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Citation: Cohen, Jason Blake, Ronald G. Prinn, and Chien Wang. "The Impact of Detailed Urban-scale Processing on the Composition, Distribution, and Radiative Forcing of Anthropogenic Aerosols." *Geophysical Research Letters* 38.10 (2011). ©2011 American Geophysical Union

As Published: <http://dx.doi.org/10.1029/2011gl047417>

Publisher: American Geophysical Union (AGU)

Persistent URL: <http://hdl.handle.net/1721.1/74005>

Version: Final published version: final published article, as it appeared in a journal, conference proceedings, or other formally published context

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The impact of detailed urban-scale processing on the composition, distribution, and radiative forcing of anthropogenic aerosols

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Received 11 March 2011; revised 18 April 2011; accepted 19 April 2011; published 26 May 2011.

[1] Detailed urban-scale processing has not been included in global 3D chemical transport models due to its large computational demands. Here we present a metamodel for including this processing, and compare it with the use of the traditional approach of dilution of emissions into large grid boxes. This metamodel is used in a global 3D model to simulate the effects of cities around the world on aerosol chemistry, physics, and radiative effects at the global scale. We show that the biases caused by ignoring urban processing on the global values of total aerosol surface concentration, the total aerosol column abundance, the aerosol optical depth (AOD), the absorbing aerosol optical depth (AAOD), and the top of the atmosphere radiative forcing (TOA) respectively are $+26 \pm 3\%$, $+51 \pm 12\%$, $+42 \pm 8^{10\%}$, $+8 \pm 16\%$, and $-0.27 \pm 0.10^{0.14} \text{ W/m}^2$. These results show that failure to consider urban scale processing leads to significantly more negative aerosol radiative forcing compared to when detailed urban scale processing is considered. **Citation:** Cohen, J. B., R. G. Prinn, and C. Wang (2011), The impact of detailed urban-scale processing on the composition, distribution, and radiative forcing of anthropogenic aerosols, *Geophys. Res. Lett.*, 38, L10808, doi:10.1029/2011GL047417.

1. Introduction

[2] Urban regions account for a large and increasing fraction of the Earth's total population and of anthropogenic aerosol emissions. However, modeling the effects of urban areas on the processing and export of anthropogenic aerosol emissions requires a detailed analysis dependent on an urban areas' geographic and meteorological properties, the amount and distribution of its emissions, and strongly nonlinear chemical and physical processing. To analyze the impact of urban aerosols in a global model, processing on spatial and temporal scales much smaller than that of the global model is required. Because of this, explicit processing of emissions at the urban scale has typically been replaced by diluting the emissions evenly over global model grids, which does not capture the real heterogeneity of urban and non-urban regions within these grids (referred to here as a "dilution" approach). Hence, urban areas account for a large amount of the variability and uncertainty in the global atmospheric spatial and temporal distributions of primary and secondary anthropogenic aerosol and gas pollutants.

[3] Two previous attempts to address this issue were made by *Calbó et al.* [1998] and *Mayer et al.* [2000]. They both derived a "parameterization" or "reduced form model" for the urban processing of gas phase chemicals relevant to the climate system. These parameterizations were derived using a simplified chemical mechanism driven by idealized meteorology. While these models were a significant step toward reality, they were limited by a constant spatial and temporal distribution of emissions on the urban scale, the requirement of relatively clean upwind conditions, uniform, non-divergent, and steady meteorology, a lack of rainfall within the urban region, and a restriction on the number of different urban areas they were designed to simulate.

[4] In this paper, we show results from a new model that has been developed to simulate this urban-scale processing [Cohen and Prinn, 2011]. This model is designed to be "two-way" interactive within a global modeling framework; that is it is capable of both being influenced by and influencing the species of interest at the surrounding model scales.

[5] This new urban modeling approach is applied here to calculate the impact of detailed urban processing on the behavior of aerosols and aerosol precursors in a global scale model and the results are compared with those from the "dilution" approximation.

2. Modeling Framework

2.1. Urban Metamodel

[6] The Comprehensive Air Quality Model with extensions (CAMx) was chosen as the parent urban chemistry model for development of the parameterized or reduced-form urban model [see, e.g., *Amiridis et al.*, 2007; *Andreani-Aksoyoglu et al.*, 2008; *Eben et al.*, 2005; *de Foy et al.*, 2007; *Lei et al.*, 2007; *Russell*, 2008; *Zunckel et al.*, 2006]. To solve CAMx, exogenous meteorology fields and emissions are required.

