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Trans-boundary transport of ozone from the Eastern Mediterranean Coast

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ABSTRACT

As part of an ongoing USAID MERC research program investigating the transport of pollutants in the Eastern Mediterranean region, a joint coordinated campaign involving scientists from Jordan and Israel took place for a three-week period in June 2009. The study was aimed at examining previous modeling results that indicated the presence of elevated ozone levels occurring in Northern Jordan, due to emissions originating in coastal Northern Israel that were transported distances of more than 100 km. In the present investigation, ozone and other pollutants were monitored at five sites in Israel and two in Jordan. The sites were located along the prevailing wind direction that tends to move air-masses eastward from the Mediterranean coast, over the Israel Valley toward the Jordan Valley, and then into Northern Jordan. Analysis of the measurements showed that air masses arriving in Jordan passed over the Israeli metropolitan coastal region, entrapping large amounts of pollutant precursors before transporting them towards Jordan under the predominately westerly wind flows causing peak maximum ozone levels of around 100 ppbv. The present findings confirmed the previous modeling results that indicated higher NO_x background levels for the inland stations, peak O₃ values at later hours depending on wind speed and distance from the Mediterranean coast and maximum O₃ levels over northern Jordan.

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1. Introduction

Ozone has been well documented not only as an urban and local pollutant but also as a regional problem (Logan and Graedel, 1996: Fishman et al., 2003; West et al., 2009). Occasionally, ozone mixing ratios measured in rural areas are even higher than those measured in large urban centers. Elevated and potentially harmful levels of ozone have been observed in several rural areas of North America (Logan, 1989), Western Europe (Hov, 1984; Lelieveld and Dentener, 2000) and over the Atlantic Ocean (Lelieveld et al., 2004). Furthermore, elevated ozone mixing ratios have been observed on several occasions at a rural site some 50 km downwind of the highly industrialized and populated coastal plain region of Israel (Peleg et al., 1994). Studies performed at a coastal site in Israel attempted to measure the pollution content in fresh incoming air masses arriving from the west (Alper-Siman Tov et al., 1997; Levy et al., 2008). Usually ozone levels ranged between 30 and 60 ppbv, however on a number of occasions, Alper et al., (1997) measured exceptionally high levels during midday which rose to a peak value

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above 200 ppbv. The reason for these unusual high episodes was due to easterly winds which swept the air parcels containing the locally emitted pollution westwards over the sea during the night and early morning. On the following day, these air masses recirculated towards the coast under the westerly wind flows, reaching in the late afternoon the Israeli coastline with levels above 200 ppbv. Since in Israel most of the industrial and urban activity occurs in the coastal region, it is expected similarly to the above recirculation episodes, that high ozone mixing ratios should occur tens of kilometers downwind of emission sources as the pollutant envelope travels inland under westerly wind flows.

To test the above hypothesis, a regional scale study was performed in Israel and Jordan in order to evaluate trans-boundary transport of ozone and related pollutants. The overall objective of the study was to evaluate the effects of transport of air pollution between Israel and Jordan. In addition the validity of previous modeling results obtained using CAMx and RAMS (Weinroth et al., 2008) was examined. To achieve these objectives, a cooperative field campaign was performed over Israel, stretching from the western coast till the eastern border, and Northern Jordan. An additional goal was an attempt to develop cooperative efforts which will allow the quantification of trans-boundary pollution transport and eventually lead to improvement in the air quality of the region.

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2. Experimental details and methods

In order to measure transport of pollutants over Israel towards Jordan, seven ambient air quality monitoring stations, situated on a west-east configuration in Israel and Jordan, were employed to continuously measure pollutant and meteorological data. This enabled the detection, identification and quantification of the pollutants as they were transported from the coastal region eastwards over Israel into Northern Jordan.

2.1. Measurement field campaigns

An intensive three-week field measurement campaign was performed during June, 2009, with one research grade monitoring station set up in Israel and two in Jordan. The Israeli monitoring station was located at Maoz-Haim (MH), a rural site, isolated from local emission sources and adjacent to the Israeli-Jordanian border. The Jordanian stations were located at Teibe and Irbid, approximately 20 and 45 km, respectively, east of MH (Fig. 1). The site in Irbid was rural and was located southeast of the town of Irbid in the Jordanian University of Science and Technology campus.

