



## Wildfires drive interannual variability of organic carbon aerosol in the western U.S. in summer

Dominick V. Spracklen,<sup>1,2</sup> Jennifer A. Logan,<sup>1</sup> Loretta J. Mickley,<sup>1</sup> Rokjin J. Park,<sup>1,3,4</sup> Rosemarie Yevich,<sup>1</sup> Anthony L. Westerling,<sup>5</sup> and Dan A. Jaffe<sup>6</sup>

Received 15 March 2007; revised 16 May 2007; accepted 16 July 2007; published 25 August 2007.

[1] Forest wildfire area burned in the western U.S. has increased in recent decades resulting in a substantial organic carbon (OC) source with large interannual variability. We derive OC emissions from wildfires using data for area burned for 1980–2004 and ecosystem specific fuel loadings. For the period 1989–2004 we analyze OC observations in the western U.S. from the IMPROVE network and use a global chemical transport model to simulate OC concentrations. Modeled and observed OC concentrations are highly correlated when we use interannually varying fire emissions ( $R^2 = 0.88$ ); the correlation is smaller with climatological emissions ( $R^2 = 0.4$ ). We estimate that the observed increase in wildfire activity after the mid 1980s has caused mean OC concentrations in summer over the western U.S. to increase by 30% relative to 1970–1984. In the coming decades, climate change will likely cause further increases in wildfires resulting in increased OC concentrations with implications for health and visibility. **Citation:** Spracklen, D. V., J. A. Logan, L. J. Mickley, R. J. Park, R. Yevich, A. L. Westerling, and D. A. Jaffe (2007), Wildfires drive interannual variability of organic carbon aerosol in the western U.S. in summer, *Geophys. Res. Lett.*, 34, L16816, doi:10.1029/2007GL030037.

### 1. Introduction

[2] Organic carbon (OC) is a major component of atmospheric aerosol in the western United States, contributing between 35 and 55% of total fine aerosol mass [Malm *et al.*, 2004]. OC aerosol has important impacts on human health, visibility and climate. Sources of OC include fossil fuel and biofuel combustion, wildfires and oxidation products from biogenic hydrocarbons. In the western U.S., summertime OC concentrations are dominated by emissions from wildfires and vegetation [Park *et al.*, 2003].

[3] Forest wildfire activity in the western U.S. is thought to have increased in recent decades. Westerling *et al.* [2006] compiled a database of large wildfires in the western U.S.

since 1970, showing a large increase in forest wildfire activity in the mid-1980s driven by increasing spring and summertime temperatures. Annually averaged area burned during 1987–2003 was six and a half times the amount recorded during 1970–1986. In this paper we explore the impact of increasing wildfire activity on OC aerosol concentrations in the western U.S.

[4] Observations of elemental carbon (EC) and OC in the U.S. have been made by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network since 1987 [Malm *et al.*, 1994]. This observation record does not include the period marked by the large increase in wildfires during the mid-1980s. However, both wildfires and summertime OC concentrations in the western U.S. have shown significant interannual variability since 1987. There is less interannual variability in EC, for which fossil fuel combustion is the dominant source, even in summer [Park *et al.*, 2003].

[5] To evaluate the extent to which interannual variability in summertime OC concentrations is caused by variability in wildfires we use a global chemical transport model (CTM) to simulate the effects of emissions of OC from wildfires in the western U.S. from 1989 to 2004. We also explore the impact of increasing forest wildfire activity on OC concentrations in the western U.S. by estimating summertime OC concentrations prior to the observation record.

### 2. Interannual Variability in Wildfire and OC Concentrations

[6] We predict carbonaceous aerosol concentrations using the GEOS-Chem global 3-D CTM [Bey *et al.*, 2001; Park *et al.*, 2003]. We use GEOS-Chem v7.04 (see <http://www-as.harvard.edu/chemistry/trop/geos>) driven by assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS)-4 at  $2^\circ \times 2.5^\circ$  horizontal resolution. In this analysis we treat EC and OC aerosol with a hydrophobic and hydrophilic fraction for each (giving 4 advected tracers). We assume that 80% of EC and 50% of OC emitted from primary sources are hydrophobic with a 1.2 day e-folding conversion from hydrophobic to hydrophilic [Cooke *et al.*, 1999; Chin *et al.*, 2002]. We assume a 10% carbon yield of OC from terpenes [Chin *et al.*, 2002]. We assume all secondary OC is hydrophilic. Terpene emissions are calculated according to Guenther *et al.* [1995] and are dependent on vegetation type and temperature. Anthropogenic emissions of OC over the U.S. are from Cooke *et al.* [1999] with the correction factor from Park *et al.* [2003]. Biofuel OC emissions are from Yevich and Logan [2003] and from Park *et al.* [2003] for the U.S. We assume there is no interannual variability in fossil

