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Diminishing Returns or Compounding Benefits of Air Pollution Control? The Case of NO$_x$ and Ozone

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ABSTRACT: A common measure used in air quality benefit-cost assessment is marginal benefit (MB), or the monetized societal benefit of reducing 1 ton of emissions. Traditional depictions of MB for criteria air pollutants are such that each additional ton of emission reduction incurs less benefit than the previous ton. Using adjoint sensitivity analysis in a state-of-the-art air quality model, we estimate MBs for NO$_x$ emitted from mobile and point sources, characterized based on the estimated ozone-related premature mortality in the U.S. population. Our findings indicate that nation-wide emission reductions in the U.S. significantly increase NO$_x$ MBs for all sources, without exception. We estimate that MBs for NO$_x$ emitted from mobile sources increase by 1.5 and 2.5 times, on average, for 40% and 80% reductions in anthropogenic emissions across the
U.S. Our results indicate a strictly concave damage function and compounding benefits of progressively lower levels of NO\textsubscript{x} emissions, providing economic incentive for higher levels of abatement than were previously advisable. These findings suggest that the traditional perception of a convex damage function and decreasing MB with abatement may not hold true for secondary pollutants such as O\textsubscript{3}.

INTRODUCTION. Estimating the health and environmental impacts of anthropogenic emissions is an important element of air quality decision-making. One measure of such impacts is marginal benefit (MB), or the incremental, monetized health or environmental benefit of reducing an additional unit (ton) of emissions. A related metric used in environmental economics that offers a reverse perspective is marginal damage (MD), or the health or environmental damage incurred by emitting an additional ton of pollutant. MB is an important decision metric in economic evaluation of air pollution policies as it provides a direct indication of the rate-of-return on potential investments made in abating emissions. Mathematically, MB/MD is the derivative of the total damage curve (i.e., the overall, monetized societal impact of air pollution) with respect to emissions. MB for criteria air pollutants is traditionally depicted to decrease as emissions are progressively reduced,\textsuperscript{1-2} indicating diminishing returns with each added ton of emission control. Such a negatively or downward-sloped MB curve with abatement is mathematically equivalent to a convex total damage curve with abatement. Convexity in this context implies that as emissions are reduced, the societal damage of air pollution initially declines rapidly, but with continued abatement, this rate-of-decrease in damage, or the accrued benefits from abatement, slows. This general behavior is attributed to the natural assimilative capacity of the environment to cleanse itself of pollution; a capacity that loses efficiency as the atmosphere becomes more polluted.\textsuperscript{2} At best, this is appropriate for some primary pollutants, but
a downward-sloped MB curve may be an oversimplification for secondary pollutants whose production depends nonlinearly on the availability of emitted precursors. The prime example is ground-level ozone (O\textsubscript{3}) formed from NO\textsubscript{x} (NO + NO\textsubscript{2}) and volatile organic compounds (VOCs). At low levels of NO\textsubscript{x} emissions, removal of each ton of NO\textsubscript{x} is very effective in reducing O\textsubscript{3}, yielding a large, positive MB. At very high NO\textsubscript{x} emission levels, and with limited VOCs, reducing NO\textsubscript{x} may be counterproductive, leading to increased O\textsubscript{3} concentrations through slower titration of O\textsubscript{3} by NO (a negative MB or disbenefit).\textsuperscript{3-6} This duality in O\textsubscript{3} response to NO\textsubscript{x} control seen in extreme chemical environments presents a specific, well-known case for non-convexity and an upward-sloped MB curve.

While non-convexity is a long-established concept in other areas of environmental economics,\textsuperscript{7-8} such as aquatic ecosystems\textsuperscript{9} or environmental aesthetics,\textsuperscript{10-11} non-convexity in air pollution impacts is treated as an exception to the general rule.\textsuperscript{12-15} Repetto,\textsuperscript{12} using results from box model simulations with limited O\textsubscript{3} chemistry, first suggested non-convexities in the response of O\textsubscript{3} to precursor controls, but with a focus on NO\textsubscript{x}-rich urban environments. Hakami et al.\textsuperscript{3} used regional, high-order forward sensitivity analysis in an air quality model to quantify local responses of O\textsubscript{3} to domain-wide precursor emission reductions. The authors found predominantly negative second-order derivatives of O\textsubscript{3} with respect to NO\textsubscript{x} emissions, indicative of a non-convex response surface. Drawing upon previous studies,\textsuperscript{4-5} Fraas and Lutter\textsuperscript{14-15} later discussed the exceptional case of non-convexity in the presence of negative MBs or disbenefits that poses challenges in implementing economically efficient policy instruments. While indications of non-convexity exist in the literature, a general lack of efficient modeling tools, data, and resources has inhibited characterization of the NO\textsubscript{x} MB curve to fully test the assumption of convexity. This work intends to characterize the NO\textsubscript{x} MB curve on a source-by-source basis, and
demonstrate that in the case of NO\textsubscript{x} and O\textsubscript{3}, non-convexity forms the general rule rather than the exception.

