

Aerosol ion concentration dependence on atmospheric conditions in Chicago

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Abstract

This study seeks to determine the influence of precursor trace gases and local meteorology, including lake breeze events, on the concentrations of secondary aerosol species in Chicago. For this, two particulate air samples per day were collected onto quartz fiber filters at the Loyola University Chicago Air Station (LUCAS) during the summer months of 2002 and 2003 and subsequently analyzed by ion chromatography for sulfate, nitrate and oxalate. In parallel, mixing ratios of ozone (O_3) and nitrogen oxides (NO and $NO_2 = NO_x$) were monitored and weather parameters were recorded. In addition, backward trajectories were obtained to estimate air mass transport to Chicago. Ozone and NO_x mixing ratios as well as sulfate, nitrate and oxalate concentrations varied substantially throughout the study, but three situations could be distinguished based on meteorology and chemistry. Case one had the lowest ozone, NO_x and ion levels due to wind directions constantly from Lake Michigan. Case two comprised of days showing the highest pollutant levels because of to predominantly southwestern air currents and warm temperatures and case three experienced an air stagnation situation in the morning leading to high NO_x mixing ratios and a subsequent lake breeze event. In the last case, elevated ozone mixing ratios and ion concentrations were observed after lake breeze onset indicating pollutant transport.

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1. Introduction

Chicago as a major metropolitan center in the Midwest presents a significant source region of emissions from traffic and industry and experiences regularly air pollution events during hot summer days. In addition, Chicago's location at the southwestern end of Lake Michigan exposes the city to lake breezes where the major air current originates

from Lake Michigan. It was found that these events might play a major role in the pollution pattern of the region (see for example: Bloomfield et al., 1996; Gatz, 1975; Lee et al., 1993; Lestari et al., 2003; Scheff et al., 1984). In order to investigate these pollution patterns in more detail, aerosol samples were collected during the summer months of 2002 and 2003 and analyzed for their ionic content. The major ions of interest comprised of sulfate, nitrate and oxalate, because these ions are considered secondary pollutants formed as a result of reactions by reactive trace gases such as sulfur dioxide (SO_2), nitrogen oxides (NO , NO_2) and volatile organic

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compounds (VOCs) with ozone (O₃) and hydroxyl radicals ([•]OH).

Studies have found that the formation of secondary pollutants does not only depend on emission sources, but is also influenced by local meteorology. For example, high ozone episodes have been observed in areas downwind of pollution sources with high temperatures and low wind speeds (Aneja et al., 1999; Bloomfield et al., 1996; Chow et al., 1998). Also, studies have linked elevated ozone levels to sea or lake breezes in regions close to large bodies of water (Cass and Shair, 1984; Gusten et al., 1988; Hastie et al., 1999; Lyons and Cole, 1976; Nair et al., 2002; Pont and Fontan, 2000). Both situations for formation of secondary aerosols have to be considered in Chicago with major emission sources primarily located in the south (S) and southwestern (SW) part of the city and lake breezes occurring at about 20% of summer days. A lake breeze is the result of a temperature gradient between land and lake surfaces. During hot summer days, the land surface heats faster than the water body creating a temperature and pressure gradient between these two. The difference in pressure leads to air mass transport from the high pressure area over the lake (cooler air) to the lower pressure area over the land (warmer air). During the night, the opposite so-called land-breeze can occur, which transports cooler polluted air from the city towards and above Lake Michigan. An air mass experiencing a lake or land breeze event is confined to a relatively small volume with little mixing and trace gases and particulates can become highly concentrated. A detailed description of the lake breeze climatology with respect to Chicago has been published by Lyons (1972).

The study presented here seeks to investigate the conditions leading to formation of the secondary aerosol species, sulfate, nitrate and oxalate, in conjunction with ozone and NO_x mixing ratios as well as local meteorology including the lake breeze in Chicago.

2. Methods

2.1. Sample collection

Atmospheric aerosol samples were collected at Loyola University Chicago's air station (LUCAS). LUCAS is situated 60m high atop a residence hall on Loyola University Chicago's lake shore campus located approximately 13 km north of downtown

Chicago and 200m west of Lake Michigan (Fig. 1). Sampling took place in 2002 (13 days) and 2003 (20 days) during the summer because the highest frequency of lake breeze events and the highest possible ozone mixing ratios and ion concentrations are expected at this time of the year. Particulates (PM_{2.5}) were collected at a flowrate of 2 m³ h⁻¹ onto quartz fiber filters (QMA, 47 mm diameter, Whatman International Ltd, Maidstone, UK) housed inside a Teflon filter holder. Samples were taken twice a day, from 07:00 h to 10:00 h (collection A), and 11:00 h to 13:00 h (collection B) local time (LT). Observations indicated that a lake breeze most likely establishes between 10:00 and 11:00 h; hence, the collection times selected allowed for the determination of particulate concentrations before and during the onset of a lake breeze. Immediately after collection, filters were frozen until analysis.

In addition to air particulate collection, continuous 24 h trace gas measurements of NO_x (NO and NO₂) and ozone were made using chemiluminescence for NO_x (Thermo Environmental Instruments, Inc. (TEI) Model 42C; US EPA designated method RFNA-1289-074, US EPA, 2005) and UV photometry (TEI Model 49C; US EPA designated method EQOA-0880-047, US EPA, 2005) for ozone, respectively. Both units were calibrated regularly with aid of the local Illinois Environmental Protection Agency's bureau of air. Weather conditions were also recorded during the sampling time from 07:00 to 13:00 h.

2.2. Sample analysis

Before analysis, filters were extracted in 5 ml of nanopure water for 15 min by ultrasonication. The obtained solution was analyzed subsequently for anions and low molecular weight organic acids by ion chromatography (IC) with chemical suppression and conductivity detection (Metrohm 761 Compact IC). The analysis procedure followed closely previously described ones with slight adaptations (Granby et al., 1997; Grosjean, 1988; Kadowski, 1986; Kato et al., 1999; Lin, 2002; Solomon et al., 1992; Tolocka et al., 2001). A Metrosep Supp5 column with 1.0mM NaHCO₃ and 3.5mM Na₂CO₃ as eluent was used to determine fluoride, acetate, formate, chloride, nitrite, bromide, nitrate, phosphate, benzoate, sulfate, succinate, and oxalate. Sulfate, nitrate and oxalate accounted for the most abundant and continuously detected anions in the aerosol samples.

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