<sup>1</sup>School of Engineering and Applied Sciences, Harvard University, Cambridge, USA.

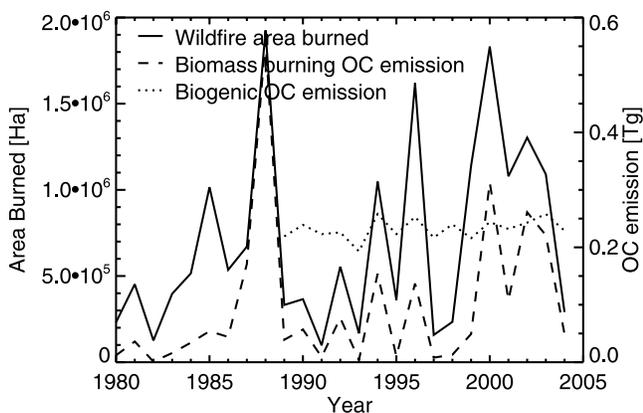
<sup>2</sup>Now at School of Earth and Environment, University of Leeds, Leeds, U.K.

<sup>3</sup>Also at Department of Earth and Planetary Sciences, Harvard University, Cambridge, USA.

<sup>4</sup>Now at School of Earth and Environmental Sciences, Seoul National University, Seoul, Korea.

<sup>5</sup>School of Engineering, University of California, Merced, California, USA.

<sup>6</sup>Interdisciplinary Arts and Sciences, University of Washington Bothell, Bothell, Washington, USA.



**Figure 1.** Summertime (June through August) wildfire area burned [Westerling *et al.*, 2003] and OC emission from wildfire and from biogenic sources in the western U.S. ( $31^{\circ}$ – $49^{\circ}$ N,  $125^{\circ}$ – $100^{\circ}$ W) for the period 1980–2004. Mean summertime OC emissions are 0.365 Tg with contributions from fossil fuel 0.018 Tg (5%), biofuel sources 0.021 Tg (6%), wildfire 0.097 Tg (26%) and biogenic sources 0.232 Tg (64%).

or biofuel emissions. A global evaluation of EC and OC is given by Chin *et al.* [2002] and a more detailed evaluation over the United States is shown by Park *et al.* [2003].

[7] We present two GEOS-Chem model simulations with different wildfire emissions in the western U.S. For the first simulation, we use monthly mean emissions from the climatological biomass burning inventory described by Duncan *et al.* [2003]. For the second simulation we use interannually varying wildfire emissions in the western U.S. ( $31^{\circ}$ – $49^{\circ}$ N,  $125^{\circ}$ – $100^{\circ}$ W) but retain climatological wildfire emissions elsewhere. We calculate emissions in the western U.S. using monthly data for wildfire area burned from Westerling *et al.* [2003]. They used reports from various agencies in the U.S. that provided the area burned on federal land, and the start and end date of individual fires, from 1980 to 2000. This database has been subsequently extended to 2004. Each fire was assumed to burn entirely in the month during which it started, and the fires were aggregated on a grid of  $1^{\circ} \times 1^{\circ}$  [Westerling *et al.*, 2003]. Monthly emissions of OC and EC were calculated using ecosystem specific fuel loadings (averaged from a  $0.025^{\circ} \times 0.025^{\circ}$  to a  $1^{\circ} \times 1^{\circ}$  grid) from the U.S. Forest Service (D. McKenzie, personal communication, 2006; <http://www.fs.fed-us/pnw/fera/fccs/maps.shtml>) and emission factors from Andreae and Merlet [2001]. We assumed wildfires occur with 25% high, 25% medium and 25% low severity and that 25% of reported area burned remains unburned, based on an analysis of the largest wildfires in 2002 in the continental U.S. [Randall, 2004].

