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Estimation of transboundary transport of pollution aerosols by remote sensing in the eastern Mediterranean

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[1] Quantifying transboundary transport of pollution is important for understanding the global distribution of pollution and pollutant burdens in regional and global scales. Using observations from the Moderate resolution Imaging Spectroradiometer (MODIS) on board the Terra and Aqua satellites, the transport of pollution sulfate aerosol was estimated in the eastern Mediterranean. Over a 150 km line west of the Israeli coast, the estimated annual sulfate flux is in a range of 0.025 to 0.062 Tg S a^{-1}. These estimates are consistent with airborne measurements which estimated an annual flux of sulfate of 0.024–0.054 TgS a^{-1}. The MODIS-based estimates are also in good agreement with estimates of seasonal and annual fluxes from the GOCART model. This case study demonstrates a feasible way to estimate transboundary transport of pollution aerosol by remote sensing means.

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1. Introduction

[2] Understanding transboundary transport of pollution plays an increasingly vital role in the understanding global pollution transport and its effects on climate, air quality and visibility worldwide [*Bertschi and Jaffe*, 2005; *Erel et al.*, 2006; *Goldstein et al.*, 2004; *Liang et al.*, 2004; *Mari et al.*, 2004; *Parrish et al.*, 2004; *Ramanathan et al.*, 2007; *Weiss-Penzias et al.*, 2006]. Examples of relevant cases of transboundary transport of pollution include dust expelled from Asia to North America and from North Africa to the North America, transport of anthropogenic pollution from North America to Europe, from east Asia to North America [*Yu et al.*, 2008] and from Europe to the eastern Mediterranean (EM) [*Wanger et al.*, 2000]. The eastern Mediterranean in particular is subject to varying meteorological conditions and to substantial fluxes of gas phase pollution and airborne particulate matter (PM) from central and eastern Europe [*Dayan and Levy*, 2002; *Erel et al.*, 2002, 2006, 2007; *Kouvarakis et al.*, 2002a; *Lelieveld et al.*, 2002], as well as from North Africa and the Saudi peninsula [*Dayan et al.*, 1991, 2008]. Estimating the flux of pollution transport is complex, and new methods which provide good temporal and spatial resolution are required.

[3] Traditional methods for estimating the flux of pollutants to a specific area are usually based on field measurements, either by ground-based monitoring stations or through extensive field campaigns that combine surface and airborne measurements of pollutant levels. The surface sampling measurements, while frequently providing excellent year-round budgets of pollution levels are, by definition, limited to ground concentration and hence may provide an inaccurate assessment of the entire flux throughout the atmospheric column. Such measurements often also suffer from limited temporal coverage. Airborne measurements can provide essential information about the transport at higher altitudes, but are scarce and provide little temporal coverage. Here we investigate a third approach for estimating the flux of particulate matter pollution which uses daily remote sensing observations of aerosol optical thickness and size parameters over a specific area. The potential advantages of remote sensing methods are the superior temporal coverage as well as integration of the entire pollution column.

[4] Most remote sensing methods derive aerosol concentrations from retrieved aerosol reflectance and spectral dependence. These parameters are used to derive aerosol optical depth (AOD), concentration and proxies of the aerosol dominant mode [*Tanre et al.*, 1996]. A number of remote sensing studies estimated aerosol mass concentrations. *Fraser et al.* [1984] used a simple multiplicative constant to relate aerosol optical depth (AOD) to mass concentration. *Kaufman et al.* [1990] used a variable proportionality constant using the Advanced Very High Resolution Radiometer (AVHRR) data. *Gassó and Hegg* [2003] applied a similar approach customized to the Moderate resolution Imaging Spectroradiometer (MODIS) applications. A number of recent studies specifically estimated fluxes of aerosols. For instance, MODIS was used for estimating African dust column concentration, transport, and deposition [*Kaufman et al.*, 2005a]. *Koren et al.* [2007] estimated the flux and deposition of dust transported

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from Africa to the Amazon forest also using MODIS. *Yu et al.* [2008] recently applied remote sensing methods to estimate flux of pollution aerosols over the Pacific Ocean using MODIS aerosol retrievals in combination with measurements of the dependence of aerosol mass extinction efficiency on relative humidity and of aerosol vertical distributions from field campaigns and available satellite observations in the region.

