Quantifying How Geophysical Correlation Aids in Estimating Lower Tropospheric Ozone, … If We Understand It *!*

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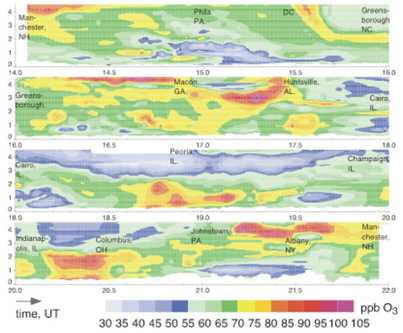
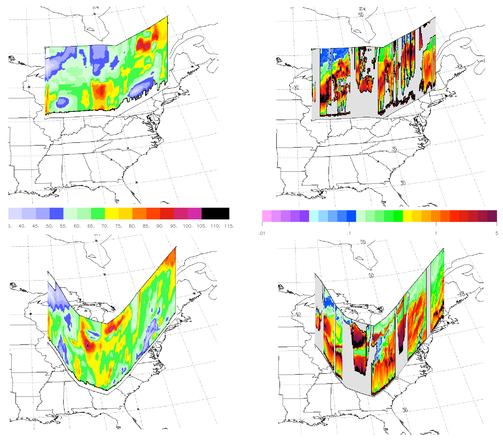
Thanks to OMI, AIRS, MLS, IONS Teams



A case-study sample of ozone variability 0–4 km asl for July 20, 2006 (INTEX survey flight aboard DC-8, thanks to E. Browell DIAL lidar group.)

Above: along-track charts of ozone, 0–4 km along the flight track shown on the right, with various nearby cities indicated. Some vertical smoothing is applied to DIAL products.

Right Center: ozone contoured above flight track, bottom curved section was outward bound from Portsmouth, NH, is below, nearly straight return flight is above. Note: although not shown, sampling time during the day also changes from morning to late afternoon.



Rightmost Panel: Aerosol backscatter (visible) along the same path. Some correlation with ozone may be evident near 3–4 km at southernmost sampling area and on the return nearing the NH airport.

Why Study Autocorrelation in the Horizontal, and the Vertical?

Some reasons to estimate and understand the autocorrelation and autocovariance of ozone:

- To define the horizontal scales required for estimates of lower-tropospheric ozone for several purposes, especially
  - best estimates of ozone when measured ozone not available
  - documentation of possible exent of significant (health-endangering) ozone features
  - nowcasts of ozone for fast-response actions (sheltering, anti-inflamatory precautions for sensitive individuals)
  - sufficienrt (best-guess) data for forecasting for next-day: initial/boundary conditions
  - sufficient of data for constraints for model-improvement cycles over years
- Potentially valuable for retrieval algorithms
- Valuable for assimilation: definition of covariance matrices

Reasons to estimate and understand the vertical autocorrelation and autocovariance of ozone:

- Valuable for proper assimilation of vertical column ozone.
- Vital in order to understand how useful thick-partial-column (0 to 4 km) and upper-layer partial column (2 to 5 km) estimates of ozone may inform us partially about boundary layer and surface ozone.
- Useful in interpreting validation of retrieval methods (e.g. UV methods) for physical vs geophysical correlation.

Autocorrelation in the Horizontal

Autocorrelation as a function of horizontal distance calculated for (a) an upper layer (1.3–2.5km, "low-cloud and regional transport" layer) and (b) a lower layer (0–1.3 km "daily-highest mixed layer").

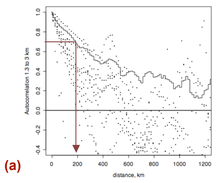
Solid lines indicate the average of all flights, individual dotted lines indicate individual flights during INTEX-NA.

The estimates were made for flights in an area slightly larger than shown for the map of the case study, south and west of Boston and north and east of Houston. DIAL ozone values were averaged within each layer, and all pairs of points along any trajectory were chosen. Only pairs sampled within 3 hours of each other were kept. Autocorrelation and autocovariance functions were calculated based on these pairs.

Autocorrelation functions are easier to visualize. When autotocorrelation drops to ~ 0.72, only half the variance can be explained by the predictor. (For practical purposes, the fact the the variance depends on the summed-square differences may exagerate the importance of a few wild cases.)

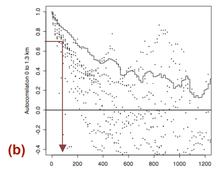
Autocovariance functions are preferred for numerical calculations, and are also provided.)

