



The effect of coal-fired power-plant SO₂ and NO_x control technologies on aerosol nucleation in the source plumes

C. R. Lonsdale¹, R. G. Stevens¹, C. A. Brock², P. A. Makar³, E. M. Knipping⁴, and J. R. Pierce¹

¹Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Canada

²NOAA Earth System Research Laboratory, Boulder, CO, USA

³Environment Canada, Downsview, Toronto, Canada

⁴Electric Power Research Institute, Palo Alto, CA, USA

Correspondence to: J. R. Pierce (jeffrey.robert.pierce@gmail.com)

Received: 17 July 2012 – Published in Atmos. Chem. Phys. Discuss.: 7 August 2012

Revised: 13 November 2012 – Accepted: 26 November 2012 – Published: 4 December 2012

Abstract. Nucleation in coal-fired power-plant plumes can greatly contribute to particle number concentrations near source regions. The changing emissions rates of SO₂ and NO_x due to pollution-control technologies over recent decades may have had a significant effect on aerosol formation and growth in the plumes with ultimate implications for climate and human health. We use the System for Atmospheric Modeling (SAM) large-eddy simulation model with the Two-Moment Aerosol Sectional (TOMAS) microphysics algorithm to model the nucleation in plumes of coal-fired plants. We test a range of cases with varying emissions to simulate the implementation of emissions-control technologies between 1997 and 2010. We start by simulating the W. A. Parish power plant (near Houston, TX) during this time period, when NO_x emissions were reduced by ~90% and SO₂ emissions decreased by ~30%. Increases in plume OH (due to the reduced NO_x) produced enhanced SO₂ oxidation and an order-of-magnitude increase in particle nucleation in the plume despite the reduction in SO₂ emissions. These results suggest that NO_x emissions could strongly regulate particle nucleation and growth in power-plant plumes. Next, we test a range of cases with varying emissions to simulate the implementation of SO₂ and NO_x emissions-control technologies. Particle formation generally increases with SO₂ emission, while NO_x shows two different regimes: increasing particle formation with increasing NO_x under low-NO_x emissions and decreasing particle formation with increasing NO_x under high-NO_x emissions. Next, we compare model results with airborne measurements made in the W. A. Parish power-plant plume in 2000 and 2006,

confirming the importance of NO_x emissions on new particle formation and highlighting the substantial effect of background aerosol loadings on this process (the more polluted background of the 2006 case caused more than an order-of-magnitude reduction in particle formation in the plume compared to the cleaner test day in 2000). Finally, we calculate particle-formation statistics of 330 coal-fired power plants in the US in 1997 and 2010, and the model results show a median decrease of 19% in particle formation rates from 1997 to 2010 (whereas the W. A. Parish case study showed an increase). Thus, the US power plants, on average, show a different result than was found for the W. A. Parish plant specifically, and it shows that the strong NO_x controls (90% reduction) implemented at the W. A. Parish plant (with relatively weak SO₂ emissions reductions, 30%) are not representative of most power plants in the US during the past 15 yr. These results suggest that there may be important climate implications of power-plant controls due to changes in plume chemistry and microphysics, but the magnitude and sign of the aerosol changes depend greatly on the relative reductions in NO_x and SO₂ emissions in each plant. More extensive plume measurements for a range of emissions of SO₂ and NO_x and in varying background aerosol conditions are needed, however, to better quantify these effects.

1 Introduction

Anthropogenic aerosols affect human health and the Earth's climate. High aerosol concentrations are known to cause

human health problems such as respiratory and cardiovascular diseases, intensification of asthma, a reduction in physical abilities and an increase in mortality rates (Arya, 1999; Stieb et al., 2002; Peng et al., 2004). Aerosols also affect the Earth's radiative properties. The direct effect of aerosols on climate is due to their ability to scatter and absorb incoming solar radiation (Charleson et al., 1992). The indirect effect of aerosols on climate is the change in the radiative properties of clouds from the altering of cloud droplet/crystal size and concentration and potentially precipitation rates by changes in the number of aerosols acting as Cloud Condensation Nuclei (CCN) (Twomey, 1974; Albrecht, 1989). Both the direct and indirect effects of aerosols have large uncertainties associated with them, partly due to uncertainties in primary anthropogenic aerosols and the nucleation of ultrafine aerosols that are initially too small to act as CCN (IPCC 2007; Spracklen et al., 2008; Makkonen et al., 2009; Pierce and Adams, 2009a; Wang and Penner, 2009).

Measurements of high concentrations of ultrafine particles have been found in the plumes of coal-fired power plants despite the reduction of emissions from installed pollution-control technology (Junkermann et al., 2011a, b). Junkermann et al. (2011b) compared the aerosol size distributions measured in plumes from modern power plants with pollution controls to those from older plants without controls. They found the modern plants to have significantly more ultrafine particles (and total particle number) than the older plants. Their hypothesis as to the source of these ultrafine aerosols is explained by in-stack formation from pollution-control technology byproducts (Srivastava et al., 2004). A fraction of these additional ultrafine particles evolve into CCN and may ultimately affect clouds and climate (Adams and Seinfeld, 2003; Pierce and Adams, 2009a; Konwar et al., 2010; Junkermann et al., 2011a, b). Junkermann et al. (2011b) describes how the shift to cleaner power plants may lead to more CCN that can impact climate. Thus, it is important to understand the various processes that affect the net number of particles formed due to power plants that are affected by pollution-control technologies. Along with the in-stack formation of new particles, changes in plume chemistry and microphysics due to the changes in emissions of sulfur dioxide (SO₂) and nitrogen oxides (NO_x = NO + NO₂) may also affect the net number of particles contributed by a power plant. This latter effect of SO₂ and NO_x emissions changes has yet to be explored.

Recent pollution-control technologies installed on power plants remove SO₂ and NO_x. SO₂ may be oxidized in the gas phase via OH to form sulfuric acid (H₂SO₄) vapors that contribute to new-particle formation and growth (Kulmala and Kerminen, 2008). Thus, a reduction in SO₂ alone would result in a reduction of particles formed in power-plant plumes. However, NO_x controls may either increase or decrease OH concentrations in the plume (depending on the environmental conditions). Thus, in many conditions a reduction in NO_x

may lead to an increase in the rate at which H₂SO₄ is formed and perhaps an increase in particle formation/growth.

In this paper, we explore the effects of SO₂ and NO_x control technologies on nucleation in the plumes of coal-fired power plants. In Sect. 2, we review the various chemical and microphysical processes affecting particle formation in plumes. In Sect. 3, we provide an overview of power-plant control technologies. The methods and the SAM/TOMAS model are described in Sect. 4. Our results, including a detailed case study of the W. A. Parish power plant, a comparison to measurements and an assessment of particle-formation changes from US coal-fired power plants, are given in Sect. 5. In Sect. 6, we present the conclusions.

2 Chemical processes in power-plant plumes

Coal-fired power plants are sources of SO₂, NO_x and primary ash particles (Zhao et al., 2008; Srivastava et al., 2004). However, due to the high efficiency of particulate controls, primary ash emissions in modern coal-fired power plants in developed countries are very low (Miller, 2010). A product of coal combustion is SO₂, which is oxidized in the gas phase by OH to form H₂SO₄ vapor under tropospheric conditions (Seinfeld and Pandis, 2006; Zhao et al., 2011). Under continental surface conditions, H₂SO₄ vapor will quickly (seconds to minutes) form either new ~1 nm aerosol particles (aerosol nucleation) or condense onto pre-existing particles (Pierce and Adams, 2009b). In the presence of clouds, SO₂ is dissolved into cloud or rain droplets and oxidized to sulfate (SO₄²⁻) by aqueous ozone (O₃) or hydrogen peroxide (H₂O₂), which reduces the H₂SO₄ concentrations in the gas phase (Zhou et al., 2012). Since SO₂ is emitted from power plants, H₂SO₄ vapor concentrations are elevated above background levels in plumes and thus power plants may be a major contributor to nucleated particles in the atmosphere under sunny and clear-sky conditions (Yu, 2010; Stevens et al., 2011). The rate of gas-phase SO₂ oxidation depends on the amount of available OH, which depends on the amount of incoming solar radiation, NO_x concentrations and volatile organic compound (VOCs) concentrations.

