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Integrated land ecosystem–atmosphere processes study (iLEAPS) assessment of global observational networks

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Long-term, continuous observations are needed for Earth system investigations and evaluation of simulations. The atmospheric and ecological communities have independently established field sites that have been running for many decades and are integrated into global networks. In the past decade, the importance of long-term observational networks focused on land ecosystem–atmosphere exchange, and the processes controlling land–atmosphere coupling, had been increasingly recognized and has led to the building of a global network of water, carbon and energy flux sites. This is an important step but further enhancements are necessary in order to quantify all of the land–atmosphere processes that need to be included in Earth system models. This paper describes the current land ecosystem–atmosphere measurement capabilities and presents the status and needs for global observational networks.

Introduction

Global environmental problems including climate change, air quality, lack of fresh water, land-cover changes, food security, biodiversity and their interconnections and feedbacks have created an urgent need to observe those changes. Therefore, humankind urgently needs reliable and precise information on present climate and environmental system change, especially for making sound policy decisions on national, regional and global level, for sustainable development. The land surface–atmosphere interface is particularly crucial for the functioning of the Earth System (ES) through interactions via mass, energy, and momentum fluxes, as well as through the biogeochemical cycles. At the same time, climate variability and atmospheric processes, such as transport and deposition of chemicals, are major constraints on biogeochemical cycles, ‘natural’ as well as anthropogenic ones. Human-driven change in land cover is likely to result in significant regional and global climate change. In turn, climate change affects terrestrial ecosystems at all spatial and temporal scales, maybe even to the extent of destabilizing large regions.

Earth system models are advancing to the point where they can begin to fully simulate the coupling between the physical, chemical, and biological processes in the climate system. This is facilitated by increases in computational power but a major limitation is the lack of suit-

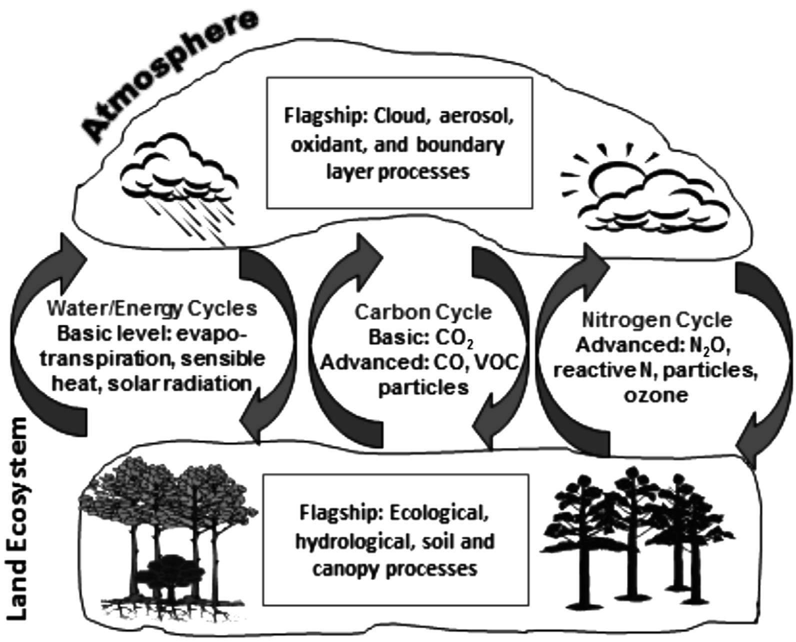

Fig. 1. Schematic of land ecosystem–atmosphere interactions and hierarchal observational levels that include basic, advanced, and comprehensive measurements at "flagship sites".

able observations for developing and evaluating the quantitative relationships needed for realistic simulations of the controlling processes. The integrated Land Ecosystem–Atmosphere Processes Study (iLEAPS, www.ileaps.org) has advocated studies of the implications of transport and transformation processes at the land–atmosphere interface to advance our understanding of Earth system dynamics that can then be incorporated into Earth system models. iLEAPS endorses interdisciplinary research that addresses key scientific questions by integrating local and regional model simulations with remote sensing, regional network observations, and canopy-to-regional-scale field studies. Regional studies and networks organized by national or regional scientific communities are enhanced by iLEAPS activities that connect regional efforts into global networks that provide knowledge transfer and link interdisciplinary observations that span the globe. The scope of iLEAPS research, particularly on coupled interactions and feedbacks, is elaborated by Arneth *et al*. (2010) and Bonan (2008). This recent progress in understanding terrestrial biogeochemical feedbacks and their linkages has led to initial estimates of the potential magnitude of biogeochemical feedbacks associated with human-mediated changes in the biosphere. Importantly, the overall magnitude of biogeochemical feedbacks could potentially be similar to that of feedbacks in the physical climate system, but there are large uncertainties in the magnitude of individual estimates and in accounting for synergies between these effects (Arneth *et al*. 2010). Continued advances in quantitative modeling require simultaneous observations of a variety of constituents that can be used to improve modeling approaches.