"low-cloud and regional transport" layer



(a)

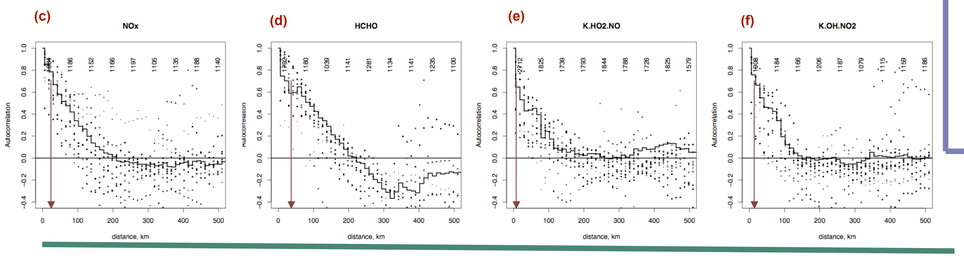
"daily-highest mixed layer"



(b)

Autocorrelation as a function of horizontal distance calculated for species closely connected to ozone production and destruction chemistry, I.e, (c) HCHO (formaldehyde) and (d) NO_x (= NO_2+NO). Autocorrelations were calculated for lower-tropospheric layers; only the 0–1.3 km layer is shown. On-board measurements due to the NCAR Fried group and the PSU Brune groups were used. Note that NO_x has narrow features, becoming uncorrelated rapidly. HCHO has both small-scale and more regional autocorrelations, due to e.g., cities and regional isoprene emission.

Two gross chemical rates important to photochemical production of O_3 in the lower atmosphere are (e) the principal chemical production term for ozone (actually, odd oxygen = O_3 + NO_2 + much smaller terms) HO_2 + NO → OH + NO_2 and (f) the prime reaction removing nitrogen oxides from the system and also substantially a sink of odd oxygen from the system (OH + NO_2 → HNO_3). Both of these rates are very highly variable in space, as calculations show. The rates of the reactions were evaluated using NO and NO_2 and the Brune-group HO_2 and OH, and averaged similarly to the HCHO and NO_x.



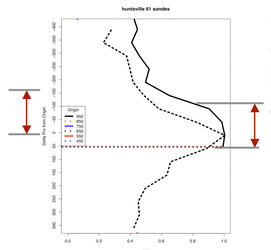
Autocorrelation in the Vertical

Vertical autocorrelation was estimated from the WODC ozonesondes database, considerably augmented by sondes taken by NASA , NOAA and Canadian agencies during recent summertime intensive periods and available through the IONS database. Only North American analyses are shown here. Most stations do not have even sampling, and tend to emphasize the months March–September. The sondes provide a wealth of information illustrated in the busy figures around the North American map shown.

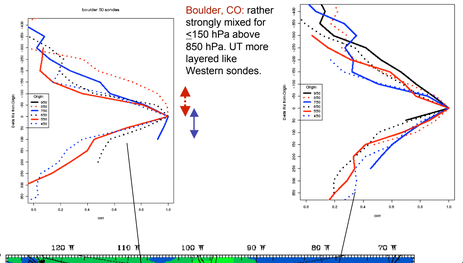
Guide: Black lines in the simplified large guide figure to the right provide a convenient introduction to all lines in all plots. The solid black line indicates the spatial autocorrelation between the 950 hPa layer and all other layers in the lower troposphere. The ozonesonde station is Huntsville, Alabama. To create estimates usefully representing transport effects, ozone concentrations were averaged for 25-hPa layers in the lower troposphere, and correlations and covariances were calculated for layers above and below.

The vertical scale indicates difference in pressure level (Δp), the horizontal scale the autocorrelation, maximizing of course at 1.0 for Δp = 0. The solid black line indicates correlation for the single layer below (around 1000 hPa) and many layers above. The dotted black line provides similar autocorrelation estimates centered around 650 hPa. (Note that the value for Δp = −300 on the dotted line equals the value for Δp = +300 on the solid line.) Huntsville shows strong correlation of 950 hPa (typical mid-PBL) ozone up to 850 hPa and then a sudden drop. Autocorrelation at Huntsville drops rapidly with for the evaluations made around 650 hPa compared to the 950 PBL. An interpretation is that the mid-troposphere exhibits less daily mixing than the PBL, and oftentimes more shearing that narrows lamina, a modified version of a phenomenon even more common in the lower stratosphere. Cloud detrainment effects may increase or decrease the stratification.