[8] Figure 1 shows area burned and OC emissions that we calculate for wildfires in the western U.S. for June–August of each year, and compares them to biogenic sources of OC. The variability in emissions does not always parallel that in area burned as the fires can occur in different ecosystems in different years, with varying fuel loads. In low fire years, biogenic emissions exceed fire emissions. The converse is true in high fire years, where wildfire

emissions can contribute up to 65% of total summertime OC emissions.

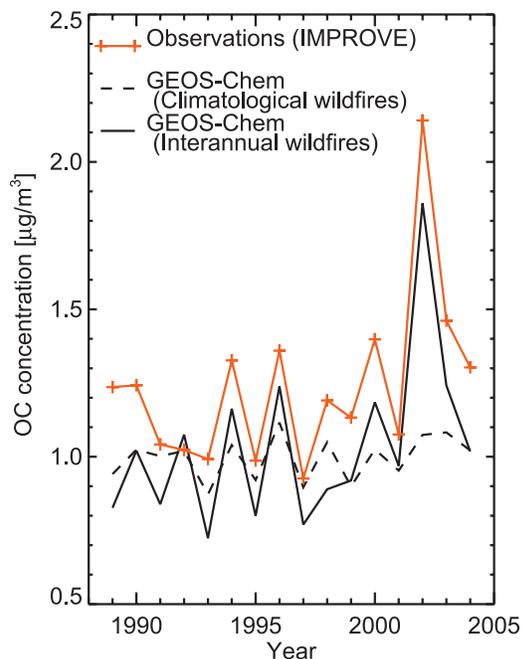
[9] We conducted GEOS-chem simulations for 1989 to 2004, the 16 year period for which both GEOS-4 analyzes and IMPROVE observations are available. This period includes years with large area burned (1994, 1996, 2000, 2002, 2003), and with small area burned (1991, 1993, 1997, 1998) and so is ideal for studying the impact of fires on the variability of atmospheric OC. Mean summertime area burned in 1989–2004 is  $0.73 \times 10^6$  Ha, with emissions of 0.093 Tg OC. This emission estimate for OC is similar to the climatological value of 0.064 Tg from Duncan *et al.* [2003].

[10] Figure 2 shows summertime mean OC concentrations in the western U.S. Observations are from the IMPROVE network [Malm *et al.*, 1994] which uses Thermal Optical Reflectance to measure 24 hour speciated OC and EC concentrations. We average over the 27 IMPROVE stations in the western U.S. (west of  $100^{\circ}$  longitude) which have observations starting in 1989 or earlier. Summertime OC concentrations show relatively large interannual variability in the western U.S. (1989–2004 observed mean  $1.24 \mu\text{g m}^{-3}$ , standard deviation  $0.29 \mu\text{g m}^{-3}$ ). Summertime EC concentrations (see auxiliary material) have smaller interannual variability (1989–2004 observed mean  $0.21 \mu\text{g m}^{-3}$ , standard deviation  $0.03 \mu\text{g m}^{-3}$ ) because wildfires contribute a smaller fraction of the total EC source [Park *et al.*, 2003].<sup>1</sup> Summertime OC concentrations are as high as  $2.1 \mu\text{g m}^{-3}$  in 2002 and as low as  $0.9$ – $1.0 \mu\text{g m}^{-3}$  in 1993, 1995 and 1997. Years with large area burned and large OC emissions in the western U.S. tend to have high summertime OC concentrations. Regression of observed summertime OC concentrations with area burned and wildfire OC emission gives  $R^2$  of 0.30 and 0.56 respectively. The higher  $R^2$  for emissions demonstrates the importance of using ecosystem specific fuel loadings to calculate OC emissions. The corresponding regression for EC concentrations yields  $R^2$  of 0.34 with area burned and 0.38 with wildfire EC emission.