[5] Although production of sulfate from the oxidation of dimethylsulphide from biogenic activity in the marine boundary layer and SO_2 from ship emissions contribute to the sulfate budget, the long-range transport of pollution from central and eastern Europe has been shown to dominate the sulfate concentration in the EM region [*Ganor et al.*, 2000; *Kouvarakis et al.*, 2002a]. The homogeneous conversion of SO_2 to particulate sulfate is rather slow, i.e., about 1–3% per hour [*Meagher et al.*, 1981]. Globally, the atmospheric lifetime of sulfate is mainly controlled by wet deposition, and it is estimated to be up to 6 days on global average [*Chin et al.*, 2000]. Because of the absence of rain and wet deposition during the summer in the EM, and the slow dry deposition velocity of sulfate aerosol (in an order of 0.01–0.4 cm s^{-1}), sulfate aerosols comprise between 50 and 90% of the total sulfur in air masses transported to the region [*Matvev et al.*, 2002]. Two additional factors favor late spring and summer sulfate particulate abundance over the EM: (1) high solar radiation flux that leads to efficient oxidation of SO_2 to SO_4 via OH radicals and (2) the prevailing summertime westerly winds [*Dayan*, 1986] that transport sulfate-rich air masses from sources over central Europe before significant removal occurs. During the spring and summer months, the most important factor affecting pollution over the EM is not necessarily atmospheric conditions leading to local air pollution buildup (shallow boundary layer and weak winds) but mainly the potential of the atmosphere to transport pollutants downwind from remote sources (central Europe) to the EM [*Erel et al.*, 2006; *Kouvarakis et al.*, 2002b]. The late spring and summer in the EM and over Israel is characterized by the predominance of a unique synoptic system originating from the Persian Gulf as an extension of the Monsoon Low known as the "Persian trough." Deepening of the Persian trough due to a temporary withdrawal of the subtropical high centered mostly over North Africa and the EM leads to steeper pressure gradients and accordingly to a strong zonal flow enabling such a transport from the source regions toward the EM [*Dayan et al.*, 1988; *Erel et al.*, 2007].

[6] In this study we demonstrate a method that applies remote sensing measurements for estimation of pollution transport to the eastern Mediterranean (EM) region. In this case study we focus on sulfate transport because field observations are mainly available for sulfate in the region. For this comparison we attempt to convert from the retrieved aerosol optical depth or corresponding volume from MODIS on board the Terra (overpassing at 1030 local time) and Aqua (overpassing at 1330 local time) platforms to sulfate mass and flux. We then compare the estimated annual transported sulfate mass with estimates from a global chemical transport model (GOCART) and with estimates based on in situ aircraft measurements that were conducted in the region over several years. While the method presented may not be directly implemented globally, we demonstrate the needed basic strategy and parameters in order to adopt it to other regions.

2. Methods

2.1. Satellite Remote Sensing

[7] The underlying assumption behind this study is that under the prevailing westerly winds in the EM, the amount of sulfur in polluted particulate matter (PM) can be estimated by remote sensing west of the coastal line of Israel, over the Mediterranean Sea where the MODIS over-ocean retrievals (Collection 4 data) of aerosols are accurate [*Remer et al.*, 2005]. By using prevailing wind speeds, the fluxes can then be estimated. Meteorological (NCEP reanalysis data) and remote sensing data for 1 year (2004) were analyzed. The analysis of the pollution transport was performed only for days categorized as those in which westerly wind flow is dominant and the flux was assumed to be zero for easterly wind flow conditions. The so-derived fluxes would represent the transport of sulfate from central Europe to the EM. Days with high cloud coverage were also not included in the analysis since on these days it was impossible to make an accurate estimate of the amount of polluted aerosol. The amount of polluted aerosol for each relevant day was calculated for $1^\circ \times 1^\circ$ locations west of the Mediterranean coast, forming a line at the eastern edge of the EM (not only those locations close to Israel). Approximately half of the cases (out of about 350 cases for Terra and for Aqua) were mixed aerosols, 45% large (dust dominated) particles, and 5% fine (pollution and smoke dominated) particles.

