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Adjoint estimation of ozone climate penalties

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[1] An adjoint of a regional chemical transport model is used to calculate location-specific temperature influences (climate penalties) on two policy-relevant ozone metrics: concentrations in polluted regions (>65 ppb) and short-term mortality in Canada and the U.S. Temperature influences through changes in chemical reaction rates, atmospheric moisture content, and biogenic emissions exhibit significant spatial variability. In particular, high-NO_x, polluted regions are prominently distinguished by substantial climate penalties (up to 6.2 ppb/K in major urban areas) as a result of large temperature influences through increased biogenic emissions and nonnegative water vapor sensitivities. Temperature influences on ozone mortality, when integrated across the domain, result in 369 excess deaths/K in Canada and the U.S. over a summer season—an impact comparable to a 5% change in anthropogenic NO_x emissions. As such, we suggest that NO_x control can be also regarded as a climate change adaptation strategy with regard to ozone air quality. **Citation:** Zhao, S., A. J. Pappin, S. Morteza Mesbah, J. Y. Joyce Zhang, N. L. MacDonald, and A. Hakami (2013), Adjoint estimation of ozone climate penalties, *Geophys. Res. Lett.*, *40*, 5559–5563, doi:10.1002/2013GL057623.

1. Introduction

[2] Ozone "climate penalty" refers to the increase in ground-level ozone concentrations within a changing climate. Studies on ozone-climate interaction have found that of all meteorological parameters, temperature exerts the strongest influence on ozone [*Steiner et al*., 2006; *Dawson et al*., 2007; *Jacob and Winner*, 2009]. Globally, surface ozone is predicted to rise by 1–12 ppb near the end of the century [*Liao et al*., 2006; *Murazaki and Hess*, 2006; *Jacob and Winner*, 2009]. Observational studies examining the effect of temperature on ozone have found that climate penalties in polluted regions are highest [*Bloomer et al*., 2009; *Jacob and Winner*, 2009], while ozone in less polluted environments is not closely correlated with temperature [*Sillman and Samson*, 1995]. *Bloomer et al*. [2009] found that ozone in the eastern U.S. responds linearly to increasing temperatures, terming the slope of the ozone-temperature response curve the climate penalty factor (CPF). In the study, CPFs throughout the eastern U.S. showed dependence on regional chemical regimes and the year of observations and ranged between 1.1 and 3.6 ppb/K.

[3] Model-based sensitivity studies have examined how ozone responds to temperature at a regional scale. *Hogrefe et al*. [2004] estimated that the daily maximum 8 h average (DM8) ozone under future climate scenarios will be 0–8 ppb higher in the eastern U.S. by 2020–2050, with an even larger increase of 7–13 ppb by 2080. *Steiner et al*. [2006] predicted changes in ozone with temperature, using brute force sensitivity analysis, ranging from 0.2 to 2 ppb/K in California. A subsequent modeling study by *Dawson et al*. [2007] found the average CPF for DM8 in the eastern U.S. to be 0.34 ppb/K, while peak hourly ozone concentrations increased by an average of 4.7 ppb/K.

[4] In the past, modeling studies examining the response of ground-level ozone to changes in temperature employed forward sensitivity analysis or scenario-based approaches. Forward sensitivity studies provide receptor-specific responses to globally or regionally rising temperatures. However, under this approach, the influences of temperature changes emanating from specific areas remain unresolved because future ozone concentrations are related to widespread, regional or continental changes in temperature rather than local changes. In this study, we use adjoint or backward sensitivity analysis to relate ozone to changes in local temperatures across Canada and the U.S. We estimate temperature influences on two different types of ozone metrics with high policy relevance: average ozone in polluted regions and short-term ozone-induced mortality. While two previous studies have briefly explored the use of adjoint sensitivity analysis for calculating the impact of temperature on ozone [*Menut*, 2003; *Hakami et al*., 2007], this is the first comprehensive effort to estimate ozone-temperature response slopes with this approach.