The ES components, interactions and feedbacks that are the focus of iLEAPS efforts include investigations of atmospheric, ecological and hydrological processes; surface fluxes of energy, aerosols, carbon dioxide (CO_2), water, and organic and nitrogen compounds; ecohydrological disturbances and other factors that control the system, followed by efforts to improve their representation in Earth system models (Fig. 1). These investigations will lead to an improved ability to quantitatively characterize the impact of land management decisions and unintended ecohydrological disturbances on biosphere–atmosphere exchange, and the associated implications for ecosystem health, air quality, and climate on time scales of months to years.

Conceptually, the ES connections and interactions (Fig. 1) are clear. However, the processes

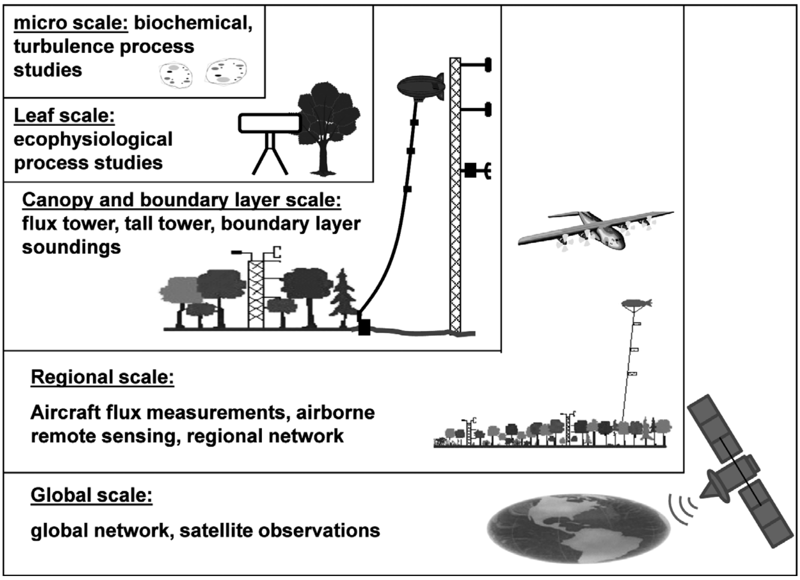

Fig. 2. Scales and observational techniques for land ecosystem atmosphere observations.

controlling this coupled system are highly uncertain and not well quantified, precluding the full incorporation of these processes into ES models. In order to understand water and biogeochemical cycles, observing fluxes between different ES compartments, like atmosphere and ecosystems, is crucial. We also need to observe the stocks and processes in the atmosphere, biosphere and soils; concentrations of greenhouse gases, reactive trace gases, aerosols. The rapid development of measuring techniques has increased our abilities to monitor climate and environmental change and to obtain information about the changes in the atmosphere and ecosystems. Digital technology and communications has made constructing and operating automatic measurement stations much easier and has improved measuring accuracy and precision.

However, no systematic measurement networks to analyze the change and the interconnections between all energy, water and biogeochemical flows in the system of atmosphere, vegetation and topsoil are available. Although independent studies of certain aspects exist, an international interdisciplinary research effort, establishing and quantifying links between these processes and potential feedbacks, is necessary to determine whether the biosphere has significant ability to control the Earth system through interactions with the atmosphere and hydrosphere. One of the major challenges is linking the small-scale observations, used to improve our fundamental understanding of land ecosystem–atmosphere processes, to the regional-scale interactions that must be represented in ES models. Approaching this requires biochemical cellular studies, plant physiology enclosure studies, above-canopy micrometeorological towers, and airborne and satellite sensors (Fig. 2). The integration of Earth observations from ground and from satellites, boundary layer measurements and modeling from the planning of measurement station location to the ES level is of importance and also requires collaboration among a variety of agencies and research communities.

The data handling procedures, involving storage in databanks, harmonization, validation, and quality control are of high priority in connection with observational networks. Methods that increase the reliability of results, both data and modeling (model-data assimilation), as well as more advanced data handling methods such as data mining (designing of algorithms capable of recognizing complex patterns in different data streams) are valuable both to experimentalists and modelers in the ES community. A global observation network will be more useful for model development and evaluation if the observation suite and site locations are determined in an iterative approach through collaborative efforts of modelers and observational scientists. Uncertainty quantification is a necessary component of an observational network and modeling studies are needed to determine which uncertainties must be reduced in order to address specific scientific questions.

Recently, Hari *et al*. (2009) proposed the development and construction of a hierarchical network of measuring stations which would produce systematic information on climate system change in forests, utilizing novel measuring techniques. Hari *et al*. (2009) proposed a three-level hierarchical system: (i) basic level, (ii) flux level, and (iii) "flagship" level. The basic stations would provide spatial information, the flux stations would provide information on fluxes in the ecosystem, and the flagship stations would provide information on processes generating the fluxes, develop instrumentation, and serve to train scientists and technical staff. To obtain global coverage, the number of basic-level stations should be around 8000, the number of flux stations around 400, and the number of flagship stations around 20 globally (Hari *et al*. 2009). Sites should be located through a strategic design that optimizes the distribution of limited resources by ensuring representativeness of key ecosystems and locating sites appropriately to address questions associated with regional interactions.