Additional air quality data was obtained from four Israeli urban sites operated by the Israeli Ministry of Environment and the Haifa District Environment Municipal Association. Two of these monitoring stations were in Haifa, one adjacent to the sea coast located in a downtown area (designated Shuk) and the second station, 5 km south east of the first site, in the Neve-Shanan (designated NS). The third station was 13 km south east of NS, in the town of Kiryat Tivon (TI), and the fourth in Afula, 20 km south east of TI.

2.2. Instruments and equipment

Measurements of O_3 , NO/NO_x and meteorological parameters were conducted continuously throughout the campaign using conventional continuous analyzers (Thermo Scientific). A detailed description of the measurement procedures have been described previously by Asaf et al. (2008; 2010). Prior to commencement of the field measurements, all the monitoring instruments were cross-calibrated. In the field, the monitors were calibrated daily for both zero and span using EPA Protocol standard NO ($\pm 1\%$) gas and an internal calibrated O_3 generator. A similar calibration system

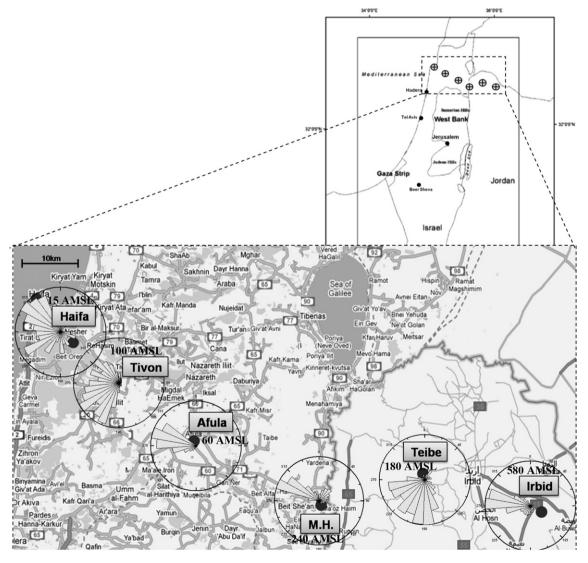


Fig. 1. Regional map showing the measuring sites (circle with inner cross) with their topographic heights and the wind direction components for each of the measuring sites in a larger scale of the region. Shuk-Haifa (32°82′N 34°96′E) and NS-Haifa (32°76′N 35°04′E) located in Haifa city, urban sites; Tivon (32°71′N 35°12′E) and Afula (32°61′N 35°29′E), semi-urban sites; Maoz-Haim (M.H., 32°50′N 35°55′E), Teibe (32°33′N 35°42′E) and Irbid (32°29′N 35°59′E), rural sites.

was also used in the daily Israeli National Air Monitoring Network calibration system. Data were measured every 10s, and recorded as 5 min averages.

Spatial NO2 was measured at MH site, using a Differential Optical Absorbance Spectrometer (DOAS) system which has been described previously by Platt (1994) and has an average detection limit of 0.1 ppby. Briefly, the instrument consists of a spectrograph and a transmitting/receiving Newtonian telescope that transmits light from a Xenon-arc lamp, focused into a parallel beam by a parabolic mirror. The transmitted light is reflected back into the telescope by an array of retro-reflectors, located at a distance of 3.5 km (total light path is 7 km). The returning light beam is transmitted into the spectrograph and then to a photodiode array. The raw data processing has been described in detail previously (Asaf et al., 2009) and is briefly summarized here. Spectra were corrected for the contributions of scattered "background" light and the spectrum of the Xe lamp, followed by a nonlinear least squares algorithm fit to the reference spectra using the analysis software MFC (Gomer et al., 1993). Finally, the optical density was quantified at the 435 nm absorption peak using the differential absorption

cross sections of NO₂ and concentrations were calculated in accordance to the procedure proposed by Platt (1994).

3. Results and discussion

The domain between the northern part of Israel and Jordan, over which presumably significant regional transport occurs, was examined through statistical analysis of observed values at different periods for each measuring site.

3.1. Wind direction analysis

Wind rose plots for all the sites as measured during the campaign are presented in Fig. 1, and exhibited a strong western component suggesting transport eastwards from the western sea coast of Israel towards the Jordan Valley and over to Jordan.

