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Perspective

Improving Estimates of Sulfur, Nitrogen, and Ozone Total Deposition through Multi-Model and Measurement-Model Fusion Approaches

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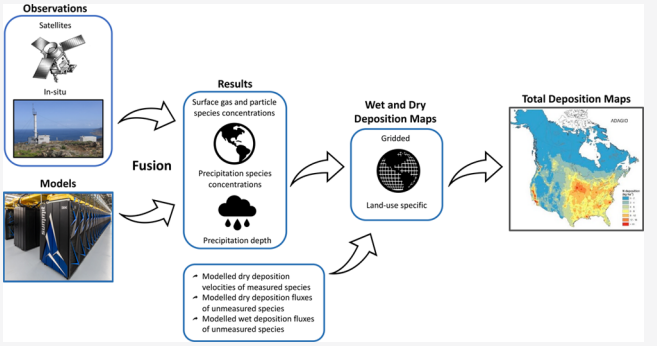
ABSTRACT: Earth system and environmental impact studies need high quality and up-to-date estimates of atmospheric deposition. This study demonstrates the methodological benefits of multimodel ensemble and measurement-model fusion mapping approaches for atmospheric deposition focusing on 2010, a year for which several studies were conducted. Global model-only deposition assessment can be further improved by integrating new model-measurement techniques, including expanded capabilities of satellite observations of atmospheric composition. We identify research and implementation priorities for timely estimates of deposition globally as implemented by the World Meteorological Organization.



KEYWORDS: *atmospheric deposition, multimodel ensemble, model-measurement fusion, earth-system modeling*

INTRODUCTION

The Earth's atmosphere contains a range of trace constituents with negative impacts upon human health, buildings, aquatic and terrestrial ecosystems, and agricultural production. Deposition is the primary mechanism by which most pollutants are removed from the atmosphere, and occurs as wet deposition, in which pollutants are dissolved or entrained within precipitation, or dry deposition on vegetation, soil, water, ice, and structures. Deposition processes are essential in the Earth System to keep our atmosphere clean and healthy, but excessive deposition of reactive nitrogen (N_r), sulfur (S), ozone (O_3), and other atmospheric pollutants can have significant negative impacts.

Driven largely by anthropogenic emissions, high N and S deposition has been observed to negatively affect ecosystem (plant and animal species) diversity[1−3] and contributes to acidification[4−7] and eutrophication[8,9] of precipitation, soil, and water. Reactive N deposition contributes to climate change by altering the interconnected biogeochemical cycling of N and carbon, which coregulates the oceanic and terrestrial uptake of carbon.[10] It also contributes to secondary emission of nitrous oxide. Acid deposition related to S and N can also corrode and damage infrastructure materials, leading to costly maintenance and repairs.[11] O_3 deposition to vegetation seriously affects the sustainable provision of food, feed, fibers, and energy globally and is estimated to cause $14−26 billion USD annually in economic damage to agriculture alone.[12] Despite the importance of atmospheric deposition, many countries lack authoritative information to account for atmospheric deposition in their planning and policy-making decisions.

Maps of atmospheric deposition of pollutants are critical in estimating the environmental impacts of pollution on ecosystems at risk, and enhancing the quality and quantity of crop production. Current global scale maps of atmospheric deposition are mainly based on chemical transport model (CTM) simulations,[13−19] due to the lack of detailed observational evidence in many regions. CTM capabilities continued to improve in the last decades with better process knowledge and enhanced computational resources enabling simulations with higher accuracy and spatial resolution. However, there remain large uncertainties in CTM simulations due to incomplete knowledge of emissions and other sources of uncertainty.[20,21] Maps based on measurements alone are incomplete due to sparse or nonexistent deposition observations in large areas of the world, such as South America, Africa,