The framework proposed by Hari *et al*. (2009) for forests can be extended to all ecosystems for characterizing land–atmosphere exchange across the Earth surface. This paper builds on the strategy proposed by Hari *et al*. (2009) and describes the current status of global land ecosystem–atmosphere networks and considers the enhancements needed to provide adequate observations.

Basic flux level: carbon dioxide, water vapor and energy fluxes

It is not surprising that until now, the implementation of a long-term land–atmosphere exchange observational network has focused on carbon dioxide, water vapor and energy. These three components are the major drivers of the climate system and an improved understanding of the processes controlling these fluxes is a high priority for improving predictions of how the Earth system will respond to human activities. Micrometeorological techniques for quantifying land–atmosphere constituent exchange are the most direct means of measuring canopy to landscape scale fluxes. Water vapor, CO_2, and sensible heat were first measured over 50 years ago (see, for instance, Monteith and Sziecz 1960) using micrometeorological flux-gradient approaches that worked reasonably well in daytime over flat grasslands but could not provide reliable measurements for forests, nighttime conditions and even moderately complex terrain. The development of fast-response sensors suitable for eddy covariance measurements provided a much improved technique for measuring these fluxes.

Long-term continuous measurements began at a few sites about 20 years ago (Wofsy *et al*. 1993, Vermetten *et al*. 1994, Baldocchi 2008) and the number of active sites steadily increased into the hundreds during the 1990s (Baldocchi *et al*. 2001). Many sites were integrated into regional networks which were joined through FLUXNET, a network of networks. FLUXNET has several important roles including archiving and distributing observations, calibration and intercomparison, and facilitating synthesis and communication within the FLUXNET community.

FLUXNET data have been used to determine net annual fluxes in important ecosystems and quantify diurnal, seasonal and interannual variability. The observations have been used to characterize ecosystem response to changing growing season, sunlight, temperature, and stand age (Baldocchi *et al*. 2001). The resulting insights into carbon, water, and energy dynamics have been used to improve the land surface components of Earth system models although comparisons of these models indicate that there are still large uncertainties associated with the representations of these processes in these models. The decadal observations of Dunn *et al*. (2007) indicate an ecosystem shifting from a carbon source in 1995 to a carbon sink in 2004 demonstrating the need for long-term observations. Investigators have also begun to directly integrate FLUXNET data with satellite observations and ecological data to produce regional- and continental-scale estimates of the magnitude and distribution of carbon (e.g., Xiao *et al*. 2008) and water (e.g. Jung *et al*. 2010). This upscaling approach has the potential to improve carbon flux estimates in an approach similar to the use of data assimilation for weather forecasting.

The compilation of a global FLUXNET dataset, the "LaThuile" dataset (*see* www.fluxnet.org), allows for a variety of conclusions regarding specific ecosystems and also on a global scale. As an example of the application of the global dataset of carbon dioxide flux measurements, Mahecha *et al*. (2010) found marked differences in the long-term fate of carbon taken up through photosynthesis, as well as in the availability of this carbon for respiration, across the sites. Another example is the Beer *et al*. (2010) observation-based global terrestrial gross primary production (GPP) estimation that shows missing feedbacks in biosphere models.

Atmospheric and ecological sciences are established scientific fields with observatories sponsored by the research and resource management agencies associated with individual disciplines in many nations but this is often not the case for multi- and inter-disciplinary land–atmosphere measurements. The global CO_2, water, and energy flux measurement network began as a collection of individual scientific activities funded by agencies with a wide range of objectives. As these measurements become somewhat routine, and more sites and longer time series are necessary to obtain publishable results, it becomes more important to identify stable funding from institutions and agencies that are able to make a commitment to long-term observations.

Advanced flux level: extending flux networks beyond CO_2, water, and energy

Water vapor, CO_2, and energy are not the only types of land–atmosphere exchange that are important for climate, air quality, and ecosystem functioning and yet there are few long-term observations of any other constituents. New harmonized networks for Earth system observations are now being developed in some regions, including northern America and Europe. The European ICOS (Integrated Carbon Observation System) is designed to observe the fluxes and concentrations of the three major greenhouse gases, carbon dioxide, methane, and nitrous oxide. The International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects and Forests (ICP Forests) has integrated forest condition and air pollution investigations by conducting measurements of carbon and water fluxes along with ozone and nitrogen fluxes at forest sites (Jochheim *et al*. 2009). The U.S. NEON (National Ecological Observatory Network) has broader aims that include biosphere–atmosphere fluxes. As these aim at harmonized long-term measurements covering major ecosystems, they may in the future provide a backbone for observational studies of biosphere–atmosphere gas exchange on these continents. In addition to these regional enhanced networks, global observational networks for additional constituents can be built by adding instrumentation to the existing FLUXNET sites.

In this section, we assess both the need for long-term observations of land–atmosphere exchange of individual Earth system constituents and the feasibility of these measurements. This includes discussion of how our understanding of the Earth system could benefit from these measurements as well as of the technical and logistical constraints associated with potential measurement techniques.