[7] The meteorological conditions were obtained from separate sets of locations from the 1995 OTAG campaign, each covering the size of a single urban area [Vukovich, 1997]. These meteorological conditions were chosen to cover typical ranges for rainfall, cloud cover, and the net mass flux of air integrated across all five boundaries of the urban air shed, permitting the simulation of more extreme conditions likely to be found in generalized urban areas [Cohen and Prinn, 2011].

[8] Since emissions vary by urban area, location, economy, and politics, the underlying inputs to the metamodel, such as emissions are based on underlying Probability Distribution Functions (PDFs). These PDFs were developed such that they are representative of cities grouped into four categories: China, India, Developed Nations, and Developing Nations. These PDFs are used to determine if a proposed region has an emission level too low to be appropriate for processing

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by the metamodel. These are designed to capture the aspects of areas with the greatest number and diversity of urban areas [Cohen and Prinn, 2011].

[9] Embedding CAMx into a global transport and chemistry model to simulate the effects of hundreds of urban areas is not computationally feasible. Therefore a parameterization of the processes contained in CAMx was developed using the probabilistic collocation method, which in turn is based on a polynomial chaos expansion [Tatang et al., 1997]. The full list of species simulated, the PDFs used to produce the full 3rd order set of orthonormal polynomials, as well as the goodness of fit of these polynomials, are described in detail by Cohen and Prinn [2011]. With 4 choices of meteorology and 4 choices of urban type there are 16 total metamodels.

[10] The ability of the metamodel to reproduce the results of the parent CAMx model has been shown to be less than 1 percent for the large majority of cases (and is always less than 10 percent) for all species of interest presented in this paper [Cohen and Prinn, 2011]. Therefore, the results presented here are significant compared to the approximation made by using the metamodel in lieu of the parent model.

2.2. Global Model

[11] To simulate chemistry and transport at the regional to global scales we utilize a 3D general circulation model [Kim et al., 2008] derived from the Community Atmospheric Model version 3.1 [Collins et al., 2006], by addition of state-of-the-art treatments of aerosol physics, chemistry, and dynamics. The model includes seven anthropogenic aerosol types, differentiated by size, chemical composition, and mixing state. This includes three sizes of sulfate (nucleation, Aitken, and accumulation (ACC) modes), external modes of black carbon (BC) and organic carbon (OC), and core-shell modes of black carbon with a sulfate shell (MBS) and organic carbon with a sulfate mode (MOS). The model uses a 2-moment approach, with each mode represented by its mass and a number density. Furthermore, the model requires a minimum concentration of H_2SO_4 to age BC to MBS or to age OC to MOS, therefore, the rate of such aging in the model is dependent on the availability of sulfuric acid gas, which in turn is dependent on their being a high enough concentration, and sufficient oxidation, of SO_2 . The model results have been previously compared with satellite, surface, and aircraft measurements, with good agreement between modeled and observed data seen in most cases [Kim et al., 2008; Wang et al., 2009].

[12] This model was used in its offline version, driven by NCEP (National Center for Environmental Prediction) reanalysis fields at a 6-hourly time resolution [Kalnay et al., 1996] and was run using the finite volume (FV) dynamical core [Rasch et al., 2006], over a 1.9 by 2.5 degrees latitude/longitude grid with 26 vertical layers up to 2.7 hPa.

[13] We use the MIT Emissions Projection and Policy Analysis (EPPA) Model [Paltsev et al., 2005; Sokolov et al., 2009], which includes detailed economics of multiple nations and agricultural sectors, and has been calibrated to agree with relevant reported emissions, to compute all relevant anthropogenic emissions: BC, OC, SO_2 , and CO. Biomass burning emissions of BC and OC are calculated as an annual average emission based on the Global Emission Inventory Activity (GEIA) (<http://www.geiacenter.org>). Emissions of isoprene and monoterpenes, from GEIA, yield a secondary production of 30.7 Tg/year of OC [Kim et al., 2008]. Finally,

annual average natural oceanic emissions of dimethylsulfide (DMS) are provided [Kettle et al., 1999]. Summing all of these sources globally leads to total annual emissions of 14.4 Tg/yr of BC, 61.5 Tg/yr of OC, 132 Tg/yr of SO_2 , 1.58 Pg/yr of CO, and 19.3 Tg/yr of DMS.

