Title: Impact of natural gas development in the Marcellus and Utica Shales on regional ozone and fine particulate matter levels

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ABSTRACT

The Marcellus and Utica shale formations have recently been the focus of intense natural gas development and production, increasing regional air pollutant emissions. Here we examine the effects of these emissions on regional ozone and fine particulate matter (PM_{2.5}) levels using the chemical transport model, CAMx, and estimate the public health costs with BenMAP. Simulations were performed for three emissions scenarios for the year 2020 that span a range potential development storylines. In areas with the most gas development, the ‘Medium Emissions’ scenario, which corresponds to an intermediate level of development and widespread adoption of new equipment with lower emissions, is predicted to increase 8-hourly ozone design values by up to 2.5 ppbv and average annual PM_{2.5} concentrations by as much as 0.27 µg/m³. These impacts could range from as much as a factor of two higher to a factor of three lower depending on the level of development and the adoption of emission controls. Smaller impacts (e.g. 0.1-0.5 ppbv of ozone, depending on the emissions scenario) are predicted for non-attainment areas located downwind of the Marcellus region such as New York City, Philadelphia and Washington, DC. Premature deaths for the ‘Medium Emissions’ scenario are predicted to increase by 200 to 460 annually. The health impacts as well as the changes in ozone and PM_{2.5} were all driven primarily by NO_x emissions.

Keywords: PM_{2.5}; ozone; shale gas; energy; air quality; chemical transport modeling; health effects

1. INTRODUCTION

The Marcellus and Utica shales are rock formations located in the Appalachian Basin; they contain large natural gas reserves and recently have been the focus of intense drilling and leasing activity.^{1,2} Natural gas development, production, and processing activities can be a significant source of air pollution.³ For example, equipment involved in setting up a well such as trucks, drill rigs, and hydraulic fracturing pumps are powered by heavy duty diesel engines that emit oxides of nitrogen (NO_x), volatile organic compounds (VOCs) and fine particulate matter (PM_{2.5}).^{4,5,6,7} Natural gas fired compressors emit NO_x and VOCs.⁸ Venting and fugitive emissions can also be a significant source of VOCs.^{5,9}

In the atmosphere, VOCs and NO_x react in the presence of sunlight to produce ozone, which causes several health problems such as asthma and decreased lung function.¹⁰ NO_x emissions also lead to the formation of PM_{2.5} nitrate that, along with primary emissions of PM_{2.5}, leads to an increase in fine particulate concentration in the atmosphere that can ultimately result in decreased life expectancy.¹¹

Field and modeling studies have also shown that the aggregate emissions from natural gas activities, including shale gas, can have important impacts on local and regional air quality. Schnell et al.¹² measured peak 1-hr ozone levels as high as 100 ppbv in Wyoming. Elevated VOC levels were also found in and around gas-producing regions of Colorado, Utah, Pennsylvania and West Virginia.^{5,13-15} Kemball-Cook et al.⁹ used a chemical transport model to predict that gas development in the Haynesville Shale could increase the maximum daily 8-hr average (MDA8) ozone levels by as much as 17 ppbv over parts of Louisiana and Texas. Many counties in and around the Marcellus region currently violate the National Ambient Air Quality

Standards (NAAQS) for ozone and PM_{2.5},¹⁶ and gas development in the Appalachian basin may complicate these existing problems.^{2,17} Litovitz et al.¹⁸ estimated that the air-quality-related damages in 2011 from Marcellus-related activity in Pennsylvania, including emissions of NO_x, SO_x, VOCs, and PM, ranged from \$7.2 million to \$32 million.

This work investigates the impacts of Marcellus and Utica (hereafter referred to as Marcellus-Utica or MU) development on regional ozone and PM_{2.5} levels by implementing an updated version of the Marcellus emissions inventory developed previously in a 3-D chemical transport model.³ Specifically, we account for impacts resulting from emissions of NO_x, VOCs, and primary PM_{2.5} from natural gas production, including condensate and wet gas, but not from oil production, which has been small thus far in the Utica formation. Different emission scenarios are evaluated corresponding to different levels of development and/or air pollution control. The health impacts of the predicted changes in air quality, namely additional mortality stemming from increases in PM_{2.5} and ozone, are evaluated using the BenMAP model. Non-mortality health end points were not considered. Emissions of air toxics and their potential health effects were not considered due to insufficient data.

2. METHODS

A series of simulations were performed with different emission inventories to assess the potential impacts of Marcellus and Utica (MU) shale gas activities on regional air quality (Table 1). All simulations were performed using 2005 meteorology for a full year to capture seasonally varying sensitivities to MU emissions. The MU emissions scenarios are based on projected MU activities for 2020. The year 2020 was chosen because it is far enough in the future that a significant amount of development activity is expected to have occurred by then and also due to the availability of scaling factors for non-MU emissions for that year. While existing regulations on non-MU emissions dramatically lower predicted ozone and PM_{2.5} concentrations in 2020 compared to 2005, this study focuses on regional air pollution caused by MU activity.

Table 1. List of simulations and emissions

Simulation name	Non-Marcellus-Utica emissions	Marcellus-Utica (tons day ⁻¹)		
		NO _x	VOC	EC*
2005	2005 emissions	0	0	0
2020 No-MU (No Marcellus-Utica)	2005 emissions scaled to 2020	0	0	0
High Emissions	2005 emissions scaled to 2020	212	282	4.2
Medium Emissions	2005 emissions scaled to 2020	112	76	2.5
Low Emissions	2005 emissions scaled to 2020	38	32	1.5
Marcellus-Utica NO _x only	2005 emissions scaled to 2020	112	0	2.5
Marcellus-Utica VOC only	2005 emissions scaled to 2020	0	76	2.5