Methane (CH_4) and nitrous oxide (N_2O)

Methane and nitrous oxide are important contributors to global atmospheric radiative forcing. Therefore, an accurate understanding of their sources and sinks, and how they might change in the future, is necessary. A major challenge associated with quantifying global land–atmosphere methane exchange is that the global total emission is comprised of many significant sources including termites, methane hydrates, wetlands, rice paddies, biomass burning, natural gas production and distribution, landfills, sewage treatment, animal waste, and enteric fermentation. Each of these sources contributes between 3% and 22% of the global total and should be considered in Earth system models. In some ecosystems, methane also comprises a significant part of the carbon exchange between the ecosystem and the atmosphere (Lohila *et al*. 2007, Rinne *et al*. 2007). The magnitude of individual sources is not constant. For example, due to the expected warming in the high latitudes, the increased release of methane from melting permafrost is anticipated.

Early studies of methane emission relied on tracer and enclosure techniques which provided an initial process-level understanding of methane source magnitudes and controlling factors. The first generation of fast response methane analyzers based on tunable diode laser absorption spectrometry (TDLAS) enabled ecosystem scale eddy covariance measurements (Suyker *et al*. 1996, Hargreaves and Fowler 1998, Kormann *et al*. 2001, Hargreaves *et al*. 2001). However, because this laser can be operated only in low temperatures, liquid nitrogen or cryo-cooling devices were necessary. Thus, this measurement technique was confined to sites with good access and infrastructure. Furthermore, only the most advanced of these instruments were stable enough to run unattended for longer periods. Thus eddy-covariance measurements of methane were usually confined to short measurement campaigns, with the exception of some longer-term measurements (Suyker *et al*. 1996, Rinne *et al*. 2007).

A new generation of fast-response instrumentation which does not need cryogenic cooling is now commercially available (*see*, for instance, Hendriks *et al*. 2008, Tuzson *et al*. 2010). These instruments are based on newer laser technology and have an optical multipass cell that is either open or closed. With these instruments, there is no need to supply the site with liquid nitrogen or with power necessary for a cryo-cooler. This enables long-term measurements at more remote locations. The choice of open-path or closed-path solution depends on application. The open-path methane analyzer is truly a low-power solution, unlike the closed-path version which may require considerable amounts of power to run the pumps. However, the operational characteristics of a closed-path system are currently better understood. Experience on long-term performance of any of the new instruments is still scarce.

Nitrous oxide is a significant greenhouse gas with a global source dominated by emissions from agricultural areas (Fowler *et al*. 2009). Nitrous oxide emissions may even determine the climatic profitability of biofuel production (Crutzen *et al*. 2008). Ecosystem-scale measurements of nitrous oxide emission have suffered from the same instrumental limitations as those of methane. The new laser technology has also enabled longer-term measurements of this compound as the new generation of instrument does not require liquid nitrogen or cryo-coolers. These closed-path instruments operate in low pressure and need powerful pumps. Thus, they require an electric power line or a generator at the measurement site (Neftel *et al*. 2010).

Networks of long-term methane and nitrous oxide eddy flux measurements in terrestrial ecosystems are important for characterizing variations in sources and sinks of these radiatively active gases and are now considered feasible. Also, measurements of methane and nitrogen are necessary in some ecosystems to close the carbon and nitrogen budgets. ICOS has identified long-term methane and nitrous oxide flux measurements as a high priority and is establishing a European network especially for these compounds. We need to expand this network to other parts of the world to characterize additional important biomes, building on the approach defined and lessons learned by this initial regional network.

Volatile organic compounds

Terrestrial ecosystems are the major source of volatile organic compound (VOC) emissions into the atmosphere (Guenther *et al*. 1996). In the atmosphere, the oxidation of VOC can influence aerosol particles, precipitation acidity, and regional ozone distributions (Guenther *et al*. 2006). Accurate predictions of biogenic VOC emissions are important for developing regulatory ozone and aerosol control strategies for at least some rural and urban areas (Karl *et al*. 2001).

Oxygenated VOC emitted from vegetation contribute to oxidant production in the upper troposphere. The capacity of VOC to produce aerosol particles is an active area of research: it is well known that VOC serve as precursors to organic aerosol particles which, in turn, have a direct (reflecting sunlight back to space) and indirect (cloud formation) effect on the global radiation balance (Kulmala *et al*. 2004b). These organic carbon emissions are also a minor but potentially significant pathway for the flow of carbon between an ecosystem and the atmosphere (Guenther 2002).

One of the great challenges associated with characterizing biogenic VOC (BVOC) is the large variety of compounds. Isoprene is the single most important BVOC with an emission that is about half of the global BVOC emission (Guenther *et al*. 2006). Many monoterpenes have been observed in the atmosphere but only a few, such as α-pinene, make a significant contribution to the global total emissions. The dominant sesquiterpenes, such as β-caryophyllene, have lifetimes of only minutes in the atmosphere and so are present at very low levels but their reaction products may be an important source of secondary organic aerosol. Oxygenated BVOC include a wide range of alcohols, aldehydes, ketones, acids, ethers, and esters but are dominated by relatively low molecular weight compounds such as methanol, acetaldehyde and acetone. Other BVOC include alkanes (e.g., heptane), alkenes (e.g., ethene), arenes (e.g., toluene), sulfur compounds (e.g., dimethyl sulfide), and nitrogen compounds (e.g., hydrogen cyanide). Observations of land–atmosphere interactions must include not only primary emissions but also the larger number of reaction products that impact atmospheric oxidants and particle formation and growth.