[8] The flux of polluted aerosol was calculated from the MODIS retrieved aerosol optical thickness and respective contributions of fine-mode and coarse-mode aerosols, as follows:

[9] 1. Fine-mode and coarse-mode aerosol volume are calculated. The MODIS average aerosol optical thickness (AOT) was obtained from an optimal selection of one fine-mode and one coarse-mode aerosol model from a total of nine aerosol models (4 for fine-mode and 5 for coarse-mode) [*Remer et al.*, 2005]. The fractional contribution of the fine-mode aerosol model to the total AOT is referred to as "aerosol fine-mode fraction." The MODIS fine-mode and coarse-mode AOT were converted to fine-mode volume (V_f) and coarse-mode volume (V_c), respectively, on the basis of the assumed aerosol size distributions for the individual aerosol models [see *Remer et al.*, 2005]. V_f and V_c have a unit of $cm^3\ m^{-2}$.

[10] 2. Contributions from maritime aerosol are deducted. Globally, AOT for pure marine air is approximately 0.06–0.07 [*Kaufman et al.*, 2001], consisting of sea salt, non-sea-salt sulfate from the oxidation of DMS, and some organic aerosols. Over the Mediterranean Sea, clean days with AOT < 0.1 have an average AOT of 0.07, which is typical for marine air in that area. For such clean days, the relationship of V_f and V_c with near surface wind speed at 850 hPa (W) was calculated respectively by a linear fit to the wind components from the NCEP reanalysis assuming that the only contribution to AOT is from sea spray. It was found that in the EM and for the maritime aerosol (a total of 2960 data points), $V_{fm} = 0.011 - 0.00037W$ and $V_{cm} =$

0.022+0.00136W. There are no observations to evaluate the derived relationships.

[11] The fine and coarse particle volumes with continental origins (i.e., pollution and dust) for each day and location, denoted as V_{fl} and V_{cl}, respectively, were calculated as follows:

$$V_{fl} = V_f - V_{fm}; V_{cl} = V_c - V_{cm} \quad (1)$$

A sensitivity test for this method shows that choosing a limit of AOT = 0.15 instead of AOT = 0.1 for a representative of maritime aerosol generates an error of less than 20% in the final pollution or dust concentration estimates.

[12] 3. Polluted aerosol volume is estimated. We assume that for each retrieval, the aerosol population can be represented as a linear combination of coarse mode and fine mode contributions; that is, V_{fl} and V_{cl} generally consist of contributions from dust and pollution, i.e.,

$$V_{fl} = aV_p + bV_d; V_{cl} = cV_p + dV_d \quad (2)$$

To determine the coefficients a, b, c, d in (2) we have classified cases of "pure dust" and "pure pollution" as following: Days in which AOT ranged between 0.4 and 1 were sorted by the fine-mode fraction (ff) of AOT. Assuming that dust has a fine-mode fraction of ~0.5 and pollution PM has a fine-mode fraction of ~0.8, the data was classified as "dust" (1 > AOT > 0.4, ff < 0.5) or "pollution" (1 > AOT > 0.4, ff > 0.8) events. These classifications are based on earlier studies. [*Gassó and O'Neill*, 2006; *Kaufman et al.*, 2005b; *Kleidman et al.*, 2005] For the pure "pollution events," we derived $a = V_{fl}/V$ and $c = V_{cl}/V$. For the pure "dust events," it was found that $d = V_{cl}/V$ and $b = V_{fl}/V$ where V, V_{fl}, and V_{cl} are the average values for each of these data sets. With these coefficients determined, the volume of pollution and dust (V_p and V_d, respectively) can be derived by inverting (2):

$$V_p = (dV_{fl} - bV_{cl})/(ad - cb); V_d = \left(V_{cl} - cV_p\right)/d \quad (3)$$

Note that the coefficients derived in equation (2) may vary with the location and season, and therefore a detailed analysis should be conducted for each case. However, for our case where annual averages are sought this is not necessary.