*Elemental carbon

2.1. Chemical transport modeling and non-Marcellus emissions

A 3-D chemical transport model, CAMx (version 5.41), was used to simulate ozone and PM_{2.5} concentrations over the continental United States. The modeling domain was discretized into a 36 km by 36 km grid with 14 vertical layers up to a height of 16 km. Details of the model are described in the CAMx User's guide.¹⁹ Briefly, CAMx simulates advection, dispersion, gas- and particle-phase chemistry, emission, wet and dry deposition, and aerosol microphysical processes. Chemistry was simulated using the Carbon Bond Chemical Mechanism 2005 (CB05).²⁰ The simulations used meteorology fields generated by the Weather Research and Forecasting (WRF) model for the year 2005.²¹ The chemical boundary conditions were taken from the output of a global chemical transport model, GEOS-Chem.²²

Emissions from all non-Marcellus and Utica (non-MU) sources are from an inventory developed by Ramboll Environ Inc. for a regulatory impact assessment conducted by the US EPA.²³ The Ramboll Environ inventory is for 2005 and takes the US National Emissions Inventory (NEI) version 2 as its starting point. CAMx has previously been evaluated for both ozone and PM_{2.5} using this inventory at a finer grid resolution (12 km x 12 km) than used here.²⁴ We supplement those results with evaluations using our 36 km x 36 km grid; details are in the online supplemental material (Section S.1). In summary, the model meets the performance standards for modeling ozone recommended by the EPA and compares favorably with other models.^{25,26} For PM_{2.5}, model performance was within the recommended thresholds of Boylan et al.²⁷ Bias and error metrics for PM_{2.5} and ozone at our 36 km resolution were very similar to those for the finer 12 km grid.

We investigated the effects of MU emissions in the year 2020. Therefore, we re-scaled the 2005 emissions inventory for all non-MU sources to 2020 using EPA NEI data and projections for the mid-Atlantic/northeastern United States compiled by the Mid-Atlantic Regional Air Management Association (MARAMA).²⁸ The scaling factors account for effects of control strategies promulgated by the US EPA, such as the phasing in of Tier 4 for the fleet of diesel engines and the Transport Rule. They are sector-specific and uniformly applied across space and time. Further details are discussed in Section S.3.

The non-MU emission scaling factors are listed in Table S.1. Between 2005 and 2020, overall non-MU area source (stationary and mobile) NO_x emissions are reduced by 56%, SO₂ emissions by 72%, and VOC emissions by 28%. Over the same period, point source emissions, NO_x emissions are reduced by 37% and SO₂ by 82%. Primary PM_{2.5} emissions are reduced by 20%. No scaling factors were applied to the biogenic emissions.

2.2 Marcellus-Utica (MU) emissions and simulation scenarios

Table 1 summarizes the simulations performed to evaluate the impact of MU activities on regional air quality. This section describes how those emissions estimates were generated. The first of the future simulations was a 2020 'No-Marcellus-Utica' (no-MU) simulation to calculate a baseline against which to compare the simulations with MU emissions. MU emissions are then added onto the No-MU 2020 inventory and the impacts of the MU emissions on ozone and PM_{2.5} concentrations were determined by difference between CAMx simulations with and without MU emissions.

The emission inventories for MU activities are based on per-unit-activity emission factors developed by Roy et al.³ for well development, gas production, and midstream processing in the Marcellus region. The interested reader is referred to that paper for additional details regarding relative contributions of specific contributing activities. Briefly, Roy et al.³ uses a bottom-up approach to estimate emissions of NO_x, VOCs, and primary PM_{2.5} for the Marcellus region from well development, gas production, and midstream activities. Per-unit-activity emission factors include contributions from several sub-categories of equipment and activities including compressor stations, wellhead compressors, trucking, fracing, drilling, completion, pneumatic devices, condensate tanks, etc. Roy et al.³ uses EPA AP-42 data for some activities but goes beyond this by including measured emission factors from in-use equipment available from published literature, deriving results specific to the Marcellus region, including an explicit uncertainty analysis, accounting for the effects of future regulations and fleet turnover on future emissions, and deriving results per unit gas production or well development. They found that drilling, hydraulic fracturing, compressor stations, and completion venting were the major sources of air pollutants from Marcellus activities.

We combined the Roy et al.³ process-level emission factors with activity data to develop three different emissions scenarios for MU activities: Low, Medium, and High Emissions (Table 1). Storylines for each scenario are described below to demonstrate their plausibility and to provide a context for their interpretation. These scenarios designed to represent a range of potential gas development activity and emissions controls for 2020. In each case, the same emissions could be obtained with other combinations of activity levels and emissions factors. As shown later, predicted ozone and PM_{2.5} concentrations vary almost linearly with changing NO_x emissions between these three scenarios. Therefore, estimates for additional emissions scenarios can be obtained by simply interpolating among the three defined here.

The Medium Emissions scenario corresponds to the median 2020 process-level emission factors from Roy et al.³ and EIA projections of natural gas production in 2020. The Roy et al.³ median emission factors account for implementation of the EPA's Oil and Gas Rule,²⁹ the Tier 4³⁰ standard for off-road diesel engines, and the 2007/2010 standard for on-road heavy-duty diesel vehicles assuming high fleet turnover -- 75% for drill rigs, frac pumps, and trucks and 98% for compressor stations and wellhead compressors. Therefore, the majority of the sources in the Medium Emissions scenario comply with the latest emission standards. The Medium Emissions scenario assumes 17.4 billion cubic foot per day (bcf) of gas production in the Marcellus shale and 4 bcf in the Utica shale in 2020, which are based on the EIA predicted¹ national gas production in 2020 and the fraction of current production in Marcellus and Utica shales (22% and 5% of total dry gas production in the United States).² The Medium Emissions scenario assumes that the number of wells drilled in 2020 in each state is the same as the average number drilled in 2012-2014. Over this period well development was relatively steady with an average of 1400 wells drilled per year in Pennsylvania, 225 in West Virginia, and 500 in Ohio.^{31,32,33}

The High and Low Emissions scenarios were designed to span a range of potential combinations of future well development, gas production, and emission controls. Both of these scenarios correspond to plausible development scenarios. For example, the High Emissions scenario would occur if there were a combination of high-level of MU activity (3000 wells drilled per year and 23.8 bcf gas produced versus 2125 wells and 21.4 bcf in the Medium Scenario) and

slower fleet turnover. This level of well development corresponds to the peak drilling observed in each state since the end of the US economic recession in 2009; this level of gas production corresponds to the “High Oil and Gas Resource” scenario defined by the EIA.¹ The High Emissions scenario has slower fleet turnover than the Medium Emissions scenario -- 20% turnover for drill rigs, frac pumps, and trucks and 30% for compressors from 2009-compliant equipment. Therefore the High-Emissions scenario has somewhat higher process-level emission factors and a higher level of MU activity than the Medium Emission scenario.