2. Methodology

2.1. Adjoint Sensitivity Analysis

[5] The adjoint method of sensitivity analysis provides location-specific information by relating changes in one chemical transport model (CTM) output to changes in many model inputs (such as temperature). An adjoint simulation calculates sensitivities or derivatives of any air quality metric of interest (termed the adjoint cost function, *J*) with respect to model inputs, as long as the metric is a scalar concentration-based function. Detailed descriptions of atmospheric adjoint modeling [*Sandu et al*., 2005; *Hakami et al*., 2007] and use of the adjoint cost function for analysis of policy metrics such as ozone-related mortality [*Pappin and Hakami*, 2013] can be found elsewhere.

[6] We use the adjoint of the U.S. EPA's Community Multiscale Air Quality (CMAQ) model v4.5.1 for gas-phase processes only [*Hakami et al*., 2007]. Our simulation

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spans over 3 months of the ozone season from 1 July through 30 September 2007. Our domain has a horizontal resolution of 36 km and a vertical structure of 34 layers extending into the stratosphere. The Weather Research and Forecasting (WRF) model is used for meteorological inputs. Projections of emission inputs for 2007 are based on the 2005 National Emissions Inventory for the U.S. and the 2006 National Pollutant Release Inventory for Canada. Emission modeling is performed using the Sparse Matrix Operator Kernel Emissions (SMOKE) model. The mean normalized error and bias of CMAQ ozone concentrations over the simulated episode are 19.8% and 2.0%, respectively. For high-concentration locations and days (>65 ppb), our simulation somewhat underestimates observed concentrations with mean normalized error and bias of 19.7% and −10.0%, respectively.

[7] We investigate the climate penalty, or the sensitivity to temperature, of two ozone metrics using the adjoint of CMAQ. We define our first adjoint cost function, J_c, as the summation of DM8 ozone concentrations (C_{DM8}; ppb) for all combinations of locations and days, (x, y, d), within Canada and the U.S. when the DM8 concentration exceeds a threshold of 65 ppb, i.e.,

$$J_c = \sum_{x,y,d} C_{DM8}(x,y,d) \qquad \forall x,y,d : C_{DM8}(x,y,d) \geq 65 \text{ ppb} \quad (1)$$

[8] We refer to these locations, over which J_c is integrated, as polluted regions (PRs) and to their temperature influence as the polluted region climate penalty (PR-CP). We use a threshold of 65 ppb as the midpoint value in the 60–70 ppb range proposed for the future U.S. ozone standard and as a value close to the recent Canadian standard of 65 ppb. Our second adjoint cost function, J_m, is defined as the number of premature deaths related to acute ozone exposure in Canada and the U.S., based on epidemiologic data from *Bell et al*. [2004] which indicates a nonthreshold ozone-mortality response of 0.052%/ppb [*Pappin and Hakami*, 2013]. Population and mortality data for inclusion were taken from the Air Quality Benefits Assessment Tool for Canada and the Centers for Disease Control and Prevention for the U.S. for 2007. We refer to the climate penalty associated with this metric as the mortality climate penalty (M-CP).

2.2. Pathways of Temperature Influence on Ozone

[9] This study only considers temperature effects related to gas-phase chemistry and does not account for meteorological pathways of temperature influence such as stagnation events and clouds, nor do we consider the impact of future changes in anthropogenic and wildfire emissions [*Jacob and Winner*, 2009]. The direct effect of temperature on ozone is manifested through accelerated photochemical reactions that occur with rising temperatures. Two other indirect effects on ozone relate to changes in biogenic emissions (VOCs and NO) and atmospheric water vapor with temperature. We examine these three pathways for temperature influences, both individually and as a whole, on the ozone metrics J_c and J_m, as in *Steiner et al*. [2006] and *Doherty et al*. [2013].