[13] 4. Sulfate aerosol fluxes are calculated. In this study, we focus on estimating sulfate fluxes to the EM. We assumed that (1) half of the polluted aerosol mass (dry) is composed of sulfate, consistent with observations from the Mediterranean Intensive Oxidant Study (MINOS) conducted in the summer of 2001 [*Lelieveld et al.*, 2002]; (2) 850 hPa is the best proxy for the altitude in which sulfate is transported to the Israel coast of the EM; and (3) aerosol-associated water in ambient conditions as observed by MODIS can be estimated by using the measured hygroscopic growth of pollution aerosol [*Hess et al.*, 1998] and the relative humidity at 850 hPa. It was found that the region is quite dry through the year. For such relatively low RH conditions, uncertainties (e.g., 10–20%) in AIRS RH retrievals would not have significant impacts on our estimates of aerosol water.

[14] As opposed to previous studies [*Fraser et al.*, 1984; *Gassó and Hegg*, 2003; *Kaufman et al.*, 1990, 2005a; *Tanre et al.*, 1996] in this work we try to derive the fine and coarse modes at the AOD level by deconvoluting the AOD to two components using the nine aerosol models described by *Remer et al.* [2005]. We adopt this approach as this is one of the operational MODIS applications. By assigning the coarse or fine modes to a specific aerosol AOD, we derive the aerosol size distribution (and later mass) from the assumed aerosol size distribution. This approach was explored in this study as a way to derive sulfate mass rather than AOD, as mass is the quantity actually measured in the field. Although the size distributions are not validated, the volume calculated is directly derived from the AOD, which is a widely used and accepted measurement. No additional assumptions are introduced in the transition to the volume. As stated above, we used volume instead of AOD in this study because the end goal is to obtain and assess sulfate transport, which can be compared with models and observations. Using estimated sulfate volume and known density, we estimate the sulfate mass rather than apply an assumed extinction efficiency to "sulfate AOD."

[15] To derive the sulfate mass, an average sulfate fraction in the aerosol was assumed. This assumption was based on MINOS measurements (in summer) [*Lelieveld et al.*, 2002] and GOCART simulations. The ratio in these simulations is 0.7–0.85 throughout the year. Given that nitrate is not included in the model, the true sulfate ratio is probably slightly lower. In order to further constrain our assumed sulfate ratio, nitrate amount was estimated from *Levin et al.* [2005] who measured a steady concentration of ~20 $\mu g\ m^{-3}$ of nitrates over the Dead Sea region. In addition, the water content of the aerosol was estimated using aerosol hygroscopic properties taken from the OPAC database [*Hess et al.*, 1998]. As a result of these adjustments we use a sulfate ratio of 0.5 in our study.

[16] The selection of 850 hPa level as an appropriate transport height for sulfate is based on previous observations and analyses [*Barnaba and Gobbi*, 2004; *Gobbi et al.*, 2004] and limited observations from the Geoscience Laser Altimeter System (GLAS) [*Spinhirne et al.*, 2005], as shown in Figure 1. The frequency of aerosol detection in a vertical layer (100 m interval) is calculated as a ratio of the number of valid aerosol backscatter retrieval to the number of laser shots in the region. The 850 hPa level was also chosen because it is typically sensitive to emissions from anthropogenic sources and is regarded as the approximate boundary between the surface wind regime and the free troposphere [*Dayan and Lamb*, 2003]. This is further corroborated by aircraft measurements of sulfate made at 350 m and the zero sulfate concentrations found at 2500 m [*Luria et al.*, 1996]. Special meteorological conditions in the EM coast during the transitional seasons often result an easterly offshore flow at a shallow tropospheric layer. We use the 850 hPa winds in order to avoid easterly offshore flow characterizing, at times, lower altitudes. In addition, it is common, especially during spring and summer months, that the wind profile is quite monotonous, and no wind shear is observed neither in speed nor in direction [see *Koch and Dayan*, 1992].