[11] As shown in Figure 2, the simulation with climatological wildfire emissions results in little year to year variability in OC concentrations (model mean  $1.00 \mu\text{g m}^{-3}$ , standard deviation  $0.07 \mu\text{g m}^{-3}$ ). The simulation using interannually varying wildfire emissions in the western U.S. mimics observed variability in OC concentrations (model mean  $1.03 \mu\text{g m}^{-3}$ , standard deviation  $0.28 \mu\text{g m}^{-3}$ ). The low model bias of about  $0.2 \mu\text{g m}^{-3}$  has been reported previously [Park *et al.*, 2003] and suggests that the model is missing a constant summertime source of OC. Recent work has demonstrated that isoprene can make a substantial contribution to organic aerosol [Henze and Seinfeld, 2006; Liao *et al.*, 2007] and can explain a low model bias of about  $0.2 \mu\text{g m}^{-3}$  during summer over the western U.S. [van Donkelaar *et al.*, 2007].

[12] The model shows elevated OC concentrations in 1994, 1996, 2000, 2002, and 2003, as do the observations. The peaks in OC concentrations in 1989, 1998 and 2004 are probably due to transport from large fires in Canada and Alaska [Stocks *et al.*, 2002; Stohl *et al.*, 2006]. These are not captured by the model due to use of climatological wildfire emissions outside the western U.S.

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2007GL030037.



**Figure 2.** Observed and modeled mean summertime concentrations of OC aerosol averaged across all IMPROVE sites in the western U.S. (west of  $100^\circ$  longitude) between 1989 and 2004.

[13] The  $R^2$  value between observed and modeled summertime mean OC concentrations in the western U.S. is 0.88 with interannual fires and 0.40 with climatological wildfire emissions. The equivalent  $R^2$  values for EC concentrations are 0.35 with interannual fires and 0.09 with climatological fires. The temperature dependence of monoterpene emissions results in a relatively large  $R^2$  for OC concentrations even when climatological fires are used in the model, in contrast to the low  $R^2$  for EC, for which there is no temperature dependent source apart from fires.

[14] These results demonstrate that the interannual variability in model transport, deposition and secondary organic aerosol (SOA) production from monoterpenes produces little interannual variability in model OC concentrations. Summertime mean monoterpene emissions in the western U.S. deviate from the mean by up to 12%. Isoprene emissions, which are not included in these model runs, may contribute somewhat to the observed variability in OC aerosol. Variations in weather and leaf area index can cause isoprene emissions to vary by up to 50% for a particular month [Guenther et al., 2006; Palmer et al., 2006]. Combining such variations with the estimate of the isoprene contribution to SOA over the western U.S. [van Donkelaar et al., 2007] implies that variability in isoprene emissions could cause interannual variability in OC concentration of only  $0.1 \mu\text{g m}^{-3}$ , much smaller than observed (see Figure 2).

### 3. Prediction of OC Concentrations Prior to 1985

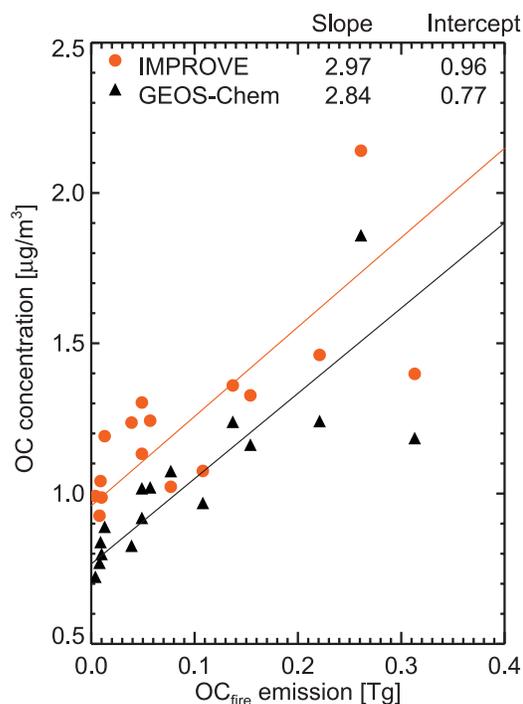
[15] Emissions of OC from wildfires in the Western U.S. were relatively low in 1980–1984, with a summertime mean of 0.02 Tg and a range of 0.001–0.04 Tg (see Figure 1). Wildfire emissions of OC were much higher in

1995–2004, with a mean of 0.13 Tg and a range of 0.01–0.32 Tg). Analysis of the frequency of large forest wildfires [Westerling et al., 2006] shows that fire activity in the 1970s was similar to that in 1980–1984, suggesting a relatively long period of low fire activity.