The Low Emissions scenario represents a lower level of activity (1350 wells drilled and 20.5 bcf gas produced) with a fleet turnover rate of 100% for all 2009-compliant equipment. Additionally, it assumes that 75% of the compressors and 40% of the drill rigs, frac pumps and trucks have adopted state-of-the-art control technology, which exceeds what is required by existing regulations. This number of wells drilled corresponds to the lowest period of well development in each state since 2009.^{31,32,33} This level of production corresponds to the “Low Oil Price” scenario defined by the EIA.² Therefore the Low-Emissions scenario would if occur if there was both less MU development and much stricter emission controls than the Medium Emission scenario.

Two additional simulations were performed to investigate the sensitivity of the model predictions to MU VOC and NO_x emissions. This was done by removing the VOC emissions entirely and only applying the Medium Emissions NO_x emissions or vice-versa.

Lacking detailed projections for the spatial distribution of future development, the MU emissions were spatially allocated assuming that future development would follow a similar spatial pattern to the development to date. First, the MU region was defined within the states of Pennsylvania, Ohio, and West Virginia. The projected 2020 MU emissions for each state were then distributed within each state based on the number of presently active wells within each grid cell with the results shown in Figure S1.^{31,32,33} Areas of highest emissions are evident in Figure S1 in eastern Ohio, southwestern Pennsylvania, and northeastern Pennsylvania, corresponding to areas that have seen significant development already. With these assumptions, 65% of the 2020 MU NO_x emissions occur in Pennsylvania and 12% and 23% occur in West Virginia and Ohio, respectively. We also performed a sensitivity simulation in which MU emissions had a uniform spatial distribution throughout the Marcellus fairway. In that simulation (not shown), PM_{2.5} and ozone impacts were broadly similar, but the maximum impacts were ~25% lower high activity areas of southwestern Pennsylvania, adjacent areas of West Virginia and Ohio, and northeastern Pennsylvania. In contrast, impacts in central Pennsylvania were similarly higher. Emissions are assumed to occur uniformly throughout the year. Speciation profiles for VOC emissions from condensate tanks are an average of profiles taken from Hendler et al.³⁴ Otherwise, speciation profiles for VOC emissions, which are primarily from pneumatics, compressor stations, completion venting, gas plant and transmission, are based on EPA’s SPECIATE database.

2.3 Air quality metrics

Ozone impacts were quantified using the Maximum Daily 8-hr average (MDA8) mixing ratio of ground-level ozone, which is calculated by taking a running 8-hr average of CAMx ozone output and then picking the maximum amongst these values for each simulation day. The current

National Ambient Air Quality Standard (NAAQS) for ozone was recently revised to an MDA8 of 70 ppb³⁵ after a review by the Clean Air Scientific Advisory Committee (CASAC) that recommended that the ozone standards be tightened to somewhere between 60-70 ppb.³⁶

Another metric considered was the ozone design value. Observed design values correspond to the 3-year average of the 4th-highest measured MDA8 value for a given monitor. Base case (2005) design values are published online by the US EPA³⁷ and were used as a starting point for estimating future design values using the relative response factor method prescribed by the US EPA.³⁸ The EPA protocol is especially useful since 3-year simulations would be computationally intensive. For each of the 2020 scenarios (No-MU, Low, Medium, and High), estimated future design values are calculated as the product of the base case (2005) observed design value and a relative response factor computed by the model. For each future scenario and monitor site, the relative response factor is the modeled ratio of average MDA8 ozone levels in that scenario to the same for the 2005 base case. Those modeled MDA8 levels are time averages over a set of 'high ozone' days defined for each monitor. 'High ozone' days were selected for all monitored locations in New York, Pennsylvania, Ohio, West Virginia, New Jersey, Maryland and District of Columbia using appropriate thresholds as described in the US EPA guidance document. This was possible since every site satisfied the minimum threshold requirement in 2005, i.e. having at least five days with MDA8 values greater than 70 ppb. A recent updated draft of the same guidance report (that is still under review) recommended selecting the ten highest days at each site regardless of a threshold.³⁹ Results from the proposed method (not shown here) predicted slightly larger changes for counties with a higher base design value, further aggravating the impacts especially in non-attainment counties.

Impacts on PM_{2.5} concentrations were quantified using mean annual concentrations of total PM_{2.5} mass. The current annual primary and secondary NAAQS for PM_{2.5} are 12 µg/m³ and 15 µg/m³ respectively.³⁵ Species-wise contributions were also studied to assist in the development of regulatory solutions.

2.4 Estimating public health effects

The premature mortality associated with PM_{2.5} and ozone formed from MU emissions of NO_x, primary PM_{2.5}, and VOCs was estimated using BenMAP-CE software.^{40,41} Emissions of air toxics and their effects were not considered. The method involves two steps: the estimation of premature deaths based on concentration-response relations from epidemiological studies and their valuation using the Value of a Statistical Life (VSL). Other health endpoints were not analyzed because mortality costs generally dominate other costs.⁴⁰ Monte-Carlo simulations are done to explore the uncertainties surrounding the concentration-response relations and VSL.

The changes in premature deaths were estimated for changes in PM_{2.5} (annual average) and O₃ (MDA8) concentrations between scenarios (No-MU, High, Medium, and Low Emissions). For PM_{2.5}, concentration-response relations from Krewski et al.⁴² and Lepeule et al.⁴³ were used, which are the latest follow-up studies from two landmark cohort studies on PM_{2.5} health effects. For ozone, two studies were used: Bell et al.,⁴⁴ an analysis of the National Morbidity, Mortality, and Air Pollution Study, and Levy et al.,⁴⁵ a meta-analysis. Other ozone concentration-response functions fall between the two used here.⁴⁶⁻⁴⁹ BenMAP population projections for 2020 were used. Baseline mortality rates for 2020 were based on a series of Census Bureau mortality projections.⁴¹ The valuation of the estimated changes in mortality relied on a Weibull distribution

of VSL based on 26 VSL studies, which is recommended by U.S. EPA.⁵⁰ Adjusted for income level in 2020 using U.S. EPA's official adjustment factors provided in BenMAP, the distribution has the mean value of \$9.6 million (in 2010 USD).