[17] Three $1^\circ \times 1^\circ$ locations near the Israeli coastline were chosen out of the entire data set. For each day and

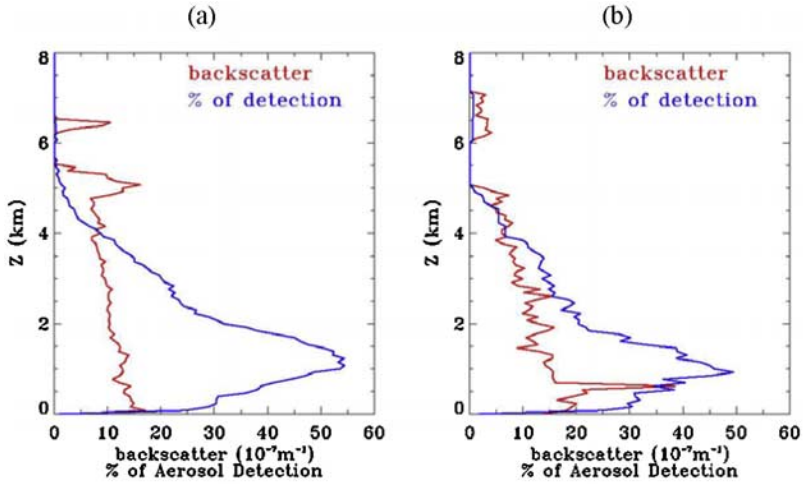

Figure 1. Profiles of GLAS average aerosol backscatter (red line) and detection frequency (blue line) during (a) 25 September to 19 November 2003 and (b) 17 February to 4 March 2004.

location the wind pattern at the 850 hPa level was obtained from the NCEP reanalysis data [*Kistler et al.*, 2001] and multiplied by the u component at 850 hPa and by the derived sulfate mass loading to obtain the flux of sulfate. Afterward a seasonal sum was calculated and compared to field measurements and model simulations.

2.2. In Situ Aircraft Data

[18] Sulfur transport to the EM was estimated in several research flights and field campaigns. Flights made within the boundary layer (300 m asl) 70 km west of and parallel to the Israeli coastline during September 1993 and June 1994 measured an average concentration of 38 ± 7 and 108 ± 63 nmol m^{-3} of particulate sulfate, respectively. Each leg was between 180 and 200 km. long. Additional flights were conducted in spring (May and June 1998, six flights) and late fall (September 1996, four flights and November 1995, four flights). The aircraft measurements indicated that the influx of sulfur compounds arriving at the Israeli coast from upwind sources located in Europe are relatively elevated during the early summer, with an average flux of 18 mg S h^{-1}. At the end of the summer, the pollutant influx rate advected toward Israel is, on average lower (~5 mg S h^{-1}). In the autumn, the EM region is usually subjected to more easterly winds. Since there are practically no pollution sources east of Israel, the contribution of sulfur from such events was, as expected, lower than the summer values (~4 mg S h^{-1}). On the basis of the above, *Matvev et al.* [2002] calculated that the yearly total sulfur influx to Israel is approximately 0.06 Tg a^{-1}.

[19] Table 1 compiles the sulfate particulate concentrations as measured and published in different field studies in Israel. Fluxes were calculated by using *Matvev et al.* [2002, equation (1)] which converts concentration of sulfur in g per cubic meter (quantity measured) to sulfur flux in mass per unit time (mg h^{-1}) taking into account the vector component of onshore wind speed, length of flight leg and the depth of the mixed layer. In order to assess the atmospheric sulfur budgets to and from Israel, the pollution flux at the western and eastern borders of central Israel was calculated by *Matvev et al.* [2002] on the basis of the flight measurements using the following equation:

$$F = C * V * L * H * 3.6/1000 \quad (4)$$

where

F sulfur flux in mass per unit time (mg h^{-1});
C concentration of sulfur (g m^{-3});
V vector component of wind speed (m s^{-1}) in eastern direction;
L length of each flight leg (m);
H mixing layer height (m).

The concentration for the sulfur compounds includes the sum of sulfur dioxide and sulfate presented as total sulfur. However, SO_2 concentrations are very low compared to that of sulfate. *Matvev et al.* [2002] measured 2–4 ppbv which are about 5–10 μg/cubic meters! The factor 3.6/1000 converts the flux from units of g/s to mg/h. The results of the flux rates for the sulfur compounds over Israel are summarized in Table 1 for the different research flights.