The autocorrelation plots show a variety of patterns significant to the estimation of PBL and hence to surface O_3. ***Please follow the brief comments starting at Houston, to the lower right of the map, then Huntsville and so counterclockwise around the map.***

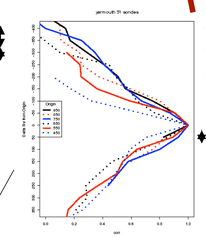


Example: Huntsville, AL: Correlation from 1000 hPa for the lowest, 950 hPa level shown on right, Δp scale from 0 shown on left. See below SE region of map.



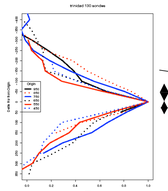
Boulder, CO: rather strongly mixed for ≤150 hPa above 850 hPa. UT more layered like Western sondes.

Egbert, ON,rural, N of Toronto : Poor correlation in lowest 0.5–1.5 km. Reason unknonwn

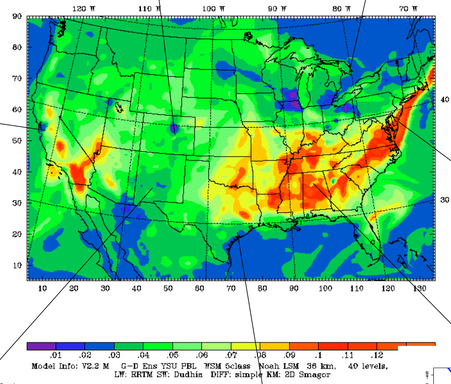


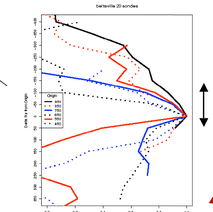
Yarmouth, NS: mixing to 900–850 mb (≤ 1.5 km).. Mid-tropos. correlations broader.

Shallow marine layer with pollution above



Trinidad Head, CA: Strong wind shear and inversion in the region above the lowest 500 m. Very narrow autocorrelation scales reflect long-distance transport, subsidence, shear, and consequent lamination in many sonde profiles. Lowermost troposphere not typical of California. See Houston lower tropsp heric profiles for closest approxmation for inland California. A limited number of Holtsville, CA profiles suggest free troposphere only weakly connected to ~500–800 m marine-influenced PBL.

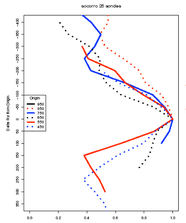


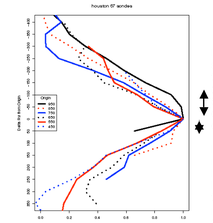


Beltsville, MD daytime PBL mixing to 850 hPa (~1.5 km) …continental convection …Warning: limited number, representativen ess of sondes

End Here

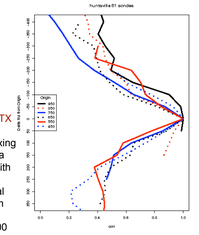
Soccorro, NM Main PBL shows mixing to 800 hPa or above (~2–2.5 km) by dry convective mixing: Warning: limited number of sondes





Houston, TX Main PBL shows mixing to 800 hPa (~2 km) with SE US continental convection

Lowest 500 m shows likely $NO+O_3$ titration, perhaps sea breeze

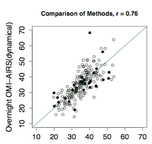


Huntsville, AL

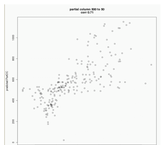
Deep daytime PBL produces mixing to 850 hPa (~1.5 km) with SE US continental convection: *BEST SITUATION FOR SENSING O_3 REMOTELY*

Start Here

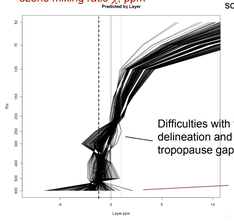
OMI–MLS estimate



New θ-χ estimate in progrss



Vertical profile of first-estimate ozone mixing ratio χ, ppm



Difficulties with tropopause delineation and the tropopause gap region

Estimations always place significant negative estimates at the lowest AIRS thermal level included. Why?

Work in Progress : Retrieving vertical information from UV total column ozone and thermal/dynamical structure of the atmosphere:

χ – θ relationships and the "curse of vertical correlation".

At Aura Science-Team meetings and at a TROPI meeting in DeBilt, we showed partial success in reproducing "tropospheric ozone columns" Ω_{200} (actually partial ozone column below 200 hPa) with approximately the skill of the Schoeberl-Zemke OMI–MLS subtraction technique. Estimates using a sophisticated statistical exploratory technique (projection pursuit regression) employed OMI total ozone and AIRS *thermal structure* but not MLS that correlated with $\Omega_{200,sonde}$ (based on summing ozonesonde layers) with equal or better skill to the OMI–MLS technique. The technique shared many characteristics of the Li et al. (2002) "GFS method" for estimating total column ozone Ω_0, but was self-updating month by month, and so promised better eventual skill.