For the plant, the production of BVOC requires a significant allocation of resources, which leads to the question of why plants would produce these compounds if they merely end up being lost into the atmosphere. We know that at least some BVOC emissions have an important biological role although there are other cases where the purpose remains a mystery. One of the best known biological roles is the use of BVOC by plants to attract pollinators and seed dispersers. Insects and animals are also known to use BVOC for a variety of other signaling activities. Some VOC are emitted from a limited number of plants or for only a limited time but emissions can be high for certain conditions and locations. Examples of this include large emission of linalool from stands of flowering plants and large emissions of methyl salicylate from stressed vegetation. Emission variations are driven by environmental conditions (light and temperature) and land-cover characteristics (foliar biomass and plant species composition) that result in variations of more than an order of magnitude for different ecosystems and for different seasons at the same location. The large variety of compounds, biological roles, and complex controlling variables make quantitative predictions of BVOC emissions a challenging task. The lack of long-term observations is a major limitation for parameterizing and evaluating existing models.

Fast-response analyzers suitable for eddy-covariance flux measurements of the most important BVOC are commercially available. They include a chemiluminescence analyzer for isoprene (Guenther and Hills 1998) and PTRMS (Proton Transfer Reaction Mass Spectrometry for a wide range of BVOC (Karl *et al*. 2001). Several studies have reported long-term BVOC eddy flux measurements (Pressley *et al*. 2005, Holzinger *et al*. 2006). These efforts have demonstrated the feasibility of long-term measurements and the value for improving understanding of the processes controlling these emissions. This is particularly important since these studies have shown that BVOC emissions are particularly sensitive to environmental and land-cover change. However, the considerable expense and expertise required for operating these direct eddy covariance measurements may limit long-term BVOC measurements to relatively few sites, such as the flagship sites described in the following section. An alternative for a widespread network is the utilization of another micrometeorological flux technique called REA (relaxed eddy accumulation). Inexpensive, low-power and reliable REA systems for measuring BVOC fluxes (Guenther *et al*. 1996, Ciccioli *et al*. 2003) could be deployed at a large number of flux tower sites. However, the samples collected by the REA systems would probably have to be shipped to a laboratory for analysis. Consequently, VOC fluxes would not be measured continuously. Given the relatively good understanding of diurnal VOC emission variations and the poor understanding of seasonal to interannual variations and regional distributions, periodic REA measurements can provide a valuable contribution to our understanding of the processes controlling VOC emissions.

Reactive nitrogen

Nitrogen (N) plays a key role in regulating plant growth and photosynthesis. In the atmosphere, nitrogen is a key mediator in many photochemical processes and plays critical roles in tropospheric ozone production, acid deposition and aerosol formation. Nitrogen deposition can occur either from direct deposition of gaseous or aerosol nitrogen (dry deposition) or dissolved within precipitation (wet deposition). Wet deposition of nitrogen is the focus of several existing long-term observation networks (Baumgaudner *et al*. 2002, Vet *et al*. 2005, Erisman 2005) and will not be discussed here. Dry deposition of N-species is rarely measured on a routine basis. The exchange of gas- and aerosol-phase nitrogen between the atmosphere and biosphere is exceedingly complex since all of the various forms of atmospheric nitrogen have their own deposition characteristics as well as different chemical production and loss mechanisms that operate on a range of time scales. Reactive nitrogen compounds can be classified into three main groups: (1) ammonia and amines, (2) aerosol-N (which includes ammonium and nitrate) and (3) oxidized-N which is often referred to as NO_y.

NO_y is typically used to refer to the sum of oxidized nitrogen species in the atmosphere. It consists primarily of NO, NO_2, HNO_3 and gas phase N-containing organics (peroxyacetyl compounds, organic nitrates, etc.). There are smaller contributions from compounds such as HONO, N_2O_5, and NO_3, but because of their short atmospheric lifetimes, these are a small portion of the concentration budget. The intrinsic problem with NO_y deposition is that each species has different surface exchange characteristics and the concentrations vary with photochemical processing in the atmosphere. Even though HNO_3 is often a small fraction of NO_y concentration (5%–25%, Williams *et al*. 1997), it is typically the major contributor to NO_y flux (Horii *et al*. 2006, Sievering *et al*. 1996). To our knowledge, there has not been a study that directly measures fluxes of all of the NO_y species; these studies are usually undertaken by either measuring concentrations coupled with inferential deposition modeling (Zhang *et al*. 2009, Trebs *et al*. 2006) or by measuring fluxes of selected species and modeling others (Horii *et al*. 2006). At present, the difficulty in measuring fluxes of all the individual NO_y species make long-term monitoring of exchanges of all the components of NO_y intractable; however, certain aspects of NO_y deposition could be targeted.

