Tropospheric ozone over the Middle East, long-term changes, chemistry dependency of trends, and feasibility study for proposed future synergetic satellite observations

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博士論文

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（中東における対流圏オゾンの長期変動と
それに含まれる化学、および対流圏オゾン
衛星観測のフィージビリティ検討）

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ABSTRACT

Regional ozone (O3) studies in the Middle East are essential for better understanding the broader puzzle of global climate change and air quality. In this study, we approach this aspect from two different perspectives, one that implements a multi-year multi-layer approach to characterize the vertical distributions, seasonal variations, and long-term changes of tropospheric ozone over the northern Middle East (25°-65° E, 30°-40° N), and the other one that studies the feasibility of a synergetic retrieval method to obtain vertically resolved tropospheric ozone profiles. The former would directly contribute to our understanding of regional ozone distributions over the Middle East, whereas the latter is an indirect approach toward finding a possible future solution to provide vertically resolved ozone profiles over the Middle East, which is a region that lacks tropospheric ozone monitoring sites and data record.

Concerning the first purpose, few available studies over the Middle East focused mostly on one tropospheric layer and did not discuss the variations in three tropospheric layers: lower, middle and upper troposphere (LT, MT, and UT). Hence, climatology of vertical and horizontal distributions of tropospheric ozone over the Middle East has been missing. In addition, these studies often focused on the region as a whole that led to paucity in regional information. Moreover, despite the availability of many studies on ozone trends over Europe, the United Stated, Canada, Japan and other regions, to our knowledge the long-term variations of tropospheric ozone over the Middle East remained unknown.

In this study, vertical profiles of ozone from: a) two ozonesonde stations, near the city of Ankara in Turkey (ANKARA: 32.88° E, 39.95° N) and Isfahan in Iran (ISFAHAN: 51.70° E, 32.51° N) available respectively over the period 1994–2012 and 1995–2011, and b) global Tropospheric Chemistry Reanalysis (TCR-1) over the period 2005–2012 were investigated for climatology, seasonal variations, and long-term changes in the three tropospheric layers. The LT and MT respectively denote 750 hPa and 460 hPa. In order to minimize the effect of stratospheric intrusion, UT level was defined above ANKARA (348 hPa) and ISFAHAN (280 hPa) based on the relationship between thermal and ozone tropopause. Statistical methods were implemented to obtain robust results. To our knowledge, this was the first time to study two nearby regions in the Middle East focusing on the regional characteristics of ozone distribution in multiple tropospheric layers.

First, the vertical profile of ozone was characterized by a strong vertical gradient near the surface in all seasons, and near the ozone tropopause at about 300 hPa that was strong in winter and spring but was non-existent in summer. These variations in vertical gradient of ozone are possibly attributed to the effect of deposition and titration by nitrogen oxide near the surface and by the stratosphere-troposphere exchange near the tropopause. The day-to-day variability from ozonesonde measurements and TCR-1 peaked below 700 hPa and near the tropopause, while it was minimized at about 615-500 hPa. Second, the seasonal variation of monthly mean ozone was characterized by a minimum in November in the UT and in December-January in the MT and LT over ANKARA and ISFAHAN, a broad spring maximum in the UT over ISFAHAN, and a relatively sharper summertime maximum in the MT and LT over ISFAHAN as well as in all the three layers over ANKARA. The ozone maxima from TCR-1 were shown one month before ozonsonde except for the MT in ANKARA and LT in ISFAHAN with an overlap. However, this lag was compensated once ozonesonde data was constrained to the same time span as TCR-1 (except in LT above ANKARA). This could implicate a shift in the month of the ozone maximum over the period of observation, irregularity in ozonesonde monthly means distribution, or

influenced by the existing model errors in TCR-1. Using TCR-1 data (2005–2012), over both ANKARA and ISFAHAN the maximum and minimum ozone anomaly from the annual mean emerged in the MT respectively in summer 2010 and winter 2005, whereas the maximum anomaly in the UT was seen in spring 2012. The seasonal ozone mean indicated greater value in the MT and UT in ANKARA (summer: 90–94 ppb) than in ISFAHAN (MT summer: 88 and UT spring: 84 ppb), whereas in the LT ozone seasonal mean had a greater value in ISFAHAN. These seasonal averages over zonal bands encompassing ANKARA and ISFAHAN demonstrated higher values compared to the seasonal averages over the whole Middle East (LT summer: 62, MT summer: 79, and UT spring: 74 ppb). This could highlight the importance of regional differences. In addition, monthly mean anomalies of NO_2 and CO were presented using TCR-1 data. NO_2 anomaly peaked from May to August over ANKARA and from June to September over ISFAHAN. CO anomaly was at a maximum in winter to spring and a minimum in summer to autumn over both locations.