[10] At any time during backward integration of the adjoint CTM, the direct impact of temperature through reaction rate constants on the adjoint cost function of choice (i.e., $\frac{\partial J}{\partial T}\big|_k$) can be calculated as

$$\left.\frac{\partial J}{\partial T}\right|_k = \sum_i \frac{\partial J}{\partial C_i} \cdot \frac{\partial C_i}{\partial T} = \sum_j \frac{\partial J}{\partial k_j} \cdot \frac{\partial k_j}{\partial T} \quad (2)$$

where T and C_i, respectively, are the temperature and concentration of species i at any location and time, and k_j is the temperature-dependent rate constant for reaction j. The first equation is the approach taken in this study and uses the calculated adjoint variables ($\partial J/\partial C$) in combination with temperature sensitivities ($\partial C/\partial T$) from the tangent linear model of the chemical solver used in CMAQ. Calculated temperature gradients can be then integrated over all chemistry time steps during backward execution of the adjoint model.

[11] The indirect impact of temperature on ozone metrics through water vapor concentrations is calculated in a similar manner as

$$\left.\frac{\partial J}{\partial T}\right|_q = \sum_i \frac{\partial J}{\partial C_i} \cdot \frac{\partial C_i}{\partial q} \cdot \frac{\partial q}{\partial T} \quad (3)$$

where q is the atmospheric water vapor concentration (i.e., specific humidity) at each location. We assume that relative humidity levels remain constant in the future climate [*Willett et al*., 2007; *Santer et al*., 2007]. We estimate the temperature dependence of specific humidity through analytical differentiation of the Clausius-Clapeyron equation for water vapor in the atmosphere [*Bohren and Albrecht*, 1998].

[12] Finally, the indirect temperature influence through increased biogenic emissions is estimated as

$$\left.\frac{\partial J}{\partial T}\right|_E = \sum_i \frac{\partial J}{\partial E_i} \cdot \frac{\partial E_i}{\partial T} \quad (4)$$

where E_i is the natural/biogenic emissions of species i (VOCs or soil NO). The temperature dependence of biogenic emissions is calculated using a central difference approximation by running perturbed simulations of the biogenic emission model in SMOKE [*Houyoux et al*., 2000; *U.S. Environmental Protection Agency*, 1995]. Perturbations are made to both surface soil and air temperatures, but we do not account for changes in soil moisture in our calculations.

3. Results and Discussion

[13] Our set of spatial plots (Figure 1) show the influence of temperature changes on (1) cumulative DM8 concentrations in PRs and (2) premature mortality related to acute ozone exposure, both in Canada and the U.S., and via different pathways (influences through soil NO emissions are not shown). In the figures that follow, a PR-CP (left), $\partial J_c/\partial T(x,y)$, where (x,y) refers to location, equal to 1 ppb/K for any model grid cell indicates that a 1 K rise in temperature in that cell on an average day would result in a 1 ppb cumulative increase in DM8 concentrations in PR grid cells. Note that such influences are likely to materialize over many (other) grid cells, and the distribution and magnitude of these influences across receptors are unknown, as adjoint sensitivity analysis lacks receptor specificity. In the same way, a M-CP, $\partial J_m/\partial T(x,y)$, equal to 1 death/K (right) indicates that a 1 K rise in temperature in that location would result in one excess death related to acute ozone exposure across Canada and the U.S. Our results are integrated over

all the model layers and thus assume a uniform change in temperature in all model layers. While this is an imperfect assumption, we consider it justified as most of the contributions to climate penalties happen within the boundary layer. For example, in the case of PR-CP, 90% of contributions through water vapor and reaction rate pathways occur in the first 2900 and 1800 m of model height, respectively.