2.3. Interaction Between the Urban and Global Models

[14] Since no global model grid box is entirely occupied by urban areas (although some have more than one urban area), the fraction of the emissions processed by the metamodel must be calculated. For this purpose, the emissions in each grid box are weighted based on the urban population in that box which is derived from an underlying $1^\circ \times 1^\circ$ global map of urban locations and populations [Center for International Earth Science Information Network, 2000]. The non-urban emissions in each grid box are handled by the usual “dilution” method.

[15] The choice of the metamodels used in each grid box at each time step is based on the grid box average rainfall and wind speed, and then an appropriate average of the outputs of the chosen meteorological metamodels is used. For the large majority of urban areas and times of day, these values lie between those corresponding to the four meteorologies, in which case a weighted linear average is used between the nearest two (if dry) or three (if raining) neighbors. For those few cases in which the values do not lay between those corresponding to the four meteorologies, the nearest neighbor meteorology is used. The location of the grid box determines which of the four city metamodels is used.

[16] In turn, for each relevant gas and aerosol species, the metamodels compute the concentrations within the urban area, and the net exports from the urban area into the global model. The net exports to the global model grid boxes are spread from the surface up to the 700 mb level, as predicted by the metamodel for each output species under each set of meteorology.

2.4. Coupled Model Runs

[17] The global model was run with and without processing of urban emissions by the urban metamodel and the results compared. The emissions, driving meteorology, urban locations, and all other inputs and configurations were otherwise identical in both cases. The model was run with a 2 year spin up (January 2000 through December 2001) and a 4 year modeling period (January 2002 through December 2005), using NCEP Reanalysis meteorology [Kalnay et al., 1996].

[18] There are 251 urban areas modeled: 91 from China, 36 from India, 50 from Developed Nations (Australia, Canada, the European Union, Japan, Singapore, South Korea, and the United States), and 74 from Developing Nations (Figure S1 of the auxiliary material).¹ Comparisons are made for each output Y by showing the bias incurred by not considering the effects of urban processing, as described by the equation:

$$\text{Bias} = (Y_{\text{Dilute}} - Y_{\text{Urban}})/Y_{\text{Urban}} \quad (1)$$

3. Results

[19] The monthly hemispheric mass burden of all aerosols show the primary effect of including urban processing is to

¹Auxiliary materials are available in the HTML. doi:10.1029/2011GL047417.