NO_x is an indirect contributor to new-particle formation rate because it affects the amount of OH available for the SO₂ → H₂SO₄ gas-phase reaction (Pope et al., 1992) (Fig. 1). At high NO_x concentrations (> 30 ppb), the reaction of OH with NO₂ becomes a significant sink for OH, and OH concentrations are reduced with increasing NO_x concentrations. For low NO_x concentrations (< 0.5 ppb), the reaction of NO₂ with OH is unimportant, and OH increases with increasing NO_x (via photolysis of NO₂, enhanced O₃ formation and increased production of O(¹D), in turn increasing the equilibrium level of OH). The peak OH concentration occurs in between these ranges at the transition of the two limiting regimes. Thus, OH concentrations may either increase or decrease for decreasing NO_x emissions (due to the addition of

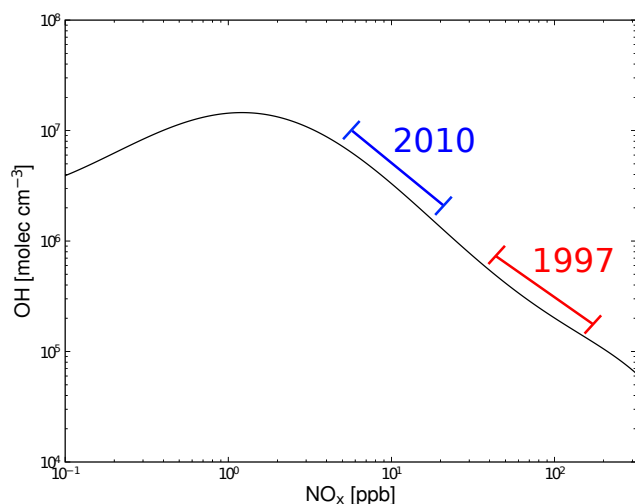


Fig. 1. [OH] as a function of [NO_x] for the background conditions on 27 September 2006. The red and blue bars show the range of NO_x concentrations found in the first 50 km of the plume for the 2010 and 1997 emissions.

control technologies), and gas-phase H₂SO₄ concentrations (and particle formation and growth rates) may subsequently increase or decrease in the plumes.

In addition to H₂SO₄ production contributing to formation and growth (via condensation) rates in the atmosphere, the presence of pre-existing particles also determines if particles will form and survive in a source plume. Condensational growth and coagulation rates depend on the concentration of pre-existing particles (Pierce and Adams, 2007). If a high condensation sink exists (due to a large amount of pre-existing aerosol surface area), H₂SO₄ will condense quickly onto these pre-existing particles, which will lower the H₂SO₄ concentrations and reduce new-particle formation and growth rates. Additionally, freshly nucleated particles are lost through coagulation with the larger pre-existing particles. Thus, large amounts of pre-existing aerosol will reduce the net number of particles formed in a plume both by reducing nucleation and condensational growth rates and by increasing coagulation. Power-plant emissions in polluted-background regions likely generate fewer new particles than in clean regions (Stevens et al., 2012). Although not investigated in this paper, the emissions of primary ash particles by power plants without particulate controls could also provide a large amount of surface area and would also greatly reduce new-particle formation and growth. These primary ash particles may be important in developing countries where particulate controls are not implemented or for historical simulations of periods prior to when these controls were commonplace in developed countries (e.g. the United States before the implementation of the Clean Air Act in 1970 (Miller, 2010)).

Finally, meteorological conditions affect nucleation and growth in the power-plant plumes (Stevens et al., 2012). Both wind and turbulence act to dilute the SO₂ and NO_x emissions from the power plant and thus particle formation. Increased relative humidity will increase the size of hygroscopic aerosols thereby increasing condensation and coagulation rates. Clouds reduce the amount of sunlight for formation of OH. Also, aqueous processes in clouds and raindrops promote the formation of sulfate in the larger, in-cloud aerosols, which reduces the amount of available SO₂ for H₂SO₄ formation and increases the size of the pre-existing particles, which reduces new-particle formation.

3 Clean control technologies in coal-fired power plants

To reduce the anthropogenic impact on the environment by coal-fired power plants, specifically acid rain, particulate matter (PM), and tropospheric ozone concentrations (Seinfeld and Pandis, 2006), pollution-control technologies have been implemented to reduce primary-particle, SO₂ and NO_x emissions (EPRI, 2008). There are numerous techniques for controlling emissions, and we will review the most common techniques here. For a more extensive discussion of pollution-control strategies, please refer to Srivastava et al. (2004) and Miller (2010).

Technologies to reduce the amount of primary particles emitted from source stacks are currently the most effective of the pollution-control technologies. Nearly all power plants in the US and Canada use baghouse filters or electrostatic precipitators (ESP) with removal efficiencies up to 99.9%. Baghouse filters contain fibrous materials that catch large particles (diameters > 1 μm) by inertial and direct impaction and smaller particles by diffusion (Miller, 2010). Because of such high removal efficiencies, emissions rates of primary particles are not included in the EPA's Clean Air Market Database used in this study, which dates back only to 1997 when ash controls were already universally implemented in the US. Therefore, primary ash particles are not included in this study.

Many power plants have switched to burning coal with low sulfur contents to allow for SO₂ emissions reductions without the need to implement new control systems. Another technique to reduce SO₂ emissions includes flue gas desulfurization (FGD, e.g. scrubbers). Different classifications of FGD include wet and dry methods, expressing the production of waste in the form of a wet slurry or dry material, respectively. A disadvantage of FGD includes the sometimes-inefficient removal of sulfur trioxide (SO₃) (which will quickly form H₂SO₄) as a by-product of the scrubbing process, and this may lead to particle formation in the stack (Junkermann et al., 2011b; Srivastava et al., 2004). This particle-formation pathway will not be considered in this paper, although its implications will be discussed.

Most coal-burning power plants now contain NO_x controls in the form of Low NO_x Burner Technology (LNBT), the over-fire air technique, or a combination of both. Both of these techniques aim to create combustion conditions that reduce the amount of N₂ (from air) oxidized to NO and generally have an efficiency up to 40–60 % (relative to uncontrolled emissions). An additional technology, selective catalytic reduction (SCR), has more recently been added, often in combination with LNBT or over-fire air, to many plants to increase removal efficiencies (> 90 % NO_x reductions). SCR involves a catalyst and the addition of a reagent, ammonia (NH₃), that promotes the reduction of NO_x to nitrogen and water. A negative attribute of SCR includes an SO₃ by-product that will quickly form H₂SO₄ (Miller, 2010). Thus, similar to FGD, SCR may also lead to particle formation in the power-plant stack (Srivastava et al., 2004; EPRI, 2008). Another disadvantage of SCR is that NH₃ may not react completely with NO_x and may be emitted to the atmosphere (EPRI, 2008; Wang et al., 2012). This process is called ammonia slip and can be somewhat controlled with careful tuning of the SCR system and regular maintenance of the catalysts (Miller, 2010; Gong et al., 2012). Since NH₃ is potentially an important enhancer of aerosol nucleation rates (Merikanto et al., 2007; Kirkby et al., 2011), this too may affect particle formation. This influence on particle formation is not explored in this paper, but is explored in Gong et al. (2012). Gaseous ammonia may also condense directly onto acidic aerosols (Seinfeld and Pandis, 2006), increasing aerosol mass and size, hence influencing subsequent condensation and coagulation rates.

4 Methods

We use the SAM/TOMAS model to explore the dynamics and aerosol microphysics (nucleation, condensation and coagulation) of power-plant plumes (Stevens et al., 2012). The host model is the System for Atmospheric Modeling (SAM), a Large-Eddy Simulation (LES) and Cloud Resolving Model (CRM) (Khairoutdinov and Randall, 2003). Stevens et al. (2012) evaluated the model's ability to predict nucleation and growth in the sulfur plumes of anthropogenic point sources through testing of several nucleation schemes and varying background conditions. With comparisons to aircraft measurements in two power-plant plumes, the model provided reasonable predictions of in-plume particle number concentration and size at distances 20–100 km downwind of the source. In this paper, we use grid-box sizes that are 400 m × 400 m horizontal resolution and two different vertical resolutions of 40 m and 80 m (depending on the boundary layer height). The simulations contain the gas-phase species of SO₂, H₂SO₄, NO_x and NH₃. OH concentrations are parametrized based on NO_x concentrations, incoming solar radiation and an assumed concentration of reactive VOCs (Stevens et al., 2012). The number and mass