[14] Rising temperatures, in general, dictate overall increases in PR DM8 ozone concentrations and mortality (Figures 1a and 1b) through faster reaction rates (Figures 1c and 1d) and higher VOC emissions (Figures 1e and 1f), despite mainly negative influences from changes in atmospheric water vapor (Figures 1g and 1h). Overall and across all three pathways, rising temperatures in the eastern U.S. and California result in large increases in both ozone metrics. For example, PR-CPs in the Los Angeles area are as high as 6.2 ppb/K, meaning that an increase of 1 K in temperature on an average day in the Greater Los Angeles area can cumulatively increase DM8 ozone concentrations in PRs by as much as 6.2 ppb. M-CPs originating from 1 K temperature increase in the Greater Los Angeles are as large as 2.4 deaths/K. M-CPs are highest in New York City, amounting to an influence of 3.2 excess deaths/K.

[15] The most important feature in Figure 1 is the spatial heterogeneity of temperature influences on either metric. This heterogeneity is in part due to the spatial distribution of PRs and populations and their importance in the formulation of J_c and J_m. Similarity between the spatial distributions of temperature influences for both metrics, however, suggests that the atmospheric chemical regime (particularly NO_x availability) plays an important role in the magnitude of climate penalties. Without exception, highly populous Canadian and American urban areas have large PR-CPs and M-CPs (Figures 1a and 1b). Similarly, areas along the power plant plumes in the Ohio River Valley appear prominently on temperature influence maps.

[16] The temperature influences through reaction rates are primarily positive (Figures 1c and 1d) and largest in urban areas (particularly for mortality). *Doherty et al*. [2013] attributed similar positive sensitivities (i.e., climate penalties) to enhanced peroxyacetylnitrate decomposition at higher temperatures. Conversely, temperature influences through the water vapor pathway are mostly negative (Figures 1e and 1f) in agreement with previous findings [*Steiner et al*., 2006; *Doherty et al*., 2013]. Important exceptions to these negative influences occur in large urban areas, where the response of ozone to temperature is subdued or becomes positive altogether. This duality of response between urban and rural (or polluted and remote) atmospheres is expected and can be explained by NO_x availability in these environments. In remote atmospheres, reaction of water vapor with $O(^1D)$ is a major chemical loss for ozone, leading to a negative response of ozone to increased temperature due to higher water vapor concentrations. However, in polluted environments with high NO_x availability, the resulting hydroxyl radical from the reaction of water vapor with $O(^1D)$ results in increased ozone due to the radical-limited nature of the chemical regime, an overall positive temperature influence.

[17] Finally, temperature influences through the biogenic emissions pathway are positive in all places and significant in urban areas and PRs. As expected, temperature influences through biogenic emissions are most significant in VOC-limited chemical regimes, with a notable example being the Ohio River Valley. On the other hand, NO_x-limited regions in the southeastern U.S. only see modest temperature influences through the biogenic emissions pathway in both ozone metrics. The contrast between urban/polluted and rural/remote atmospheres is more pronounced in the water vapor pathway for the PR-CP and in the biogenic emissions pathway for the M-PR.

[18] These results further elaborate the findings of *Bloomer et al*. [2009] about the observed trend in ozone-temperature response slopes over time. They noted a sizable decrease in CPFs in the eastern U.S. (except in the southeast) and attributed this decrease to widespread power plant NO_x control. Our results illustrate a significant spatial component in temperature influences that emphasizes major polluters and urban areas. Large differences between temperature influences from polluted and less polluted areas suggest that NO_x control will have the added benefit of reducing climate penalty. Our findings are of particular interest for urban areas, where NO_x emission reductions often have negative influences on ozone mortality [*Pappin and Hakami*, 2013]. Potential short-term disbenefits from NO_x emission reductions in urban areas such as New York or Los Angeles are likely to be compensated in part by reductions in mortality in a warming climate (Figure 1b).