[20] An examination of Table 1 shows that the highest fluxes from the west were for the early summer time, averaging 18.4 mg S/h. During this season, the levels were more than three times higher than averages for late summer (5.3 mg S h^{-1}) and almost five times the average levels measured for the fall (3.9 mg S h^{-1}).The local pollution contribution during the early and late summer periods were similar (close to 25 mg S h^{-1}), and almost triple the autumn sulfur level, 9.1 mg S h^{-1}. The wide range in fluxes derived is due to the distance from the polluted coastline and the distance from the center to cleaner atmosphere in the northern part of the country and in the south in the Negev Desert.

3. Results and Discussion

[21] To evaluate the pollution flux calculation, we estimated the amount of sulfur flux using MODIS onboard both Terra and Aqua that have been independently calibrated. These flux estimates were further compared with flux calculations made by the GOCART model and those de-

Table 1. Compilation of Sulfate Particulate Concentrations and Yearly Fluxes as Measured in Different Studies in Israel

Region	Measurement Periods	Sulfate Concentration Average (nmole m^3)	Sulfate Yearly Flux[a] (Tg a^{-1})
Etzion (mountains)[b]	Jul–Aug 1984, 1986	86	0.08
Jerusalem (mountains)[b]	Jul–Aug 1987, 1988	103	0.09
Etzion (mountains)[b]	May–Jun 1989	70	0.06
Jerusalem (mountains)[b]	Jul–Aug 1990	128	0.12
Jerusalem (mountains)[b]	May, Jul 1990, 1991	85	0.08
West Kinnereth[b]	Aug–Sep 1993	87	0.03
East Kinnereth[b]	Dec 1993	71	0.07
Caesarea (northern coast)[b]	Jun 1993	106	0.12
Eastern Mediterranean[c]	Sep 1993	38	0.08
Eastern Mediterranean[c]	Jun 1994	108	0.22[d]
Eastern Mediterranean[e]	Jun 1998	105	0.16
Eastern Mediterranean[e]	Sep 1996	26	0.04
Eastern Mediterranean[e]	Nov 1995	21	0.03

[a]Conversion of measured concentrations in (nmole m^3) to yearly fluxes was along *Matvev et al.* [2002] while taking into account the vector component of onshore wind speed, length of flight leg and the depth of the mixed layer.
[b]*Luria et al.* [1996].
[c]*Wanger et al.* [2000].
[d]Flight during June 1994, which was a month with extremely high pollution in Israel.
[e]*Matvev et al.* [2002].

rived from the field experiments discussed in the previous section. GOCART results are independent of the MODIS instruments and it uses its own inventory of emission sources for anthropogenic sulfur and of modeled atmospheric chemical reactions.

[22] The global model GOCART is driven by assimilated meteorological fields at 3-h interval from the Goddard Earth Observing System Data Assimilation System (GEOS DAS) [*Chin et al.*, 2002, 2004]. GOCART simulates the major aerosol types: sulfate, dust, black carbon (BC), organic carbon (OC), and sea salt. Emissions from anthropogenic, biomass burning, biogenic, and volcanic sources, windblown dust and sea salt are included in the model. Processes represented in GOCART are chemistry, convection, advection, boundary layer mixing, dry and wet deposition, gravitational settling, and hygroscopic growth of aerosol particles. The size distributions for individual components are prescribed and coagulation and condensation processes are not considered. Individual components do not interact with each other. Details of GOCART and evaluation of its results against observations are documented in a number of publications [*Chin et al.*, 2000, 2004; *Ginoux et al.*, 2001, 2004].

[23] For this study, the GOCART model was run twice for 2004, one with all sources of sulfate aerosol and the other without anthropogenic sources. A difference between the two runs is used as a proxy for the anthropogenic sulfate coming from Europe. Model calculated sulfate mass is converted to that of ammonium sulfate $(NH_4)_2SO4$ for calculating sulfate fluxes on mass basis which can be compared with the aircraft measurements and the MODIS retrievals. To be consistent with the MODIS-based estimate and those derived from field experiments, GOCART sulfate fluxes are also calculated only for days when wind flow at about 850 hPa level is westerly but assumed to be zero for cases of easterly conditions. Comparison was done on the entire length as the research flights. GOCART values were taken for one point (because of its resolution), which are comparable to 2–3 MODIS pixels.

