ATMOSPHERIC CHEMISTRY

Ozone pollution from near and far

Tropospheric ozone is generated from precursor pollutants, but can be blown far afield. Satellite observations show rising ozone levels over China — and almost stable levels over western North America despite stricter regulations.

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Understanding changes in tropospheric ozone levels is imperative for policymakers concerned with surface air quality and near-term climate change. Ozone in the lower atmosphere, the troposphere, acts as a greenhouse gas. Near the surface, ozone is also an air pollutant associated with respiratory health conditions and premature mortality. Changes in regional ozone levels are mainly influenced by two processes: changes in local emissions of ozone precursor compounds, and changes in baseline ozone levels entering the region with the winds. Over the past few decades, anthropogenic emissions of ozone precursors have fallen in North America and Europe as a result of air-quality legislation, while they have risen in East Asia[1]. Writing in *Nature Geoscience*, Verstraeten and colleagues[2] report observations of an increase in free tropospheric column ozone over China between 2005 and 2010, but no significant change in free tropospheric column ozone over the western United States: they attribute this lack of change over the western United States to the combined influence of rising emissions in China and increasing downward transport of ozone from the stratosphere.

Tropospheric ozone is a secondary pollutant, formed by atmospheric chemical reactions involving nitrogen oxides (NO_x), carbon monoxide and volatile organic compounds. Most of these species have multiple anthropogenic and natural emission sources. Once formed, ozone has a lifetime of several weeks in the troposphere. In that time, it can be transported by the prevailing airstreams between mid-latitude continents, for example from East Asia to western North America. Ozone is therefore a hemispheric pollutant, and reductions in local or national emissions do not always produce an analogous decrease in atmospheric concentrations. In particular, although emissions of most ozone precursors have levelled or decreased in the past few decades over western North America, free tropospheric ozone levels have increased in springtime[3,4]. Rising East Asian emissions have been implicated in both regional tropospheric ozone increases over Asia[5] and in the increase in springtime tropospheric ozone levels over western North America[4,6]. They have also been associated with exceedances of ozone air-quality standards there[7].

In winter and spring, the transport of East Asian pollution typically occurs via ascent in airstreams within mid-latitude storms, and subsequent descent in dry airstreams embedded within the cyclone over western North America[7,8] (Fig. 1). In summer, however, mid-latitude cyclones are less frequent, and the transport of ozone and its precursors generally has a lesser impact on downwind air quality[8].

In addition to these sources from human activities, downward transport of naturally generated ozone from the stratosphere is an important source of tropospheric ozone in mid-latitude regions. This transport occurs primarily in the descending dry airstreams of mid-latitude cyclones, and therefore also has its maximum impact on free tropospheric ozone in springtime. This component of the ozone budget has contributed significantly to Northern Hemisphere interannual variability in tropospheric ozone levels[9] as well as to exceedances of ozone standards in the western United States[10,11] in spring.

Verstraeten and co-authors[2] add to this body of work by presenting a unique study of annual changes in tropospheric column ozone between 2005 and 2010 over China and the western United States, using a combination of satellite data and a chemical transport model. The satellite data are based on observations from the Tropospheric Emission Spectrometer on NASA's Aura satellite. Over China, they report an increase of 1.08% (or 0.23 Dobson units) per year in partial (or free) tropospheric column ozone between altitudes of 3 and 9 kilometres. Over the same period, they derive increases of 21% in emissions of the ozone precursor

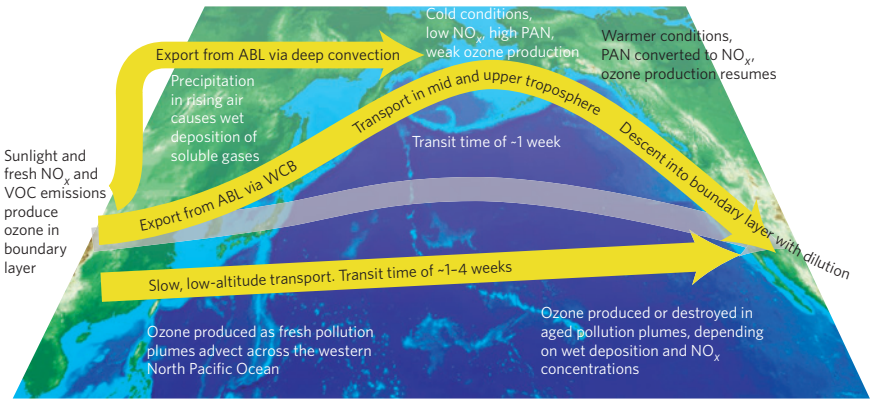

Figure 1 | Intercontinental transport processes. In winter and spring, the transport of pollution from East Asia to western North America typically occurs via mid-latitude cyclones that loft surface ozone and its precursors to the middle and upper troposphere, followed by descent in dry airstreams embedded within the cyclone over western North America. In summer, deep convection is typically the dominant mechanism for lofting pollutants from the surface over East Asia, but weaker descent and enhanced photochemical destruction mean that East Asian pollution has less impact in surface air over western North America. Slower, low-altitude transport also occurs in all seasons. Verstraeten and colleagues[2] show that over the western United States, reductions of free tropospheric ozone as a result of tighter regulations have been compensated by a combination of ozone transported from China and downward transport from the stratosphere. VOC, volatile organic compounds; ABL, atmospheric boundary layer; PAN, peroxyacetile nitrate; WCB, warm conveyor belt. Figure reproduced with permission from ref. 13, United Nations.

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species NO_x in China. In contrast, over the western United States, they find no significant trend in free tropospheric ozone concentrations between 2005 and 2010, despite a 21% decline in NO_x emissions.

Their simulations with a chemistry transport model isolate the impacts of natural downward transport from the stratosphere from changes in anthropogenic NO_x emission on free tropospheric ozone changes in each of the two regions. They show that changes in downward transport of naturally generated stratospheric ozone account for an increase in tropospheric ozone of about 0.5% per year over both regions. Interestingly, over China contributions from both sources to tropospheric ozone trends are roughly equal.

For the western United States, the simulations by Verstraeten *et al.* suggest that the increases in stratosphere-to-troposphere ozone transport and the change (both regional and remote) in ozone-precursor NO_x emissions together result in an insignificant trend in free tropospheric ozone, broadly consistent with earlier work[4,6,7,10,11]. But the changes in NO_x emission over China alone would have led to a rise in tropospheric ozone levels over the western United States. Hence they suggest that changes in NO_x emissions over China have partly offset the impact of domestic emissions reductions in the western United States.

It is important to note that the deseasonalized trends reported by Verstraeten *et al.*, although considerable in relative amounts, are small in absolute terms. It would be interesting to see how deseasonalized trends derived from other satellite observations compare to those from Tropospheric Emission Spectrometer. In addition, the time series so far is short. Longer-term observations could confirm whether the change observed in this 6-year record is a robust trend or whether the observed changes are largely accounted for by interannual variability. Finally, China itself lies downwind from India and other parts of Asia, and it remains to be established how the free tropospheric ozone trend over China is in turn influenced by emissions upwind.

These uncertainties aside, the findings of Verstraeten *et al.*[2] present a unique quantitative assessment of causes of free tropospheric ozone trends over China. The findings add to the evidence[12] that suggests that effective control of ozone air quality in any downwind continent requires national controls to be augmented with international agreements. Currently, there are no international agreements that address the intercontinental transport of tropospheric ozone and its precursors. ❐

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