[16] There are no direct observations of the impact of the increased fire activity on atmospheric OC concentrations as IMPROVE observations of OC did not start until 1987. We estimate OC concentrations prior to 1985 from the relationship between observed OC concentrations and OC emissions from fires in the western U.S. (Figure 3). The slope of the regression line for observed OC concentrations (slope = 2.97) is well captured by the model (slope = 2.84). Using the regression between observed OC concentrations and wildfire OC emissions in the western U.S. ( $R^2 = 0.56$ ) we predict that mean summertime OC concentrations for 1980–1984 were  $1.02 \mu\text{g m}^{-3}$  and ranged from 0.96 to  $1.08 \mu\text{g m}^{-3}$ . We use these results to estimate that mean summertime OC concentrations in the 1970s and early 1980s were  $1.0 \mu\text{g m}^{-3}$ , considerably smaller than the mean in 1995–2004,  $1.3 \mu\text{g m}^{-3}$ .

### 4. Implications

[17] We have shown that the variability in summertime OC concentrations in the western U.S. can to a large part be explained by variability in western U.S. wildfire emissions. As this work only considers area burned in the western U.S. we have restricted our analysis to carbonaceous aerosol tracers which have relatively short lifetimes. Earlier work has shown that CO concentrations north of  $30^\circ\text{N}$  are



**Figure 3.** Model and observed summertime OC concentrations in the western U.S. plotted against summertime emission of OC from wildfires. Solid red and solid black lines are the reduced major axis regression for observations and model respectively.

correlated with area burned by forest fires in North America and Russia [Wotawa et al., 2001; Kasischke et al., 2005].

[18] We estimate that increases in wildfire area burned in the western U.S. after the mid-1980s has caused long-term mean summertime OC concentrations in the western U.S. to increase by 30% (from  $1.0 \mu\text{g m}^{-3}$  between 1970 and 1984 to  $1.3 \mu\text{g m}^{-3}$  in 1995–2004). Based on an analysis of the IMPROVE data, we find that the contribution of organic carbon mass to the total fine aerosol concentration is about 40% in low fire years, but increases to about 55% in high fire years. The increase in wildfires and OC concentrations we describe here will therefore have important implications for visibility degradation.

[19] Increasing temperature over the next few decades in the western U.S. could drive further increases in wildfire activity with potential impacts on OC concentrations. Further degradation in visibility due to increasing wildfires will make it increasingly difficult to meet targets set by the U.S. Environmental Protection Agency [2003] Regional Haze Rule. The impact of future climate change on forest fires in North America needs to be quantified to allow us to estimate the effects on future air quality.

[20] **Acknowledgments.** We thank the anonymous reviewers for their helpful comments on the original manuscript. This work was funded by the Environmental Protection Agency (grant RD-83227501-0), the National Science Foundation (ATM-0554804), and the National Aeronautics and Space Administration (NASA-MAP grant NNG06GB48G).