[24] Under the above mentioned set of assumptions and analyses, the following results were obtained:

[25] 1. We find good agreement between MODIS-derived sulfur fluxes (both Aqua and Terra) and estimates based on field measurements, for a 150 km line west of the Israeli coast. The Terra estimate of the annual sulfate flux is 0.153 Tg a^{-1} and the Aqua estimate is 0.159 Tg a^{-1}, corresponding to 0.038 and 0.040 Tg S a^{-1}, respectively. On the basis of field measurement data, *Matvev et al.* [2002] estimate an annual flux of 0.06 Tg a^{-1} of (dry) sulfur over the 150 km flight transect. Considering that the observed ratio of sulfate to total sulfur is 40–90% in the region [*Sciare et al.*, 2003; *Matvev et al.*, 2002] the annual flux of sulfate based on field measurements is 0.024–0.054 Tg S a^{-1}. Clearly, the MODIS-based estimates fall in the middle of the range.

[26] 2. We also find that MODIS-based estimates of the sulfate flux agree reasonably well with GOCART model simulations of anthropogenic sulfate, as shown in Figure 2 for seasonal averages. Compared to Terra, the GOCART estimates are about 85% and 30% higher in winter and spring, respectively, but 10–25% lower in summer and fall than the MODIS-based estimates. The annual sulfate flux from the GOCART model is 0.181 Tg a^{-1}, about 18% larger than the MODIS estimate of 0.153 Tg a^{-1}. For Aqua, GOCART simulations are in excellent agreement with MODIS estimates in summer and fall, but about 50% higher in winter and spring. The GOCART simulated annual flux of 0.201 Tg a^{-1} is about 25% higher than the MODIS/Aqua estimate of 0.159 Tg a^{-1}. Several factors could contribute to the GOCART-MODIS differences. Higher sulfate concentration in GOCART may be associated with cloudy conditions as a result of aqueous phase chemistry, thereby causing the GOCART average to be biased higher. The higher GOCART calculations may also result from an overestimate of aerosol emissions in Europe [*Chin et al.*, 2004]. On the other hand, assumptions in the MODIS-based estimate are likely to contribute to the MODIS-GOCART difference. For example, the assumption of sulfate mass fraction is based on MINOS measurements

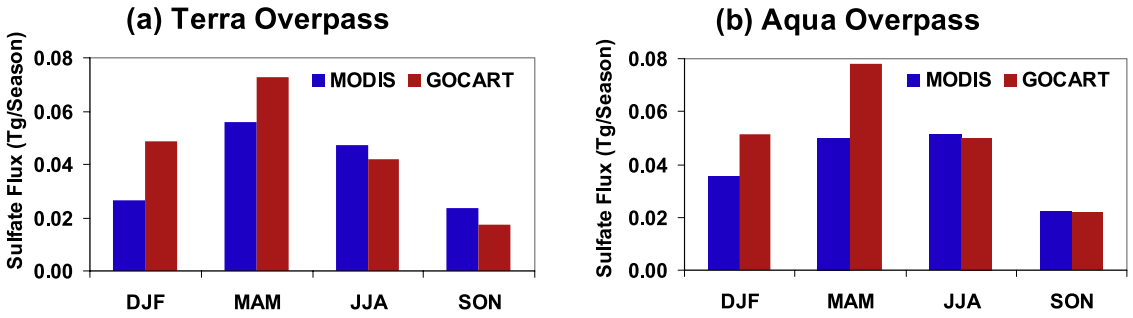

Figure 2. Comparison of MODIS and GOCART estimates of seasonal flux (Tg/season) of dry sulfate to the EM along the 150 km coastline for (a) the Terra overpass and (b) the Aqua overpass.

in summer, which may not be correct for winter and spring.

[27] On the basis of the model results, the comparison of the two instruments, and the consistency with the aircraft measurements, we suggest that either MODIS instrument can be used for estimating the flux of pollution on the basis of their daily retrievals.