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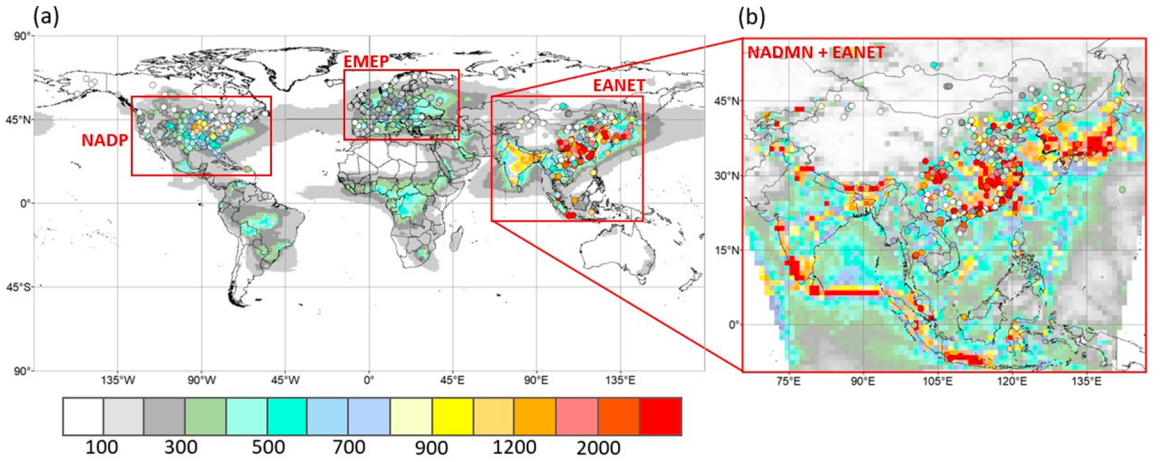

Figure 1. Modeled wet deposition of TNO_3 ($HNO_3 + NO_3^-$) (contours) from HTAP II MMM in 2010 compared with observations (circles) from (a) NADP sites in U.S., EMEP sites in Europe and EANET sites in East Asia (b) from MICS-Asia MMM compared with observations (circles) from NADMN sites in China and EANET sites in East Asia. The values are annual accumulated deposition fluxes in mg (N) m^{-2} yr^{-1}.

Asia, Australia, the polar regions, and oceans.[22] To overcome these deficiencies, new methods have been developed to create more accurate deposition maps through measurement-model fusion (MMF) approaches that merge best-available measurement data and CTM results, and they have been applied successfully at national to continental scale. These methods have not yet been applied on a global scale. Together with new opportunities from satellite observations, the scientific community is now positioned to merge measurement data with model outputs to produce high-quality estimates of atmospheric pollutant concentration and their fluxes at national to global scales. These deposition products will allow government agencies and nongovernment user communities to assess the impacts of deposition of atmospheric pollutants on terrestrial and marine ecosystems, agricultural lands, and climate change, using data of known quality.

This paper presents the necessary steps toward the goal of provision of high quality depostion maps and the intended role of Global Total Atmospheric Deposition (MMF-GTAD) project coordinated by the World Meteorological Organization's Global Atmosphere Watch Programme (WMO GAW) in providing operational stake-holder oriented services.

CURRENT STATE OF ESTIMATING REGIONAL-GLOBAL DEPOSITION

Current global assessments of deposition are almost entirely based on CTMs. CTMs utilize emission inventories and physical, chemical, and meteorological process models to simulate the transformation, transport, and deposition of atmospheric pollutants at regional and global scales. CTMs of varying complexity have been developed utilizing various combinations of process algorithms and parametrization for specific applications. An individual CTM may excel at simulating certain variables but not others. To better understand the capabilities and limitation of CTMs, multimodel studies have been performed. In this paper we utilize results from four recent studies; that is, HTAP,[13] AQMEII,[23] MICS,[19] and ACCMIP.[14]

