J.A. de Gouw¹, D.D. Parrish, G.J. Frost¹, M. Trainer

NOAA Earth System Research Laboratory, Boulder, CO, USA

Abstract

Since 1997, an increasing fraction of electric power in the U.S. has been generated from natural gas. Here, we use data from continuous emissions monitoring systems (CEMS), which measure emissions at the stack of most U.S. electric power generation units, to investigate how this switch affected the emissions of CO₂, NOx and SO₂. Per unit of energy produced, natural gas power plants equipped with combined cycle technology emit on average 44% of the CO₂ compared with coal power plants. As a result of the increased use of natural gas, CO₂ emissions from U.S. fossil-fuel power plants were 23% lower in 2012 than they would have been, if coal had continued to provide the same fraction of electric power as in 1997. In addition, natural gas power plants with combined cycle technology emit less NOx and far less SO₂ per unit energy produced than coal power plants. The increased use of natural gas has therefore led to emissions reductions of NOx (40%) and SO₂ (44%), in addition to those obtained from the implementation of emissions control systems on coal power plants. These benefits to air quality and climate should be weighed against the increase in emissions of methane, volatile organic compounds and other trace gases that are associated with the production, processing, storage and transport of natural gas.

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1002/eft2.196

¹ Also with: Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO

1. Introduction

Accepted Articl

Over the last decade, natural gas in the U.S. is being produced in increasing amounts from shale and tight sand plays made possible through advances in directional drilling and hydraulic fracturing [Fichman, 2011]. Concerns have been raised about the impact of atmospheric emissions associated with these production methods and their effects on climate and air quality. Notably, it has been suggested that the net greenhouse gas emissions of shale gas are higher than those from coal [Howarth et al., 2011]. There has been significant discussion of these findings [Cathles III et al., 2012; Howarth et al., 2012], partly because the emissions are highly uncertain. For example, recent measurements in Colorado [Petron et al., 2012] and in Utah [Karion et al., 2013] have suggested that a significant fraction of shale and tight sand gas can escape to the atmosphere during production and transport, while measurements by others have suggested the emissions to be lower [Allen et al., 2013]. The effect on air quality has been most apparent in the Upper Green River basin in Wyoming and the Uintah basin in Utah, where emissions associated with oil and gas production can accumulate under wintertime inversions and react to form high concentrations of surface ozone [Schnell et al., 2009; Carter and Seinfeld, 2012; Edwards et al., 2013]. Additionally, emissions associated with natural gas production in the Denver-Julesburg basin provide the majority of atmospheric hydrocarbon reactivity in the Colorado Front Range metropolitan area [Gilman et al., 2013; Swarthout et al., 2013]. The direct health effects of the emissions were studied in Garfield County in Colorado [McKenzie et al., 2012]. The study of the emissions associated with shale gas production and their effects on climate and air quality is ongoing. However, for a comprehensive assessment of the impact of increasing shale gas production, the environmental effects of the end use of natural gas must be considered in addition to the effects of shale gas extraction.

An important use of natural gas in the U.S. is electric power generation: according to the Energy Information Administration (EIA), 32% of the U.S. natural gas consumption in 2011 was for electric power, 34% for industrial, 13% for commercial and 20% for residential use [*Fichman*, 2011]. The number use of natural gas for electric power generation has grown over the last decade [*Fichman*,

2011], which has led to significant reductions in the emissions of CO₂, NOx and SO₂ [Venkatesh et al., 2011; Lu et al., 2012a; Lu et al., 2012b; Venkatesh et al., 2012]. U.S. power plant emissions are usually measured at the stack using continuous emissions monitoring systems (CEMS) required by the EPA. Previous studies using airborne measurements have shown these CEMS measurements to be accurate [*Frost et al.*, 2006; *Peischl et al.*, 2010], and the inclusion of these emissions data into air quality models has led to more accurate representations of photochemical ozone formation and annual trends therein [*Kim et al.*, 2006]. The CEMS data were also used in developing a CO₂ emissions inventory with a high spatial and time resolution [*Petron et al.*, 2008]. Here, we use annual emissions from all point sources included in the CEMS database to quantify the changes in CO₂, NOx and SO₂ emissions that have resulted from the switch from coal to natural gas by U.S. power plants over the period 1995 – 2012.