METHODS. NO\textsubscript{x} MBs are partly driven by the sensitivity of O\textsubscript{3} to NO\textsubscript{x} that depends on the relative abundance of precursor species in the atmosphere. Characterization of the NO\textsubscript{x} MB curve necessitates the use of atmospheric models that adequately describe the nonlinear pathway from NO\textsubscript{x} emissions to O\textsubscript{3} concentrations, and can do so on a ton-by-ton basis. Furthermore, sources differ in their public health impacts based on their proximity to population centers and the atmospheric conditions conducive to local and downwind O\textsubscript{3} formation. This combination of factors indicates that the same ton of NO\textsubscript{x} control for various sources may have different health impacts. Estimating MBs on a source-by-source level would thus yield invaluable information for air quality decision-making.

To estimate source-specific MBs, we use adjoint (or reverse) sensitivity analysis in a regional air quality model. The “reverse” or backward characteristic of the adjoint method indicates that influences on various receptors are traced back to individual sources through an auxiliary set of equations that govern source-receptor relationships. To accomplish this, the analysis entails backward (in time and space) integration of adjoint equations after forward simulations are carried out. More details about adjoint sensitivity analysis and its applications in health benefits assessment can be found elsewhere.\textsuperscript{6} Our approach simultaneously calculates sensitivities of model output with respect to a large number of input parameters.\textsuperscript{16-17} Achieving the same level of detail with traditional modeling approaches limits analyses to a few sources or groups of sources,\textsuperscript{4,18-19} or else requires simplifications of nonlinear chemical processes that may lead to underestimations of NO\textsubscript{x} MBs.\textsuperscript{5,20-21} Adjoint sensitivity analysis is an ideal tool for the purpose of this study as it allows for estimating MBs for a multitude of polluters across different
locations, sectors, and times while accounting for nonlinear atmospheric processes.\textsuperscript{6,22} We note that while the adjoint method offers sensitivity information on a source-by-source basis, it cannot feasibly provide information about the distribution of impacts across receptors (a question more suitable to forward methods of sensitivity analysis). Adjoint sensitivity analysis is most appropriate for applications where a collective measure of policy effectiveness is sought, such as the total health or environmental damage of emissions, as in the case of seeking to estimate MBs.

We construct MB curves for mobile and point sources using the adjoint of the U.S. EPA’s Community Multiscale Air Quality model, or CMAQ.\textsuperscript{23} The gas-phase CMAQ-adjoint model used in this study is based on CMAQ v4.5.1 with the SAPRC-99 chemical mechanism.\textsuperscript{24} The adjoint of CMAQ has been validated previously\textsuperscript{25} and used in various health impact studies.\textsuperscript{6,22,26-27} We use the standard U.S. EPA domain spanning the continental U.S. at a 36-km horizontal grid resolution with 34 vertical layers extending into the stratosphere. Our CMAQ-adjoint simulations are conducted over the O\textsubscript{3} season of 2007 (May 1 – September 30). Emissions are based on the National Emission Inventory (NEI) for the U.S. and the National Pollutant Release Inventory (NPRI) for Canada, and are generated using the Sparse Matrix Operator Kernel Emissions (SMOKE) model.\textsuperscript{28} Meteorological inputs are from the Weather Research and Forecasting (WRF) model,\textsuperscript{29} processed using the Meteorology Chemistry Interface Processor (MCIP). Performance evaluation of observed and simulated hourly O\textsubscript{3} concentrations for the 2007 O\textsubscript{3} season indicate a mean fractional error (MFE) of 16\% and mean fractional bias (MFB) of 2.5\%. Comparison of observed daily maximum 8 h average (DM8A) O\textsubscript{3} concentrations with simulated DM8As (used for health impact estimation) yields a MFE of 15\% and MFB of 9.5\%.
We define MB as the monetary societal benefit ($) of reducing NO\textsubscript{x} emissions by 1 ton from a given mobile or point source. We focus our analysis on the MB of NO\textsubscript{x} emission reductions, as NO\textsubscript{x} has by far the largest impact on population exposure to O\textsubscript{3} of all precursor species.\textsuperscript{6} Our estimations of MB account for averted mortality in the U.S. population resulting from reduced short-term O\textsubscript{3} exposure. We consider only acute O\textsubscript{3} exposure mortality, and not acute morbidity, as mortality has a high monetary value and is the largest contributor to the monetized health benefits of emission reductions.\textsuperscript{30} We do not account for environmental impacts, as we focus our analysis on population health damages. We consider only acute exposure mortality without consideration for mortality from long-term exposure to O\textsubscript{3} based on the weight of epidemiological evidence for causal associations between O\textsubscript{3} and mortality.\textsuperscript{31} As the overall behavior of NO\textsubscript{x} MBs is driven largely by the chemistry of O\textsubscript{3} production, we believe that the generality of our approach or results are not lost in exclusion of other O\textsubscript{3} damage endpoints. We note that NO\textsubscript{x} emissions also contribute to NO\textsubscript{2} exposure and inorganic PM formation, and that our MB estimates do not capture the full spectrum of impacts seen through species other than O\textsubscript{3}.