These studies, described in detail in the supplement, show that gradual progress has been made in capabilities to predict deposition, with important remaining uncertainties related to emission inputs and parametrization of processes used in models. For instance, uncertainty in emissions range between 8−14% for SO_2 and a factor of 2 for road vehicle NO_x; while for volatile organics it can be even higher.[24] Model uncertainties are also related to the accuracy of modeled meteorological parameters such as winds and precipitation, the deposition and scavenging parametrizations, and the accuracy with which mass closure is achieved. Particle deposition velocity parametrization results in roughly between 10 and 30% differences compared to observations and is found to be most sensitive to the aerosol size and the land cover data set used.[25] Differences of Nr ($N_r = NH_x + NO_y$) including NH_x (ammonia and ammonium) and reactive nitrogen compounds ($NO_y = NO + NO_2 + HNO_3 + HONO$ + organic nitrates + particulate nitrates) gas deposition in CTMs and inferential models, can reach a factor of 2 to 3, thereby exceeding the spatial variability of observed fluxes. Nonstomatal deposition pathways are an important factor in these differences.[26]

In addition, incompleteness of chemical schemes used in models can introduce uncertainties in the deposition fluxes. For instance, omission of organic nitrogen in chemistry schemes can lead to an underestimate by about 20−30% of the total nitrogen deposition.[17,18,27]

Methods to Develop a Multimodel Ensemble. Ensembles of deposition estimates based on mean/median of multiple models, are generally more accurate than any individual model.[28,33−36] Deposition results from multimodel community efforts (see Supporting Information), are the backbone of MMF-GTAD maps, in particular in data poor regions. Nevertheless, improvements in the quality control, such as mass balance requirements, statistical evaluation protocols and skill scores, removal of unintended outliers, and appropriate weighting of models can further improve the accuracy of ensembles, as described in the supplement.

Figure 1 shows examples of multimodel mean (MMM) global maps of total wet NO_3^- deposition for 2010, using results from two model intercomparisons, and several regional deposition networks.

The MMM maps capture important features in observed deposition and accurately identify the global hot spots of deposition (Figure 1a). The regional total wet NO_3^- MMM Mics-Asia deposition estimate in Figure 1(b) shows a greater correspondence with a new set of observations, National Acid Deposition Monitoring Network (NADMN)[29] in China. It is possible today to build global maps of atmospheric deposition

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using MMM from global models and stitching in higher resolution MMM estimates from regional models. We note, however, that a scarcity of observations prevents rigorous evaluation of results in many other world regions.

MMF of Models and Surface Observations. Significant biases in deposition remain even when using MMM ensembles. While efforts continue toward reducing these biases, approaches that use available observations to adjust model output, rather than just evaluate it, can provide improved estimates of deposition. It requires the development of benchmark data sets included in MMF to improve the estimate of the final MMF deposition.

In the U.S.,[30] Canada,[31] and Sweden,[32] regional-scale MMF deposition mapping has been developed, with a conceptual methodology shown in Figure 2. MMF involves bias correcting

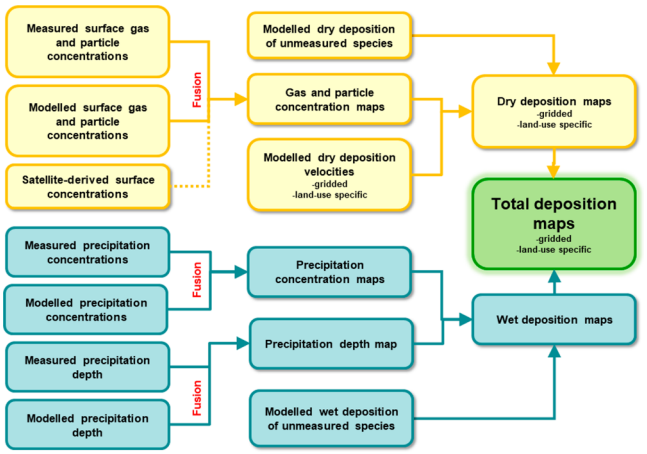

Figure 2. A general methodology for producing global total deposition maps.

