



Tropospheric ozone production regions and the intercontinental origins of surface ozone over Europe

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HIGHLIGHTS

- Intercontinental origins of surface ozone in Europe have been identified.
- North America contributed 10–16 ppb across Europe and Asia 3–5 ppb.
- Intercontinental ozone transport influences seasonal cycles.

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ABSTRACT

Ozone tagged labelling schemes have been implemented in a global Lagrangian chemistry–transport model to identify the intercontinental origins of surface ozone in Europe. Stratosphere–troposphere exchange gave rise to between 3 and 5 ppb across Europe, whereas the mid-latitudes of the Middle East, Asia and the Pacific Ocean region contributed 6–8 ppb. Surface ozone levels of 10–16 ppb were associated with the mid-latitudes of North America and the North Atlantic Ocean regions. Appreciable intercontinental ozone production occurred downwind of continental regions and above the surface layer. Intercontinental ozone formation and transport from tropical regions contributed about 4 ppb and was much less efficient compared with that from mid-latitudes. There were approaching 60 chemical processes driving intercontinental ozone formation, of which the $\text{HO}_2 + \text{NO}$, $\text{CH}_3\text{O}_2 + \text{NO}$ and $\text{CH}_3\text{COO}_2 + \text{NO}$ reactions were the most important. Ozone production appeared to be driven by OH oxidation of secondary reaction products rather than the oxidation of primary emitted VOCs. The largest intercontinental ozone contributions amounted to about 20 ppb from North America to European baseline stations, 14 ppb from Asia to North American baseline stations and 10 ppb from Asia to European baseline stations. It is possible that changing intercontinental ozone production and transport could have led to seasonal ozone trends and shifts in seasonal cycles at northern hemisphere mid-latitude baseline ozone monitoring stations.

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1. Introduction

Tropospheric ozone (O_3) is of major policy concern because ozone is an important air pollutant that at elevated concentrations damages human health and vegetation (WHO, 1987). It is also an important radiatively active trace gas that contributes to climate change (IPCC, 1990). Observations of tropospheric ozone, whether at the surface (Oltmans et al., 2006) or from ozonesondes and

commercial aircraft (Logan et al., 2012) when taken together, present a picture of increasing tropospheric ozone levels during the second half of the 20th century (Parrish et al., 2012). Global chemistry transport models can explain qualitatively the increase in surface ozone levels in terms of increasing man-made emissions of ozone precursors, changes in meteorology and in natural emissions (Stevenson et al., 2013). However, quantitative explanations remain elusive (Pozzoli et al., 2011; Parrish et al., 2014). Furthermore, Parrish et al. (2014) suggest that a systematic investigation of modelled seasonal cycles may enlighten a search for the drivers of the ozone increase and this suggestion has provided the stimulus for this modelling study.

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We begin our investigation of seasonal cycles in this modelling study by implementing a tagging or labelling scheme to identify whether the ozone at the surface in Europe is of stratospheric or tropospheric origins and where tropospheric photochemical ozone production actually takes place. We characterise whether surface ozone across Europe was formed locally within Europe or whether it has been transported from other northern hemisphere continental source regions. Source attribution studies for ozone have typically employed tagging or labelling schemes where ozone molecules formed in specific regions are tagged or labelled and then tracked in the model using additional model variables (HTAP, 2010). All ozone molecules, whatever their labels, are given the same local, instantaneous removal rates. Wang et al. (1998) performed global scale ozone tagging and reported that transport from the stratosphere accounted for about 30% of midlatitude ozone in winter, 10% in summer and 5% in the tropics. Production of ozone in the upper, middle and continental lower troposphere made significant (10–50%) contributions to tropospheric ozone concentrations. Sudo and Akimoto (2007) used a tagged tracer approach to characterise ozone production from 14 polluted source regions in the atmospheric boundary layer and 8 regions in the free troposphere, in addition to the stratosphere. They found that transport from polluted continental source regions accounted for about 40% of the ozone in remote locations. In addition, there have been other studies that tagged stratospheric ozone and used sensitivity studies to infer the tropospheric origins of ozone (Auvray and Bey, 2005; Fiore et al., 2003). Auvray and Bey (2005) focussed on the annual ozone budget over Europe whilst Fiore et al. (2003) provided intercontinental source attributions for 8 surface locations in North America. The ozone labelling or tagging scheme employed in this study adopts the framework laid out by Wang et al. (1998) and extends it further because the Lagrangian framework of STOCHEM allows a much larger number of tracers to be carried in the CTM. Our aim is to understand the atmospheric chemical processes that drive the intercontinental transport of ozone into Europe from North America and Asia.