Knowing the total amount of oxidized nitrogen entering an ecosystem via dry deposition would be useful information for modeling of ecosystem productivity. Some existing instruments are capable of measuring eddy covariance fluxes of total NO_y. Whereas some of these are fairly specialized (Farmer *et al*. 2006), some are commercially available with some minor modifications (Munger *et al*. 1996, Turnipseed *et al*. 2006). The work of Munger *et al*. (1996) also demonstrated the feasibility of using this modified-commercial system for long-term measurements over several years. The technique is based on the familiar ozone-induced chemiluminescence detection of NO. The key for using this method for NO_y is to rapidly convert all of the different NO_y species to NO at the inlet of the instrument (close to the path of the sonic anemometer for eddy covariance measurements) by means of a heated gold catalyst. NO can then be transported through tubing to a commercial NO instrument. These analyzers typically have adequate sensitivity and can be modified for the fast sampling rates (1 to 10 Hz) necessary. These measurements do require significant power (~500 watts) and expertise. Thus, they may not be feasible at all locations.

Fluxes of certain species of NO_y can be measured by eddy covariance. NO and NO_2 (collectively known as NO_x) have long been of interest to micrometeorologists since they rapidly interconvert on a scale of a few minutes (similar to that of local turbulence). Therefore, any direct micrometeorological flux measurement of either of these species is affected by the photochemical partitioning. In terms of total nitrogen deposition, the surface fluxes of NO and NO_2 often offset each other to some degree: NO is emitted from soils whereas NO_2 is taken up via stomata in vegetation. However, owing to their fast photochemical interconversion and different source/sink vertical profiles, the surface exchange of these compounds are often quite complicated. There are very few long-term studies of NO/NO_2 surface exchange (Rummel *et al*. 2002, Farmer *et al*. 2006, Horii *et al*. 2006); many studies to date have used long-term NO and NO_2 concentrations and then inferred deposition (Zhang *et al*. 2009, Trebs *et al*. 2006).

As mentioned above, NO can be measured at high sampling rates using its gas-phase chemiluminescent reaction with ozone. Although several techniques exist for the detection of NO_2 (laser flurorescence, etc.), the most common (and commercially available) approach is to convert NO_2 to NO via either photolysis or a heated molybdenum catalyst. The NO produced is then detected via chemiluminescence, so that a single instrument can be used for both NO and NO_2.

Recent work has demonstrated that fluxes of peroxynitrate compounds can be measured by eddy covariance (Turnipseed *et al*. 2006) using a Chemical Ionization/Mass Spectrometric (CIMS) technique (Slusher *et al*. 2004). Wolfe *et al*. (2009) used this technique to measure EC fluxes of several peroxynitrate compounds (focusing primarily on peroxyacetylnitrate or PAN) over an entire season. This does imply that long-term flux studies are feasible; however, this instrumentation is not commercially available and the technique does require considerable power and expertise that is not available at most network flux tower sites.

Nitric acid (HNO_3) is of utmost importance for understanding the dry deposition of nitrogen. Along with ammonia, HNO_3 is deposited very rapidly to nearly all surfaces and typically constitutes a majority of the deposited NO_y. However, the same properties that make HNO_3 important in total N-deposition make it very difficult to measure. There is currently no method specific to HNO_3 that has been used successfully to determine eddy-covariance fluxes. Therefore, micrometeorological methods that rely on integrated samples, such as the gradient or relaxed-eddy accumulation techniques, are the only viable alternative (Pryor *et al*. 2002). However, these methods are often labor-intensive and as such, not very suitable for long-term continuous measurements.

A long-term reactive nitrogen land–atmosphere flux network is necessary to quantify nitrogen inputs to ecosystems and to understand biogenic contributions to atmospheric reactive nitrogen. The ICP Forest Programme has demonstrated the value of these observations with the use a simple canopy budget model to assess total nitrogen deposition (Staelens *et al*. 2009). Instrumentation exists for using eddy covariance for measuring the sum of some reactive nitrogen species although additional effort is necessary to characterize which species are being measured. NEON has identified long-term NO_y flux measurements as a priority and is including NO_y eddy flux measurements as a component of NEON. The experience gained by NEON will be valuable in determining whether and how to expand this measurement to a global reactive nitrogen flux network.

Ozone

Ozone is one of the most important atmospheric oxidants and plays a key role in atmospheric oxidation processes, so understanding all of its formation and loss processes (including deposition) is important for understanding tropospheric photochemistry. It also inhibits plant growth via its uptake into stomata of vegetation. Depending upon concentration level, these effects can be either acute or chronic. Acute symptoms of plant stress are seldom seem in natural ecosystems at typical ozone levels; however, chronic effects develop over long time scales (years) and thus, long-term monitoring is required to assess this type of plant damage.

Monitoring ozone concentrations is not sufficient for assessing ozone damage since it does not account for the amount of ozone that is taken up by a plant. Direct micrometeorological fluxes of ozone measure the sum of the uptake via the stomata as well as to other surfaces (cuticles, bark, soils). The stomatal uptake is the only component associated with ozone damage so assessments typically use models of stomatal conductance, along with measured ozone concentration, to estimate the stomatal deposition fraction. The parameters needed to model ecosystem-level stomatal conductance (vapor pressure deficit, latent heat flux, etc.) and the O_3 concentration are the necessary inputs for these models.