of sulfate aerosol, ammonium and water are tracked in 15 size sections spanning 3 nm to 10 μm using the TOMAS microphysics scheme (Adams and Seinfeld, 2002; Pierce and Adams, 2009a). Power-plant emissions of SO₂ and NO_x are obtained from the Clean Air Market emissions inventory (Clean Air Markets – Data and Maps, 2012). Primary-particle emissions are assumed to be negligible in model simulations. Emissions are assumed to be well-mixed in the emissions grid-box. Other inputs to the model include background SO₂, NO_x, NH₃ and aerosol size distributions acquired from Texas Air Quality Studies (TexAQS) field campaigns near Houston, Texas during 2000 and 2006. The large-scale meteorological forcing of the SAM simulations are provided by vertical profiles of mean winds, temperature, relative humidity and surface sensible heat, latent heat and momentum fluxes from the National Center for Environmental Prediction (NCEP) North American Regional Reanalysis (NARR) data (Mesinger et al., 2006). The reanalysis data were provided by the National Oceanic and Atmospheric Administration (NOAA)/Outstanding Accomplishments in Research (OAR)/Earth System Research Laboratory (ESRL) Physical Sciences Division (PSD), Boulder, Colorado, USA, from their website at <http://www.esrl.noaa.gov/psd/>. The NCEP NARR data has a spatial resolution of 32 km by 32 km and 3-h time resolution. Although SAM resolves turbulent flows on spatial scales smaller than the NCEP NARR data, any systematic (non-turbulent) variability in meteorology (e.g. flow around hills or buildings) will not be captured by the SAM model, and this could lead to uncertainties in the plume dispersion. Aerosol nucleation is calculated using the empirical activation nucleation scheme (Kulmala et al., 2006), where nucleation rates are equal to $A[\text{H}_2\text{SO}_4]$ and $A = 10^{-7} \text{ s}^{-1}$. Various nucleation schemes were tested in Stevens et al. (2012), and the activation scheme compared best against observations from the TexAQS2006 field campaign.

In these simulations, we do not consider the oxidation of SO₂ within clouds (the meteorology on the tested days contained cloud-free or nearly cloud-free boundary layers). Also, we do not consider the loss of NO_x by chemical reactions. The direct effect of the NO₂+OH reaction on OH is implicit in the OH parameterization; however, this does not result in the reduction of NO_x concentrations. Early in the plume, the mixing of the plume with background air will dominate the reduction of NO_x concentrations in the plume, so the chemical loss of NO_x is less important than later in the plume. NO_x lifetime in high [OH] conditions is only a few hours during the day, with the summertime lifetimes being shorter as compared to winter (Beirle et al., 2011). Thus, we expect to over-predict NO_x concentrations with increasing distances from the source. VOC concentrations were assumed fixed in the OH parameterization in our simulations (150 pptv of isoprene). Changes in VOCs may affect OH concentrations resulting in different SO₂ oxidation rates (Stevens et al., 2012). The formation of Secondary

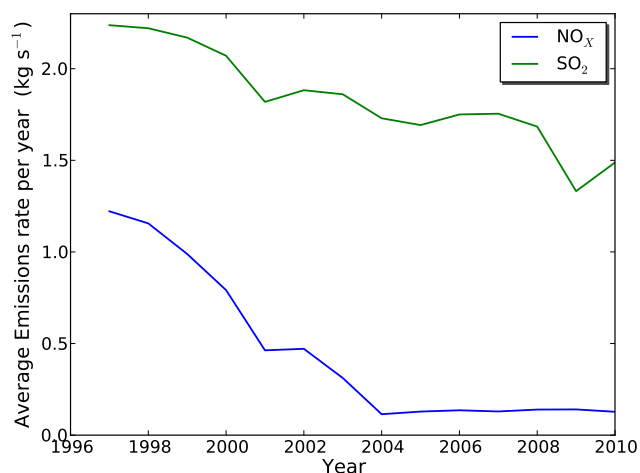


Fig. 2. The yearly average emissions (kg s⁻¹) of SO₂ (green) and NO_x (blue) from the 4 coal-burning units (summed together) at the W. A. Parish Power Plant.

Organic Aerosol (SOA) is not considered in this study. Our assumption is that H₂SO₄ formation will dominate growth in the SO₂-rich power-plant plumes, and we were able to reproduce most of the growth in two plumes in Stevens et al. (2012) (also see Figs. 7 and 8). However, the lack of SOA is an additional uncertainty in this study. We also do not consider any primary ash particles (which would result in a decrease in nucleation rates) or particles that may have formed in the stack because of high SO₃ formation from FGD or SCR (which would increase the number of particles in the plumes (Junkermann et al., 2011b)). This formation of SO₃ by the pollution control technologies is an additional way in which pollution controls can lead to increased numbers of ultrafine particles. However, this formation pathway is not explored in this paper.

5 Results

5.1 Changes in the W. A. Parish plume between 1997 and 2010

We have compared the model to observations (Stevens et al., 2012) and shown that it captures the essential features of aerosol size distributions in anthropogenic plumes. In this section, we discuss the sensitivity of the model to changes in SO₂ and NO_x emissions rates due to pollution-controls from the W. A. Parish coal-fired power-generation facility between 1997 and 2010.

The annually averaged SO₂ and NO_x emissions from the W. A. Parish power plant from 1997 to 2010 are shown in Fig. 2. In 1997, four of the eight total units were coal-fired sources (units 5, 6, 7 and 8). All four of these units contained particle controls in the form of baghouse filters. NO_x controls were installed on unit 5 (LNBT) and unit 8 (over-fire air)

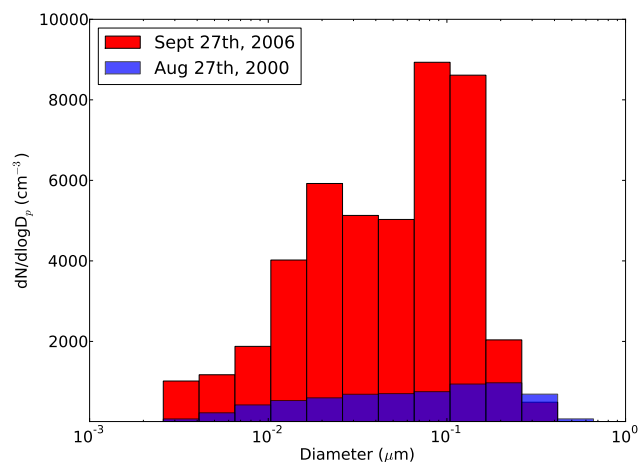


Fig. 3. The measured background particle size distributions by the NOAA P-3 aircraft on 27 September 2006 (red) and the NCAR Electra aircraft 27 August 2000 (blue) near the W. A. Parish Power Plant. The condensation/coagulation sink was $\sim 4\times$ larger for the 2006 case.

prior to 1997. There is no record of NO_x controls on units 6 and 7 in 1997. By 2004, all units contained LNBT with either over-fire air or combinations with SCR, and by 2005 all units contained SCR. For SO₂ controls, only unit 8 is recorded to have Wet Lime FGD for all years, and the other three units have no record of SO₂ controls for any years. From the Clean Air Markets database, we derived that in 1997 the average NO_x and SO₂ emissions for the W. A. Parish power plant were 1.22 kg s⁻¹ and 2.24 kg s⁻¹, respectively. In 2010, these emissions were reduced to 0.128 kg s⁻¹ and 1.49 kg s⁻¹ for NO_x and SO₂, respectively (Fig. 2). Even though changes in SO₂ control implementations were not recorded, there is an indication of emissions reduction in the database, and this may be due to reduced sulfur content in the coal. The emissions of both gaseous species decreased over the 13-year period; however, NO_x rates decreased by $\sim 90\%$ while SO₂ only decreased by $\sim 30\%$.