2. Data

CEMS data were taken from the EPA Air Markets Program Data website (http://ampd.epa.gov/ampd/). Annual data were downloaded at the boiler/stack unit level rather than for each monitoring location. Power plants with separate units, which could use different fuels and have different pollution control technology, are included as separate entries in the database. The total data set includes 72481 individual entries for the period 1995-2012. CEMS data include electric utilities as well as some industrial generating units. No distinction between the two is made in this work.

The CEMS data set includes measurements of CO_2 , NOx and SO_2 emissions as well as gross load and heat input. Gross load is the measured output from the electrical generator. The net output of a power plant is the gross load minus the energy consumption of the plant itself, which is typically a few percent of the gross load and dependent on pollution control equipment. Net output, which might be a more appropriate measure of electric power generation, is not included in CEMS. Also included in the data set are the name, location, operator, fuel and some details on emissions controls for each source. The data are used as downloaded with the exception of four entries that had unrealistic values for gross load.

3. Results

Figure 1 shows a correlation plot of the CO_2 emissions versus the gross load for all the entries included in the CEMS data set used here. These are the entries for the period 1997 through 2012; gross loads were not reported for 1995 and 1996. Each individual power plant unit is typically associated with multiple data points in Figure 1, i.e. one for each year.

The data in Figure 1 are separated according to different types: coal, natural gas, natural gas with combined cycle technology, and other sources (mostly Diesel and residual oil). In combined cycle power plants, two heat engines are used in tandem to convert a higher fraction of heat into first mechanical and then electrical energy. Coal power plants have clearly some of the highest gross loads and CO₂ emissions. The CO₂ emission intensities, defined here as the CO₂ emissions divided by gross load, are determined from the slopes of linear regression fits to the data with the intercept forced to zero. The average CO₂ emission intensities between 1997 and 2012 were 915.0±0.8 g(CO₂)/kWh for coal, 549.4±1.1 g(CO₂)/kWh for natural gas, 436.0±1.4 g(CO₂)/kWh for natural gas with combined cycle technology, and 784±2 g(CO₂)/kWh for other fuels. These are average numbers derived from 16 years of data and the uncertainties are the 1- σ errors in the slope from the regression fits. Below, we will quantify how the CO₂, NOx and SO₂ emission intensities for individual sources are distributed and how they have changed since 1995.

Figure 2 shows the annual trends in total gross load, and total emissions of CO_2 , NOx and SO_2 from fossil fuel power plants in the U.S., again separated by source type. The total gross load increased between 1997 and 2007, then decreased in 2009 likely due to the economic recession and has since stabilized at levels below the peak production in 2007. The contributions from different sources to the total gross load have markedly changed since 1997. In 1997, the total gross load was dominated by coal (83%). Since 1997, the relative contributions from coal, natural gas without combined cycle and other sources have all decreased. Over the same time, the relative contribution of

natural gas with combined cycle technology steadily increased and amounted to 34% of the total gross load in 2012. The contribution from coal to the total gross load had decreased to 59% in 2012.

The gross loads from CEMS were compared with the primary production of electric power from the EIA [*Fichman*, 2011]. The data from CEMS and the EIA agree within 2% for coal, 14% for natural gas (with and without combined cycle; the EIA does not make the distinction) and 10% for other fuels. The EIA data are not systematically higher than CEMS. From this we conclude that the CEMS data include almost all of the emissions associated with electric power generation from fossil fuels in the U.S. and can be regarded as a representative data set for U.S. fossil-fuel power plants.

In general accord with gross load, the total CO_2 emissions from fossil-fuel power plants in the U.S. increased until 2007, but then decreased more rapidly between 2008-2012. Since the CO_2 emission intensity of coal is so much higher than that of natural gas, particularly with combined cycle technology, a significant fraction of this decrease in CO_2 emissions can be attributed to the switch from coal towards natural gas. The exact fraction will be quantified further below.

The lower two panels of Figure 2 show the decreases in NOx and SO₂ emissions from fossilfuel power plants in the U.S. since 1995. The overall emissions of NOx and SO₂ are dominated by coal power plants, much more so than for CO₂. The decreases in the NOx and SO₂ emissions from power plants are due in part to the implementation of emissions controls, as enacted under various clean-air programs of the U.S. Environmental Protection Agency. In addition, the switch from coal to natural gas has also contributed to reductions in emissions of NOx and SO₂. The relative importance of these two will be quantified further below.