Adjoint estimation of NO\textsubscript{x} MBs is based on the definition of a scalar adjoint cost function, \( J \), as follows.

\[
J = V_{SL} \sum_t \sum_{t'} M_{0\omega} P_{\omega} \left( 1 - e^{-\beta \Delta C_{t\omega}} \right) \quad (1)
\]

Detailed description of the application of equation (1) in the adjoint model is provided elsewhere.\textsuperscript{6} For MB estimation, \( J \) is the monetized mortality count in the U.S. population attributable to short term O\textsubscript{3} exposure over May 1 – September 30, i.e., the 2007 O\textsubscript{3} season ($); \( V_{SL} \) is the value of a statistical life, estimated to be $7.9 million in 2008 USD;\textsuperscript{32} \( M_{0\omega} \) is the 2007 all-age, non-accidental mortality rate in location \( \omega \) (yr\textsuperscript{-1}, scaled to a daily rate); \( P_{\omega} \) is the 2007 all-age population in location \( \omega \), both of which are reported by the Centers for Disease Control...
and Prevention (CDC) at the county level; $\beta$ is the effect estimate derived from epidemiological studies; and $\Delta C_{o\omega t}$ is the change in DM8A O$_3$ concentration at time $t$ and location $\omega$, with respect to a reference concentration of zero. We apply a $\beta$ of $4.27 \times 10^{-4}$ ppb$^{-1}$ for DM8A O$_3$ due to its wide coverage of populations across the U.S.$^{33}$

We construct MB curves for 1 ton of emitted NO$_x$ using various U.S.-wide emission abatement scenarios. We use emission inventories for the O$_3$ season of 2007 as our baseline of comparison. Abatement scenarios are defined by U.S.-wide, fixed-percentage reductions in (a) mobile (onroad and nonroad) or point source emissions (e.g., a 20% reduction in all mobile source emissions only), or (b) both mobile and point source categories simultaneously (e.g., a 20% reduction in all mobile and point source emissions). Scenarios of 20, 40, 60, 80, and 100% reductions in emissions of all species from either source category are used. For each scenario, 2007 emissions are perturbed by a specified percentage in the forward CMAQ model. Concentration outputs from the forward model are used to calculate a new set of adjoint forcing terms (details are available elsewhere)$^6$ and for calculating adjoint-based MBs in the backward model.