To estimate how the emissions changes between 1997 and 2010 may have affected particle formation in the plume, we performed two simulations: one with the 1997 emissions and another with the 2010 emissions, each using meteorology and chemical background conditions from 27 September 2006 during the TexAQS2006 field campaign as tested in Stevens et al. (2012). This day had clear skies and a boundary-layer height of 1000 m. The mean boundary-layer winds were northward at 5 m s⁻¹ avoiding Houston, Texas with somewhat polluted remote-continental background aerosol concentrations (red bars in Fig. 3). Figure 4 shows the predicted Net Particle Contribution (NPC) rate as a function of distance from the stack. We define the NPC rate as the net number of particles that the power plant has contributed per unit time at a given distance from the stack accounting for both particle gains by new particle formation

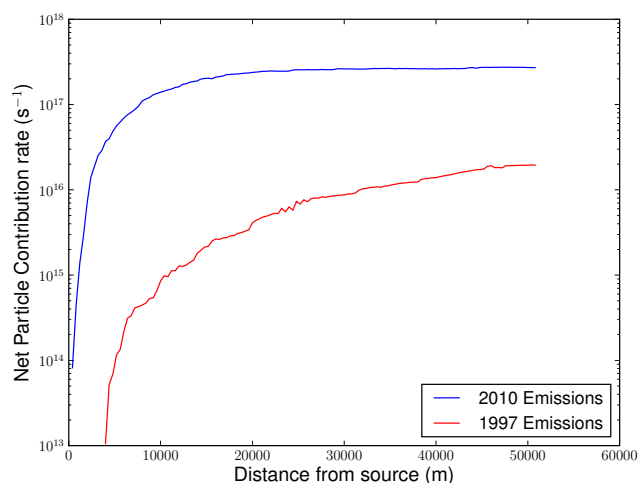


Fig. 4. The Net Particle Contribution (NPC) versus distance downwind of W. A. Parish Power Plant for the 2010 (blue) and 1997 (red) emissions scenarios. Meteorology and background conditions were for 27 September 2006 for both cases.

and loss by coagulation. The NPC rate is the effective emissions rate of particles from the power plant once chemistry and physics in the plume has been accounted for. It depends on the distance from the stack because both nucleation and coagulation shape the aerosol distribution as the plume moves further from the stack.

In Fig. 4, it can be seen that there is over an order of magnitude increase in the predicted NPC rate between 1997 and 2010 (with background conditions fixed). This time range includes the historical record of NO_x and SO₂ emissions for W. A. Parish provided by the Clean Air Markets database. This increase occurs even though SO₂ emissions have decreased by 30%. The increase is due to the large reduction of NO_x emissions that has caused an increase in the predicted OH in the plume. The OH and NO_x changes for the corresponding years are shown in Fig. 1. The predicted OH concentrations have increased by ~10x in the plume between 1997 and 2010. Thus, SO₂ is oxidized ~10x more quickly in 2010 leading to larger H₂SO₄ concentrations early in the plume even though SO₂ emissions rates were reduced by ~30%.

The predictions shown here are limited to a single set of background conditions. As we will show later (in Sect. 5.3), the sensitivity of the NPC rate to changes in emissions depends greatly on the background conditions. In the limit of no or low sunlight (e.g. night) or high background particle concentrations, no nucleation will take place regardless of emissions (Stevens et al., 2012), and the NPC rate will have no sensitivity to emissions (it will be zero).

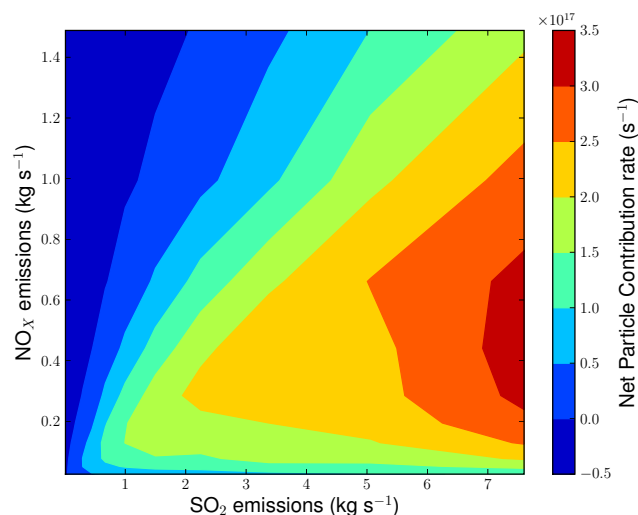


Fig. 5. The interpolated NPC rates (50 km downwind from source) from 110 model simulations versus NO_x and SO₂ emissions. Meteorology and pre-existing particle background concentrations for all model runs were for 27 September 2006 near the W. A. Parish power plant.

5.2 Net Particle Contribution rates versus SO₂ and NO_x

To better understand how NPC rates depend on SO₂ and NO_x emissions, Fig. 5 shows the NPC rates at 50 km downwind of a source stack against NO_x emissions ranging from 0–1.4 kg s⁻¹ and SO₂ emissions ranging from 0–7.5 kg s⁻¹ (these ranges encompass most power plants in the US). Similar to Fig. 4, the background conditions are set to those for 27 September 2006 at the W. A. Parish power plant. 110 individual SAM/TOMAS simulations are used to populate the figure. The NPC rate ranges from over 10¹⁷ s⁻¹ in high-SO₂ emissions cases to zero particles in very-low-SO₂ emissions cases. Increasing SO₂ emissions rates generally increases the NPC rate. As would be expected based on Fig. 1, increasing NO_x emissions leads to an increase in the NPC rate if NO_x emissions are low (OH increases with increasing NO_x), and increasing NO_x emissions leads to a decrease in the NPC rate if NO_x emissions are high (OH decreases with increasing NO_x). These results indicate that for power plants with high NO_x emissions (> ~0.6 kg NO_x s⁻¹ according to Fig. 5), SO₂ concentrations must be reduced by a larger fractional amount than NO_x in order for the net particle production in the plume to decrease. Similar to the previous section, these results are only for a single background. The magnitude of the predicted NPC rates will depend greatly on the background concentrations (Stevens et al., 2012), which will be shown in the following section.

5.3 Observational evidence and comparison of model to measurements

Aircraft-based measurements of coal-fired power-plant plumes using the suite of instruments required to set-up and test the model (e.g. SO₂, NO_x, fast aerosol size distribution measurements) are quite sparse. To ideally identify the effect of pollution-control technologies from observations, many aircraft-based measurements of power-plant plumes would be needed both before and after pollution controls were implemented. A large number of measurements would be required in order to average over day-to-day variability in background meteorology and aerosol concentrations. These before/after measurements are even more sparse, though we have found one set of measurements that we will analyze here.

The NCAR Electra aircraft and NOAA P-3 aircraft made transects through the W. A. Parish power plant in the TexAQS2000 and TexAQS2006 field campaigns, respectively. For each campaign, we have identified a measurement case where the power-plant plume was able to evolve with minimal additional anthropogenic emissions mixing into the plume (e.g. the plume must avoid the city of Houston). The TexAQS2000 flight measurements were on 27 August 2000 and the TexAQS2006 measurements were on 27 September 2006 (described in Sect. 5.1). There were significant changes in NO_x and SO₂ emissions between these dates (Fig. 2). Although the change in emissions is smaller between 2000 and 2006 than it is between 1997 and 2010 (as tested earlier), NO_x emissions were still reduced by 83% and SO₂ emissions were reduced by 15% between 2000 and 2006. The boundary layer height on 27 August 2000 was recorded to be at a maximum of ~2000 m above the surface with northward winds ranging from 3–6 m s⁻¹ (Brock et al., 2003). Fair-weather boundary-layer cumulus clouds were occasionally present on this day; however, during the time of flight, low-cloud coverage was less than 10% according to the NCEP North American Regional Reanalysis. As mentioned earlier, in-cloud oxidation of SO₂ was not included in the model; however, the reduction of downward short-wave radiative flux by the clouds was accounted for in the model inputs.