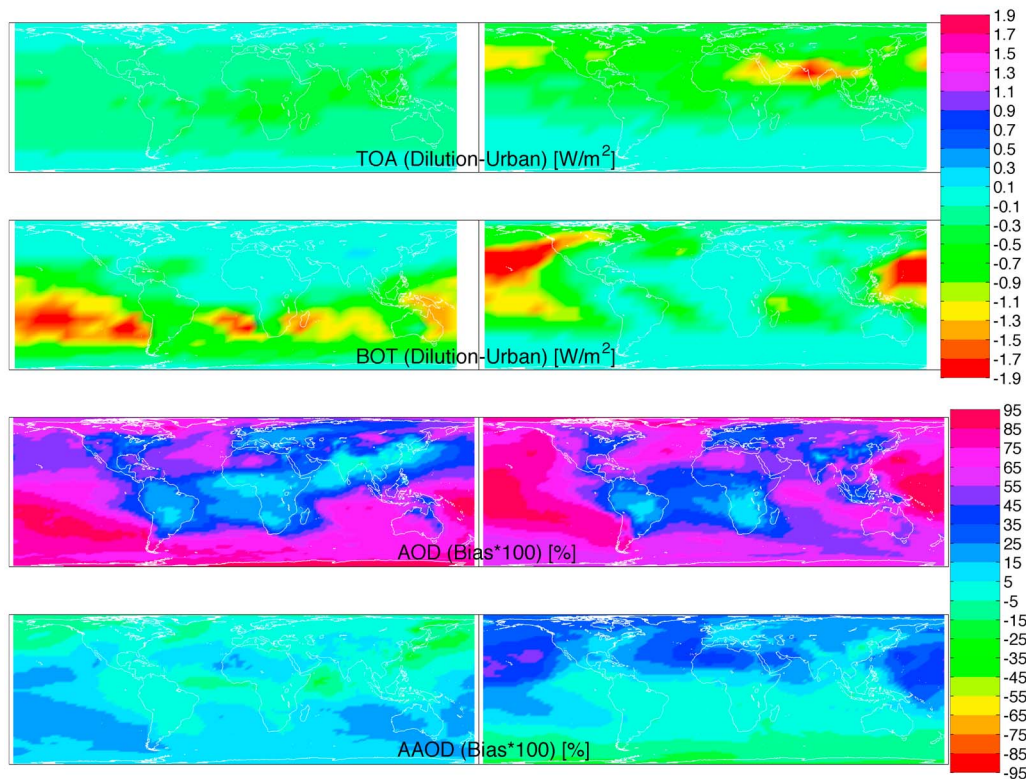


Figure 1. From top to bottom, the absolute bias (Dilution - Urban) due to ignoring urban processing for TOA and BOT, and the fractional bias (see equation (1)) for AOD and AAOD. (left) January 2002 and (right) July 2002.

reduce the total atmospheric mass burden of aerosol. This reduction is larger in the Northern Hemisphere (NH) than the Southern Hemisphere (SH) and is greater in the summer than the winter. Furthermore, urban processing leads to an increase in the primary aerosol modes, BC and OC, and a decrease in the secondary aerosol modes, MBS, MOS, and ACC. In the summer these changes are greater than the winter (Figure S2).

[20] The zonally averaged total aerosol concentrations for January and July of 2002 show two points that support the above overall conclusions. Firstly, there is significant secondary production of aerosol in the free-troposphere during the summer, when the oxidation of SO_2 is greatest. Secondly, a significant amount of extratropical SH total aerosol mass is secondary aerosol transported from the NH during the summer (Figure S3). Due to the dependence of the season on the results, all error bars in this paper refer to this interannual variability.

[21] We have found that when excluding urban processing, the globally averaged surface concentrations of OC and BC are underestimated, and those of MBS, MOS, and ACC are overestimated, respectively, as compared to the results when we include urban processing. The monthly average surface concentration biases for BC, OC, MBS, MOS, and ACC respectively are $-1.1 \pm 3.7\%$, $-4.5 \pm 1.4\%$, $+90 \pm 21\%$, $+71 \pm 12\%$, and $+59 \pm 7\%$. Analyzing only those model grid boxes which contain urban areas, when excluding urban processing, all species are overestimated due to more efficient urban removal. The biases for BC, OC, MBS, MOS, and ACC respectively are computed to be $+27 \pm 2\%$, $+12 \pm 3\%$, $+130 \pm 28\%$, $+110 \pm 22\%$, and $+79 \pm 18\%$ (Figure S4). The

less positive bias for OC compared with BC in urban areas is due to secondary OC production.

[22] Furthermore, when excluding urban processing, we have found that the globally averaged total column burdens for OC and BC, are underestimated, while those for MBS, MOS, and ACC are overestimated, respectively compared to when urban processing is included. The monthly average global total column burdens biases for BC, OC, MBS, MOS, and ACC respectively are $-32 \pm 6\%$, $-32 \pm 3\%$, $+97 \pm 28\%$, $+64 \pm 13\%$, and $+85 \pm 5\%$. Analyzing the subset of model grid boxes that contain urban areas, when we exclude urban processing, the column values for BC and OC are less underestimated relative to all grid boxes, with their respective biases being $-22 \pm 10\%$ and $-30 \pm 5\%$. These results show that there is a second effect of urban processing, occurring over longer temporal and spatial scales, which compensates for the initial short-range higher efficiency of urban removal of primary aerosols. The impact of these long-range effects are further demonstrated by the large biases in the column loadings of MBS and MOS (Figure S5).

[23] The AOD is related to the column burden, and since excluding urban processing leads to reductions in ACC, MBS, and MOS that are greater than the increases in BC and OC, the global average AOD is overestimated compared to when we include urban processing. The overall AOD bias is computed to be $+42 \pm 8\%$, with the contribution from each species bias being $-28 \pm 8\%$ for BC, $-28 \pm 4\%$ for OC, $+85 \pm 21\%$ for MBS, and $+80 \pm 6\%$ for scattering aerosols (MOS + ACC). The larger values in the NH Summer are both due to the larger column loading biases and the more intense NH radiative flux (see Figure 1).