CTM-computed variables using observational data sets to create fused deposition maps. To calculate wet deposition, the concentrations of ions in precipitation from model and observations are fused to create maps of concentrations. Similarly, modeled and measured precipitation depths are fused. These two fused products are then combined to calculate wet deposition fluxes for observed species. Wet deposition of additional chemicals (e.g., sparsely measured species such as nitrate radicals (NO_3), nitric anhydride (N_2O_5), nitrous acid (HONO), and organic nitrogen) is derived solely from the CTM and is added to the final estimate of MMF wet deposition. Dry deposition is calculated by merging simulated and observed species surface concentrations, and multiplying with model-only dry deposition velocities, as observations of dry deposition velocities are not routinely available. While not included in current MMF approaches above, the generalized method in Figure 2 recognizes the potential for use of satellite-derived concentrations for wet and dry deposition. Depending on the CTM used, deposition velocities may be available as grid-averaged or land-cover specific values,[33] relevant for estimating deposition to sensitive ecosystems.

Figure 3 shows example outputs of the MMF approaches used in the U.S., Canada, and Sweden, which vary in the fusion methods employed, the observational data included and the underlying CTMs. It is notable that the domains of the Canadian Atmospheric Deposition Analysis Generated from optimal Interpolation from Observations (ADAGIO)[34] product and the U.S. National Atmospheric Deposition Program Total Deposition Science Committee (TDep)[35] product overlap, and we are able to see that, despite differences in methodology, the approaches provide very similar results. These examples illustrate the state-of-science of MMF-GTAD mapping on a regional basis and represent the jumping-off point for developing an operational global deposition mapping system through the WMO's MMF-GTAD Initiative.

Uncertainties in MMF Products Depend on Quality of Observations. The availability, representativity and quality of observations, and the models used and fusion methodology all have uncertainties that propagate into the final MMF maps. The inaccuracies associated with observations are incrementally reduced by the use of well-defined observational protocols and state-of-the art instrumentation, harmonized between regional and global networks. This is especially the case for chemical speciation measurements in precipitation.[15,36] Near-surface atmospheric ozone observations are comparable across the world, following the observational, instrument, and calibration guidelines from WMO (2013).[37] For sulfur and nitrogen measurements in air, there are larger variations with different methods like continuous monitors, filter pack samplers, and passive samplers. Further, there are inherent

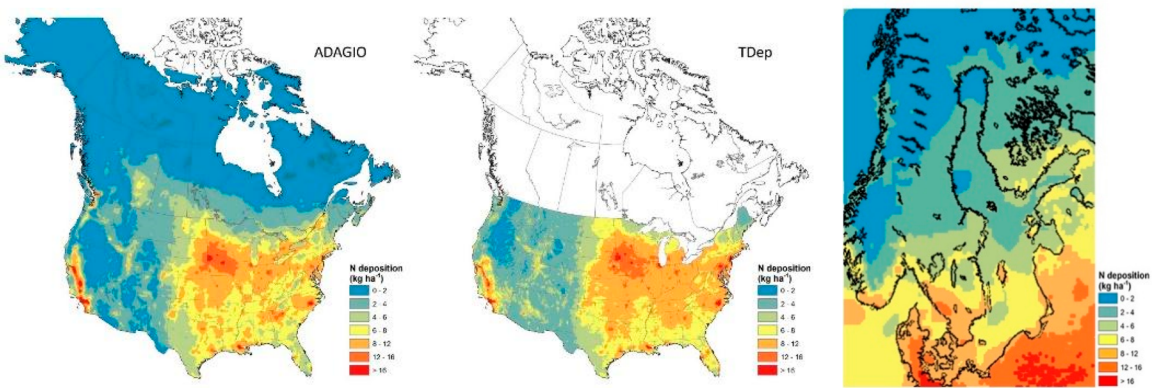

Figure 3. Current MMF maps of total (wet+dry) annual atmospheric deposition. (Left) 2010 nitrogen total deposition produced by Environment and Climate Change Canada, (middle) 2010 nitrogen total deposition produced by the United States Environmental Protection Agency[31] and (right) 2010 nitrogen total deposition in Sweden and adjacent countries produced by the Swedish Meteorological and Hydrological Institute.[32] Unit: kg N ha^{-1} yr^{-1}.