We categorize MBs for NO$_x$ emitted from (1) any mobile source and (2) any point source in a given grid cell in the domain. We report MBs for 1 ton of NO$_x$ emitted over the O$_3$ season according to the spatiotemporal (i.e., day-to-day and layer-by-layer) distribution of emissions for any given source location. Mobile source MBs are thus calculated for surface-level emissions only, while point source MBs are proportionally integrated over all vertical model layers whose emissions are non-zero, according to

$$MB_{o\omega} = \frac{\sum_t \sum_z \frac{\partial J}{\partial e_{o\omega t}} e_{o\omega t}}{\sum_t \sum_z e_{o\omega t}}$$

(2)
where \( MB_\omega \) is the NO\(_x\) MB for a mobile or point source in location \( \omega \). MBs for a given grid cell are emission-weighted averages over all vertical model layers, \( z \), and all simulation times, \( t \), in that location. Adjoint sensitivities, \( \partial f / \partial e_{\omega zt} \), are outputs of the CMAQ-adjoint model and indicate the sensitivity or response of U.S.-wide mortality to NO\(_x\) emitted in location \( \omega \), at layer \( z \), for time \( t \). Adjoint sensitivities are scaled to amount to the influence of 1 ton of NO\(_x\) emitted over the O\(_3\) season. We note that equation (2) applies only to the first layer for mobile sources, but is integrated over all vertical model layers for point sources. As the adjoint method provides sensitivity information specific to each location, layer, and time of emission release, the distinction between mobile and point source MBs comes from emission weighting in equation (2). When depicting point source MBs, we apply a filter to exclude small point sources (NO\(_x\) emissions < 100 ton/season).

We note that adjoint-based MBs indicate how emissions generated in one location contribute to a change in nationwide O\(_3\) exposure somewhere along their trajectory, but that the adjoint method cannot specify where such changes in exposure occur within the boundaries of the U.S. (as defined by the adjoint cost function in equation (1)). Our estimations of NO\(_x\) MBs are therefore representative of the nation-wide public health benefit in the U.S. attributed to a 1 ton reduction in NO\(_x\) from a given source.

RESULTS AND DISCUSSION. Mobile and point-MBs across the U.S. are estimated to average $13,200 and $14,100/ton, respectively, at baseline 2007 emission levels (Figure 1A-B). For mobile sources, MBs at baseline 2007 emission levels range from -$86,000/ton to $87,000 per ton of NO\(_x\) emitted near New York, NY and upwind of Los Angeles, CA, respectively (Figure 1A). For point sources, NO\(_x\) MBs range from -$20,000 to $39,000/ton at baseline (Figure 1B). Our estimates at baseline are comparable to those found by others using various photochemical
modeling tools and approaches. For example, Mauzerall et al. used forward sensitivity analysis in an air quality model and found O$_3$-based MBs of $10,700-$52,800/ton for large point sources in the eastern U.S.

Our estimates of mobile- and point-MBs are spatially heterogeneous and show similar behavior despite differences in the vertical layers of emission release (i.e., surface vs. elevated layers). Our findings therefore suggest that location is a stronger predictor of O$_3$-based NO$_x$ MB than source category. We find that positive MBs in Figure 1 are widespread across low-NO$_x$ environments in the U.S. Negative MBs, or disbenefits, are localized in various urban areas and are due to the chemistry of O$_3$ production in NO$_x$-rich (or NO$_x$-inhibited) environments.

The dominant feature in Figure 1 is the widespread increase in NO$_x$ MBs towards higher levels of abatement. Without exception, positive MBs become more positive and MBs that are initially negative (i.e., disbenefits) become less so – and eventually positive – with U.S.-wide reductions in emissions. In other words, as the relative abundance of NO$_x$ declines with added controls, each additional ton of NO$_x$ reduction carries larger benefits than the previous ton. This trend exists at all locations across the domain for both source categories. Such behavior is due to the role of NO$_x$ availability in O$_3$ production. When NO$_x$ is abundant, competition between NO$_x$ molecules is high, yielding a small impact of increased NO$_x$ availability on O$_3$. As less NO$_x$ becomes available for reactions to produce O$_3$, additional NO$_x$ molecules face little competition and have higher O$_3$ formation efficiency, yielding larger MBs.