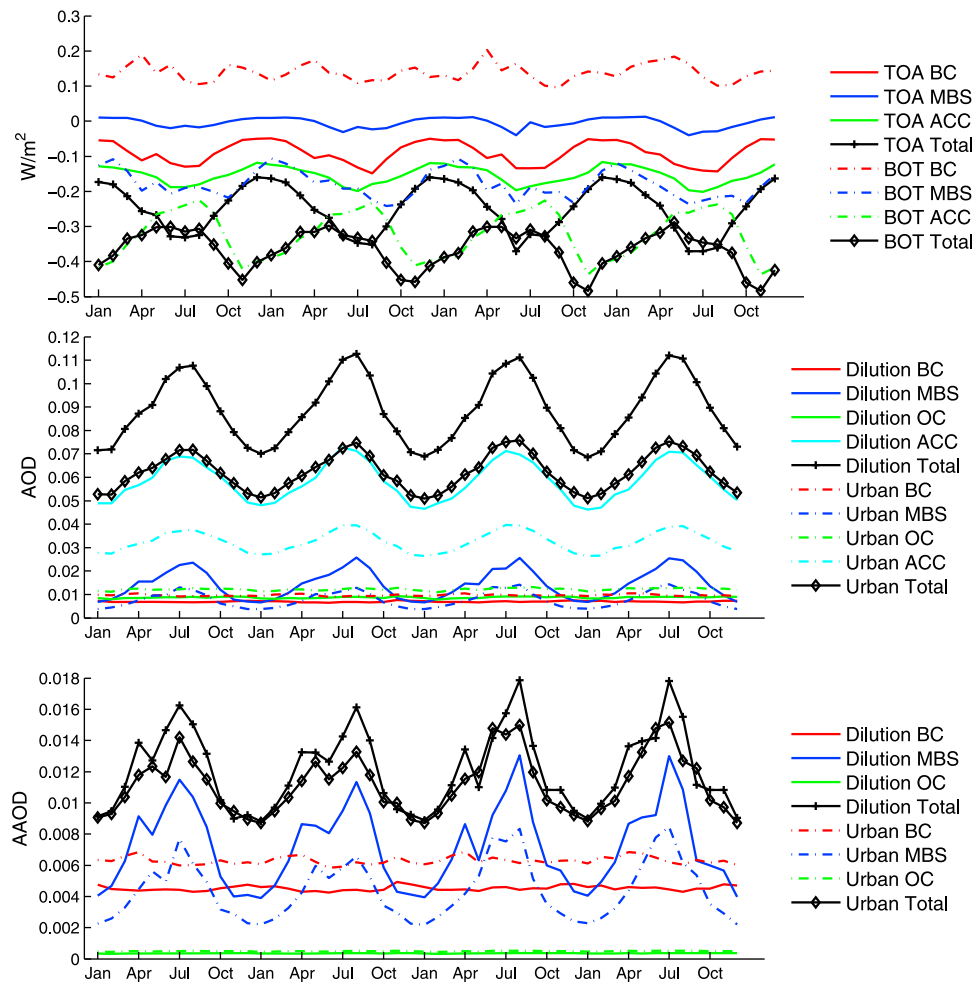


Figure 2. (top) Monthly average (January 2002 through December 2005) ACC, BC, and MBS contributions to the TOA and BOT biases (Dilution - Urban) in $[\text{W}/\text{m}^2]$. (middle) ACC, BC, MBS, and OC contributions to the global average AOD, for both the Dilution and Urban cases. (bottom) ACC, BC, and MBS contributions to the AAOD, again for both the Dilution and Urban cases.

[24] The AAOD is $\text{AOD} \cdot (1 - \omega)$ (where ω is the single scattering albedo). The total global average AAOD bias, and the biases for the individual components BC, OC, and MOS respectively are $+8 \pm 18\%$, $-28 \pm 8\%$, $-28 \pm 4\%$, and $+70 \pm 36\%$. The larger summertime bias is due to the relative summertime increase in the AOD of MBS to BC, as shown in Figure 2.

