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Tracking potential sources of peak ozone concentrations in the upper troposphere over the Arabian Gulf region



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HIGHLIGHTS

- For the first time temporally highly resolved O₃ sondes in the Middle East region.
- Upper tropospheric ozone in the Arabian Gulf region is on average 80 ± 13 ppbv.
- O₃ in the 6–12 km range is higher when air masses came from the Mediterranean.
- High pressure may cause subsidence of O₃ from upper troposphere/lower stratosphere.
- Convective activity and associated lightning can substantially increase O₃.

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ABSTRACT

In August 2013, the Qatar Environment and Energy Research Institute (QEERI), was the first to launch temporally highly resolved ozonesondes in the Middle East region. The data from 20 launches consistently show changes in meteorological parameters at about 5.5 km above the surface, which are more pronounced following a change in synoptic conditions on 15 August 2013, including temperature inversions, corresponding change in potential temperatures, relative humidity, and significant wind shear. These changes are typically associated with a large scale subtropical subsidence layer in accordance with previous aircraft studies in this region. Below the inversion layer, the ozone follows typical patterns for lower tropospheric measurements, starting in the surface layer up to 0.5 km above the ground level around noon at about 66 ± 15 ppbv. However, above the subsidence inversion, ozone mixing ratios begin to increase to 80 ± 13 ppbv between 6 and 12 km with maximum values ~100 ppbv around 8 km, then decreasing again before reaching the stratosphere.

Three-day HYSPLIT back trajectories indicate that ozone levels are typically about 17% lower in the 6 -12 km range under wind flow conditions from the East than in cases when trajectories came from the Mediterranean. High pressure may lead to subsidence of ozone from the upper troposphere/lower stratosphere and eventually cause an increase of ozone mixing ratios by ~18% above average between 6 and 7 km, i.e. slightly above subtropical subsidence layer. Under the impact of regional convective activity and associated lightning, ozone mixing ratios can increase by more than 35% averaged over the 9 -12 km altitude range. In both cases maximum ozone in the mid to upper troposphere reached more than 100 ppbv.

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1. Introduction

Ozone (O₃) in the troposphere is of interest because of its negative effects on human health and global warming (US

Environmental Protection Agency, 2012). Tropospheric ozone is primarily formed photo-chemically through the break-down of nitrogen oxides (NO_x) by sunlight in the presence of volatile organic compounds (VOCs) (e.g. Lelieveld et al., 2009). Contrary to the tropospheric ozone, ozone in the stratosphere is formed through photolysis of oxygen and forms a barrier protecting the Earth from the sun's ultra-violet radiation; however, it is sometimes mixed into the troposphere through Stratospheric–Tropospheric Exchange (STE) (e.g. Lelieveld et al., 2009).

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There are very few studies of ozone in the Arabian Gulf region (Li et al., 2001; Lelieveld et al., 2009; Zanis et al., 2014). While they all concur that there is unusually high ozone in the mid to upper troposphere in the Middle East region, they do not agree on the reasons for it.

Li et al. (2001) focus on a study in July 1997. They conducted sensitivity simulations to determine contributions to ozone at various levels within the troposphere, as well as distinguish between the influence of anthropogenic and lightning sources. Li et al. claim that there are unusually high summertime levels of ozone, over 80 ppbv in the mid to upper troposphere in the Middle East region. The reason for this is the anti-cyclonic circulation over the Arabian Peninsula and the Indian sub-continent "funneling" in pollution from Europe and nitrogen oxides (NO_x) generated by lightning outflow from the Indian Monsoon. Pollution from eastern Asia transported in the Easterly Tropical Jet Stream is said to contribute as well. Results indicate that the largest source of ozone (35–50%) is due to production in the upper troposphere and largescale subsidence in the region. Losses in these areas due to midlevel outflow are only little according to the Li et al. study. They estimate that 20–30% of the tropospheric ozone column has been caused by anthropogenic sources and 10-15% is from lightning. According to the paper, stratospheric ozone is not thought to be a major contributor, although it seems to have a greater impact than in other regions of the world.

Li et al.'s study used the GEOS-CHEM (Goddard Earth Observing System) global 3-D model of atmospheric chemistry in conjunction with vertical ozone profiles collected by the MOZAIC (Measurement of Ozone and Water Vapour on Airbus in-service Aircraft; MOZAIC, 2014) program over the regions of Tel-Aviv, Dubai, and Tehran. Data from the NASA (National Aeronautics and Space Administration) Data Assimilation Office for the years 1993—1997 were used as input for the model along with an anthropogenic base emissions inventory from 1985, adjusted for the study period, which included NO_x, Non-Methane Hydrocarbons (NMHC), and Carbon Monoxide (CO). Lightning-NO production associated with deep convection was parameterized following Price and Rind (1992).