Depiction of MB as a function of emission reduction (abatement) level (a MB or MD curve) yields insight about the predicted benefits of added controls. A mix of MB curves for select, individual urban areas and point sources in the U.S. demonstrate the spectrum of behavior seen across different chemical environments in varying proximity to population centers (Figure 2).
Mobile source MBs (Figure 2A-B) and point source MBs (Figure 2C-D) rise invariably, monotonically, and nonlinearly as nation-wide emission levels decline from the 2007 baseline. MBs increase by 2-30 times their initial value, and by as much as $169,000/ton with continued abatement of both source categories. The nonlinearity in the total damage function implied in these plots indicates a changing atmospheric regime as the abundance of NO$_x$ progressively declines. Such a shift can eventually amount to a change in MB sign (i.e., from negative to positive) for environments that are initially NO$_x$-inhibited. One example is Los Angeles (LA in Figure 2A-B), whose mobile-MB at baseline is estimated to be -$17,000/ton and grows rapidly to $152,000/ton with 100% abatement (Figure 2B). Given that vehicles are by far the dominant source of anthropogenic emissions in and around LA, its MB is very sensitive to mobile source abatement (Figure 2A). MB behavior depicted for LA is among the most extreme of any source across the U.S. due to (1) the initially NO$_x$-inhibited environment necessitating a transition through the O$_3$ ridge into a NO$_x$-limited regime with abatement, (2) the large populations in and downwind of LA, and (3) the lack of large point sources in the region that lends little change in MB with point source abatement (Figure 2A). The spectrum of behavior depicted in Figure 2 shows, without exception, that as NO$_x$ approaches background levels, changes in MBs become more drastic with each additional unit of abatement.

System-wide average MB curves represent the overall response of O$_3$ health damages in the U.S. population to a 1 ton reduction in NO$_x$ from an average emitter. We calculate system-wide average mobile- and point-MB curves for separate and combined reductions in source categories (Figure 3). System-wide average MBs are calculated using emission-weighted averaging of MBs in Figure 1 for all sources. On an aggregate level, MBs are positive, upward-sloping, and rise from baseline monotonically and nonlinearly with NO$_x$ emission controls of increasing intensity.
Contrary to traditional depictions of MB curves, NO\textsubscript{x} MBs increase substantially as background concentrations are approached, indicating a heightened sensitivity of pristine environments to any added NO\textsubscript{x}. With combined reductions in both mobile and point source categories, NO\textsubscript{x} MBs increase roughly 3-4 times (from $13,000 to $51,000/ton for mobile-MB, and from $14,000 to $45,000/ton for point-MB) after 100% emission abatement. Mobile-MBs are more sensitive to abatement of mobile source emissions, while point-MBs are similarly affected by either type of control.

The prevalent presumption of a downward-sloping MB curve in the environmental economics literature is akin to convexity of the cumulative or total damage curve with respect to abatement. Total damage in this context is the monetized U.S. health burden from O\textsubscript{3} exposure at a given abatement level. Our estimations of MB curves indicate a consistently concave NO\textsubscript{x} total damage curve with compounding benefits towards lower levels of emissions (Figure 4). In other words, the total damage depicted in Figure 4 declines more rapidly towards higher levels of abatement. Past studies\textsuperscript{20,34-35} have assumed that MBs for a specific source do not change with NO\textsubscript{x} emissions, and estimated total damage by multiplying fixed MBs and emissions. This linear approximation of the total damage curve is prone to underestimation as it neglects its curvature as emissions change. Our finding of a strictly concave total damage curve applies to all sources, rather than to specific cases of sources with negative MBs at baseline as suggested previously.\textsuperscript{15}

Further, our findings suggest a smooth and gradual transition in O\textsubscript{3}-based NO\textsubscript{x} benefits across chemical regimes, contrary to discontinuities or instantaneous changes suggested by others.\textsuperscript{1,36}

Closer examination of Figures 3-4 illustrates an important point about nonlinearity and curvature of the total damage function. The benefits of controlling both mobile and point source categories together (solid line in Figure 4) are larger than the summation of benefits incurred
from controlling these sources separately (long-dashed line in Figure 4). This nonadditivity is a result of the concave nature of the NO\textsubscript{x} total damage curve that becomes more pronounced as the overall abundance of NO\textsubscript{x} declines. Combined reductions in both mobile and point sources, together rather than separately, results in a more extreme NO\textsubscript{x}-limited environment where each additional ton of NO\textsubscript{x} gains higher efficiency for O\textsubscript{3} production. In the presence of regional-to-national scale emission controls from many polluters across different sectors, a simple addition to estimate the overall benefits of abatement is likely to underestimate the combined effect. We note that the quantitative results shown in this work are based on emission reduction scenarios that apply nation-wide, fixed percentage reductions in point and/or mobile source emissions. For a specific policy targeting only a subset of sources (e.g., on-road gasoline vehicles), NO\textsubscript{x} MBs would increase with abatement, but at a lower rate. Our results also show that evaluating such policy options in isolation from the larger emission reduction landscape is likely to (significantly) underestimate the benefits of abatement.