3. RESULTS

3.1 Ozone Impacts

3.1.1 Spatial impacts on MDA8 ozone

Figure 1 shows maps of predicted MDA8 ozone mixing ratios averaged across the entire ozone season (March – October). Figures 1(b)-1(d) illustrate the impacts of various MU emission scenarios on ozone levels in 2020, each map showing the difference in ozone-season (March – October) average MDA8 between each MU scenario and the 2020 No-MU simulations. Impacts of at least 0.2 ppbv are predicted in all three scenarios in most of Pennsylvania, southern New York, eastern Ohio and northern West Virginia. The largest increases in ozone levels are predicted to occur in northeastern and southwestern Pennsylvania and eastern Ohio where most

of the MU-related activity is taking place, but some impacts occur downwind in major East Coast cities. Within the modeling domain, the greater sensitivity of rural areas to emissions from MU activity is likely due to their NO_x-limited nature.

Figure 1: Mean MDA8 ozone mixing ratios (ppbv) during the ozone monitoring season (March - October) in the MU shale region for: (a) the No-MU case in 2020. Panels (b) - (d) show the differences between the No-MU base case simulation and the three MU emission scenarios: (b) Low Emissions (c) Medium Emissions (d) High Emissions.

As seen in Figure 1(c), the Medium Emissions scenario increases the ozone season average MDA8 ozone by 0.5-0.8 ppbv across much of the Marcellus Fairway. Figure 1(b) shows that the maximum ozone impacts for the Low Emissions scenario over the same area are ~0.3 ppbv, thus highlighting the potential air quality benefits of lower levels of MU development in the region and tighter emission controls. Similarly, the ozone impacts for the High Emissions scenario are nearly twice as severe as those in the Medium Emissions.

The ozone impacts shown in Figure 1 are considerably less severe than those predicted by Kembell-Cook et al.⁹ for the Haynesville region. This difference is likely due to a number of factors. First, there are several notable differences in how they developed their emissions inventory versus the one used here. For example, they did not assume any controls on midstream sources like compressor stations, which contribute a significant fraction of NO_x emissions. Additionally, Kembell-Cook did not account for the effects of the EPA's Oil and Gas Rule²⁹ since it was not implemented at that time. The Kembell-Cook low and high emissions scenarios for 2012 have 61 and 140 tons day⁻¹ of NO_x from gas development activities, which is similar to the 129 tons day⁻¹ emitted in our Medium Emissions (2020) scenario. However, the Kembell-Cook emissions are more concentrated in the Haynesville region, which is more than 10 times smaller than the MU. Moreover, we expect that the impact of NO_x emissions depends strongly on the pre-existing VOC/NO_x ratios resulting from the baseline (without gas development) emissions.

3.1.2 Impacts on ozone design values

To understand the predicted ozone impacts from a regulatory perspective, future ozone design values were estimated for selected urban and rural sites (Figure 2) using the methods described above. The projections were done for the 2020 No-MU case and for the three MU emission scenarios. By definition, changes in design values are larger than ozone-season MDA8 changes because they represent effects during high ozone episodes when ozone levels are more sensitive to changes in emissions. Overall, the results indicate that MU development could result in pronounced ozone changes in regions with significant gas development. Long-range transport from the MU region could still affect downwind nonattainment regions including New York, Philadelphia and Washington, DC.

The greatest impacts occur in counties located in and around regions with high MU activity, such as northeastern and southwestern Pennsylvania and eastern Ohio. For the Medium Emissions scenario, design values for counties in the Pittsburgh, Cleveland, and Charleston areas are predicted to increase by 1-2 ppbv due to MU activities in 2020 (Figure 2(a)). For the High Emissions scenario, this increases to 2-4 ppbv in the most-affected regions. In Allegheny

County, which is located in southwestern Pennsylvania, the impact of the Medium Emissions case on the ozone design value is 1.7 ppbv while the impact of the High and Low Emissions scenarios are approximately a factor two higher and lower. Smaller impacts, 0.1-0.5 ppbv, depending on the emissions scenario considered, are predicted for non-attainment areas such as New York City, Philadelphia and Washington, DC that are located downwind of the MU region.

Although the rural regions experiencing the most MU development presently meet existing ozone standards, the effect of MU-related activities on ozone levels is predicted to be noticeably larger in these regions than in urban areas. For example, rural counties in southwestern Pennsylvania and eastern Ohio such as Greene County in PA and Washington County in OH show design value

Figure 2: (a) Impacts of Marcellus emissions on future design values for different MU emissions scenarios. Plotted results show the difference between the estimated future design value for the Low, Medium, and High emissions scenarios and the No-MU case in 2020. (b) Projected design values (ppbv) for selected monitored counties shown as the height of the black bar for the No-MU case in 2020. The yellow bars correspond to the change in design value caused by the Medium Emissions scenario. The solid horizontal line represents the current attainment standard of 70 ppbv while the dotted line represents the stricter of the CASAC recommended standards. Both panels are vertically divided into seven metropolitan areas (numbered from left to right): Pittsburgh, Cleveland/Youngstown, Charleston/Morgantown, Buffalo/Rochester, Philadelphia, Washington, DC and New York. Red numbers correspond to regions that are in non-attainment of ozone as of 2015. The eighth panel displays some of the most highly impacted counties. impacts between 1.9 and 2.3 ppbv for the Medium Emissions scenario, while adjacent urban counties such as Allegheny County in PA and Washington county in PA have lower impacts of around 1.5 ppbv. Marcellus development would contribute to non-attainment status in several of these rural counties if a future regulatory review lowered the ozone standard to 60 ppbv, a value that was considered in the most recent review. Figure S2 and Table S6 in the Supplemental Information show the predicted MU impacts on ozone design values for all monitored counties in and around the MU region.