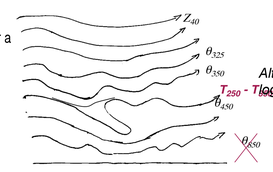
There were difficulties and opportunities remaining. Ozonesondes were limited in number and spatial distribution, and imprecise. Additionally the OMI–MLS technique was itself not extremely precise, and so the whole process brought in uncertainties tied to other techniques. There were also opportunities. it is very desirable to estimate a quantities like Ω_{350}, avoiding the frequent and severe stratospheric contamination found in Ω_{200}, or even better, estimating Ω_{500}, approaching an estimate for a lower-tropospheric ozone partial column.

We sought to make a statistical estimate based firmly on underlying meteorology of the stratosphere and upper troposphere.

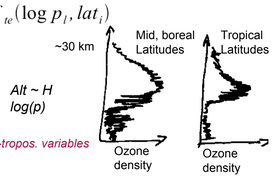
We may write that we are seeking an estimate of total column ozone based on this equation:

$$\widehat{\Omega_0} = \sum_{n=Sfc}^{top} \chi_{l,i}\, p_l(\Delta \log p)_l \sim \sum_{n=Sfc}^{top} \alpha\, \theta_{l,i}\, f_{te}(\log p_l, lat_i, t)\, p_l(\Delta \log p)_l$$

i.e, suggesting that the appropriately weighted sum of ozone mixing ration in the vertical was proportional to an identically weighted sum of potential temperature, θ, with appropriate proportionalities represented by α and f_{te}. This was based on the observation that in the stratosphere and indeed the uppermost troposphere, that both ozone mixing ratio and potential temperature changed slowly, that is, approximately conserved just enough that statistical correlation techiniques could unwind their proportionalities. It is important to note that the relationship is a function of vertical position (log *p*), latitude, and has a slow, often linear trend with time for periods of a month or so. Estimates that were made approximately monthly should allow for corrections such as chemical change and very large-scale motions. If so, we may expect that there must also be a detailed equivalence,



$$\widehat{(\chi_{l,i})} = \alpha\ \theta_{l,i}\, f_{te}(\log p_l, lat_i)$$



Results based on the "physical" method are instructive and suggestive if incomplete. Total ozone column is reproduced to approximately r = 0.87, or r^2 = 0.76, not quite as good as the less physically based statistical estimates. Ozone above 500 hPa, Ω_{500} , is reproduced to only r = 0.71, although Ω_{200} reaches r = 0.86, and Ω_{350} is close. The correlation plot shows Ω_{500}.

A plot of estimated ozone mixing ratio vs log pressure demonstrates the difficulty: meteorological correlation of the troposphere and the stratosphere. The upper part of the figure (the middle stratosphere) shows reasonable values, 6 to 10 ppm. The large portion of ozone in the lower stratosphere is close to 1 ppm; the technique inherently involves some uncertainty in the 0 ppm baseline since the regression includes a constant term. Our models consistently place large negative values at the lowermost level of the troposphere for which AIRS thermal structure is included, and these values must be included for the total column to be estimated correctly.

We have three tentative conclusions:

(a) Stratosphere-troposphere correlations must be better understood in order for a "physical" method to work.

(b) Estimates must more wisely include a sharp tropopause, probably using the AIRS temperature minimum.

(c) Uncertainties in estimating ozone above 30–50 hPa must be resolved in order for a small lower tropospheric partial column to be estimated.

Conclusions:

Geophysical correlation of ozone in the horizontal and vertical, i.e., responding to physical and chemical processes is always with us.

Strong correlation may help by easing our horizontal and vertical sensing requirements. Regional variation is the rule: Rocky Moutain Southwest and Southeastern US have the deepest mixing of ozone and thus easiest remote sensing problems. Significant horizontal autocorrelation of indicators of ozone chemistry is as small as 10–20 km.

Correlations may complicate our understanding of retrieval physics when geophysical, not physical, factors contribute to our validation measures.

Covariance estimates are preferred for numerical calculations, but correlations are useful to contribute basic understanding.

GEO-CAPE's science definition task has a good start in defining horizontal and vertical correlations of ozone in the lower troposphere, but time auto-correlation estimates are spotty at best. Multi-day correlation estimates may be tractable.

Adequate accounting for time-of-day and seasonal effects in our current ozonesonde and aircraft dataset is not acceptable for lower tropospheric questions. A larger measurement program, assisted by much more careful analysis of existing datasets is indicated.