Figure 3 shows the distribution in CO_2 emission intensities for U.S. power plants in four different years. Several changes occurred over the 1997 - 2012 period. The number of coal power plants gradually decreased. While their median CO_2 emission intensities were relatively constant, the distribution had a longer tail towards higher emission intensities in 1997 than in 2012. The number of natural gas power plants grew strongly over this same period. In 2002, some of these plants were equipped with combined cycle technology, but the average CO_2 emission intensities of these plants were the same as those of natural gas power plants that did not use combined cycle. In 2007, the number of natural gas power plants with combined cycle had further grown and the majority of them

now showed improved CO_2 emission intensities relative to conventional natural gas power plants. This trend continued through 2012. It should be noted that the distributions in Figure 3 only describe the number of power plants and not the electric power generated by these plants. While the number of natural gas power plants with and without combined cycle was similar in 2012 (Figure 3), a far greater fraction of electric power was generated by natural gas power plants that use combined cycle (top panel of Figure 2). The number of natural gas power plants far exceeded the number of coal power plants in 2012 (Figure 3), but the coal power plants still generated about twice the amount of electric power (top panel of Figure 2).

Figure 4 shows how CO₂, NOx and SO₂ emission intensities have changed since the start of the CEMS measurements. The emission intensities are determined here from regression fits to the data for each year, analogous to the determination of CO₂ emission intensities from all data in Figure 1. The error bars in Figure 4 represent the 1- σ uncertainties from the regression fits. From 1997-2012, the CO₂ emission intensity of coal power plants decreased slightly. The CO₂ emission intensity of natural gas power plants with combined cycle technology decreased by about one-third. Due to the bimodal distribution in CO2 emission intensities of natural gas power plants with combined cycle (Figure 3), the error bars for this source in Figure 4 are relatively large before 2000. In 2012, most of the natural gas power plants with combined cycle had CO₂ emission intensities below 400 $g(CO_2)/kWh$. The continued operation of a number of less efficient plants results in an average value of 404.0±0.9 g(CO₂)/kWh in 2012, i.e. 44% of the average CO2 emission intensity of a coal power plant in 2012. This value is close to the 47% value in the latest IPCC report (469 for natural gas vs. 1001 g(CO₂)/kWh for coal) [Moomaw et al., 2011]. It should be noted that the IPCC values are lifecycle GHG emissions and include GHG emissions associated with the production and transport of the fuels to the power plants. However, these additional emissions are small contributions to the total [Howarth et al., 2011] and affect the ratio between CO₂ emission intensities even less.

Due to the implementation of emissions controls, the NOx and SO₂ emissions intensities for coal and natural gas power plants all decreased between 1997 and 2012. For coal, the average decreases in NOx (72%) and SO₂ (71%) emissions intensities were steady but important, since coal power plants emit the majority of NOx and SO₂ associated with electric power generation. The NOx

emissions intensities for natural gas power plants with combined cycle decreased rapidly between 1997 and 2012. In 2012, the average NOx emission intensity of a natural gas power plant with combined cycle was 7% of that of a coal power plant. The NOx emissions from a modern natural gas power plant with combined cycle can be much more efficiently controlled than those from a coal power plant. The SO₂ emissions intensities of natural gas power plants with combined cycle are very low, due to the low sulfur content of natural gas, and did not show a decrease between 1997 and 2012. In 2012, the average SO₂ emission intensity of a natural gas power plant with combined cycle was 0.2% of that of a coal power plant.

4. Discussion

We estimate here how much the CO_2 , NOx and SO_2 emissions from fossil fuel power plants in the U.S. have decreased solely as a result of the switch away from coal towards natural gas with combined cycle. The decrease in emissions is calculated from:

$$\Delta Emissions = GL_{combined \ cycle} \times (EI_{coal} - EI_{combined \ cycle}), \tag{1}$$

where *GL* stands for gross load and *EI* for emission intensity. The calculation assumes that the new capacity from natural gas power plants with combined cycle has been installed to replace coal power plants rather than conventional natural gas power plants. This is justified by that fact that the number of coal power plants has indeed decreased since 1997, whereas the number of conventional natural gas power plants has increased (Figure 3). Equation (1) is calculated for each year and the results are shown in Figure 5 as a percentage of the total power plant emissions. By doing the calculation using the measured emission intensities for each year, any improvements in time of the technology do not contribute to the reductions in Figure 5. In other words, the reductions in Figure 5 can be attributed to the switch from coal to natural gas only.