Figure 3: Mean annual $\text{PM}_{2.5}$ concentration ($\mu\text{g}/\text{m}^3$) in the MU shale region for the (a) No-MU case in 2020. Panels (b) - (d) show the differences between the No-MU 2020 simulation and the three MU emission scenarios: (b) Low Emissions (c) Medium Emissions (d) High Emissions.

Cumulative distribution functions (CDFs) of the MU ozone impacts in select locations are shown in Figure S1 to further illustrate the temporal variability of increases in MDA8 values during high ozone days for different MU related emissions scenarios. While the largest impacts are in northeastern and southwestern Pennsylvania and eastern Ohio, similar trends are predicted across the entire MU region. For the Medium Emissions scenario, the largest increases are in the range of 2-2.5 ppbv. Another clear trend is the substantial reduction in the severity of impacts for the Low Emission scenario which corresponds to a future with tighter emissions controls and less MU development.

3.2 $\text{PM}_{2.5}$ Impacts

Figure 3 shows the predicted annual-average $\text{PM}_{2.5}$ concentrations in 2020 for the No-MU emission scenario as well as the $\text{PM}_{2.5}$ increases due to Low, Medium, and High Emissions scenarios. Emissions from MU activities are predicted to appreciably increase $\text{PM}_{2.5}$ levels. For example, the Medium Emissions scenario predicts increases of $\sim 0.25 \mu\text{g}/\text{m}^3$ in parts of northwestern Pennsylvania and eastern Ohio. The maximum impacts range from ~ 0.1 to $\sim 0.4 \mu\text{g}/\text{m}^3$ for the Low Emissions and High Emissions case, respectively, highlighting the substantial effects of the scale of activity and emission controls. Outside of the MU development region, $\text{PM}_{2.5}$ increases tend to be less than $0.1 \mu\text{g}/\text{m}^3$. Nitrate, ammonium, and elemental carbon account for all of the predicted increases in total mean $\text{PM}_{2.5}$ mass concentrations (Figure 4). Across most of the MU region and downwind areas, ammonium nitrate accounts for 80-90% of the predicted $\text{PM}_{2.5}$ increase. Around Pittsburgh and northern West Virginia, elemental carbon accounts for 40-55% of the predicted increase in $\text{PM}_{2.5}$, but the impacts of directly emitted elemental carbon are much less elsewhere. The increases in elemental carbon are due to primary emissions from diesel engines. The $\text{PM}_{2.5}$ nitrate is due to atmospheric oxidation of NO_x emissions from diesel and natural gas fired engines and turbines, the latter occurring at the well pad and within the gathering network. The model predicts substantial increases in summertime $\text{PM}_{2.5}$ nitrate concentrations (see Section S.2).

Figure 4: Speciation of MU-related $\text{PM}_{2.5}$ impacts in the Medium Emissions scenario. Panels (a)-(c) display the fractional contribution of each species to the overall $\text{PM}_{2.5}$ increase for (a) elemental carbon (PEC) (b) nitrate and (c) ammonium.

4. DISCUSSION AND CONCLUSIONS

4.1 Estimated public health costs

Estimated annual premature deaths associated with $\text{PM}_{2.5}$ and ozone increases from MU emissions and their economic values are presented in Tables S4 and S5, respectively. Estimates based on Krewski et al.⁴² and Bell et al.⁴⁴ are presented as lower bounds and those based on Lepeule et al.⁴³ and Levy et al.⁴⁵ as upper bounds. MU emissions are projected to increase premature deaths by 200 to 460 annually for the Medium Emissions scenario, by 350 to 800 for the High Emissions scenario, and by 73 to 170 for the Low Emissions scenario. The economic value of the annual premature deaths caused by MU emissions are estimated to be \$1,600 million to \$3,700 million for the Medium Emissions scenario and proportionally higher and lower in the High and Low Emissions scenarios. Depending on the concentration-response relation, ozone health effects account for 2% to 21% of the total economic value.

Figure 5: Assessment of the relative contributions of MU VOC and NO_x emissions to the overall MU impacts on MDA8 ozone in the Medium Emissions scenario. (a) Fractional contribution of VOC emissions assessed as MU ozone increases in the Marcellus-Utica VOC only scenario normalized by those in the Medium Emissions scenario. (b) Same but for NO_x emissions using the Marcellus-Utica NO_x only scenario.

Our estimates, ranging from \$590 million to \$6,900 million, are approximately two orders of magnitude larger than those of Litovitz et al.¹⁸ The differences are due to multiple factors: emissions scenario, air quality modeling, dose-response, and economic valuation. First, the Litovitz et al.¹⁸ analysis was done for 2011, which had lower emissions than the 2020 emissions used here (e.g. Litovitz et al.¹⁸ estimated 17,000-28,000 tons of NO_x in 2011 versus 14,000 to 77,000 tons here). A second factor is the methodology used to estimate air quality impacts from MU emissions. Litovitz et al.¹⁸ used a reduced-form model, APEEP,^{51,52} to value health damages per unit emissions. APEEP relies on a Gaussian dispersion model with a simplified representation of $\text{PM}_{2.5}$ formation from NO_x emissions compared to CAMx, the air quality

model used here. Other comparisons have shown that, all other factors being equal, CAMx predicts PM_{2.5} formation from NO_x emissions in Pennsylvania that is ~5 times higher than APEEP.⁵³ In addition, the overall emissions landscape is rapidly changing, which alters the sensitivity of ambient concentrations to changes in emissions.⁵⁴ For example, Holt et al.⁵⁵ found that sensitivity to NO_x emissions increased by as much as a factor of two due to emissions changes between 2005 and 2012. This is important as APEEP calculates impacts using 2002 as a baseline year whereas the values presented here account for recent and projected changes in emissions from non-MU sources. Recent updates from APEEP modeling show NO_x social costs increasing by ~50% in Pennsylvania between 2002 and 2008 (Nick Muller, personal communication). A third factor is the concentration-response function. Our lower bound is based on Krewski et al., which is a very similar concentration-response function to the APEEP default,⁵⁶ but our upper bound is more than two times higher.⁴³ Finally, we use VSL to monetize premature mortality here, which is about three times higher than the age-adjusted VSL values from APEEP used in Litovitz et al.¹⁸ Other differences in methodology include changes in population distributions and adjustments to VSL for income growth or inflation, but these factors only play a minor role in the differences.

