

Atmospheric variability and emissions of halogenated trace gases near New York City

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ABSTRACT

Elevated mixing ratios of chlorofluorocarbons (CFC-11 and CFC-12), and sulfur hexafluoride (SF₆) have been observed at Lamont-Doherty Earth Observatory (LDEO), located approximately 25 km north of New York City (NYC). Emissions and transport of these gases are of interest because of their global warming potential, the role of CFCs in depletion of stratospheric ozone and information they provide on the transport of atmospheric pollutants. Comparison of trace gas time series with meteorological data indicates that both NYC and the region to the southwest (New Jersey and the Philadelphia–Washington DC area) are significant sources of CFCs, and confirms that NYC is an unusually large source of SF₆. From 1996 to 2005 the elevation of CFC-12 mixing ratio above that of the remote (well mixed) atmosphere has decreased on average by 5.2 ± 0.6 ppt y⁻¹, whereas that of CFC-11 has not changed significantly (0.0 ± 2.0 ppt y⁻¹). From 1998 to 2006, the elevation of SF₆ mixing ratios above that of the remote atmosphere declined by 0.4 ± 0.1 ppt y⁻¹. Time series of the same gases measured at Harvard Forest, 205 km northeast of LDEO, demonstrate transport of air masses with elevated levels of these gases from their source region to central Massachusetts. Emissions in the local area around LDEO were quantified through analysis of diurnal cycles. Local CFC-12 emissions decreased ca. 95% between 1996 and 2005 while CFC-11 emission decreased ca. 51% during the same period. Local SF₆ emissions decreased by 47% between 1998 and 2005.

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1. Introduction

The chlorofluorocarbons, CFC-11 and CFC-12, are man-made gases that have been used in large quantities as refrigerants, foam blowing agents and propellants. Production and use of CFCs were controlled under the Montreal Protocol (adopted in 1987) and subsequent amendments, due to their contributions to stratospheric ozone destruction. As a result, global emissions of CFC-11

are estimated to have decreased by 98% between 1987 and 2000 and CFC-12 emissions by 94% (AFEAS, 2008). Current CFC-12 emissions result largely from its use as a refrigerant gas, whereas CFC-11 emissions are mainly from releases from previously manufactured closed cell foam (AFEAS, 2008). Due to decreasing global emissions, remote atmospheric mixing ratios of all CFCs were decreasing by 2002 (NOAA/CMDL, 2004). Sulfur hexafluoride (SF₆), like CFCs, is a compound of predominantly anthropogenic origin. Its primary use is as an electrical insulator in high voltage switchgear. It has not been significantly regulated, although voluntary emission reductions have been reported (e.g. EPA, 2010), and its mixing ratio is rising in the remote atmosphere by 0.2 to 0.3 ppt per year (e.g. Harnisch and Hohne, 2002; Levin et al., 2010).

Atmospheric time series provide useful information on concentrations and emissions strength of gases of environmental concern, and can also provide useful data for calibration and

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validation of atmospheric models (e.g. Prather et al., 1987; Peters et al., 2004). Independent emission estimates based on time series analysis can serve as a useful check of emission estimates based on reported production and records of use on a variety of geographical scales. For CFCs, observation of atmospheric mixing ratios is the only direct way to determine emissions as new production approaches zero. For SF₆, emission estimates can be made from records of use and production for some geographic areas (e.g. Smythe, 2004; EIA, 2008) and estimates based on atmospheric observations can supplement and provide a cross check for these records (e.g. Levin et al., 2010). Many studies of SF₆ and CFCs have used time series collected at remote locations to investigate the budget of these gases in the global troposphere (e.g. Elkins et al., 1993; Cunnold et al., 1997; Geller et al., 1997; Maiss and Brenninkmeijer, 1998). More recently, time series from locations that are intermittently influenced by pollution from urban areas have been used to investigate regional emissions (e.g., Bakwin et al., 1997; Biraud et al., 2000; Barnes et al., 2003; Li et al., 2005) while others have used measurements from aircrafts to derive emission estimates for these and other gases (e.g. Hurst et al., 2006; Millet et al., 2009).

Atmospheric measurements within or near a metropolitan area provide insights on local emissions and transport, and such time series have been reported for CFCs and SF₆. Ho et al. (1998) and Ho and Schlosser (2000) describe elevated CFC-11, CFC-12 and SF₆ mixing ratios near New York City (NYC). Time series data have also been used to characterize elevated levels of CFCs and other halocarbons in and around urban areas including a Taiwanese industrial park (Chang et al., 2001), Krakow, Poland (Lasa and Sliwka, 2003) and Bristol, UK (Khan et al., 2009). Buchmann et al. (2003) observed elevated levels of CFCs and other halocarbons at a site outside Zurich, Switzerland and used them to estimate emissions. Flask samples have also been used to document elevated levels of CFCs and other halocarbons. For example, Barletta et al. (2006) reported observations from 45 cities in China. These studies show the promise of using atmospheric samples to understand and estimate emissions and atmospheric transport in urban areas.