There was a large difference in the background aerosol size distributions between the campaign days as seen in Fig. 3. The condensation/coagulation sink of the background aerosol for the 2000 case was only 0.25 of the 2006 case. The total mass of particles smaller than 600 nm (PM_{0.6}) for the 2000 and 2006 background was 6.8 μg m⁻³ and 9.3 μg m⁻³ respectively. The difference between the PM_{0.6} values of the two cases are more similar than the difference between the condensation sink values because the size distribution for the clean-2000 case is skewed towards larger diameters (see Fig. 3). Thus, we expect the differences in the condensation and coagulation sinks to have a significant effect on the NPC rates, so unfortunately the measured effect of the changes in power-plant emissions between 2000 and 2006

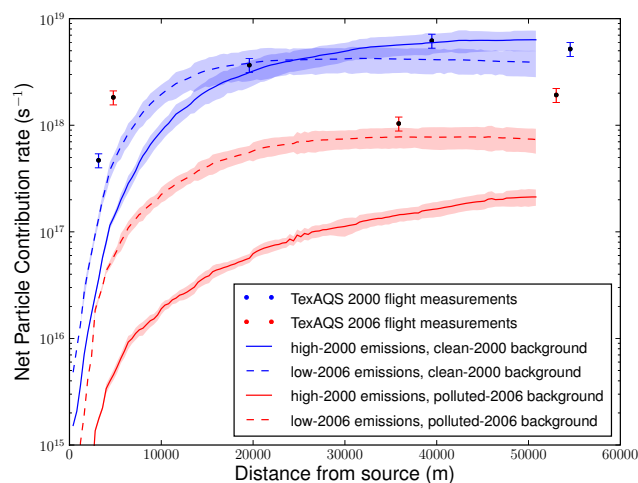


Fig. 6. NPC rate as a function of distance downwind of the W. A. Parish power plant for the two sets of measurements and the model simulations with the four combinations of emissions and background conditions for the 2000 and 2006 cases. The shaded regions correspond to the standard deviation in the model-predicted NPC with time. The error bars correspond to the estimated effect of measurement error (Brock et al., 2002) on the measured NPC rate.

cannot be isolated in the measurements alone. In this section, we will use the model as a means of interpreting the measured response to the emissions changes by isolating the emissions and background-aerosol changes. We present 4 simulations: one with clean-2000 background concentrations and meteorology with high-2000 NO_x and SO₂ emissions, one with the clean-2000 background concentrations and meteorology with low-2006 emissions, one with polluted-2006 background concentrations and meteorology with high-2000 emissions, and one with the polluted-2006 background concentrations and meteorology with low-2006 emissions.

Figure 6 shows the NPC rate for the four simulations and the measurements. The model is generally within a factor of 2 for NPC at distances beyond 20 km from the stack for both the 2000 and 2006 cases when the corresponding emissions and backgrounds were simulated. However, the model underpredicts the NPC rate for the first transect in both cases. We will explore the reasons and possible sources of these additional particles below.

The effect of the background aerosol concentrations on the simulated NPC rate in Fig. 6 was significant, consistent with Stevens et al. (2012). The difference between the clean-2000 background cases (blue) and the polluted-2006 background cases (red) was about a factor of 5–20 when the emissions were held fixed. Thus, the differences in background likely contributed significantly to the difference in the measured NPC rates between 2000 and 2006. The model also showed that the difference between the 2000 and 2006 measurements could be even larger if the emissions were held fixed under the polluted-2006 background conditions. Thus, if the

model-predicted dependence of the NPC rate on the background concentrations is correct, the change in background between the 2000 and 2006 cases resulted in a clear decrease in NPC (that is larger in magnitude than the effect of the emissions change).

The model predicts that the sensitivity to changing emissions under fixed-background conditions is significantly larger during the polluted-2006 conditions (difference between the red lines) than during the clean-2000 conditions (difference between the blue lines). Under the clean-2000 background conditions, nucleation proceeds quickly in both emissions cases (although more quickly initially in the case of 2006 emissions). The nucleation mode quickly becomes the dominant condensation and coagulation sink (the pre-existing particles do not contribute greatly to these sinks). In the low-2006 emissions case (with the clean-2000 background conditions), the initial condensation and coagulation sink formed in the plume is somewhat larger than the high-2000 emissions case (with the clean-2000 background conditions), and this provides a negative feedback on the NPC rate due to slower nucleation and faster coagulation. In addition, the low-2006 emissions of NO_x in combination with the abnormally large boundary-layer height of the clean-2000 background (~4 km) and the clean (low-NO_x) background conditions allow NO_x concentrations within the plume to fall below 1 ppb. NO_x concentrations below 1 ppb result in decreased OH concentrations and decreased H₂SO₄ formation within the plume. This causes nucleation within the plume to slow dramatically beyond 20 km from the stack in the low-2006 emissions case, while nucleation continues within the plume in the high-2000 emissions case that never predicts NO_x concentrations below 1 ppb. Thus, the NPC rate for the low-2006 emissions case falls slightly below the rate for the high-2000 emissions case after 25 km (for the clean-2000 background). For the polluted-2006 background conditions, the pre-existing particles continue to be the dominant condensation and coagulation sink throughout the plume (unlike under the clean-2000 background conditions). Thus, the total condensation and coagulation sink did not depend greatly on the SO₂ and NO_x emissions. Unlike the clean-2000 background conditions, there is no strong negative feedback in the polluted-2006 background cases that reduce the new-particle formation rates and increase the coagulation rates for the low-2006 emissions case, and this facilitates a large fractional difference in the NPC rate between the low-2006 and high-2000 emissions cases. In addition, the more shallow boundary-layer of the polluted-2006 background conditions does not dilute the emissions to the same extent as in the clean-2000 cases. Thus, as mentioned earlier, the sensitivity of the NPC rate to emissions is strongly sensitive to the background conditions.

The particle size distribution of the model simulations and flight measurements are shown in Figs. 7 and 8. Figure 7 contains the simulated (solid lines) and aircraft-measured (dashed lines) size distributions for the 2000 case (high-2000

emissions and clean-2000 background in the model) with the size distribution averaged across the plume at the four transects made by the plane; 3.2 km (blue), 19.6 km (red), 39.5 km (green) and 54.5 km (purple) from the source stack. Figure 8 contains the model and aircraft-measured size distributions for the 2006 case (low-2006 emissions and polluted-2006 background in the model) at the three transects made by the plane; 4.8 km (blue), 35.9 km (red) and 53.0 km (green) from the source stack. It should be noted that the model domain ends at 50 km down wind of the stack, and the size distributions at this edge of the model domain are used for comparison to the furthest aircraft transects. Similar to the NPC rates in Fig. 6, the model does better at predicting the particle distributions further downwind in the plume. In both cases the model does not capture a measured mode with a median diameter around 20 nm at the closest transects. It is these 20-nm particles that are the source of the low model bias in Fig. 6. This initial peak may be particles that formed in stack due to SO₃ emissions (Junkermann et al., 2011b; Srivastava et al., 2004). SO₃ emissions may be due to the FGD on one of the four units, or they may be due to the SCR on all four units, although the SCR was only installed for the 2006 case and would not influence the 2000 case. It is also possible that photolysis of HONO emitted by the power plant is contributing to enhanced OH concentrations near the source (Gonçalves et al., 2009; Elshorbany et al., 2010). The model does not simulate HONO and would miss this extra OH. We plan to investigate the role of SO₃ and HONO in future work. In both cases, the model captures the evolution of the size distributions increasingly well with later transects showing that the in-plume processes simulated by the model are dominating the size distribution outside of the first 10–20 km. However, the initial 20-nm mode, while diminished due to coagulation, grows to enhance ~100-nm particles, and this enhancement is not captured by the model.

This sensitivity to background conditions is particularly important because power plants may correlate (or anti-correlate) with highly polluted regions. For example, the power plants in the Ohio River Valley will exist in generally polluted air. Thus, future work will be done using 3D chemical transport models to explore these correlation effects.

5.4 US emissions statistics and estimated particle formation

The increase in particle production of a single coal-fired power plant may not be indicative of broader trends in power plants; therefore, we explored how particle production may have changed for each plant in the US based on their SO₂ and NO_x emissions changes. Using the Clean Air Markets database, the 1997 and 2010 SO₂ and NO_x emissions were found for each of the available 330 coal-fired power plants in the US (that was operational in each of these years). Figure 9 shows the ratio of emissions between 2010 and 1997 for SO₂ and NO_x emissions for these plants. Nearly all power plants

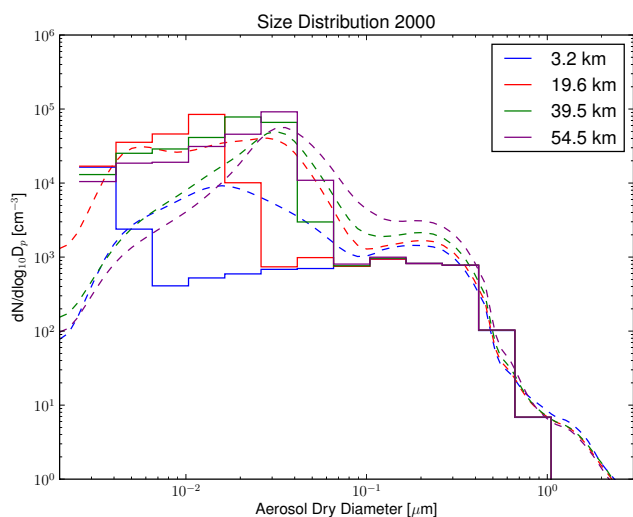


Fig. 7. The average particle size distributions across the 2000 W. A. Parish plume at each transect of the TexAQS2000 measurements for both the aircraft measurements and the model (using both 2000 emissions and background). Solid lines represent the model and dashed lines represent the flight measurements at 3.2 km (blue), 19.6 km (red), 39.5 km (green) and 54.5 km (purple) downwind of the source stack. The purple solid line represents the average size distribution of the model at 50 km downwind.