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biases in some of the methods, especially in the gas/particle separation of nitrogen species,[38,39] causing an added uncertainty in using these data.

In general, wet deposition fluxes using chemical concentration measurements in rainwater and precipitation depth are more accurate than those of dry deposition fluxes.[40] Nevertheless, the spatial variability in the precipitation amount can be very large, especially in hilly landscapes and along coastlines, and taking advantage of the much larger spatial coverage of the (national) meteorological service networks, as opposed to only the regional air pollution networks improve the representation of the spatial variability of wet deposition in MMF routines.

Currently, dry deposition fluxes are observationally derived directly from eddy covariance measurements[25,41] or from atmospheric concentrations combined with model-calculated deposition velocities[42] for the location and time period of the observations. Despite the very high accuracy of eddy covariance measurements for many chemical species, such difficult measurements remain scarce both geographically and temporally and are therefore more useful for mechanism/parametrization development than for routine calibration. MMF dry deposition flux maps based on fused observed and modeled atmospheric concentrations combined with model-derived deposition velocities remain therefore currently the only feasible method to provide complete coverage in space and time (Figure 2). We note that there remains some unexplored potential to use satellite based estimates of land cover[43] and vegetation activity (e.g., the essential climate variable faPAR, Fraction of Absorbed Photosynthetically Active Radiation) to improve estimates of dry deposition.

Improving Global MMF Using Satellite Observations. Regional MMF analysis of wet deposition has been limited to deposition over land. There are practically no in situ observations of precipitation depth over the oceans due to logistic constraints; this gap is filled by satellite observations (like the Global Precipitation Climatology Center data reanalysis, Schneider et al. 2016[44]), which can provide information on the precipitated water, but not on its ionic composition (see next section). Also over land satellite observations of precipitation depths can provide useful information. Over Southeast Asia, models in MICS-Asia phase III had difficulty capturing wet deposition during dry and wet seasons. By adjusting the simulated precipitation using the Tropical Rainfall Measuring Mission (TRMM) measurements, both the simulated wet deposition amount and its seasonal pattern were improved.[44] The advantage of using climatological or satellite precipitation data as part of the wet deposition estimation methodology should be weighed against the impact of decoupling those fields from the ion concentrations in that precipitation. To address how to best use precipitation data, a pilot study using one year of global CTM output (or an ensemble) and a global precipitation data set is needed before an operational system is implemented.

The representativeness of measurement sites is an important factor to consider for MMF, and it may vary between components depending on local influences such as traffic, farming, and industry. Further, there are regions of the world with very limited information from in situ observations where it is difficult to apply the MMF method.

In data-sparse areas satellite observations, in particular those with high spatial resolution products, like TROPOMI[45] for NO_2 and CrIS[46] for NH_3 can be converted into spatially continuous satellite-derived estimates of surface concentrations, which may be combined with inferential dry deposition models to calculate dry deposition fluxes. Satellite-based surface concentration estimates have been derived using geophysical modeling approaches,[47−53] or using empirical/statistical relationships with in situ observations,[54] which introduces some error caused by interpretation between them that will impact vertical distribution, for instance. Deposition estimates of constituents not directly observed by satellite have also been estimated from satellite observations using statistical relationships between the observed and modeled[55] constituents and several processes of interest, for example, wet NO_3^- or NH_4^+ deposition.[56,57] Furthermore, the next expected generation geostationary satellites may offer higher accuracy and lower detection limits, which can expand usable information to low-intermediate polluted regions.

DATA ASSIMILATION AND INVERSE MODELING

There are significant advancements in chemical data assimilation as a means to improve model predictions of atmospheric composition.[58] Better constraints on near surface ozone may, for example, help reduce the uncertainty in deposition estimates. However, we note that biases in surface concentrations may be partly caused by biased deposition velocities. Therefore, a coupling between models and observations by data assimilation could misrepresent the model mass balance between the input emissions, the atmospheric content, and the deposition flux.[59] A review of some relevant methods and current limitations in deriving surface concentration and deposition estimates from satellite observations can be found in Liu et al. 2020b.[60] Direct assimilation of satellite observations of atmospheric composition in atmospheric chemical forecast models[61] also has the potential to provide improvements into near-real time deposition predictions, but this technique has thus far not been explored for deposition.