[25] The difference in global average radiative forcing between the dilution and urban cases is computed at the TOA, the surface (BOT), and for the amount absorbed by the atmosphere (ABS), using a delta-Eddington approximation in connection with the size distribution and mixing state of the aerosols. A lookup table based on the results of Mie theory is used to determine the values of k , w , and g , as explained in more detail by Kim *et al.* [2008]. The monthly average difference between the dilution and urban cases, for TOA, BOT, and ABS, respectively are: $-0.27 \pm 0.10 \text{ W}/\text{m}^2$, $-0.43 \pm 0.08 \text{ W}/\text{m}^2$, and $+0.16 \pm 0.18 \text{ W}/\text{m}^2$. The individual contributions most important to the TOA bias are ACC and BC, which respectively contribute $-0.15 \pm 0.04 \text{ W}/\text{m}^2$ and $-0.09 \pm 0.06 \text{ W}/\text{m}^2$. The individual contributions most important to the BOT bias are ACC, MBS, BC, and MOS, which respectively contribute $-0.32 \pm 0.11 \text{ W}/\text{m}^2$, $-0.18 \pm 0.07 \text{ W}/\text{m}^2$,

$+0.14 \pm 0.06 \text{ W}/\text{m}^2$, and $-0.10 \pm 0.02 \text{ W}/\text{m}^2$. These results are shown in Figure 2.

4. Discussion and Conclusion

[26] Several factors contribute to the biases incurred through using a simple dilution approach. Firstly, urban regions are more efficient at oxidizing and removing the primary emitted substances BC, OC, and SO_2 . Furthermore, this more efficient oxidation of primary species leads to an increased generation of the secondary aerosols and their precursors OC, sulfate, and H_2SO_4 , at the urban scale. Finally, by efficiently reducing the SO_2 emitted from urban areas, over longer temporal and spatial scales, the aging of BC, OC, and sulfate to MBS, MOS, and ACC is reduced.

[27] The first two effects are exemplified by the more efficient removal of BC, SO_2 , and OC both in the urban mass fluxes to the grids containing urban areas, as well as in the concentrations of these species in the global grids containing urban areas. SO_2 is chemically oxidized to sulfate by both gas and liquid phase processing, with a portion of this new urban sulfate deposited in the urban area, and another portion escaping as new sulfate aerosols. The only exception

is that secondary production of OC sometimes leads to a small net increase near urban areas, when compared with the dilution method.

[28] The third effect is exemplified by the differences between the column values and the surface values, or more generally, the distance downwind from urban areas. Due to more efficient urban removal of SO₂, there is less available in regions not near urban areas, to be oxidized to H₂SO₄ and sulfate. Therefore, the global lifetimes of BC and OC increase since their chemical aging is dependent on the concentration of H₂SO₄.

[29] Note that there are several processes affecting these factors which are formulated in our model as depending non-linearly on aerosol precursor concentrations. Some of these include the aging of BC and OC by sulfuric acid gas to form mixed MBS and MOS, and the competition between various aerosols (MBS, MOS, ACC, and smaller-sized sulfate) for consuming sulfuric acid through condensational growth and new particle nucleation. The outcome is very different if the more realistic urban processing method is adopted given these non-linear processes.

[30] Excluding urban processing leads overall to an overestimation of the AOD, due to the ACC, MBS and MOS overestimates outweighing the BC and OC underestimates. The AAOD is usually also overestimated, although this is seasonally variable. The underestimates of BC (and to a much lesser extent OC) are outweighed by the overestimate of MBS. This leads to the conclusion that in the case including urban processing, the TOA and BOT are both less negative than in the dilution case, whereas the ABS bias depends on the spatial and seasonal balance between the BC and MBS column changes.

[31] **Acknowledgments.** This research was supported in part by DOE Grant # DE-FG02-94ER61937, U.S. NSF (AGS-0944121), the federal, industrial, and foundation sponsors of the MIT Joint Program on the Science and Policy of Global Change <http://globalchange.mit.edu/sponsors/> and in part by the Singapore National Research Foundation (NRF) through the Singapore-MIT Alliance for Research and Technology (SMART) Center for Environmental Sensing and Monitoring (CENSAM). We would also like to acknowledge Phillip Rasch for his help with setting up and running the offline version of CAM. Finally, we would like to acknowledge the two anonymous reviewers for their thoughtful and helpful comments and critiques.

[32] The Editor thanks two anonymous reviewers for their assistance in evaluating this paper.

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