The top panel shows the emissions reductions as a percentage of the total power plant emissions. The reductions in emissions grew rapidly over the last decade and amounted to 23% for CO₂, 40% for NOx and 44% for SO₂ in 2012. The bottom panel shows the emissions reductions as a fraction of all U.S. emissions. Total emissions of NOx and SO2 are taken from the air pollutant emissions trend data from **EPA** National Emissions the Inventory (<u>http://www.epa.gov/ttn/chief/trends/index.html</u>); total emissions of CO₂ from the National Greenhouse Gas Emissions Data (http://www.epa.gov/climatechange/ghgemissions/usinventoryreport.html). The emissions reductions have been particularly significant for SO_2 (26% in 2012), since electric power generation is the dominant source nationally. The emissions reductions have been smaller but also significant for CO_2 (6% in 2011) and NOx (6% in 2012), because other sources (industry, heating, motor vehicles) are also important.

The other significant change in electric power production in the U.S. has been the strong increase in wind power [*Fichman*, 2011]. According to the EIA, wind power increased from 3 TWh in 1997 to 120 TWh in 2011, whereas natural gas increased from 496 to 1017 TWh. The addition of wind power has had a smaller effect on reductions in total CO₂, NOx and SO₂ emissions than the increased use of natural gas power plants with combined cycle.

The emissions reductions in CO₂, NOx and SO₂ due to the increased use of natural gas power plants with combined cycle must be weighed against emissions reductions in other species such as mercury that are not included in the CEMS measurements, as well as the increased emissions of methane and volatile organic compounds (VOCs) associated with the production of natural gas. In particular for shale gas, significant emissions of methane [*Petron et al.*, 2012; *Allen et al.*, 2013; *Karion et al.*, 2013] and VOCs [*Gilman et al.*, 2013; *Swarthout et al.*, 2013] have been observed. Howarth et al. [2011] compared the greenhouse footprint of shale gas versus coal. These authors concluded that the emissions of methane from shale gas production more than outweighed the reductions in CO₂ emissions. It should be noted that these authors compared the direct CO₂ emissions of shale gas and coal on the basis of the heat content of the two fuels. Compared on this basis, the CO₂ emission intensity of natural gas is 60% of that of coal. For electric power generation, we find a significantly more favorable value of 44%. This difference is due to the fact that electric power is more efficiently generated from natural gas than it is from coal. This issue was suggested to be a shortcoming in the Howarth et al. [2011] analysis [*Cathles III et al.*, 2012], but was justified based on the fact that more natural gas is used for heating than for electric power generation [*Howarth et al.*, 2012]. An updated analysis of the greenhouse gas footprint of shale gas awaits comprehensive estimates of methane leak rates from all important production areas in the U.S. The updated analysis should also separate the use of natural gas for heating versus electric power generation, as their CO₂ emissions intensities differ.

5. Conclusion

Over the last decade the increased use of natural gas power plants with combined cycle technology has significantly decreased the atmospheric emissions of CO_2 , NOx and SO_2 associated with electric power generation in the U.S. Further reductions in these emissions can follow by converting a larger fraction of U.S. electric power production to natural gas, and by ensuring that all natural gas power plants are equipped with the latest combined cycle technology. These results illustrate some of the advantages to both climate and air quality that result from the switch from coal to natural gas. These advantages must be considered in the perspective of the environmental impacts of natural gas production including methane and hydrocarbon leakage to the atmosphere that await more comprehensive quantification.

