

Particle Growth in Urban and Industrial Plumes

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Particles are formed within the plumes from industrial facilities from condensation of the oxidation products of SO₂ and other compounds such as organics. A quantitative understanding of the relationship between emissions of precursor gases and downwind aerosol properties is needed for mitigation efforts.

During the Southern Oxidants Study 1999 and the Texas 2000 Air Quality Study, airborne measurements of particle size distributions were made in a variety of power plant, petrochemical, and urban plumes. Data from a 5-channel condensation particle counter, a laser optical particle counter, and a white light optical particle counter were combined to determine particle size distributions from 0.004-8 μm diameter with 1 second resolution. This study also uses measurements of photochemically reactive gases and tracer compounds to identify plumes, estimate dilution, and evaluate the mass budget of condensable compounds.

Measurements were made in the plumes downwind of large fossil-fueled power generation stations, petrochemical facilities (which were often located near power generation units and transportation centers), and the urban areas of Nashville, Atlanta, and Houston. Downwind of these sources, substantial enhancements in particle mass were found in only those plumes rich in SO₂. Even in the plumes downwind of petrochemical complexes with large tabulated emissions of NO_x and aromatic volatile organic compounds (VOCs), particle enhancements were not detected in the absence of SO₂, while nearby, modest sources of SO₂ produced detectable enhancements in particle volume.

Large point sources of SO₂ often produced particle number concentrations that were at a maximum a few km downwind of the sources. Particle number was sometimes highest at the edges of young plumes, in regions where numerical models of plume photochemistry predict high concentrations of OH. The sensitivity of particle nucleation to thermodynamic and chemical properties and the tightly coupled interactions between aerosol microphysics and chemistry and plume dynamics suggest that quantitative prediction of particle number concentration from point sources will be extremely difficult.

Particle volume (mass) fluxes measured in the oxidized plumes downwind of large SO₂ point sources were substantially greater than those produced by the urban areas of Nashville, Atlanta, and Houston. The increased particle volume associated with these plumes was often detectable >100 km downwind from the sources at plume ages >6 hours. Although substantial particle mass growth was found only in plumes rich in SO₂ throughout the SOS 1999 and TexAQS 2000 projects, the largest

mass fluxes found during the experiments were observed in the VOC-rich plume downwind of the petrochemical complexes located along the Houston ship channel just east of downtown Houston. Comparisons with a two-dimensional numerical model of plume chemistry and mixing indicate that the observed particle growth could not be accounted for by SO₂ oxidation. These results suggest that oxidation of VOCs and conversion of the gas-phase oxidation products to the particulate phase contributes to ~50% of the observed volume growth. In contrast, in SO₂-rich plumes from coal-fired power plants that were not emitted into VOC-rich environments, the numerical model calculated SO₂ oxidation rates that slightly low, but still consistent within experimental uncertainty, with the observed particle growth.

Our results indicate that known photochemistry can explain observed rates of particle growth within the plumes from large SO₂ point sources in dry, summertime conditions in the absence of large sources of VOCs. Co-condensation of nonsulfate compounds appears to be important in VOC-rich environments such as the plume of the petrochemical complexes in the Houston ship channel area. However, since substantial volume growth was never observed in the absence of SO₂, these observations also point to the oxidation of SO₂ as the primary process regulating particulate mass formation from these sources. We recommend studies of the aerosol-forming potential of mixtures of reactive VOCs (particularly aromatics), NO_x, and SO₂ to extend previous work on NO_x-VOC systems (*e.g.*, Odom *et al.*, 1997; Jacobson *et al.*, 2000).