4. Error Estimations

[28] MODIS AOT over ocean is quite accurate with an estimated uncertainty of ±0.05 AOD about 10%. Fine mode fraction has a larger uncertainty. *Tanre et al.* [1996] claim fine-mode fraction accurate to within 0.25. *Kleidman et al.* [2005] shows an over prediction of ~0.2 for dust dominated aerosols and underprediction of 0.05–0.1 for very heavily polluted cases. The overall error for our study will therefore depend on the frequency distribution of fine mode fraction of the cases. The theoretical maximum would be an error of 0.2 if all the cases were dust. A more likely number is somewhat lower since we have a mixture of cases, probably ~0.15. Here we assume that these uncertainties will transformed to an error of about 30% for the derived pollution volume, according to *Kaufman et al.* [2005a].

[29] We have also conducted an analysis of the possible errors that can be induced by changes in the values for the coefficients that transform the AOD to actual mass. We find that by varying the coefficients in equation (3) by one standard error of the fit, the coefficients for the fit in V_p change by 1.5%. The error for the fine fraction estimate in the marine aerosol leads to an error of 10%, and a change of the threshold in the classification of pollution particles leads to an error of 20%. By far, the largest error lies in the assumptions of the sulfate mass fraction in the aerosol, and in the mass of the wet aerosol. Because of lack of measurements to assess the assumptions made in this study, we assume such error is about 40%.

[30] If we assume that the individual errors discussed above are independent, which itself is difficult to assess, the overall uncertainty for the annual sulfate flux is estimated at about 55%. Correspondingly the MODIS estimated sulfate flux ranges from 0.025 to 0.062 Tg S a^{-1}. Note that this range is likely to be a conservative estimate because some uncertainties discussed above may be dependent and several uncertainties cannot be quantified. For example, we have used aerosol retrievals in cloud-free conditions to calculate transport flux of sulfate aerosol. If dry aerosol mass in cloudy conditions is significantly different from that observed in cloud-free conditions, the estimated sulfate flux may have large uncertainties or biases. We do not know how to quantify such differences and whether an overall bias exists in the estimated pollution fluxes [*Yu et al.*, 2008]. The use of 850 hPa wind in calculating the flux was based on previous observations and analysis of the general pollution aerosol vertical displacement [*Barnaba and Gobbi*, 2004; *Gobbi et al.*, 2004] and limited data from the Geoscience Laser Altimeter System (GLAS) [*Spinhirne et al.*, 2005]. A full evaluation of the assumptions and associated uncertainties can be done in the future with the emerging and continuous observations of aerosol vertical profiles from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard CALIPSO [*Winker et al.*, 2003].

[31] While the MODIS-based estimates are consistent with estimates from field experiments in the region and GOCART model simulations, we admit that these comparisons do not constitute the evaluation and validation of the satellite-based method. The accuracy of flux estimates based on field experiments and model simulations depends on a number of assumptions, as discussed earlier. It is also noted that the fluxes may have changed during the years of the measurements and the year of the MODIS analysis. However, the airborne measurements (see Table 1) do not show a consistent trend throughout the 90s. Several other pieces of evidence also point to a lack of significant fluctuations in the particulate sulfate concentrations over the eastern Mediterranean, aside from the seasonal fluctuation. Ambient particulate sulfate measurements have been intermittently performed at a rural site in Israel over a period more than 2 years. The measured concentration of sulfate observed during both consecutive summers were about 10–20 $\mu g\ m^{-3}$ [*Luria et al.*, 1989].

5. Conclusions

[32] This study demonstrates that combining observations of fine and coarse aerosols by the MODIS instrument on the Aqua and Terra satellites over ocean with the average winds from the NCEP reanalysis can be used to estimate pollution sulfate fluxes to Israel. On an annual basis, we estimated that 0.025–0.062 Tg S a^{-1} of anthropogenic sulfate was transported to the region. The estimates are found to be in accord with fluxes calculated by GOCART model simulations, and are in line with estimates based on several field

campaigns. The results also show that, at least in the EM region, both Terra and Aqua measurements provide fairly similar estimates (within 20%), indicating that there may be no obvious difference in pollution transport between morning and afternoon in this region. Major sources of uncertainty in the estimated flux come from estimates of maritime aerosol, fraction of sulfate in aerosol, and aerosol water and assumption of transport height. To reduce these uncertainties, an integration of multiple satellite sensors, field experiments, monitoring stations, and modeling is desired.

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