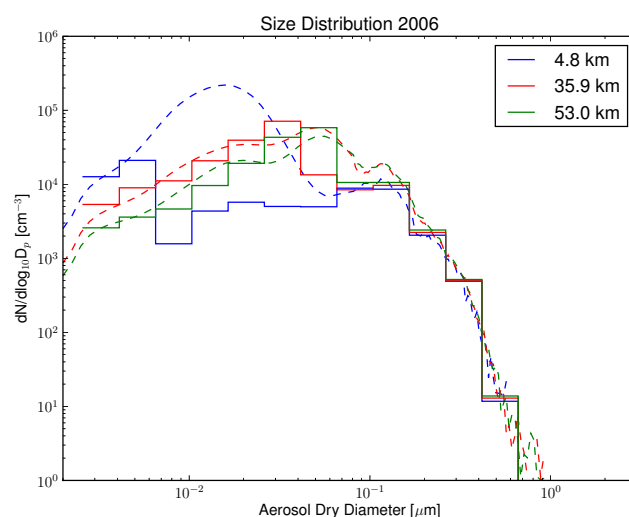


Fig. 8. The average particle size distributions across the 2006 W. A. Parish plume at each transect of the TexAQS2006 measurements for both the aircraft measurements and the model (using both 2006 emissions and background). Solid lines represent the model and dashed lines represent the flight measurements at 4.8 km (blue), 35.9 km (red) and 53.0 km (green) downwind of the source stack. The green solid line represents the average size distribution of the model at 50 km downwind.

show decreases in both species between 1997 and 2010 with a median ratio of 0.64 and 0.48 for SO₂ and NO_x respectively.

In the case of the W. A. Parish plant explored throughout this paper, NO_x emissions reductions are significantly greater than the SO₂ emissions reductions. Thus, the emissions changes for the plant are not representative of the general US population of coal-fired power plants. We estimate how the NPC rate from each power plant would change between 1997 and 2010 under the fixed meteorological and background chemical conditions of the W. A. Parish power plant on 27 September 2006 by interpolating the NPC rates in Fig. 5. Figure 10 shows the ratio of the predicted NPC rate between 2010 and 1997 for the 330 plants under the fixed background conditions. Based on these plants, we estimate that there is a net reduction of particle formation between the two years with a median decrease of 19 %, when background conditions are held constant. This overall decrease could be due, in part, to NO_x emissions from smaller power plants being low enough such that the plume is not in the high-NO_x regime. For these power plants, reductions in NO_x do not lead to large increases in OH and may even lead to decreases in OH.

It appears that the W. A. Parish power plant investigated throughout this paper may be an extreme case relative to the median of the US power plants because of its stronger NO_x controls than SO₂ controls. Again, we stress that the results for the sensitivity of NPC rate to emissions changes will depend on the background meteorology and chemistry. How-

ever, the results show that the median change in the NPC rate across the 330 power plants is small. This result is likely robust to changes in background conditions because changes in the background will generally not change the sign of how emissions change the NPC rate. Because the individual 330 plants show a wide range of increases and decreases in NPC rate that largely cancel out (Fig. 10), this cancellation should not change with the background. However, a full analysis of the effect of power-plant emissions changes on aerosol concentrations using regional and global chemical transport models (using a parameterized version of sub-grid nucleation based on results from the SAM-TOMAS model) is planned for future work, which will allow for emissions changes to be considered over the full range of atmospheric conditions and aerosol properties to be predicted further from the sources.

6 Conclusions and discussion

In this paper, we explored how power-plant controls may have affected new-particle formation in coal-fired power-plant plumes. Using the meteorology and background conditions from NCEP/NARR datasets and TexAQS aircraft measurements, we simulated the nucleation of particles at the W. A. Parish coal-fired power plant for 1997 and 2010 emissions with the SAM/TOMAS model. The decreasing emissions rates of SO₂ (30 %) and NO_x (90 %) due to pollution-control technologies during this time were simulated, and model results indicated a significant increase in aerosol

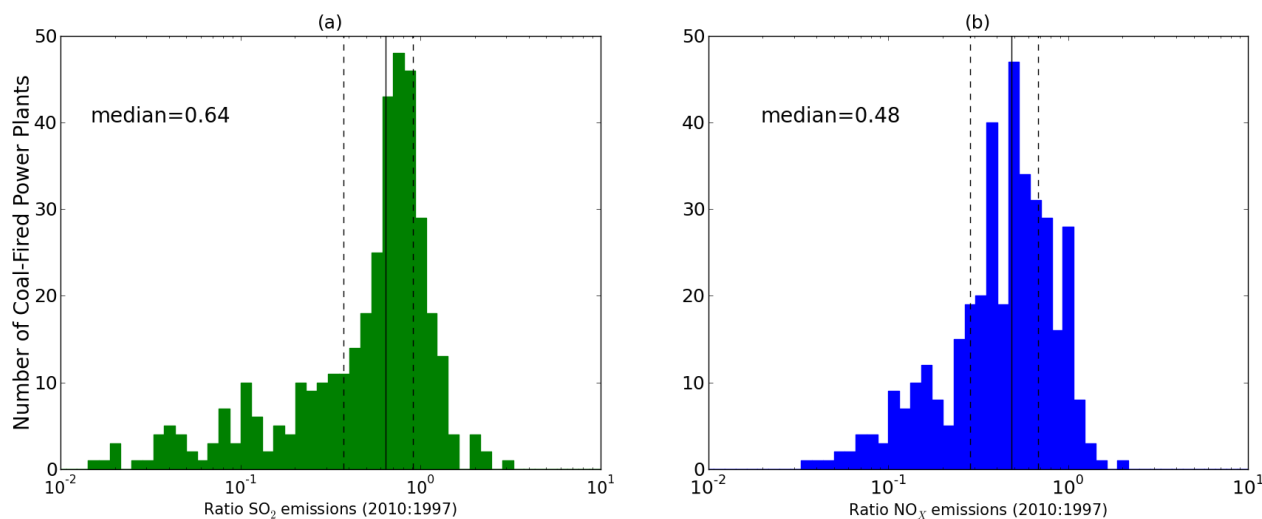


Fig. 9. Histograms showing the ratio of (a) SO₂ and (b) NO_x emissions changes between 2010 and 1997 for all (330) coal-fired power plants in the US registered to the CAM database. The mean ratio (solid line), 25 % and 75 % (dashed-lines) quartiles are plotted. There is an overall decrease in emissions with a median ratio of 0.64 SO₂ and 0.48 NO_x for 2010:1997.

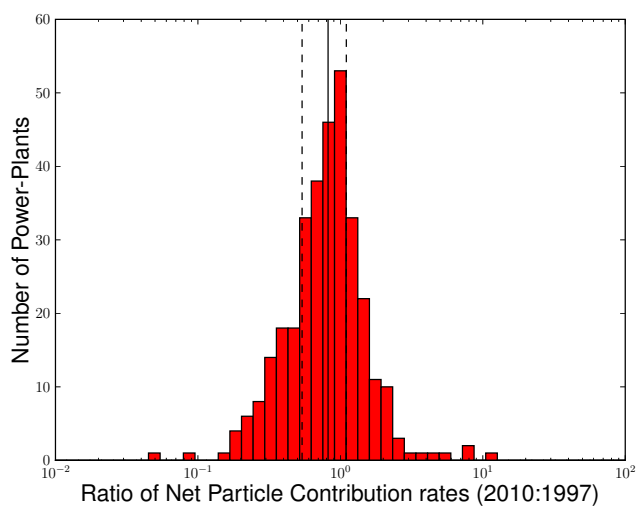


Fig. 10. Histogram showing the ratio of predicted NPC rates between 2010 and 1997 for all US power plants. The plot was made using the NO_x and SO₂ emissions from each of the years and the predicted NPC rates from these emissions shown in Fig. 5. The NPC rates were calculated for the background conditions of 27 September 2006 and produced a median ratio of 0.81. The solid line represents the median and dashed lines represent the inter-quartile ranges.

formation in the plume. The model showed an order-of-magnitude increase in Net Particle Contribution rate (NPC, the effective emissions of ultrafine particles from the plant taking into account formation and coagulation in the plume) 50 km downwind of the plant during this time period. This predicted increase in NPC rate was due to a decreasing in

NO_x emissions, which caused an increase in OH and the oxidation rate of SO₂ to H₂SO₄.