The western quadrant dominated (>90% of the time) between 08:00 and 00:00 (GMT+2), indicating the importance of daytime eastward transport. This tendency becomes more pronounced during the afternoon hours (Fig. 2) when the Mediterranean Sea

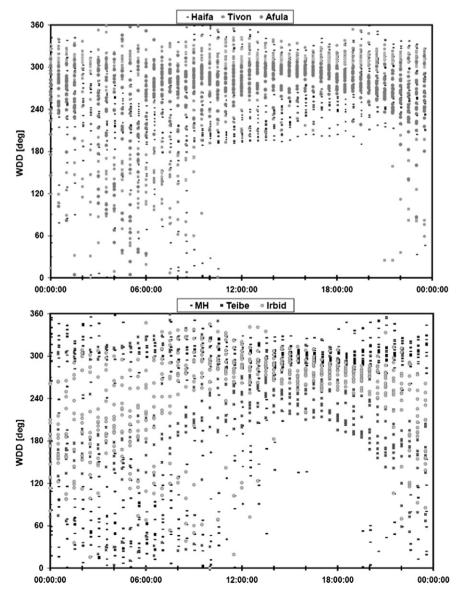


Fig. 2. Diurnal variation of the wind direction in half hourly intervals for the measuring sites, during the entire campaign.

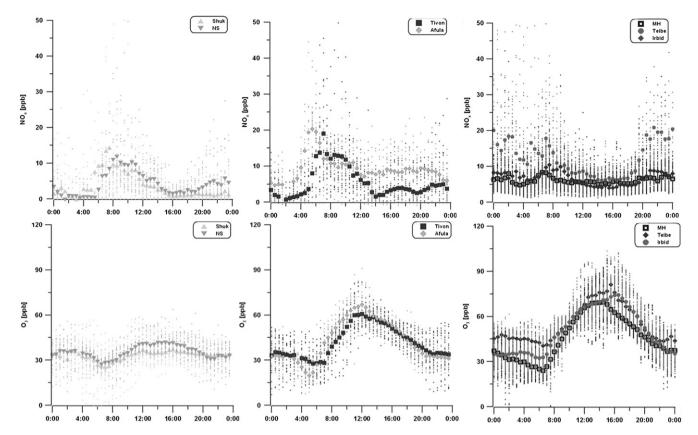


Fig. 3. Diurnal variation for NO_x and O₃ concentration and half hourly averages (marked by the bold marker) at the seven measuring sites, for the entire campaign. Initials <NS> for Neve Shanan and <MH> for Maoz Haim.

breeze affects the inland sites. During the night (22:00–00:00) until the morning hours (08:00) the wind demonstrated an opposite pattern with air masses arriving from the west for a limited period of time. Nevertheless, it is clearly demonstrated for every one of the sites, starting from the coast (at Haifa) and all the way to Irbid, that during day time, wind predominantly flows from the west and northwest quadrant.

The topography in the region allows direct eastward pollution transport through a valley corridor constrained in the north and south by elevated terrain. In the north the Galilee Mountains and from the south the Shomron mountain ridges leaving the Izrael and Beit She'an Valleys in between, followed by the Jordanian plateau, creating a valley which can funnel the air masses towards Jordan.

3.2. Time of day analysis

The photochemical processes responsible for ozone formation are the oxidation of volatile organic compounds in the presence of nitrogen oxides. Nitrogen oxides and ozone concentrations as a function of time of day are shown (Fig. 3) for all the seven sites extending from west to east.

3.2.1. Nitrogen oxides

A significant rise in NO_x levels during the morning traffic hours was observed as expected for Shuk and NS sites, following a decrease to mean levels below 2 ppbv until the evening hours in accordance with the drop in traffic pollution. Similar patterns were observed for the Tivon and Afula sites. At MH, the diurnal pattern was monotonous, indicative of background NO_x levels for this site.

Irbid showed a diurnal pattern very similar to that of MH, while Teibe demonstrated distinct morning and afternoon peaks, due to pollution emitted from transport on the roads south of the site. As can be observed from Fig. 3, the NO_X mean levels observed between 12:00–19:00 increase progressively towards the east, with the western stations at Shuk, NS and Tivon having lower mean levels than the eastern stations (MH, Teibe and Irbid), at parallel hours.