Lelieveld et al. (2009) postulate that the Middle East is an ozone "hotspot" due to long-range transport of pollutants as indicated by the tracer CO, unusually strong STE, substantial natural upwind sources of NO_x such as lightning, the lack of precipitation, and contribution from local emissions such as those from oil and gas refineries combining to create ideal conditions for ozone production and entrainment. They find that there is a distinct ozone maximum between the surface and 750 hPa, which is even more pronounced in the summertime when conditions favor photochemistry. The average ozone mixing ratio in the mid to upper troposphere in the summer is around 80 ppbv. A comparison with other subtropical areas showed that the diel variation of midtropospheric ozone in Bahrain was related more to the longrange transport of pollutants than to local production, and that after removing anthropogenic sources from the model, the region still had higher ozone than the other areas. Unlike Li et al. (2001), this study suggests that stratospheric ozone does have a major impact on the regional tropospheric ozone column, making up about two thirds of it in winter and one quarter in the summer.

The Lelieveld study uses the EMAC (European Center—Hamburg 5th generation model MESSy [Modular Earth Submodel System] Applied to Atmospheric Chemistry) model in conjunction with satellite-retrieved ozone imagery from the Tropospheric Emission Spectrometer (TES, 2014) and Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY, 2014) to identify where the highest ozone concentrations are. The model is nudged toward the meteorological conditions in 2006 based on re-

analysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) and uses EDGAR 3.2 (Emissions Database for Global Atmospheric Research 2000) for inputs of anthropogenic emissions. The model includes a stratospheric ozone tracer as well and follows its transport and destruction in the troposphere. However, it does not include recycling processes of that stratospheric ozone tracer in the troposphere.

The most recent study by Zanis et al. (2014) indicates that the enhanced tropospheric summer ozone levels in the Eastern Mediterranean and Middle East are the result of stratospheric ozone being transported to the troposphere through subsidence induced by high pressure systems in the region. While local photochemical formation is still the dominant contributor to ozone concentration, stratosphere to troposphere transport (STT), a type of STE, plays a critical role in places with favorable conditions.

The Zanis et al. (2014) research was conducted in a similar way to the previous Lelieveld et al. (2009) study, using the EMAC model with ECMWF inputs and a stratospheric ozone tracer, and comparing the results to TES satellite data, but this time for a 12 year climatological study from 1998 to 2009. Both the model and satellite show pools of higher ozone concentration in the upper and middle troposphere over the Eastern Mediterranean, with the stratospheric ozone tracer indicating a 40–45% contribution from the stratosphere to the middle troposphere. This is attributed to the large-scale subsidence and limited outflow resulting from the anticyclonic motion of the high pressure systems prevalent in the area during the summer months.

Reid et al. (2008) report about dust profiles obtained through aircraft measurements, mostly over the area of the United Arab Emirates and vicinity, and also give a detailed overview of meteorological patterns in the region, including the mention of a subtropical subsidence inversion. While the focus of the paper was on aerosols and dust in the region, their flights encountered polluted air masses from Europe, India, and possibly Africa as well.

All of these publications agree that the Middle East area is of specific interest due to some unique meteorological conditions and presumably elevated ozone levels which may have a large-scale impact and contribute to global warming as the ozone is exported from the region. Due to the scarcity of data sets in that area, they all call for more in situ measurements.

In this paper we present data analysis from ozonesondes launched from Doha, Qatar, in August 2013 and explore potential source areas for ozone in the upper troposphere.

2. Methods

2.1. Data collection

Electro Chemical Concentration (ECC) ozonesondes (Droplet Measurement Technologies, 2013) in conjunction with iMet-1 radiosondes (InterMet Systems, 2008) attached to a 1.2 kg Kaymont brand weather balloon were used. Sondes were equipped with GPS (Global Positioning System). The iMet-1 radiosonde system (InterMet Systems, 2013) provides data for pressure (accuracy: 0.5 hPa), temperature (accuracy: 0.2 °C), relative humidity RH (accuracy: 5% RH), wind speed (accuracy: 0.1 m s^{-1}), wind direction (accuracy: $\leq 5^{\circ}$ for wind speeds $< 14 \text{ m s}^{-1}$, and $\leq 2^{\circ}$ for wind speeds >14 m s⁻¹), altitude (accuracy: 15 m), and position (accuracy: 5 m). The principle of ECC sondes are described thoroughly in Russell III et al. (1998). ECC sondes were used because they routinely outperform other types of ozonesondes such as the Brewer-Mast and KC96 Carbon Iodine Cell, as demonstrated in the 1996–2000 Jülich OzoneSonde Intercomparison Experiment (JOSIE) (Smit and Kley, 1996; Smit et al., 2007) and are currently the most commonly used world-wide (Global Atmospheric Watch, 2013).

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