References

- Allen, D.T., et al. (2013), Measurements of methane emissions at natural gas production sites in the United States, *PNAS*, doi:10.1073/pnas.1304880110.
- Carter, W.P.L., and J.H. Seinfeld (2012), Winter ozone formation and VOC incremental reactivities in the Upper Green River Basin of Wyoming, *Atmos. Environ.*, *50*, 255–266.
- Cathles III, L.W., L. Brown, M. Taam, and A. Hunter (2012), A commentary on "The greenhouse-gas footprint of natural gas in shale formations" by R.W. Howarth, R. Santoro, and Anthony Ingraffea, *Climatic Change*, 113, 525-535.
- Edwards, P.M., et al. (2013), Ozone photochemistry in an oil and natural gas extraction region during winter: simulations of a snow-free season in the Uintah Basin, Utah, *Atmos. Chem. Phys.*, *13*, 8955-8971.

Fichman, B.T. (2011), Annual Energy Review 2011, U.S. Energy Information Administration.

- Frost, G.J., et al. (2006), Effects of changing power plant NOx emissions on ozone in the eastern United States: Proof of concept, J. Geophys. Res.-Atmos., 111, D12306, doi:10.1029/2005JD006354.
- Gilman, J.B., B.M. Lerner, W.C. Kuster, and J.A. de Gouw (2013), Source signature of volatile organic compounds from oil and natural gas operations in northeastern Colorado, *Environ. Sci. Technol.*, 47, 1297-1305, doi:10.1021/es304119a.
- Howarth, R.W., R. Santoro, and A. Ingraffea (2011), Methane and the greenhouse-gas footprint of natural gas from shale formations, *Clim. Change*, *106*, 679-690, 10.1007/s10584-011-0061-5.
- Howarth, R.W., R. Santoro, and A. Ingraffea (2012), Venting and leaking of methane from shale gas development: response to Cathles et al., *Climatic Change*, *113*, 537-549.
- Karion, A., et al. (2013), Methane emissions estimate from airborne measurements over a western United States natural gas field, *Geophys. Res. Lett.*, doi:10.1002/grl.50811.
- Kim, S.W., et al. (2006), Satellite-observed US power plant NOx emission reductions and their impact on air quality, *Geophys. Res. Lett.*, 33, L22812, doi:10.1029/2006GL027749.

- Lu, X., M.B. McElroy, G. Wu, and C.P. Nielsen (2012a), Accelerated reduction in SO2 emissions from the U.S. power sector triggered by changing prices of natural gas, *Environ. Sci. Technol.*, 46, 7882-7889.
- Lu, X., J. Salovaara, and M.B. McElroy (2012b), Implications of the recent reductions in natural gas prices for emissions of CO2 from the US power sector, *Environ. Sci. Technol.*, 46, 3014-3021.
- McKenzie, L.M., R.Z. Witter, L.S. Newman, and J.L. Adgate (2012), Human health risk assessment of air emissions from development of unconventional natural gas resources, *Sci. Tot. Environ.*, in press, doi:10.1016/j.scitotenv.2012.02.018.
- Moomaw, W., P. Burgherr, G. Heath, M. Lenzen, J. Nyboer, and A. Verbruggen (2011), Annex II: Methodology. In IPCC Special Report on Renewable Energy Sources and Climate Change Mitigation, Cambridge University Press, Cambridge, UK and New York, USA.
- Peischl, J., et al. (2010), A top-down analysis of emissions from selected Texas power plants during TexAQS 2000 and 2006, *J. Geophys. Res.-Atmos.*, *115*, doi:10.1029/2009jd013527.
- Petron, G., et al. (2012), Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study, *J. Geophys. Res.-Atmos.*, *117*, D04304, doi:10.1029/2011JD016360.
- Petron, G., P. Tans, G. Frost, D. Chao, and M. Trainer (2008), High-resolution emissions of CO2 from power generation in the USA, J. Geophys. Res.-Biogeosci., 113, G04008, doi:10.1029/2007JG000602.
- Schnell, R.C., S.J. Oltmans, R.R. Neely, M.S. Endres, J.V. Molenar, and A.B. White (2009), Rapid photochemical production of ozone at high concentrations in a rural site during winter, *Nature Geosci.*, 2, 120-122.
- Swarthout, R.F., R.S. Russo, Y. Zhou, A.H. Hart, and B.C. Sive (2013), Volatile organic compound distributions during the NACHTT campaign at the Boulder Atmospheric Observatory: Influence of urban and natural gas sources, J. Geophys. Res.-Atmos., in press, doi:10.1002/jgrd.50722.