Most of these parameters are routinely measured at the nearly 500 flux tower sites designed to monitor energy, water and CO_2 exchange (*see* previous section). The technology for monitoring O_3 concentration using UV-absorbance is well developed, relatively inexpensive and requires little maintenance. Several new O_3 monitors now exist that operate at very low power, which means that O_3 concentration measurements could be added to nearly all of the existing flux sites at a relatively low cost and effort.

Quantification of the total atmospheric loss of ozone is necessary for understanding the photochemistry of ozone and requires a direct measurement of the total deposition flux. Furthermore, recent studies have also shown that the non-stomatal portion of the flux may, in part, be due to chemical reactions with highly reactive species (NO, organics) that are emitted from either soils or vegetation (Kurpius *et al.* 2005). Rapid oxidation of high-molecular-weight organics by ozone may then contribute to aerosol formation. Therefore, both the total flux and the parameters necessary for modeling of the stomatal contribution need to be measured.

Several previous studies have reported on long-term continuous monitoring of ozone flux (Mikkelson *et al.* 2004, Fares *et al.* 2010, Munger *et al.* 1996, Hogg *et al.* 2007). Many techniques based on chemiluminescence are capable of sampling rates amenable to using eddy covariance. Note that all of the long-term studies mentioned above used a different method of ozone detection. All of these techniques have their advantages and disadvantages (Kalnajs and Avallone 2010, Muller *et al.* 2010), but all require more effort to run as continuous long-term measurements than the familiar CO_2 and H_2O instrumentation currently available. Furthermore, all of these techniques must be run in parallel with a more stable, slow-response ozone analyzer (typically a UV-absorbance monitor) for calibration purposes. Muller *et al.* (2010) also showed that with "dry" chemiluminescence detectors (where ozone reacts with a dye impregnated on a solid substrate), the way the calibration is carried out can affect the derived flux. Recently developed fast-response instruments based on UV-absorbance (Kalnajs and Avallone 2010) could alleviate many of the calibration and stability issues related to chemiluminescence instruments; however, these are still in development and not commercially available.

A long-term land–atmosphere flux network for ozone is necessary for quantifying the influence of this chemical on ecosystem health. A network would be particularly valuable for improving Earth system models if it could provide measurements of stomatal and non-stomatal components of ozone deposition. NEON has identified long-term ozone flux measurements as a priority and is including ozone flux-gradient measurements as a component of NEON. This could be a worthwhile approach for a global flux network although continued improvements in UV-absorbance techniques for ozone eddy covariance measurements could provide a better alternative.

Aerosol particles

Aerosol particles have important roles in both climate and air quality. However, they are among the most difficult to accurately represent in Earth system models because of their highly complicated production and loss mechanisms and complex atmospheric effects. Long-term measurements of particle fluxes, including size resolved measurements of chemical composition and physical properties, are necessary for understanding how atmospheric distributions and ecosystem inputs will respond to changes in the Earth system. Land ecosystems are a major source of atmospheric particles, including both direct emissions (dust, pollen, etc.) and the atmospheric formation and growth of particles from the surface emission of precursor gases (gas-to-particle conversion, Kulmala *et al.* 2004a, 2004b). Dry and wet deposition to land surfaces is also a major loss process for atmospheric particles.

Particle number fluxes have been derived with the eddy covariance technique since the late 1970s (e.g. Wesley *et al*. 1977). Characterizing the chemical composition or resolved size distributions is more challenging but the eddy covariance approach has now been applied for this as well (Pryor *et al*. 2008, Nemitz *et al*. 2008). Relatively low-cost and low-power fast-response sensors suitable for measuring total particle number using the eddy covariance technique with condensation particle counters or optical particle counters are commercially available. Size-resolved aerosol particle fluxes have been measured by eddy covariance with optical particle counter (Katen and Hubbe1985) or electric low-pressure impactor (Schmidt and Klemm 2008, Damay *et al*. 2009), or by REA method (Gaman *et al*. 2004, Grönholm *et al*. 2007).

Recently, long-term observations of particle concentrations and their formation and growth rates have been performed all around Europe (Manninen *et al*. 2010). Long-term eddy flux measurements of particle numbers are feasible but have not been implemented in a regional or global network. Eddy covariance methods for quantifying size resolved chemical composition, particle fluxes and the chemical composition of particle fluxes would be particularly valuable for improving Earth system understanding but the available approaches require instrumentation that are currently suitable only for the flagship sites described below.

Other constituents

There are other constituents exchanged between terrestrial ecosystems and the atmosphere that have significant roles in the Earth system. These include carbon monoxide (CO), sulfur dioxide (SO_2), reduced sulfur gases, halogens, and particles containing elements such as phosphorus that can be a limiting nutrient in some ecosystems. Fast-response sensors suitable for eddy flux measurements have been developed for some of these, and could be developed for others, but their relatively high cost and operational constraints limit long-term flux measurements. Additional studies are required to determine if a long-term observational network of any of these land–atmosphere processes would be beneficial.