We determined generally how SO₂ and NO_x emissions changes affect NPC under fixed atmospheric conditions. For power plants with high NO_x emissions, SO₂ concentrations must be reduced by a larger fractional amount than NO_x concentrations in order for particle production to decrease in the plume. Using these results, the US-median NPC rate was estimated to have a 19 % decrease between 1997 and 2010 assuming fixed background conditions. However, the results of this paper are limited by the comparison to only two background conditions, and thus the magnitude of the NPC changes may, on average, be different from these predictions.

Model predictions were compared to aircraft measurements from the TexAQS 2000 and 2006 campaigns. This analysis showed that the change in plume particle concentrations were likely influenced more strongly by differences in the pre-existing particle concentrations between measurement days than by differences in emissions between the years. Meteorological conditions will also have a large influence on the plume microphysics. Thus, it is difficult to quantify the impact of emissions controls on aerosol formation in the plumes from measurements alone without a large number of measurements both before and after the controls were added. In addition, the simulations predict that the sensitivity to changing emissions under fixed background conditions is much larger under more polluted conditions (2006) than during the less polluted conditions (2000).

Junkermann et al. (2011b) has shown evidence of increased particle concentrations in plumes of coal-fired power plants with new pollution-control technologies measured in Germany, Inner Mongolia and Southern Australia. They

attributed the increased particle concentrations to the in-stack formation of SO₃/H₂SO₄ from Flue Gas Desulfurization and Selective Catalytic Reduction technologies (which may lead to particle formation in/near the stack). They did not investigate the potential for changes in plume chemistry and nucleation as we do here. On the other hand, we did not address the in-stack processes in this paper since emissions data of SO₃, H₂SO₄ or ultrafine particles formed in the stack is not readily available for US power plants. However, a significant mode of ~20-nm particles were evident in aircraft measurements within 5 km of the W. A. Parish stack in both 2000 and 2006 that was not resolved by the model. This is likely due to the in-stack processes discussed in Junkermann et al. (2011b). While we find that in-plume (as opposed to in-stack) nucleation can account for most of the measured particles beyond 20 km downwind of the stack, the unintended formation of SO₃/H₂SO₄ could cause significant changes in the particle concentrations in plumes under some conditions and will be investigated in future work. Additionally, NH₃ slip from SCR may lead to increased nucleation rates in a plume. We estimated the possible effects of NH₃ slip in Gong et al. (2012) but do not address this here as NH₃ emission data is also not readily available for US power plants. The aqueous oxidation of particles in clouds was also not considered in this work due to little to no plume/cloud interaction in the test cases. For cases where the plume enters clouds, there would be a decreased NPC rate in the plume due to the aqueous removal of SO₂, the addition of sulfate to activated particles, as well as reduced OH because of the reduction in sunlight. VOC concentrations were assumed fixed in our simulations, and changes in VOCs may affect OH concentrations resulting in different SO₂ oxidation rates for fixed NO_x emissions and backgrounds. Finally, growth of the particles via SOA was not explored, and may also contribute to growth, even in the sulfur-rich plume.

This study has shown that an unintended result of pollution-control technologies may be occurring with the addition or loss of particles produced within power-plant plumes. In order to accurately estimate the cumulative effect of aerosols on climate and health, primary emissions, nucleation and growth of particles in plumes of power plants must be understood. With the continuing construction of coal-fired power plants and the changes in pollution-control technologies, there is a strong need for additional modeling and measurements of power-plant plumes in order to quantify this impact on a global scale. Future work will involve data analysis from additional flight tracks through power-plant plumes, upon availability, for further testing against model simulations of particle production as well as the additional investigation of in-stack particle formation. Additionally, we are creating a sub-grid plume parameterization for three-dimensional regional and global models to more accurately quantify aerosol microphysics in coal-fired power-plant plumes and the effects of pollution-control technologies. Using this parameterization, the impact of power-plant

controls on downwind CCN production can be studied using regional and global models. Regional changes in nucleation and growth are also expected due to changes in pollution controls (Jung et al. 2010; Fountoukis et al., 2012), and these effects must be considered along with the in-plume nucleation changes considered here.

Acknowledgements. This research was funded by Environment Canada and the Electrical Power Research Institute (EPRI). We would like to thank Annmarie Carlton of Rutgers University for her suggestions.

Edited by: B. Vogel

References

- Adams, P. J. and Seinfeld, J. H.: Disproportionate impact of particulate emissions on global cloud condensation nuclei concentrations, *Geophys. Res. Lett.*, 30, 1239, doi:10.1029/2002GL016303, 2003.
- Albrecht, B.: Aerosols, cloud microphysics, and fractional cloudiness, *Science*, 245, 1227–1230, 1989.
- Arya, S. P.: *Air Pollution Meteorology and Dispersion*, Oxford University Press, Inc., New York, USA, 309 pp., 1999.
- Beirle, S., Boersma, K. F., Platt, U., Lawrence, M. G., and Wagner, T.: Megacity Emissions and Lifetimes of Nitrogen Oxides Probed from Space, *Science*, 333, 1737–1739, 2011.
- Brock, C., Trainer, M., Ryerson, T., Neuman, J., Parrish, D., Holloway, J., Nicks, D., Frost, G., Hübler, G., Fehsenfeld, F., Wilson, J., Reeves, J., Lafleur, B., Hilbert, H., Atlas, E. L., Donnelly, S., Schauffler, S. M., Stroud, V. F., and Wiedinmyer, C.: Particle growth in urban and industrial plumes in Texas, *J. Geophys. Res.* 108, 4111, doi:10.1029/2002JD002746, 2003.
- Canadian Environmental Protection Act Federal-Provincial Advisory Committee Working Group on Air Quality Objectives and Guidelines, National Ambient Air Quality Objectives for Particulate Matter, Science Assessment Document, Health Canada and Environment Canada, Ottawa, Ontario, Canada, 1999.
- Clean Air Markets: Data and Maps, available at: <http://ampd.epa.gov/ampd>, last access: April 2012.
- Charlson, R. J., Schwartz, S. E., Hales, J. M., Cess, R. D., Coakley, J. A., Hansen, J. E., and Hofmann, D. J.: Climate forcing by anthropogenic aerosols, *Science*, 255, 423–430, 1992.
- Electric Power Research Institute: Estimating Total Sulfuric Acid Emissions from Stationary Power Plants, Report 1016384, Palo Alto, CA: Electric Power Research Institute, 2008.
- Elshorbany, Y. F., Kurtz, R., Wiesen, P., Lissi, E., Rubio, M., Villena, G., Gramsch, E., Rickard, A. R., Pilling, M. J., and Kleffmann, J.: Oxidation capacity of the city air of Santiago, Chile, *Atmos. Chem. Phys.*, 9, 2257–2273, doi:10.5194/acp-9-2257-2009, 2009.
- Fountoukis, C., Riipinen, I., Denier van der Gon, H. A. C., Charalampidis, P. E., Pilinis, C., Wiedensohler, A., O'Dowd, C., Putaud, J. P., Moerman, M., and Pandis, S. N.: Simulating ultrafine particle formation in Europe using a regional CTM: contribution of primary emissions versus secondary formation to aerosol number concentrations, *Atmos. Chem. Phys.*, 12, 8663–8677, doi:10.5194/acp-12-8663-2012, 2012.