Venkatesh, A., P. Jaramillo, W.M. Griffin, and H.S. Matthews (2011), Uncertainty in life cycle greenhouse gas emissions from United States natural gas end-uses and its effects on policy, *Environ. Sci. Technol.*, 45, 8182-8189.

Venkatesh, A., P. Jaramillo, W.M. Griffin, and H.S. Matthews (2012), Implications of changing natural gas prices in the United States electricity sector for SO2, NOX and life cycle GHG emissions, *Environ. Res. Lett.*, 7, 034018, doi:10.1088/1748-9326/7/3/034018.

Figure 1: Correlation plot of annual CO_2 emissions versus the annual gross load of individual units of fossil-fuel power plants in the U.S. Data were obtained from the continuous emissions monitoring systems (CEMS) for the period 1997-2012. The dashed lines show results of linear regression fits to the data.

Articl Accepted

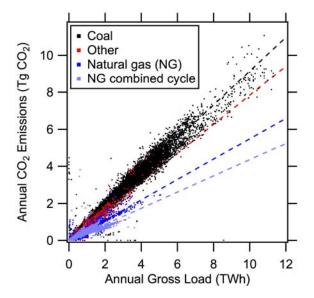


Figure 2: Annual total gross loads and emissions of CO₂, NOx and SO₂ from power plants in the U.S. between 1995 and 2012. Gross loads were not reported in the CEMS database in 1995 and 1996.

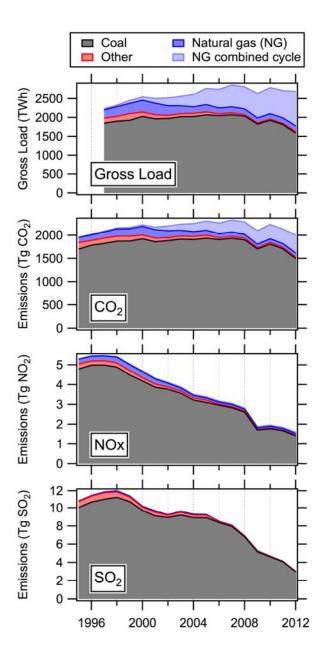


Figure 3: Distribution of CO_2 emission intensities for U.S. power plants in 1997, 2002, 2007 and 2012.

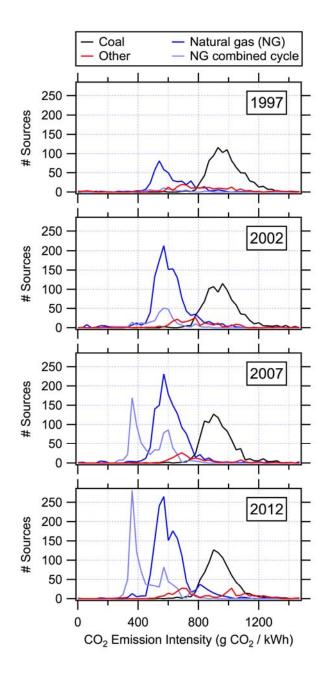


Figure 4: Average emission intensities (in units of g/kWh) of CO₂, NOx and SO₂ from U.S. power plants between 1995 and 2012. Error bars give the 1- σ in the averages.

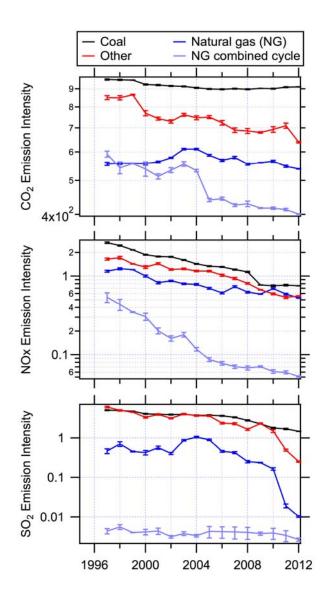


Figure 5: Reductions in the U.S. emissions of CO_2 , NOx and SO_2 as a result of the introduction of natural gas power plants with combined cycle technology. The top panel compares the reductions with the total fossil-fuel power plant emissions. The bottom panel compares the reductions with the total U.S. emissions from all energy sectors.

TTI C Acceptec