Here, we present 8–9 years of CFC-11, CFC-12 and SF₆ mixing ratios from Lamont-Doherty Earth Observatory (LDEO) located ~

25 km north of NYC (Fig. 1). These time series are extensions of those presented by Ho et al. (1998) and Ho and Schlosser (2000), and we examine them for the time periods between June 1996 and June 2005 for CFCs and August 1998 to May 2006 for SF₆. CFC data from July 2005 on were compromised by a failure in the analytical system and are not included in this analysis. Monitoring of both CFCs and SF₆ ceased at LDEO in May 2006. Temporal variability in each time series is explored and interpreted in the context of emissions and transport of these gases within the NYC area and the larger northeastern US region.

2. Methods

Atmospheric CFC-11, CFC-12 and SF₆ mixing ratios were measured at LDEO at approximately 15-min intervals. Samples were pumped into the laboratory from an inlet located approximately 100 m from buildings at the site and 10 m above ground level, and measured by an automated gas chromatograph with two electron capture detectors (GC-ECD). For details on sample collection and analysis, see Ho et al. (1998) and Ho and Schlosser (2000). Analytical precision for CFC-11, CFC-12 and SF₆ were usually better than 1%. Trace gas data from Harvard Forest (HF) in Petersham, MA, approximately 205 km northeast of NYC, are also analyzed. Barnes (2000) and Barnes et al. (2003) provide details of sample collection and analysis for this site. CFC mixing ratios from both sites are reported in this work on the SIO 98 scale (Prinn et al., 2000) whereas SF₆ measurements for both sites are reported on the NOAA 2000 scale (NOAA/CMDL, 2004). Daily wind speed and direction data from Central Park within NYC are also used in the analysis of trace gas data (NOAA/NCDC). Wind data from this location was chosen because NYC is expected to be a significant source of these gases. Wind direction at Central Park and Black Rock Forest (~ 80 km north of NYC) are generally in good agreement (Ho et al., 1998) indicating that winds from Central Park describe transport in the lower Hudson region encompassing LDEO reasonably well.

Emissions were estimated from the diurnal cycles at LDEO using a method derived from Buchmann et al. (2003). Buchmann et al. (2003) observed the slope of increasing mixing ratios due to the

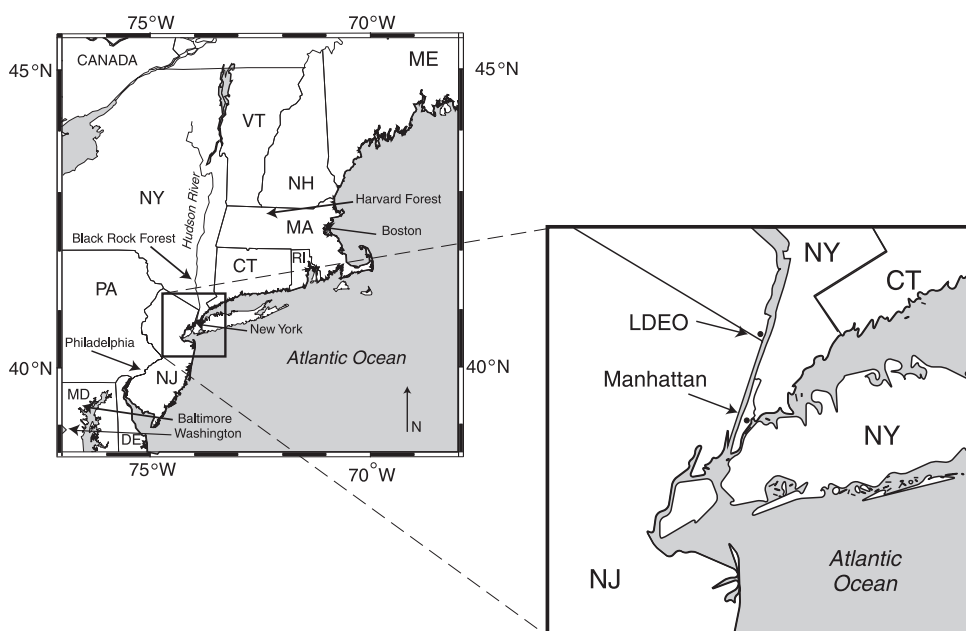


Fig. 1. Map of the northeastern United States, showing the locations of LDEO, Black Rock Forest, and HF.

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