[19] Integrating temperature influences in Figure 1a over the domain (Table 1) yields an average climate penalty if temperatures increase uniformly in all locations, a number more comparable to studies done thus far [e.g., *Bloomer et al*., 2009]. Our estimate of a national average PR CPF of 1.34 (Table 1, all pathways) is lower but comparable to the range of 1.4–2.4 ppb/K reported for the eastern U.S. by *Bloomer et al*. [2009]. Two points about this comparison are worth mentioning. First, our adjoint estimates of CPFs are for polluted regions (i.e., location and days with DM8 ozone concentrations >65 ppb), which prevents a direct comparison with observed ozone-temperature slopes. Second, since we do not account for indirect meteorological impacts of temperature, as suggested in *Jacob and Winner* [2009], our overall CPFs are expected to be underestimated compared to those reported by *Bloomer et al*. [2009].

[20] Past work has estimated that reducing NO_x emissions across all anthropogenic sources in Canada and the U.S. by 10% would reduce mortality related to short-term ozone exposure by 730 deaths between July and September 2007 [*Pappin and Hakami*, 2013]. Here we estimate that for every 1 K rise in temperature nationally, more than half that amount, or 369 excess ozone-related deaths, occur over the same period. In other words, the mortality impact from each degree increase in temperature is roughly equivalent to a 5% reduction in anthropogenic NO_x emissions. More research is needed to quantify climate-related benefits of reduced NO_x emissions, but our results are in line with those of *Doherty et al*. [2013] suggesting that reductions in ozone precursor emissions in excess of 20% may be needed to offset rising temperatures in a North American climate that are, on average, 4° warmer toward the end of the century.

4. Conclusions

[21] We show that backward/adjoint sensitivity analysis can help delineate local and regional influences on collective ozone metrics. These results can be combined with regional