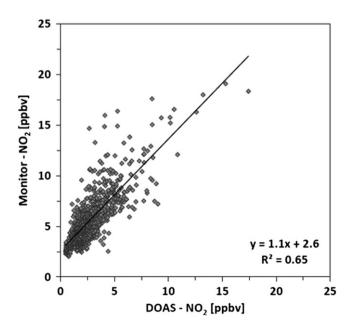


Fig. 4. A scatter diagram showing the ratio between the standard monitor to the DOAS technique for NO₂ measurements during May 31–June 18, 2009.

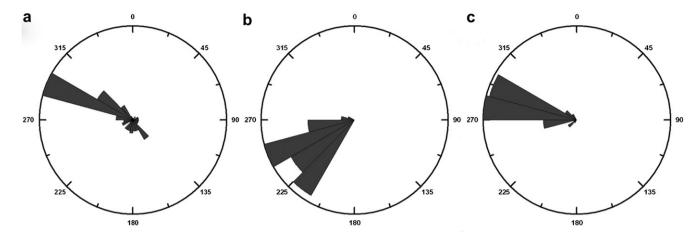


Fig. 5. (a) Maoz-Haim, (b) Teibe and (c) Irbid wind rose frequency plots for the maximum O₃ events defined as μ+2σ, plotted as a function of wind direction.

The relatively higher background NO_X levels (average of ~ 5 ppbv for noon and nighttime periods) for the eastern sites as compared to the western sites (especially at Haifa), appear to indicate eastwards transport of polluted air masses.

The relationship between the DOAS spatial measurements of NO_2 at MH site and the monitor values as shown in Fig. 4 shows a good correlation with higher values for the monitor by approximately 10%. This is to be expected since the monitor technique measures the difference between the total higher oxidized nitrogen species (NO_x) and NO, while the DOAS technique measures NO_2 directly. The good correlation further indicates the background nature of the air masses.

3.2.2. Ozone

The two stations in Haifa, Shuk and NS showed the lowest O₃ concentrations and mean values, due to morning titration from the traffic emitted NO (Fig. 3). O₃ peaked relatively early at approximately noon time (12:00) with maximum values of 55 ppbv, and average maximum levels up to 35 ppbv for Shuk and 40 ppbv for NS. Inland at Tivon, O₃ peaks at the same time as Haifa but has much higher values of 60 ppbv and 80 ppbv for maximum and average maximum levels, respectively. The fourth site, Afula, peaked even later during the day, shortly after noon (12:30), with even higher concentrations of almost 90 ppbv and an average maximum of 65 ppbv. The MH site showed the highest concentrations for the Israeli side, peaking at around 14:30 and reaching up to 100 ppbv with an average maximum level above 70 ppbv.

As the air masses continue to move inland, the O₃ levels continue to rise crossing the border towards Jordan border, peaking at 15:30 in Teibe with values above 100 ppbv and a mean maximum between 75 and 80 ppbv. The Irbid site O₃ levels were similar to Teibe, with a maximum above 100 ppbv, and a mean maxima of 75 ppbv, occurring however even later in the day, at 16:30.

The demonstrated pattern of the increase in O_3 concentrations with increasing inland distance, combined with the delay in the daily maximum peak time, clearly indicates a pattern of pollution transport inland from Israel towards Jordan. These elevated O_3 levels can be attributed to strong photochemical O_3 production occurring during inland transport, combined with the dominant S/SW/W wind direction patterns taking place during the late morning and noon time.

A plot of O_3 maximum events (defined as mean $(\mu)+2$ standard deviations $(2\ \sigma)$) as a function of wind direction is shown in Fig. 5. The figure clearly demonstrates that the O_3 peak events for MH, Teibe and Irbid all originated from the west. While the general wind

direction for Teibe demonstrated a strong southern component (Fig. 1), the maximum O₃ levels were recorded when the wind flow was from the west (see Fig. 5), presenting additional evidence that these events are mainly due to transportation of polluted air masses arriving from the west.

The O_3 statistics from the seven sites are presented in Table 1, demonstrating a constant rise in the absolute values of the O_3 mean, median and especially, maximum values with increasing inland distance. Generally, the coastal sites (Haifa) have the lowest O_3 levels, followed in order by those measured at Tivon, Afula, MH, and the Jordanian sites, further supporting the fact that O_3 is being produced during inland travel of the air parcels.