4.2 VOC/NO_x sensitivity and control strategies

Figure 5 illustrates the sensitivity of ozone concentrations to MU VOC and NO_x emissions for the Medium Emissions scenario. NO_x emissions account for more than 80% of the impact on regional ozone across most of the region while VOC emissions only play a limited role in some urban regions. These patterns are consistent with the well-known spatial patterns of ozone formation, namely that rural ozone levels tend to be NO_x-limited.⁵⁷ The 36 km resolution used in this study probably does not fully resolve the VOC-limited nature of urban core regions; therefore, geographically small but high-population downtown areas may be more sensitive to VOC emissions than these results predict.

The major sources of MU NO_x emissions are drill rigs, truck traffic, and compressor stations.³ Our simulations suggest that tighter controls on these sources would reduce the MU ozone impacts. The Low Emissions scenario (Figure 1(b)) shows that implementing state-of-the-art controls on these sources (equivalent to more than a 90% reduction) strongly reduces the impact of MU activities on ozone levels. Controls on VOC emissions benefit smaller regions compared to NO_x controls, but these are generally urban areas with the most severe ozone problems and the highest population.

Control of NO_x emissions will also reduce the majority of the PM_{2.5} impacts as more than 80% of the predicted PM_{2.5} increases in most of the domain are associated with NO_x emissions with the balance due to direct primary emissions (Section 3.2).

4.3 Impacts of emission controls

The three emissions scenarios also provide insight into potential emission controls options. For example, the Low and High Emissions scenarios can also be derived using the same activity level as the Medium scenario level coupled with varying degrees of emissions controls. Therefore, the High and Low Emissions scenarios illustrate the benefits of both existing regulations and possible further reductions.

The Low Emissions scenario is probably most interesting. It can be derived using the Medium Scenario activity coupled with full implementation of state-of-the-art control technology on all sources and equipment (e.g. selective catalytic reduction units for NO_x control on all engines and turbines). Therefore the differences between the Medium and Low Emissions scenarios illustrate the predicted ozone and PM_{2.5} reductions achievable given full implementation of stringent control technologies at an intermediate level of MU activity.

Similarly, the High Emissions scenario can be constructed if one assumes that no new controls are added to 2009-compliant equipment in 2020 combined with intermediate activity. This would represent a hypothetical future scenario in which there was very little equipment turnover or the Oil and Gas Rule, Tier 4 standards, and on-road vehicle standards were not implemented in the oil and gas sector. Thus comparing the differences between High and Medium Emissions scenario illustrate the benefits of these recently promulgated regulations for an intermediate level of MU activity.

4.4 Potential benefits of reduced end-use emissions

At point of use, natural gas generally has lower emissions than other fossil fuels, especially coal.⁵⁸ However, the analysis presented here accounts only for the emissions associated with the development and production of MU gas on regional ozone and PM_{2.5} levels. A broader assessment would also account for changes in emissions associated with the end uses of MU gas. For example, regional NO_x emissions may decrease if a portion of the MU gas is used to repower or replace older coal-fired power plants in the region because gas-fired power plants emit approximately one tenth of the NO_x per unit MWh compared to coal-fired plants.⁵⁹ In this scenario, fuel switching from coal to gas could bring significant benefits to regional ozone and PM. However, these hypothetical emission reductions must be evaluated in the larger regulatory context. The Cross-State Air Pollution Rule (CSAPR) has set strict caps for NO_x emissions from power plants in the eastern US. It is not clear if fuel switching from coal to MU gas will reduce regional emissions below the CSAPR caps. For example, fuel switching from coal to MU gas is likely being done by companies in lieu of other control strategies such as the installation emission control devices. Therefore, switching to natural gas likely provides the required emissions reductions at lower cost than adding emission controls to coal-fired power plants, but it does not necessarily mean that the overall regional emissions from the electricity sector will be lower than the CSAPR cap. Alternatively, if abundant gas lowers energy prices such that overall energy use grows, then an increase in NO_x emissions beyond that projected here may occur.⁵⁴ These sorts of scenarios are difficult to forecast and therefore beyond the scope of the work presented here. Nevertheless, our results indicate that the direct impacts of gas development alone will increase ozone and PM_{2.5} levels significantly and therefore merit regulatory attention in their own right.