Benefit-cost assessment relies on estimates of MB and the cost-per-ton of emission reduction (referred to as marginal [abatement] cost, or MC) as decision-making metrics. Based on economic equilibrium theory, the net societal benefit of a given policy item is highest when MB equals MC (A\textsuperscript{*} in Figure 5).\textsuperscript{1,37-38} At lower abatement levels than this equilibrium point (to the left of A\textsuperscript{*} in Figure 5), there is incentive to further control emissions as the incremental benefit exceeds the cost. At higher abatement levels than A\textsuperscript{*}, rising costs are prohibitive and no longer compensated in full by expected returns. Traditional depictions of this equilibrium point assume a downward-sloped MB curve and an upward-sloped MC curve with abatement. Our findings of an upward-sloping and monotonic NO\textsubscript{x} MB curve challenge the conventional scheme presented in Figure 5 in two important ways. First, if the MB curve is upward-sloping and nonlinear, as in
Figures 2-3, the uniqueness of the equilibrium point, as often presumed in the environmental economics literature, is not guaranteed and will depend on the shape of the total damage and cost curves. Second, in the presence of an upward-sloped MB curve, an economically viable abatement policy at baseline (i.e., MB>MC), would yield a new intersection point that lies at a higher abatement level (A* in Figure 5) than that suggested by a conventionally convex total damage curve. Our results, in most cases, are therefore in support of more stringent emission reduction targets than previously thought to be economically efficient. One example is the emission cap of the U.S. cap-and-trade program. Upward-sloping MB curves, such as those found in this study, would provide economic incentive for a lower system-wide emission cap than previously envisioned. The general shape of the MB curve in Figure 5 is taken from our results (i.e., Figure 3); however, we emphasize that it is a qualitative depiction. Though strictly qualitative, Figure 5 demonstrates that a shift in the economic paradigm, from convexity to non-convexity, would entail an important change in the MB curve, and a correspondingly significant shift in the point of economic equilibrium.

We note that our conclusions apply generally to the overall system and not necessarily to each source individually, as the shapes of MB and MC curves differ from source to source. We also recognize that the MB curves presented here are based on a series of U.S.-wide emission reductions and capture responses of MBs to national rather than local changes in emission patterns. Reductions in emissions from single sources, in most cases, would have little tangible impact on the ambient availability of NOx in the system when other emissions are kept constant. MB curves for single sources are thus expected to be relatively flat compared to the curvature seen in Figures 2-3. Changes in sectoral emissions, such as mobile or electricity generating sources, seldom happen in isolation and commonly materialize within a broader, nationwide
context. As such, we believe that our depiction provides a more realistic and relevant view of MB behavior for decision-making. In the particular case of regulating sources with negative MBs, a broader consideration of system- or sector-wide abatement and resulting benefits is preferable to isolating the impacts of abatement of individual sources. Information garnered from the total damage/benefit curve, such as those in Figure 4, can yield important insight into the cumulative benefits of widespread emission control policies.