A histogram showing the average maximum O_3 levels, median values and time of maximum levels for each of the sites is shown in Fig. 6. It clearly demonstrates the delay in time of the O_3 events with distance inland. Tivon showed a maximum at 11:00 am with values above 70 ppbv for the elevated events. Afula demonstrated elevated levels above 75 ppbv with peak events occurring an hour later, at 12:00. Taking into account the average wind speed of 20 km h $^{-1}$ at noon together with the distance between the sites (20 km) gives a time lag of 1 h, supporting evidence of O_3 transportation. On the Israeli-Jordanian border, at Maoz-Haim, a further distance of 25 km east, the O_3 elevated levels highest frequency occurred at 14:00, 2 h later than Afula, with values above 78.5 ppbv. The measured wind speed during these hours (mean 17 km h $^{-1}$) between the sites

Table 1Summary of the ozone mean, median, standard deviation, time of maximum events and the value of the average daily maximum for the seven measuring sites, presented in the order from west to east for the entire campaign. Units are given in ppb. In addition, per-cent increase in the ozone average daily max, relative to the coastal sites, is presented for the current campaign and the previous modeling results (Weinroth et al., 2008).

	Haifa- Coastal		Tivon	Afula	Maoz- Haim	Teibe	Irbid
	Shuk	NS					
Mean	33.8	35.7	42.8	43.3	45.9	55.8	51.3
Median	33.9	35.9	43.4	42.8	45.3	52.0	50.5
Std	9.2	8.0	13.8	15.7	16.3	14.2	16.9
Average daily max	43.7	47.9	63.5	72.7	78.8	88.5	83.6
Time of max			11:00	12:00	14:30	15:00	16:30
Relative increase in the average	-	-	~40%	~60%	~75%	~95%	~87%
daily max							
Model estimated relative increase in the average daily max			~40%	~50%	~80%	~90%	~90%

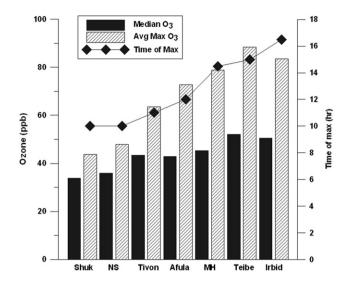


Fig. 6. Time of day histogram of the average maximum $(\mu+2\sigma)$ and median O_3 concentrations for Tivon, Afula, Maoz-Haim, Teibe and Irbid.

correlated with the time delay of the maximum peak events. At Tiebe, peak events above 84 ppbv occurred between 14:30–15:30, and based on the wind speed and distance between the sites, a distinct correlation is observed with the transport time. O_3 levels at Irbid, peaked in the late afternoon, later than 16:00, with the highest frequency of elevated levels (above 82 ppbv), correlating with average wind speeds of 23 km h⁻¹ and a travel distance of 25 km.

3.3. Trajectory analysis

While the time of day analysis provides important insights in understanding the observations, it does not explicitly takes into account the physical transport processes. To assess the effects of the synoptic-scale atmospheric transport patterns on observations at

the different sites, air mass pathways were analyzed. During the campaign, the synoptic conditions were typical of the season (Dayan, 1986; Dayan and Levy, 2002) dominated by the Persian trough. This synoptic configuration generates northwest winds, influencing the north and the central regions of Israel.

The source regions of the air masses were determined using the NOAA HYSPLIT model (Draxler and Rolph, 2003) with the GDAS meteorological dataset. 72 h back trajectories were generated every 3 h for each campaign day, and applied for the Irbid site. The results indicated a decisive trend of a west-northwest wind regime for air masses passing over the polluted coastal plain of Israel. More than 90% of the air-trajectories originated in the east and south of Europe and passed over the western sea shore of Israel before reaching Irbid, picking up fresh pollutants on their way, thus explaining the elevation in the local pollutant background levels as the air masses moved eastwards.

Typical 24 and 72 h back-trajectories arriving to Irbid are shown for June 6th (Fig. 7) demonstrating the pathway of the air masses, originating from south of Europe and passing over the metropolitan coast of Israel. Consequently, the air masses that arrive at Maoz-Haim and Irbid, are associated with local emissions located in coastal Israel. Fig. 7 (inset) clearly demonstrates the rise in O₃ concentrations accompanied by the delay in the time of the peak, and transport inland of the air masses from the coast of Israel towards northern Jordan.