Flagship level: comprehensive measurements

One of the most exciting aspects of the projects advocated by iLEAPS are the opportunities for scientific interaction among biologists, ecologists, hydrologists, micrometeorologists, atmospheric chemists and physicists, and other scientists. Previously, multi-disciplinary collaboration was often limited to studies led by scientists of one discipline with participants from other disciplines in a relatively minor supporting role. Future advances in understanding land ecosystem–atmosphere interactions will likely stem from collaborative studies where participants tackle key scientific issues from the point of view of several different disciplines. One of the most effective means of accomplishing this is through the development of flagship sites with a comprehensive suite of long-term multi-disciplinary measurements that can provide supporting information for intensive campaigns focused on a wide range of biological, physical and chemical processes. Among the core measurements for such sites are stable isotope measurements, ecosystem structure and functioning, mass spectrometer flux systems, boundary layer and cloud properties, and instrumentation for characterizing oxidants and particles and their precursors.

Since flagship stations are based on comprehensive, continuous measurements simultaneously observing greenhouse gases, reactive trace gases, and optical, physical and chemical properties of aerosol particles, their hygroscopicity and ability to act as cloud condensation nuclei (CCN), they can provide detailed data on different radiative forcing components. In addition, the data obtained from flagship stations can be utilized for investigating different feedbacks and linkages (Kulmala *et al*. 2004b). In order to connect observations to the regional scales where feedbacks can occur, these sites should be linked to networks, e.g. SpecNet (http://specnet.info/) and CEOS/CalVal (http://calvalportal.ceos.org) for up-scaling ground observations through satellite remote sensing.

An example of a flagship station is the SMEAR II station (Station for Measuring Forest Ecosystem–Atmosphere Relations) in southern

Finland (Kulmala *et al*. 2001, Hari and Kulmala 2005). This station has operated continuously since 1996, and it continues to provide comprehensive data sets in the fields of atmospheric chemistry and physics, soil chemistry, and forest ecology, all produced with an inter- and multidisciplinary approach. The power of long-term continuous measurements has been shown, for instance, in the study comparing new particle formation over solar cycles with cosmic-ray-induced ionization (Kulmala *et al*. 2010). The observation program at the SMEAR II station includes air ions (their mobility and composition), and composition and fluxes of VOC using time-of-flight mass spectrometers (Ehn *et al*. 2010).

Detailed measurements of ecosystem structure and functioning are another necessary component of a long-term land ecosystem–atmosphere measurement site. This includes information of variables such as the leaf area index (LAI), above- and below-ground biomass, plant species composition, and stable isotopes. Repeated airborne remote sensing using LIDAR and imaging spectrometers are necessary to provide a time series of detailed three-dimensional distribution of these variables across a site. Stable-isotope techniques can improve our understanding of the sources and sinks of CO_2, water vapor, reactive trace gases, and particles including the partitioning of fluxes into individual components (transpiration and evaporation components of water vapor fluxes and photosynthesis and respiration components of CO_2 fluxes). Ecosystem manipulation studies (controlled drought, warming, CO_2 enrichment) are an important activity for comprehensive land ecosystem–atmosphere research sites.

Conclusions

The community investigating land–atmosphere interactions faces a daunting number of complex processes controlling transport and transformation at the land–atmosphere interface as well as an enormous diversity among the Earth's ecosystems. Quantifying these processes with the precision necessary for parameterizing and evaluating Earth system models requires intensive campaigns focused on specific processes as well as long-term observation networks providing continuous and high-frequency time series of fluxes and driving variables. The ecological, hydrological and atmospheric communities have separately developed networks of ecological field stations, instrumented hydrological watersheds and atmospheric monitoring networks. A major step towards integrating these efforts has been accomplished by the FLUXNET global network of eddy covariance tower sites measuring fluxes of carbon dioxide, water vapor and energy between land ecosystems and the atmosphere. FLUXNET has established an initial framework for a global land ecosystem–atmosphere observational network to allow integration of data across sites and time and by designing data systems that facilitate the exchange of scientific information.

A continuation of these advances is necessary to obtain a global observational network that can transform our understanding of land ecosystem–atmosphere interactions and feedbacks that will improve the ability of Earth system models to address global environmental problems. This should include the following activities:

1. Stable long-term funding should be secured in order to continue the established FLUXNET activities.
2. At a subset of FLUXNET sites, measurements should be extended to include fluxes of particles, ozone and biogenic volatile organic compounds (BVOC).
3. Regional networks, such as ICOS and NEON, that include long-term flux measurements of methane, nitrous oxide, and NO_y, should be extended to other regions.
4. "Flagship" level sites representing the major global biomes should be maintained or established with a comprehensive suite of long-term multi-disciplinary measurements providing sufficient information for investigating the complex linkages between biological, physical and chemical processes.
5. A strategy should be determined for locating sites in representative locations and for developing an optimal balance for distributing resources among basic, advanced and flagship sites.

The support of surface, boundary layer, and satellite Earth observations and integration with Earth system models is essential for obtaining new and reliable knowledge for scientists and policy makers to the benefit of society. This requires increased collaboration, development and advancement of the land ecosystem–atmosphere networks that are vital to monitoring, understanding and predicting the earth system.

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