Data assimilation can be extended to constrain surface fluxes (also known as inverse modeling). In regions where fluxes are dominated by emissions, such approaches can be used to improve emission inventories. These updated surface emissions may then provide more consistent constraints on deposition calculated by chemical transport models.[62] The latter approach has the benefit of providing improvements in predictions of chemical species that are not directly observed by satellite but which are related by chemistry (e.g., improving NO_x emissions estimates to simultaneously improve modeled deposition estimates of NO_2, HNO_3, peroxyacyl nitrates, particulate NO_3^-, and even O_3).

Emerging machine learning techniques commonly employed in other disciplines, can also be applied for the mapping of deposition fluxes provided that sufficient observations are available for training and validation.[63]

Model mass balance is also susceptible to the coupling between models of various geographic scales. Significant subgrid variability in land-cover and land-use as well as in meteorology/precipitation also exists to make a level of uncertainty of mass balance[64] that challenges the coupling of in situ (point) observations with regional and global models. In all cases, it requires validation data sets not used for generation of MMF products. Finally, further advancement of Earth System Models that couple biosphere/atmosphere processes offer the hope of better estimates of deposition processes.

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ROADMAP FOR FUTURE WORK

Better management of the earth-land-ocean-atmosphere system in a sustainable development context requires improved and regularly updated information on atmospheric deposition. The MMF of surface observations and models is a promising step forward to provide deposition estimates in regions with sufficient model and observational capacity.[48,49,52,53] To produce global measurement-model fusion products for deposition, several components of the methodology need to be explored and developed and cross-validated. Some fusion techniques may be more appropriate than others for regions where data are sparse, or to account for the lower spatial resolution of global models. For instance, due to in situ measurement data sparsity, the inclusion of broad-scale satellite observations will be a major step forward for the future global mapping.

Better use of the combined information in models and observations through MMF is a promising means by which this can be accomplished. Increasing the accuracy and reducing the uncertainty in global estimates of atmospheric deposition can be achieved by (i) expanding existing MMF approaches into more regions where observations allow to do so, (ii) using multimodel ensemble estimates in regions where MMF cannot be carried out due to lack of observations, and (iii) expanding MMF approaches to include satellite observations and the improved deposition models, as well as more formal data assimilation and inverse modeling. The new WMO GAW's MMF-GTAD Initiative aims to develop best-possible global and regional atmospheric deposition maps of known quality following the approaches described in this paper. The project will implement a comprehensive operational system incorporating the gathering of additional measurement data and modeling output, expanded application of MMF techniques, quantification of uncertainty, and development and delivery of stakeholder-driven products and services. International and national stakeholders that raise awareness of the importance of deposition include governmental and nongovernmental organizations targeting biodiversity and ecosystems, agriculture, human health, and climate. In the Supporting Information (Figure S1), a short summary of the MMF-GTAD implementation plan is presented.

To what extent and in which way singular or multiple data sets can be merged into superior global deposition products will depend on the specific use, for example, the data set, error characteristics of models and observations. To operationally deliver state-of-the-art deposition maps and their associated products into the future, MMF components and methodologies must be incrementally developed and improved, as guided by both research and input from users. Such developments and improvements with time will include the following:

1. Extending observational coverage to under-sampled regions of the world (Africa, Australia, India, South America, and oceans).
2. Developing and adopting new measurement capabilities such as additional/improved satellite observations of atmospheric composition, including utilization of next-generation geo-stationary satellite data.
3. Advancing CTM predictive capabilities (e.g., higher spatial resolution, improved precipitation predictions, updated emissions and higher resolution land use data with improved characterization of vegetation parameters such as stomatal resistance measurements) to improve the final MMF deposition estimates.
4. Developing benchmark data sets for inclusion in MMF and collecting independent observations for evaluation of model accuracy. Data exchange from regional and global networks should follow the findability, accessibility, interoperability, and reuse of digital assets (FAIR) principles.
5. Building a platform for results sharing and updates, including both observation and model results and documentation (i.e., data access, format, and data sharing policy).
6. Expanding products to meet user needs, for example, providing long-term trend of deposition estimates of uncertainties in MMF deposition maps when users utilize the generated maps for their applications and decision making.
7. Reaching out to environmental impact and ESM communities, raising awareness of policy organization for the importance of deposition in SDGs.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c05929.

Additional information as noted in the text including Figure S1 (PDF)

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Author Contributions

J.S.F, G.R.C., and F.D. conceived the research. All authors wrote, reviewed, and edited the paper.

Notes

The authors declare no competing financial interest.

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Joshua S. Fu is John D. Tickle Professor and James G. Gibson Professor in the Department of Civil and Environmental Engineering at the University of Tennessee and Joint Appointment Professor in Computational Earth Sciences Group in Computational Sciences and Engineering Division at Oak Ridge National Laboratory. He has served Vice-Chair and member of the WMO Model Measurement Fusion Initiative, contributed as a coauthor of the Final Report of the Hemispheric Transport of Air Pollution for the UN ECE Hemispheric Transport of Air Pollution, and reviewing committee member for air quality status in East Asia for the EANET located in Japan. He also was a coauthor of Technical Report, Climate Change and Infrastructure, Urban System, and Vulnerability, to the U.S. Department of Energy in the support of the National Climate Assessment in 2012. He has received numerous awards from national and international associations and a Fellow of the AAAS and of the A&WMA and Board-Certified Environmental Engineering Member from AAEES and other distinguished and endowed professorship awards. Joshua has published more than 180 refereed journal articles and 110 peer-reviewed conference proceedings. Joshua obtained his PhD from North Carolina State University, MS from UCLA, and BS from Taiwan's National Cheng Kung University.

Gregory R. Carmichael is the Karl Kammermeyer professor of chemical and biochemical engineering at the University of Iowa, and codirector of the Center for Global and Regional Environmental Research. His research is focused on air pollution and climate change, where he uses comprehensive computer models and big data to simulate the interactions of air pollutants with weather and climate, and to estimate resulting environmental impacts. His models are also used to evaluate effectiveness of various air pollution and climate change mitigation strategies. He has authored or coauthored over 380 scientific publications. He has received numerous awards, including the American Institute of Chemical Engineer's Lawrence K. Cecil Award for outstanding chemical engineering contribution and achievement in the preservation or improvement of the environment. He is a Fellow of the American Institute of Chemical Engineers and of the American Geophysical Union. He serves on numerous international advisory boards including serving as chair of the UN World Meteorological Organization's Environmental Pollution and Atmospheric Chemistry Scientific Steering Committee.

Frank Dentener has worked for 30 years in the field of climate change, agriculture, and atmospheric pollution. Frank is a senior expert and group leader at the European Commission's Joint Research Centre. He completed a Ph.D. in Physics with Nobel prize-laureate Paul Crutzen at Utrecht University. He is a member of the scientific advisory board of the JPI FACCE, and cochair of WMO's Scientific Advisory Group on near-real-time applications, member of the WMO Model Measurement Fusion initiative, and has served on a variety of international commissions. He has (co-) authored more than 200

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peer-reviewed publications and four IPCC reports, Hirsch factor 94, and is a Clarivate highly cited scientist since 2015. Frank held assistant professorships at Wageningen and Utrecht University, The Netherlands, PrivatDozent at the ETH in Switzerland, and is currently affiliated with the Politechnical University of Torino. He supervised and examined ca. 10 Ph.D. and Habilitation candidates. Frank is an expert in atmospheric chemistry−land interactions, including ozone impacts on agriculture, agricultural production systems, climate change, and mitigation and adaptation.

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