- Gonçalves, M., Dabdub, D., Chang, W. L., Saiz, F., Jorba, O., and Baldasano, J. M.: The impact of different nitrous acid sources in the air quality levels of the Iberian Peninsula, *Atmos. Chem. Phys. Discuss.*, 10, 28183–28230, doi:10.5194/acpd-10-28183-2010, 2010.
- Gong, L., Lewicki, R., Griffin R. J., Tittel, F. K., Lonsdale, C. R., Stevens, R. G., Pierce, J. R., Malloy, Q. G., Travis, S. A., Bobmanuel, L. M., Lefer, B. L., and Flynn, J. H.: Atmospheric ammonia measurements in summer and implications for particulate matter formation in Houston, TX, submitted to *Atmos. Environ.*, 2012.
- Jung, J., Fountoukis, C., Adams, P. J., and Pandis, S. N.: Simulation of in situ ultrafine particle formation in the eastern United States using PMCAMx-UF, *J. Geophys. Res.*, 115, D03203, doi:10.1029/2009JD012313, 2010.
- Junkermann, W., Hagemann, R., and Vogel, B.: Nucleation in the Karlsruhe plume during the COPS/TRACKS – Lagrange experiment, *Q. J. Roy. Meteorol. Soc.*, 137, 267–274, 2011a.
- Junkermann, W., Vogel, B., and Sutton, M. A.: The climate penalty for clean fossil fuel combustion, *Atmos. Chem. Phys.*, 11, 12917–12924, doi:10.5194/acp-11-12917-2011, 2011b.
- Khairoutdinov, M. F. and Randall, D. A.: Cloud resolving modeling of the ARM Summer 1997 IOP: model formulation, results, uncertainties, and sensitivities, *J. Atmos. Sci.*, 60, 607–625, 2003.
- Kirkby, J., Curtius, J., Almeida, J., Dunne, E., Duplissy, J., Ehrhart, S., Franchin, A., Gagné, S., Ickes, L., Kürten, A., Kupc, A., Metzger, A., Riccobono, F., Rondo, L., Schobesberger, S., Tsagkogeorgas, G., Wimmer, D., Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Dommen, J., Downard, A., Ehn, M., Flagan, R. C., Haider, S., Hansel, A., Hauser, D., Jud, W., Junninen, H., Kreissl, F., Kvashin, A., Laaksonen, A., Lehtipalo, K., Lima, J., Lovejoy, E. R., Makhmutov, V., Mathot, S., Mikkilä, J., Minginette, P., Mogo, S., Nieminen, T., Onnela, A., Pereira, P., Petäjä, T., Schnitzhofer, R., Seinfeld, J. H., Sipilä, M., Stozhkov, Y., Stratmann, F., Tomé, A., Vanhanen, J., Viisanen, Y., Vrtala, A., Wagner, P. E., Walther, H., Weingartner, W., Wex, H., Winkler, P. M., Carslaw, K. S., Worsnop, D. R., Baltensperger, U., and Kulmala, M.: Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation, *Nature*, 476, 429–433, doi:10.1038/nature10343, 2011.
- Konwar, M., Maheskumar, R. S., Kulkarni, J. R., Freud, E., Goswami, B. N. and Rosenfeld, D.: Suppression of warm rain by aerosols in rain-shadow areas of India, *Atmos. Chem. Phys. Discuss.*, 10, 17009–17027, doi:10.5194/acpd-10-17009-2010, 2010.
- Kulmala, M. and Kerminen, V.: On the formation and growth of atmospheric nanoparticles, *Atmos. Res.*, 90, 132–150, doi:10.1016/j.atmosres.2008.01.005, 2008.
- Makkonen, R., Asmi, A., Korhonen, H., Kokkola, H., Järvenoja, S., Räisänen, P., Lehtinen, K. E. J., Laaksonen, A., Kerminen, V.-M., Järvinen, H., Lohmann, U., Bennartz, R., Feichter, J., and Kulmala, M.: Sensitivity of aerosol concentrations and cloud properties to nucleation and secondary organic distribution in ECHAM5-HAM global circulation model, *Atmos. Chem. Phys.*, 9, 1747–1766, doi:10.5194/acp-9-1747-2009, 2009.
- Merikanto, J., Napari, I., Vehkamäki, H., Anttila, T., and Kulmala, M.: New parameterization of sulfuric acid-ammonia-water ternary nucleation rates at tropospheric conditions, *J. Geophys. Res.*, 112, D15207, doi:10.1029/2006JD007977, 2006.
- Mesinger, F., Dimego, G., Kalnay, E., Mitchell, K., Shafran, P. C., Ebisuzaki, W., Joviæ, D., Woollen, J., Rogers, E., Berbery, E. H., Ek, M. B., Fan, Y., Grumbine, R., Higgins, W., Li, H., Lin, Y., Manikin, G., Parrish, D., and Shi, W.: North American Regional Reanalysis: A long-term, consistent, high-resolution climate dataset for the North American domain, as a major improvement upon the earlier global reanalysis datasets in both resolution and accuracy, *B. Am. Meteorol. Soc.*, 87, 343–360, 2006.
- Miller, B. G.: *Clean Coal Engineering Technology*, Elsevier Inc, Oxford, 375–481, 2010.
- Peng, R. D., Dominici, F., Pastor-Barriuso, R., Zeger, S. L., and Samet, J. M.: Seasonal Analyses of Air Pollution and Mortality in 100 US Cities, *Am. J. Epidemiol.*, 161, 585–594, 2004.
- Pierce, J. R. and Adams, P. J.: Efficiency of cloud condensation nuclei formation from ultrafine particles, *Atmos. Chem. Phys.*, 7, 1367–1379, doi:10.5194/acp-7-1367-2007, 2007.
- Pierce, J. R. and Adams, P. J.: Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates, *Atmos. Chem. Phys.*, 9, 1339–1356, doi:10.5194/acp-9-1339-2009, 2009a.
- Pierce, J. R. and Adams, P. J.: A computationally efficient aerosol nucleation/condensation method: pseudo-steady-state sulfuric acid, *Aerosol Sci. Technol.*, 43, 1–11, 2009b.
- Poppe, D., Wallasch, M., and Zimmermann, J.: The Dependence of the Concentration of OH on its Precursors under Moderately Polluted Conditions: A Model Study, *J. Atm. Chem.*, 16, 61–78, 1992.
- Seinfeld, J. H. and Pandis, S. N.: *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, 2, John Wiley & Sons, Inc., Hoboken, New Jersey, USA, 2006.
- Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L. (Eds.): *Climate change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, IPCC, Cambridge University Press, Cambridge, UK, and New York, NY, USA, 2007.
- Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Sihto, S.L., Riipinen, I., Merikanto, J., Mann, G. W., Chipperfield, P. M., Wiedensohler, A., Birmili, W., and Lihavainen, H.: Contribution of particle formation to global cloud condensation nuclei concentrations, *Geophys. Res. Lett.*, 35, L06808, doi:10.1029/2007GL033038, 2008.
- Srivastava, R. K., Miller, C. A., Erickson, C., and Jambhekar, R.: Emissions of sulfur trioxide from coal-fired power plants, *Air Waste Manage. Assoc.*, 54, 750–762, 2004.
- Srivastava, R. K. and Jozewicz, W.: Flue Gas Desulfurization: The State of the Art, *Journal of the Air & Waste Management Association*, 51, 1676–1688, 2001.
- Stevens, R. G., Pierce, J. R., Brock, C. A., Reed, M. K., Crawford, J. H., Holloway, J. S., Ryerson, T. B., Huey, L. G., and Nowak, J. B.: Nucleation and growth of sulfate aerosol in coal-fired power plant plumes: sensitivity to background aerosol and meteorology, *Atmos. Chem. Phys.*, 12, 189–206, doi:10.5194/acp-12-189-2012, 2012.
- Stieb, D. M., Judek, S., and Burnett, R. T.: Meta-analysis of time-series studies of air pollution and mortality: effects of gases and particles and their influence of cause of death, age and season, *J. Air Manage. Assoc.* 52, 470–484, 2002.

- Twomey, S.: Pollution and planetary albedo, *Atmos. Environ.*, 8, 1251–1256, 1974.
- Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, *Atmos. Chem. Phys.*, 9, 239–260, doi:10.5194/acp-9-239-2009, 2009.
- Wang, S. W., Zhang, Q., Streets, D. G., He, K. B., Martin, R. V., Lamsal, L. N., Chen, D., Lei, Y., and Lu, Z.: Growth in NO_x emissions from power plants in China: bottom-up estimates and satellite observations, *Atmos. Chem. Phys.*, 12, 4429–4447, doi:10.5194/acp-12-4429-2012, 2012.
- Zhao, Y., Wang, S., Duan, L., Lei, Y., Cao, P., and Hao, J.: Primary air pollutant emissions of coal-fired power plants in China: Current status and future prediction, *Atmos. Environ.*, 42, 8442–8452, 2008.
- Zhou, W., Cohan, D. S., Pinder, R. W., Neuman, J. A., Holloway, J. S., Peischl, J., Ryerson, T. B., Nowak, J. B., Flocke, F., and Zheng, W. G.: Observation and modeling of the evolution of Texas power plant plumes, *Atmos. Chem. Phys.*, 12, 455–468, doi:10.5194/acp-12-455-2012, 2012.