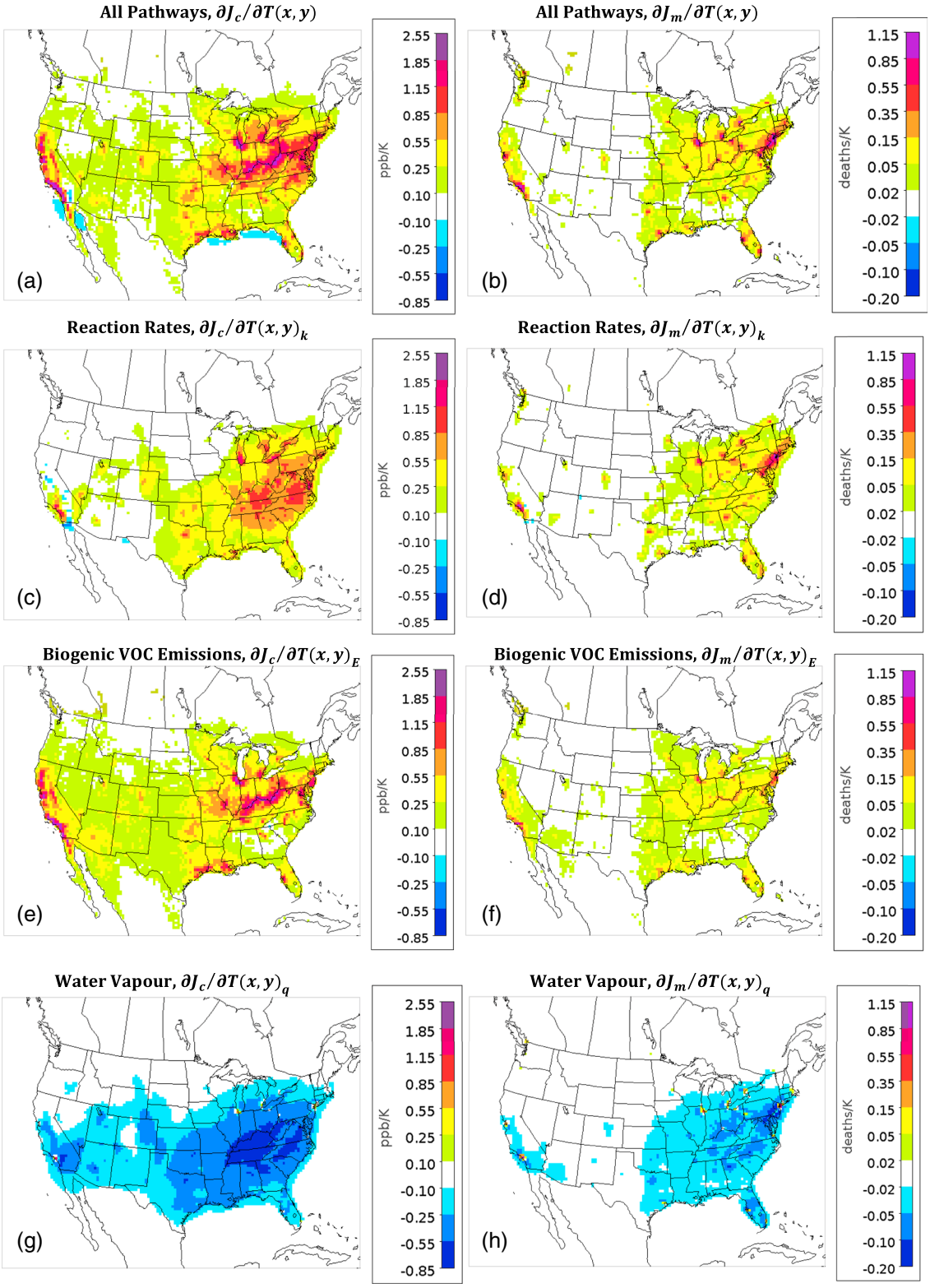

Figure 1. Climate penalty factors or temperature influences for (left) cumulative DM8 ozone in PRs (PR-CP) and (right) ozone mortality (M-CP) in Canada and the U.S. Temperature influences are shown for (a and b) all pathways and separately for three pathways: (c and d) reaction rates, (e and f) biogenic VOC emissions, and (g and h) water vapor. Influences are scaled for a uniform 1 K change in temperature in all atmospheric model layers and are integrated over 1 July to 30 September 2007.

climate model predictions to provide a more resolved picture of the effects of climate on future air quality. Our results have potential policy implications. Specifically, our findings suggest the following.

[22] 1. Emission controls for anthropogenic NO_x, and, in particular, for urban NO_x, are likely to be effective measures for reducing the vulnerability of future air quality to rising temperatures. As such, urban NO_x control can be regarded

Table 1. Average DM8 Ozone and Mortality CPFs

Temperature Pathway	Average PR CPF (ppb/K)[a]	Total Mortality (deaths/K)[a]
Reaction rates	0.93	260
Water vapor	−0.97	−198
Biogenic VOC emissions	1.26	278
Soil NO emissions	0.13	28
Total	1.34	369

[a]Sensitivities are integrated spatially to give the overall influence of a 1 K temperature change everywhere in the domain.

as a potential adaptation strategy with regard to future air quality. Obviously, this is in addition to the direct benefits associated with reduced NO_x emissions.

[23] 2. In areas where urban NO_x emission reductions incur a health disbenefit, inclusion of the temperature-related benefit can, at least in part, compensate the short-term disbenefit.

[24] 3. Considering the large magnitude of urban temperature influences, there may be significant health and nonattainment burdens attributable to the urban heat island effect. Quantifying this impact requires further research.

[25] 4. In epidemiological studies of the impact of heat or ozone on health outcomes, quantified local temperature influences can help resolve the confounding relationship between ozone and temperature, which may, in turn, result in more refined epidemiological models.

[26] 5. Future research is required to quantify how emission controls may affect the ozone-temperature response and its impacts on health outcomes and regulatory attainment objectives, but our initial findings suggest that significant interplay exists between future emission policies and air quality consequences of a warming climate.

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