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References

- (1) *Annual Energy Outlook 2015 with projections to 2040*; U.S. Energy Information Administration; U.S. Department of Energy, Washington, DC, 2015; [http://www.eia.gov/forecasts/aeo/pdf/0383\(2015\).pdf](http://www.eia.gov/forecasts/aeo/pdf/0383(2015).pdf).
- (2) *Drilling Productivity Report*; U.S. Energy Information Administration; U.S. Department of Energy, Washington, DC, 2015; <http://www.eia.gov/petroleum/drilling/pdf/dpr-full.pdf>.
- (3) Roy, A. A., Adams, P. J., & Robinson, A. L. Air pollutant emissions from the development, production, and processing of Marcellus Shale natural gas. *Journal of the Air & Waste Management Association* **2014**, 64(1), 19-37.
- (4) Field RA, Soltis J, Murphy S: Air quality concerns of unconventional oil and natural gas production. *Environ Sci Process Impacts*. **2014**, 16: 954-969. 10.1039/c4em00081a.
- (5) Vinciguerra, T., Yao, S., Dadzie, J., Chittams, A., Deskins, T., Ehrman, S., & Dickerson, R. R. Regional air quality impacts of hydraulic fracturing and shale natural gas activity: Evidence from ambient VOC observations. *Atmospheric Environment* **2015**, 110, 144-150.
- (6) Moore, C. W., Zielinska, B., Petron, G., & Jackson, R. B.. Air impacts of increased natural gas acquisition, processing, and use: a critical review. *Environmental science & technology*, **2014**, 48(15), 8349-8359.
- (7) Grant, J., Parker, L., Bar-Ilan, A., Kemball-Cook, S., Yarwood, G., 2009. Draft Report: Development of Emissions Inventories for Natural Gas Exploration and Production Activity in the Haynesville Shale. Environ International Corporation, CA. http://www.netac.org/UserFiles/File/NETAC/9_29_09/Enclosure_2b.pdf
- (8) USEPA: Natural Gas-Fired Reciprocating Engines. 2000, Washington, DC: United States Environmental Protection Agency, AP-42 Section 3.2. <http://www.epa.gov/ttn/chief/ap42/ch03/final/c03s02.pdf>
- (9) Kemball-Cook, S., Bar-Ilan, A., Grant, J., Parker, L., Jung, J., Santamaria, W., ... & Yarwood, G. Ozone impacts of natural gas development in the Haynesville Shale. *Environmental science & technology* **2010**, 44(24), 9357-9363.
- (10) Levy, J. I., Carrothers, T. J., Tuomisto, J. T., Hammitt, J. K., & Evans, J. S. Assessing the public health benefits of reduced ozone concentrations. *Environmental Health Perspectives* **2001**, 109(12), 1215.
- (11) Pope III, C. A., Ezzati, M., & Dockery, D. W. Fine-particulate air pollution and life expectancy in the United States. *New England Journal of Medicine* **2009**, 360(4), 376-386.
- (12) Schnell, R. C., Oltmans, S. J., Neely, R. R., Endres, M. S., Molenaar, J. V., & White, A. B. Rapid photochemical production of ozone at high concentrations in a rural site during winter. *Nature Geoscience* **2009**, 2(2), 120-122.
- (13) *Southwestern Pennsylvania Marcellus Shale Short-Term Ambient Air Sampling Report*; Pennsylvania Department of Environmental Protection, Bureau of Air Quality; 2010. http://www.dep.state.pa.us/dep/deputate/airwaste/aq/aqm/docs/Marcellus_SW_11-01-10.pdf.

- (14) Helmig, D., Thompson, C. R., Evans, J., Boylan, P., Hueber, J., & Park, J. H. Highly elevated atmospheric levels of volatile organic compounds in the Uintah Basin, Utah. *Environmental science & technology* **2014**, 48(9), 4707-4715.
- (15) Brantley, H. L., Thoma, E. D., & Eisele, A. P. Assessment of VOC and HAP Emissions from Oil and Natural Gas Well Pads Using Mobile Remote and Onsite Direct Measurements. *Journal of the Air & Waste Management Association* **2015**, (just-accepted).
- (16) The Green Book Nonattainment Areas for Criteria Pollutants; <http://www.epa.gov/oaqps001/greenbk/>.
- (17) Adgate, J. L., Goldstein, B. D., & McKenzie, L. M. Potential public health hazards, exposures and health effects from unconventional natural gas development. *Environmental science & technology*, **2014**, 48(15), 8307-8320.
- (18) Litovitz, A., Curtright, A., Abramzon, S., Burger, N., & Samaras, C. Estimation of regional air-quality damages from Marcellus Shale natural gas extraction in Pennsylvania. *Environmental Research Letters* **2013**, 8(1), 014017.
- (19) *CAMx User's Guide, version 5.40*; ENVIRON International Corporation, Novato, CA; 2011; http://www.camx.com/files/camxusersguide_v5-40.aspx
- (20) *Updates to the Carbon Bond Chemical Mechanism: CB05*; Yocke & Company, Novato, CA; 2005; http://www.camx.com/publ/pdfs/cb05_final_report_120805.aspx.
- (21) Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Duda, M.G., Huang, X-Y, Wang, W., Powers, J.G., 2008. A description of the Advanced Research WRF Version 3. http://www2.mmm.ucar.edu/wrf/users/docs/arw_v3.pdf (accessed December 24, 2012).
- (22) *GEOS-Chem v10-01 Online User's Guide*; Atmospheric Chemistry Modeling Group, School of Engineering and Applied Sciences, Harvard University; 2015; <http://acmg.seas.harvard.edu/geos/doc/man/>.
- (23) *Regulatory Impact Analysis for the Proposed Federal Transport Rule*; US Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC; 2010; http://www.epa.gov/ttnecas1/regdata/RIAs/proposaltrria_final.pdf.
- (24) *Air Quality Modeling Final Rule Technical Support Document*; US Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC; 2011.
- (25) *Guidance for Regulatory Application of the Urban Airshed Model (UAM)*; US Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC; 1991.
- (26) Solazzo, E., Bianconi, R., Vautard, R., Appel, K. W., Moran, M. D., Hogrefe, C., & Galmarini, S. Model evaluation and ensemble modelling of surface-level ozone in Europe and North America in the context of AQMEII. *Atmospheric Environment* **2012**, 53, 60-74.
- (27) Boylan, J. W., & Russell, A. G. PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. *Atmospheric environment* **2006**, 40(26), 4946-4959.
- (28) *MARAMA Future Year Inventories Version 3*; Mid-Atlantic Regional Air Management Association, 2012; <http://www.marama.org/technical-center/emissions-inventory/2007-emissions-and-projections/future-year-inventory-version-3>.
- (29) *Oil and natural Gas Air Pollution Standards*; <http://www.epa.gov/airquality/oilandgas/>.