Third, a frequency analysis of the backward trajectories exhibited that the air masses reaching ANKARA and ISFAHAN were dominantly originated from Europe, North Africa and North America. However, a different pattern during summer showed air masses originated from Asia in the UT and MT. An eight-year climatology (2005–2012) of the horizontal wind, geopotential height and vertical motion was presented. It illustrated that in summer ANKARA is located in the eastern flanks of the South Asian High, where the dominant signature is the strong descent of air, while ISFAHAN is situated in the center of the Arabian anticyclone near the Zagros mountains in Iran, where the influential signature is mountain venting. Moreover, a correlation study was conducted between ozone and its summertime anomaly and four meteorological parameters, that are, geopotential height, westerly wind, vertical motion, and relative humidity. A relationship between increasing ozone, ascending air and increasing relative humidity was found in ISFAHAN, in contrast to ANKARA. Over ISFAHAN, maximum ozone anomaly in summer in the MT was correlated with stronger easterlies and in the UT with ascent of the air. Over ANKARA, positive ozone anomaly was correlated with negative anomaly of the geopotential height and vice versa. A sensitivity analysis for 2006 illustrated the differences in regional precursor emission contribution to the ozone buildup over ANKARA and ISFAHAN. The large contribution of European emission to the LT ozone production was highlighted particularly over ANKARA but also over ISFAHAN. To our knowledge, this was not addressed by the previous studies in the Middle East. The summertime ozone maximum coincided with increased and decreased NO_2 and CO levels, respectively. O_3–CO correlation was negative in the LT and MT and positive in the UT. O_3–NO_2 correlation was positive at the three altitude levels.

Furthermore, long-term trends were investigated after smoothing the time series of tropospheric ozone from ozonesonde measurements during 1994–2012 over ANKARA and 1999–2011 over ISFAHAN to attenuate the effect of seasonality. Above ANKARA, ozone trends were significant and positive in the LT (0.77%/yr), MT (0.59%/yr), and UT (0.56%/yr) over the period of the observation. Seasonal trends above ANKARA were positive and significant. Nonetheless, there was a turning point in 2004 after which the trends became significantly negative. For each season, ozone trend in the LT was found to be the highest in value compared to the MT and UT trends. The highest MT and UT trends were observed in winter and in summer, respectively. Above ISFAHAN, the only significant trend was the positive trend in the UT (0.86%/yr) that was possibly influenced by strong stratospheric intrusion over some years. However, the LT trend became significant once split into 1999–2006 positive and 2006–2011 negative trends. The significant positive LT trend in summer over

ISFAHAN represented an increase of 1.5%/yr since 1999. Moreover, using multiple species data from the TCR-1, a parsimonious Multiple Linear Regression (MLR) model was created at LT, MT, and UT to describe the variations in tropospheric ozone with respect to the regional variables (precursor emissions and transported ozone). We indicated that in the MT over ANKARA, where no individual correlation >20% was found between ozone and each variable, the combination of the regional variables resulted in a 62% correlation or predictability. However, at ISFAHAN, where the individual correlations were of a greater magnitude, the combination of predictors in MLR resulted in a greater predictability of about 80–90%.

This study could connect the regional characterization of tropospheric ozone over the vicinity of the two cities Ankara and Isfahan in the Middle East to current literature to help create a broader picture of tropospheric ozone vertical and horizontal distribution and variation over this region. This could consequently contribute to understanding ozone pollution (in LT), transport (in MT), and radiative forcing (in UT) over the Middle East.

Concerning the second purpose, to the best of our knowledge, no study attempted to show how measurements in microwave (MW) range in a combination with ultra violet (UV) and thermal infrared (TIR) could improve the retrieval of tropospheric ozone. Therefore, error estimation was performed as part of the feasibility study on a synergetic retrieval method that implemented combination of the three separate wavelength ranges with the aim of obtaining a vertically resolved ozone profile in the troposphere. Observation geometries of nadirs for the UV and TIR measurements and limb for the MW measurement were considered from low orbit at a height of 300 km from the International Space Station. Twenty atmospheric profiles at Central East China (CEC) and Eastern China Sea (ECS) in June and December 2009 were used for the simulation. Sensitivities of ozone retrieval were presented in the three vertical regions (UT (215–383 hPa), MT (383–749 hPa) and LMT (>749 hPa)) in terms of Degree of Freedom for Signal (DFS), Partial Column Error (PCE), and averaging kernel (A) according to the optimal estimation method. The additional MW measurement was most effective at improving the sensitivity in the UT when combining several wavelength ranges. Adding the MW measurement to the UV+TIR measurements increased the DFS value in the UT by a factor of two. The average DFS value of the TIR measurement for all of the 20 profiles increased to more than unity by adding either the UV or MW measurements. The UV and TIR measurements were dominant in the ozone retrieval in the LMT. The value of DFS in the LMT strongly depended on ozone concentration levels and the peak of A. Adding MW measurements to the combination of UV and TIR improved the sensitivity not only in the UT but also in the MT and LMT. The DFS values increased by 95%, 23%, and 40% respectively in the UT, MT, and LMT by adding the MW measurements to the UV+TIR combination. One of the important applications of this method if implemented in the future satellites could be providing more accurate tropospheric ozone observations over the regions with insufficient in-situ measurement records such as the Middle East.