2. Methodology

2.1. The STOCHEM model

The chemistry-transport model CTM used in this study is the UK Meteorological Office 3-D tropospheric chemistry-transport model, known as STOCHEM. The model uses a Lagrangian approach in which the troposphere is divided into 50,000 air parcels which are advected every three hours by fine resolution ($0.83^\circ \times 1.25^\circ \times 19$ vertical levels) winds from the Meteorological Office numerical weather prediction model. A full description of the advection and dispersion processes used in STOCHEM is given by Collins et al. (1997). The chemical mechanism in the original STOCHEM model of Collins et al. (1997) named STOCHEM-OC was replaced with an entirely new mechanism (Common Representative Intermediates CRI v2-R5) based on the condensation and reduction of a much larger explicit Master Chemical Mechanism (Jenkin et al., 2008; Watson et al., 2008) and this new version is named STOCHEM-CRI. STOCHEM-CRI addresses 220 chemical species taking part in 609 chemical reactions, treating 22 emitted organic compounds. This increased chemical complexity has the advantage of dealing with first and second generation oxidation products which turn out to play an important role in the intercontinental formation and transport of ozone. Further details of STOCHEM-CRI are given elsewhere (Utembe et al., 2010, 2011; Cooke et al., 2010). The CRI mechanism is available to view and download at <http://mcm.leeds.ac.uk/CRI/>.

2.2. Emissions

The emissions data employed in STOCHEM model are either 2-D or 3-D in distribution. Anthropogenic, biomass burning, vegetation, soils, oceans and 'other' surface emissions, detailed in Table 1 are based on two-dimensional source maps on a resolution of 5° longitude by 5° latitude. Stratospheric sources of O_3 and HNO_3 are also two dimensional but only contribute to the top model layer. Details of the emissions data and their handling are given elsewhere (Collins et al., 1997; Utembe et al., 2010; Derwent et al., 2012).

2.3. STOCHEM output

STOCHEM output imposes grid square dimensions of $5^\circ \times 5^\circ$ which implies an average occupancy of approximately two air parcels per Eulerian grid box. Each simulation was conducted with meteorology from 1998 for a period of 24 months with the first 12 months allowing the model to spin up. In addition to the above standard gridded output fields, we have access to the Lagrangian mixing ratios carried by the individual air parcels. At the end of each 3-h advection time step, the trace gas mixing ratios held by all air parcels within a region, defined by a solid angle of 1° radius, around each location of interest were stored. Further details are given in Derwent et al. (2012).

Over the years, STOCHEM-OC and STOCHEM-CRI model results have been compared with observations in numerous studies (Collins et al., 1999; Johnson et al., 2002; Sanderson et al., 2003; Utembe et al., 2011; Derwent et al., 2012), including some multi-model comparisons (Stevenson et al., 2006; Young et al., 2013) and the comparisons have been generally favourable. To build confidence in the model's ability to account for the origins of the ozone seen at the surface in Europe, we present a comparison of STOCHEM-CRI ozone predictions with observations from the European Monitoring and Evaluation Programme (EMEP) rural ozone monitoring network (Hjellbrekke, 2000). This comparison is available in the Supplementary Information.

2.4. The tagged ozone scheme

The ozone labelling or tagging scheme employed in this study adopts the framework laid out by Wang et al. (1998) and further details are given in the Supplementary Information attached to this paper.

We begin with the chemical tendency of ozone in the basic STOCHEM model which we can write as:

$$dO_3/dt = P - L[O_3], \quad (1)$$

where P is the production rate of ozone from chemistry and stratosphere–troposphere exchange and L is the instantaneous ozone loss rate coefficient due to chemistry and surface uptake. Then following Sudo and Akimoto (2007), we transport separate O_3 tracers tagged by their region of formation i as:

$$dO_3^i/dt = P^i - L[O_3^i]$$

where

$$P^i = k_{HO_2NO} \cdot HO_2 \cdot NO + \sum k_j \cdot RO_2 \cdot NO: \text{ inside the region } i$$

and

$$P^i = 0: \text{ outside the region } i.$$

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