- (30) *Clean Air Nonroad Diesel Rule*; US Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC; 2004;
<http://www.epa.gov/otaq/documents/nonroad-diesel/420f04032.pdf>.
- (31) Pennsylvania Department of Environmental Protection - Oil and gas reporting website;
<https://www.paoilandgasreporting.state.pa.us/publicreports/Modules/DataExports/DataExports.aspx>.
- (32) West Virginia Geological and Economic Survey – Marcellus and other shales;
<http://www.wvgs.wvnet.edu/www/datastat/devshales.htm>.
- (33) Ohio Department of Natural Resources – Shale well drilling and permitting;
<http://oilandgas.ohiodnr.gov/shale>.
- (34) Hendler, A., Nunn, J., Lundeen, J., McKaskle, R., 2009. VOC emissions from oil and condensate storage tanks.
<http://files.harc.edu/Projects/AirQuality/Projects/H051C/H051CFinalReport.pdf>
- (35) National Ambient Air Quality Standards;
http://www.epa.gov/ttn/naaqs/standards/ozone/s_o3_history.html.
- (36) *Review of the EPA's Second Draft Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards*; Clean Air Scientific Advisory Committee; 2014;
[http://yosemite.epa.gov/sab/sabproduct.nsf/5EFA320CCAD326E885257D030071531C/\\$File/EPA-CASAC-14-004+unsigned.pdf](http://yosemite.epa.gov/sab/sabproduct.nsf/5EFA320CCAD326E885257D030071531C/$File/EPA-CASAC-14-004+unsigned.pdf).
- (37) Air Trends – Design Values; <http://www.epa.gov/airtrends/values.html>
- (38) *Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, PM_{2.5}, and Regional Haze*; US Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC; 2007; <http://www.epa.gov/scram001/guidance/guide/final-03-pm-rh-guidance.pdf>.
- (39) *Modeling Guidance for Demonstrating Attainment of Air Quality Goals for Ozone, PM_{2.5}, and Regional Haze*; US Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC; 2014;
http://www.epa.gov/ttn/scram/guidance/guide/Draft_O3-PM-RH_Modeling_Guidance-2014.pdf.
- (40) *Final Ozone NAAQS Regulatory Impact Analysis*; EPA-452/R-08-003; U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Health and Environmental Impacts Division, Air Benefit and Cost Group (C439-02): Research Triangle Park, NC, 2008.
- (41) RTI International. *Environmental Benefits Mapping and Analysis Program – Community Edition: User's Manual*; Data and Tools; Prepared for Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, 2015.
- (42) Krewski, D.; Jerrett, M.; Burnett, R. T.; Ma, R.; Hughes, E.; Shi, Y.; Turner, M. C.; Pope, C. A., III; Thurston, G.; Calle, E. E.; et al. *Extended follow-up and spatial analysis of the American Cancer Society study linking particulate air pollution and mortality*; Research Report 140; Health Effects Institute: Boston, MA, 2009.
- (43) Lepeule, J.; Laden, F.; Dockery, D.; Schwartz, J. Chronic Exposure to Fine Particles and Mortality: An Extended Follow-up of the Harvard Six Cities Study from 1974 to 2009. *Environ. Health Perspect.* **2012**, 120 (7), 965–970.

- (44) Bell, M. L.; McDermott, A.; Zeger, S. L.; Samet, J. M.; Dominici, F. Ozone and Short-term Mortality in 95 US Urban Communities, 1987-2000. *JAMA J. Am. Med. Assoc.* **2004**, 292 (19), 2372–2378.
- (45) Levy, J. I.; Chemerynski, S. M.; Sarnat, J. A. Ozone Exposure and Mortality: “An Empiric Bayes Metaregression Analysis.” *Epidemiology* **2005**, 16 (4), 458–468.
- (46) Bell, M. L.; Dominici, F.; Samet, J. M. A Meta-Analysis of Time-Series Studies of Ozone and Mortality with Comparison to the National Morbidity, Mortality, and Air Pollution Study. *Epidemiology* **2005**, 16 (4), 436–445.
- (47) Ito, K.; Leon, S. F. D.; Lippmann, M. Associations between Ozone and Daily Mortality: Analysis and Meta-Analysis. *Epidemiology* **2005**, 16 (4), 446–457.
- (48) Schwartz, J. How Sensitive Is the Association between Ozone and Daily Deaths to Control for Temperature? *Am. J. Respir. Crit. Care Med.* **2005**, 171 (6), 627–631.
- (49) Huang, Y.; Dominici, F.; Bell, M. L. Bayesian hierarchical distributed lag models for summer ozone exposure and cardio-respiratory mortality. *Environmetrics* **2005**, 16 (5), 547–562.
- (50) *Guidelines for Preparing Economic Analyses*; National Center for Environmental Economics, Office of Policy, U.S. Environmental Protection Agency, 2010b.
- (51) Muller, N. Z.; Mendelsohn, R. Measuring the damages of air pollution in the United States. *J. Environ. Econ. Manag.* **2007**, 54 (1), 1–14.
- (52) Muller, N. Z.; Mendelsohn, R. Efficient Pollution Regulation: Getting the Prices Right: Corrigendum (Mortality Rate Update). *Am. Econ. Rev.* **2012**, 102 (1), 613–616.
- (53) Heo, Jinhyok. *Evaluation of Air Quality Impacts on Society: Methods and Application* (Doctoral dissertation). Carnegie Mellon University, 2015.
- (54) Ansari, A. S., & Pandis, S. N. Response of inorganic PM to precursor concentrations. *Environmental Science & Technology* **1998**, 32(18), 2706-2714.
- (55) Holt, J., Selin, N. E., & Solomon, S. Changes in inorganic fine particulate matter sensitivities to precursors due to large-scale US emissions reductions. *Environmental Science & Technology*, **2015**, 49, 4834-4841.
- (56) Pope, C. A., Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., & Thurston, G. D. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *Journal of the American Medical Association*, **2002**, 287(9), 1132-1141.
- (57) Tsimpidi, A. P., Karydis, V. A., & Pandis, S. N. Response of fine particulate matter to emission changes of oxides of nitrogen and anthropogenic volatile organic compounds in the Eastern United States. *Journal of the Air & Waste Management Association* **2008**, 58(11), 1463-1473.
- (58) De Gouw, J. A., Parrish, D. D., Frost, G. J., & Trainer, M. Reduced emissions of CO₂, NO_x, and SO₂ from US power plants owing to switch from coal to natural gas with combined cycle technology. *Earth's Future* **2014**, 2(2), 75-82.
- (59) US EPA, 2013c. eGRID2012 Version 1.0 data files for year 2009, <http://www.epa.gov/cleanenergy/energy-resources/egrid/index.html>, (accessed September 26, 2013).