The concave (or nonconvex) behavior demonstrated here is for a NO\textsubscript{x} damage function based only on mortality from short-term O\textsubscript{3} exposure. More comprehensive estimation of NO\textsubscript{x} MBs would consider non-fatal health and other environmental impacts of O\textsubscript{3}, particularly endpoints related to long-term exposure. In addition to influencing O\textsubscript{3} production, NO\textsubscript{x} also contributes to secondary PM formation. We note that our O\textsubscript{3}-based MB estimates are comparable in magnitude to estimates based on PM\textsubscript{2.5}. Fann et al.\textsuperscript{4} used reduced-form air quality modeling to estimate NO\textsubscript{x} MBs that account for chronic PM\textsubscript{2.5} exposure mortality and morbidity in the U.S. population. In the study, NO\textsubscript{x} MBs averaged $10,000/ton for mobile sources and $9,700-15,000/ton for point sources over 9 urban areas of the U.S. A later, more detailed study by Fann et al.\textsuperscript{19} employed source apportionment in an air quality model to estimate MBs (termed benefits-per-ton) and found lower estimates averaging $4,500/ton and $3,700/ton for mobile sources and power plants in the continental U.S. Although the overall public health burden of PM\textsubscript{2.5} is larger than that of O\textsubscript{3}, our comparison suggests that NO\textsubscript{x} emissions may incur as much or more damage through O\textsubscript{3} in the short term as in the long term through PM\textsubscript{2.5}. MBs that include long-term health impacts of O\textsubscript{3}\textsuperscript{41} are likely to be significantly larger than our estimates, and would thus have a dominant share of the total benefits of NO\textsubscript{x} control. Non-convexity induced by O\textsubscript{3} would therefore likely extend to non-convexity in the overall damage curve.
Though no studies have fully tested the assumption of convexity as applied to PM health damages, indications of two forms of non-convexity exist in the literature. The first is non-convexity due to the role of chemical equilibrium in formation of secondary inorganic PM constituents from NO\textsubscript{x}. Fann et al.\textsuperscript{19} reported consistently higher MBs for all inorganic PM precursor emissions under a 2016 abatement scenario compared to estimates for 2005 emission levels. It is noteworthy that the authors found (slightly) increased MBs even for primary emissions of PM, possibly due to nonlinearity induced by other species through aerosol growth and dynamics. Holt et al.\textsuperscript{42} compared PM sensitivities to NO\textsubscript{x}, SO\textsubscript{2}, and NH\textsubscript{3} emissions in 2005 and 2012 and found that for SO\textsubscript{2} and NO\textsubscript{x}, sensitivities increase with emission controls. Zhang et al.\textsuperscript{43} used the high-order direct decoupled method (HDDM)\textsuperscript{44} to estimate 2\textsuperscript{nd} order derivatives of PM with respect to precursor emissions, including NO\textsubscript{x}. They found mostly negative 2\textsuperscript{nd}-order HDDM sensitivities, indicative of a concave response surface.

In addition to non-convexity in the atmospheric response of PM to NO\textsubscript{x}, recent studies have suggested that unlike O\textsubscript{3},\textsuperscript{45} a non-linear and concave concentration-response function may be more suitable for PM\textsubscript{2.5},\textsuperscript{46-47} implying an epidemiologically induced non-convex damage curve.\textsuperscript{48-50} A supralinear or concave curve implies a large slope, or high incremental risk per unit concentration, at low levels of exposure that diminishes towards higher concentrations. Such a shape of the concentration-response function indicates a heightened sensitivity of populations to PM in cleaner environments. Combined with the likely non-convex atmospheric response of PM to NO\textsubscript{x}, persisting, or even enhanced concavity, may be expected with inclusion of PM in the damage function. Future research is required to disentangle the interactions between these two sources of non-convexity for PM.
Our MB estimates are affected by uncertainties stemming from atmospheric modeling and emissions characterization, population demographics, epidemiological concentration-response relationships, and economic valuation of damage endpoints. Firstly, we estimate MB curves at a 36-km horizontal resolution that may not capture fine spatial gradients in O$_3$ exposure, particularly over urban and suburban areas. Second, we use population and mortality data for 2007 without considering dynamic changes in population that may become relevant into the future. Third, we apply a uniform effect estimate to the entire U.S. population, while recognizing that effect estimates may vary by region. We also assume a linear, no-threshold response of mortality to O$_3$ exposure based on the current epidemiological literature. Alternate forms of the concentration-response function would affect NO$_x$ MB estimation along the abatement trajectory. Fourth, while we assign a uniform value of a statistical life for valuating public health impacts, this willingness-to-pay may differ among subgroups of the population and shift as pollution levels and consumer preferences change. We note that we use 2007 emissions as our reference point, and MBs at current emission levels, or those under planned policies, may differ from estimates reported here, particularly given the progressive post-2007 emission reductions that have taken place. We also note that our estimates of NO$_x$ MBs consider the impact of NO$_x$ control on the U.S. population only. In reality, emissions generated within the U.S. may also impact public health in other nations, and thus marginal reductions in emissions may have additional monetary benefits not captured here. Interpretation of our results should consider these uncertainties and limitations of our analysis.

Our findings suggest compounding benefits for progressive NO$_x$ emission reductions. The benefit of urban NO$_x$ control has been debated for cities with negative MBs at current emission levels, where localized emission reductions appear unfavorable in the short-term.
Compounding benefits with added NO$_x$ control on a broader scale support continued NO$_x$ abatement in the longer term for urban air quality management. Strictly concave total damage functions and upward-sloping MB curves with abatement, such as those found here, suggest larger yet unexplored economic incentives for more aggressive emission reductions.