## References

- Andreae, M., and P. Merlet (2001), Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cycles*, *15*, 955–966.
- Bey, I., et al. (2001), Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, *J. Geophys. Res.*, *106*, 23,073–23,096.
- Chin, M., et al. (2002), Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sunphotometer measurements, *J. Atmos. Sci.*, *59*, 461–483.
- Cooke, W., C. Lioussé, H. Cachier, and J. Feichter (1999), Construction of a  $1^\circ \times 1^\circ$  fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM-4 model, *J. Geophys. Res.*, *104*, 22,137–22,162.
- Duncan, B. N., R. V. Martin, A. C. Staudt, R. Yevich, and J. A. Logan (2003), Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, *J. Geophys. Res.*, *108*(D2), 4100, doi:10.1029/2002JD002378.
- Guenther, A., et al. (1995), A global model of natural volatile organic compound emissions, *J. Geophys. Res.*, *100*, 8873–8892.
- Guenther, A., et al. (2006), Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, *6*, 3181–3210.
- Henze, D. K., and J. H. Seinfeld (2006), Global secondary organic aerosol formation from isoprene oxidation, *Geophys. Res. Lett.*, *33*, L09812, doi:10.1029/2006GL025976.
- Kasischke, E. S., E. J. Hyer, P. C. Novelli, L. P. Bruhwiler, N. H. F. French, A. I. Sukhinin, J. H. Hewson, and B. J. Stocks (2005), Influences of boreal fire emissions on Northern Hemisphere atmospheric carbon and carbon monoxide, *Global Biogeochem. Cycles*, *19*, GB1012, doi:10.1029/2004GB002300.
- Liao, H., D. K. Henze, J. H. Seinfeld, S. Wu, and L. J. Mickley (2007), Biogenic secondary organic aerosol over the United States: Comparison of climatological simulations with observations, *J. Geophys. Res.*, *112*, D06201, doi:10.1029/2006JD007813.
- Malm, W. C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill (1994), Spatial and seasonal trends in particle concentration and optical extinction in the United States, *J. Geophys. Res.*, *99*, 1347–1370.
- Malm, W. C., B. A. Schichtel, M. L. Pitchford, L. L. Ashbaugh, and R. A. Eldred (2004), Spatial and monthly trends in speciated fine particle concentration in the United States, *J. Geophys. Res.*, *109*, D03306, doi:10.1029/2003JD003739.
- Palmer, P. I., et al. (2006), Quantifying the seasonal and interannual variability of North American isoprene emissions using satellite observations of the formaldehyde column, *J. Geophys. Res.*, *111*, D12315, doi:10.1029/2005JD006689.
- Park, R. J., D. J. Jacob, M. Chin, and R. V. Martin (2003), Sources of carbonaceous aerosols over the United States and implications for natural visibility, *J. Geophys. Res.*, *108*(D12), 4355, doi:10.1029/2002JD003190.
- Randall, D. (2004), Strawman-approach for technical refinement of large fires and fire complexes for WRAP Phase 1 fire emissions inventory, Project 178-6, technical memorandum, Air Sci. Inc., Portland, Ore.
- Stocks, B. J., et al. (2002), Large forest fires in Canada, 1959–1997, *J. Geophys. Res.*, *107*, 8149, doi:10.1029/2001JD000484 [printed 108(D1), 2003].
- Stohl, A., et al. (2006), Pan-Arctic enhancements of light absorbing aerosol concentrations due to North American boreal forest fires during summer 2004, *J. Geophys. Res.*, *111*, D22214, doi:10.1029/2006JD007216.
- U.S. Environmental Protection Agency (2003), Guidance for estimating natural visibility conditions under the regional haze rule, *Rep. EPA 454/B-03-005*, Research Triangle Park, N. C.
- van Donkelaar, A., et al. (2007), Model evidence for a significant source of secondary organic aerosol from isoprene, *Atmos. Environ.*, *41*, 1267–1274, doi:10.1016/j.atmos.env.2006.09.051.
- Westerling, A., et al. (2003), Climate and wildfire in the western United States, *Bull. Am. Meteorol. Soc.*, *84*, 595–604.
- Westerling, A., H. Hidalgo, D. Cayan, and T. Swetnam (2006), Warming and earlier spring increases western U.S. forest wildfire activity, *Science*, *313*, 940–943.
- Wotawa, G., P. Novelli, M. Trainer, and C. Grainer (2001), Inter-annual variability of summertime CO concentrations in the Northern Hemisphere explained by boreal forest fires in North America and Russia, *Geophys. Res. Lett.*, *28*, 4575–4578.
- Yevich, R., and J. Logan (2003), An assessment of biofuel use and burning of agricultural waste in the developing world, *Global Biogeochem. Cycles*, *17*(4), 1095, doi:10.1029/2002GB001952.

D. A. Jaffe, Interdisciplinary Arts and Sciences, University of Washington Bothell, Bothell, WA 98011–8246, USA.

J. A. Logan, L. J. Mickley, and R. Yevich, School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA.

R. J. Park, School of Earth and Environmental Sciences, Seoul National University, Seoul 151-747, Korea.

D. V. Spracklen, School of Earth and Environment, University of Leeds, Leeds LS2 9JT, UK. (dominick@env.leeds.ac.uk)

A. L. Westerling, School of Engineering, University of California, Merced, P.O. Box 2039, Merced, CA 95344, USA.