to the atmospheric boundary layer need to be quantified, but also the size and hygroscopicity of the aerosols and the controlling factors (e.g., light and temperature) of their emissions must be characterized.

Topics related to aerosol–cloud interactions that need to be understood better are cloud drop formation and aqueous chemistry. In particular, studies investigating the effect of organic species in CCN on the formation of cloud drops should be continued. Laboratory research has already yielded much information but has proven to be particularly challenging because of the physical and chemical complexity of the organic aerosols. Studies on aqueous-phase sulfate formation and its impact on the CCN size distribution need to continue, particularly for sulfate production catalyzed by transition metal ions. However, emerging needs are determining what organic chemistry is occurring in the aqueous phase and how such chemistry is affecting the CCN. For example, it is known that methylglyoxal, which is formed in the gas phase from oxidation of alkenes and aromatics, can be oxidized in the aqueous phase to form pyruvic acid, which then can become part of the CCN because of its low volatility. Organic aqueous chemistry should be assessed with laboratory, field, and modeling studies.

Other topics of importance to our hypothesis should also be addressed. The magnitude, the controlling conditions, and the gas and aerosol chemical mechanisms of secondary organic aerosol (SOA) formation should be determined. The organic compounds in aerosols need to be identified, and the optical properties of water-soluble organic compounds in aerosols need to be quantified. Studies must continue on determining the composition and physical characteristics of atmospheric CCN and the time scales for CCN growth. The calculation of the wet deposition of a variety of gas-phase

species needs to be improved. Investigations into the onset of precipitation in clouds and how CCN characteristics may affect precipitation formation must continue. Ice microphysics, particularly the nucleation of ice, needs to be understood better. Evaluating which ecosystems are more important to the coupling of clouds and biogenic aerosols should be pursued. The need for a well-coordinated, integrated field program to measure the importance of interactions between ecological, hydrological, and atmospheric systems should be evaluated. Assessment of the impact of altered land use, both natural and anthropogenic, on the coupled cycle should be encouraged.

The individual processes of the coupled cycle and the coupling of those processes need to be addressed by laboratory, field, and modeling studies from the nanoscale to the global scale. One approach that we plan to pursue is to assess coupled feedbacks between the biogeochemical and the atmospheric hydrologic cycles via a simple model. Estimates of the emissions of organic aerosol precursors and their formation of secondary organic aerosols can be used to determine the organic aerosol mass, number concentration, and optical depth. These results can then be implemented into calculations of cloud drop activation and precipitation formation, which then can influence the emissions of the precursor gases.

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