As discussed earlier, our findings of non-convexity related to NO$_x$ and O$_3$ are likely to extend to inorganic PM and its precursors such as SO$_2$. Given the challenges that O$_3$ and PM pose to air quality management in North America and the world over, we believe that the notion of generally non-convex behavior for secondary pollutants such as O$_3$ and inorganic PM has important policy implications. Reported emission trends from the U.S. EPA suggest that anthropogenic NO$_x$ emissions have decreased by more than 30% from 2007 to 2014. Based on our results, this level of reduction could place us on the onset of an important point in time and on the MB curve, where NO$_x$ MBs can increase significantly in the near future (Figure 4). In such a policy context, adhering to the traditional view of convexity and disregarding the compounding nature of NO$_x$ control benefits does not appear to be a prudent option.
Figure 1. Simulated MBs for NO\textsubscript{x} emitted from mobile sources (left panel) and point sources (right panel) across the U.S. MBs are shown for baseline 2007 emission levels (A-B) and for U.S.-wide abatement of all species emitted from both mobile and point source categories in amounts of 40% (C-D), 60% (E-F), 80% (G-H), and 100% (I-J). MBs are for 1 ton of NO\textsubscript{x} emission allocated over the 2007 O\textsubscript{3} season (May-September) according to the spatiotemporal
distribution of emissions. MB values are only shown for point sources (B, D, F, H, J) whose emissions are more than 100 ton/season at baseline.
**Figure 2.** Simulated NOx MBs as a function of U.S.-wide abatement level for a sample of source locations. Mobile-MBs (A-B) are the benefits associated with reductions in NOx emitted from mobile sources within the specified city (Atlanta (ATL), New York (NY), Detroit (DET), Los Angeles (LA)). Point-MBs (C-D) are the benefit associated with reductions in NOx emitted from an anonymous, major point source in the specified state (NM, PA, AL, TX). Hatched and dashed lines (A, C) depict MBs for 0-100% abatement of all species emitted from mobile or point sources, respectively, across the U.S., as compared to 2007 levels. Solid lines (B, D) show the same for simultaneous reductions in both mobile and point sources. For example, the “LA-point” dashed line in (A) shows mobile-MBs at different levels of U.S.-wide point source abatement.

**Figure 3.** Average U.S.-wide mobile- (A) and point-MB curves (B) for various levels of U.S.-wide abatement of all species emitted from mobile and point sources. Average MB curves are depicted as a function of mobile source abatement (hatched line) and point source abatement.
(dashed line) separately. Solid lines depict the combined rise in MBs from controlling both source categories simultaneously. MBs shown here are emission-weighted averages over the U.S.

**Figure 4.** Total U.S.-wide damage as a function of U.S.-wide abatement of mobile sources (hatched line), point sources (short-dashed line), and both simultaneously (solid line). Total damage is the monetized health burden of mortality attributable to short-term O$_3$ exposure of the U.S. population, calculated at each abatement level. Total benefit shown on the secondary axis is the avoided health damage in moving from the 2007 baseline to lower emission levels. The long-dashed line depicts total benefits as the summation of benefits incurred from controlling mobile and point source emissions separately (i.e., the summation of benefits for the short-dashed and hatched lines). Total damage accounts for O$_3$ exposure during the 2007 O$_3$ season (May-September).
**Figure 5.** Depiction of the economic equilibrium point ($A^*$) between MB and MC (dashed line) based on traditional forms of MB curves (hatched line) and our findings (solid line). We consider 2007 as our baseline and the starting point for MB curves. Curves shown here are qualitative and for demonstrative purposes only and are based on the general shape of system-wide average curves in Figure 3. Note that the baseline-level MC is often less than the MB, and changes in the shape of either curve will affect where the points of equilibrium lie.

**ASSOCIATED CONTENT**

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ABBREVIATIONS

MB, marginal benefit; MD, marginal damage; VOC, volatile organic compound; CMAQ, Community Multiscale Air Quality model; NEI, National Emission Inventory; NPRI, National Pollutant Release Inventory; SMOKE, Sparse Matrix Operator Kernel Emissions model; WRF, Weather Research and Forecasting model; MCIP, Meteorology Chemistry Interface Processor; MFE, mean fractional error; MFB, mean fractional bias; DM8A, daily maximum 8 h average; CDC, Centers for Disease Control and Prevention; MC, marginal cost; PM, particulate matter.

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