3.4. Comparison to modeling studies

The effect of local air pollution emission sources on O_3 formation downwind of coastal Israel has been examined in a previous modeling study (Weinroth et al., 2008) which employed a two linked models system: the mesomet RAMS 6.0 model developed by Pielke et al. (1992) and the photochemical Eulerian CAMx 4.31 model of ENVIRON (2002) and Zhang et al. (2004). Simulated O_3 values and expected cycles were generally well reproduced by the models and showed good comparison to the observed O_3 maxima

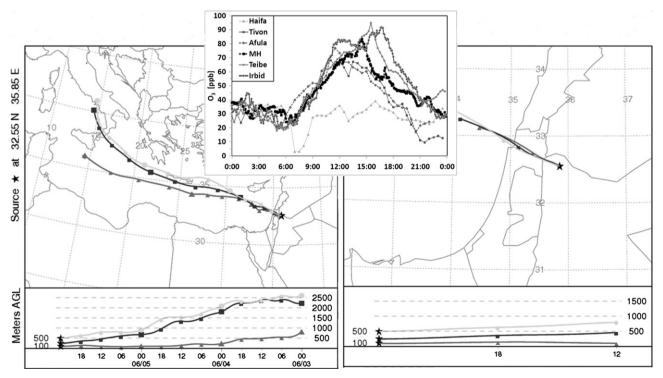


Fig. 7. Typical air mass back-trajectories for June 6th, 2009 demonstrating the distinct trend of a west-northwest wind regime.

for different sites in Israel. CAMx O₃ values at surface and 300-m AGL over central inland Israel presented a southwest to northeast ridge of high values from Jerusalem extending to Irbid, Jordan. Elevated levels were predicted by the model for the Jordan Valley (Maoz-Haim) with absolute maximum O₃ level, predicted for Irbid, Jordan during the afternoon. Since no measurements were available at that time for these areas, the simulations could not be validated

Comparing between the predicted model trends with the current campaign results yields good agreement for the multiple sites investigated. While the model under predicted by 15% max values (Weinroth et al., 2008) as compared to measured results (Table 1), a similar trend for the average daily maximum was verified. The constant rise predicted by the model results, especially the predicted afternoon $\rm O_3$ maximum over Irbid, was validated in the current campaign yielding a maximum relative increase in $\rm O_3$ levels of 90% between Haifa and Irbid. Furthermore, the measurements showed very clearly that elevated levels by $\sim 75\%$ were measured for the Jordan Valley relative to the coastal sites, correspondingly to the calculated model values.

The results presented in the current study suggest that the high O_3 values observed at the sites were indeed as predicted by the previous modeling simulations, due to pollution transport. This transport originated from the polluted north-western coastal plain of Israel, validating the model simulations.

4. Summary and conclusions

Under suitable meteorological conditions of prevailing westerly winds, elevated levels of photochemical pollution of anthropogenic origin are transported over long distances (up to more than 100 km). Thus continental emissions from Israel can provide a major contribution to photochemical pollution in Jordan. During such periods, photochemical pollution is essentially a regional rather than a local problem with elevated concentrations of photochemical ozone being observed simultaneously over Jordan and Eastern Israel.

The present study indicates that inland areas of Israel and Jordan can be strongly affected by elevated ozone concentrations which are produced inside air masses that have entrapped pollutants during their passage over the metropolitan coastal region of Israel. Previous modeling results have been validated by showing that peak ozone values were observed at later hours depending on distance and wind speed from the Mediterranean coast and that maximum ozone levels can be observed over Northern Jordan.

In the case of the present study, multiple political jurisdictions were included (i.e., Israel and Jordan). Thus the geographic patterns of pollutants often do not coincide with the boundaries of political jurisdictions. Consequently, impacts of air pollution in one political jurisdiction may be caused, in part, by emissions from another jurisdiction, making cooperation necessary to achieve pollution control. A successful preliminary collaboration infrastructure was established between the participant authorities, such as coordinated campaigns and data sharing. These led to further successful collaborations with additional research groups in the Middle East and opened a pathway for future campaigns. Additionally, in order to control elevated ozone levels, the two countries will need to cooperate if improvement in air quality is to be achieved.

Acknowledgments

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