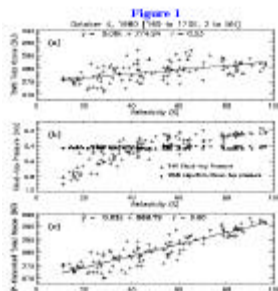


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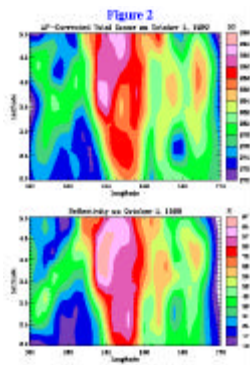
Motivation

- Often, TOMS ozone over a convective region ($R \geq 80\%$) reports more ozone than over nearby clear regions.
- Large difference between the TOMS-assumed and THIR cloud-top pressure.
- Correct the added climatological ozone using THIR (defined as ΔP correction), even though the TOMS climatology might not be right.
- Investigate the sources of cloudy ozone excess.



ΔP correction

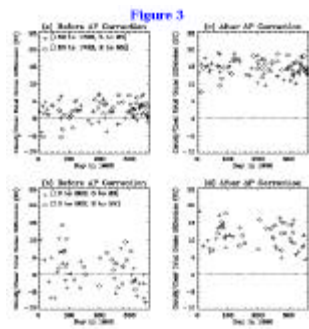
- Three errors induced by cloud height: radiation interpolation errors due to the forward calculation of radiances from look-up table, ozone retrieval errors in the added ozone below clouds, and ozone retrieve errors above clouds.
- Correcting these errors based on TOMS algorithm. The ΔP error is dominated by ozone retrieval error below cloud, and the other two are usually within 2 DU for high reflectivity clouds.
- Figure 1 show a) uncorrected total-ozone/reflectivity, b) measured (THIR) and algorithm cloud top pressure, and c) ΔP corrected total ozone.
- Figure 2 is a ΔP -corrected example of total ozone and Reflectivity, 15 DU more ozone over cloudy areas.



1

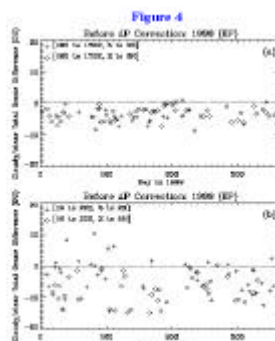
Science question

- Is this occurrence of more ozone over cloudy areas frequent?
- Why is there more ozone over cloudy areas compared to neighboring clear areas?
 - Inappropriate climatology in the added ozone below clouds?
 - TOMS calibration errors?
 - Algorithm ozone retrieval errors?
 - Actual geophysical phenomena?
- How these ozone retrieval errors affect tropospheric derivation using the CCP and CCD methods?



Is this occurrence frequent?

- We analyzed two regions: (1) the western Pacific Ocean (WPO) (2) Equatorial Africa (EA).
- After the ΔP correction, always more ozone over cloudy areas in both regions in the N7 TOMS data in 1980 (Figure 3).
- On average, there is -15 DU more ozone over WPO and -11 DU more ozone over EA.
- In EP TOMS data, no THIR is available to correct ΔP error.
- Without ΔP correction, the cloudy/clear difference in EP TOMS is -6.7 DU smaller than in E7 TOMS (Figure 4).



2

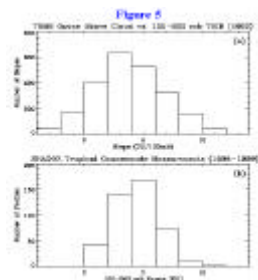
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Due to incorrect ozone climatology?

- Over WPO, the added ozone below clouds of ~26 DU is ~7 DU larger than SHADOZ measurements at Fiji and Java.
- Over EA, the added ozone below clouds of ~24 DU, is ~7 DU smaller than TRACE-A measurements at Brazzaville (about same location).
- Considering imperfect retrieval efficiency in the lower troposphere, the ΔP correction overestimates (underestimates) by ~5 DU over WPO (EA) on an annual average.
- Considering incorrect ozone climatology cannot fully account for the cloudy/clear difference over WPOA and exacerbate the difference over EA.

Chemical or dynamical effects?

- Time periods associated with clouds are too short for chemical production to explain the cloudy ozone excess.
- High convective complex penetrating the stratosphere tends to reduce the stratospheric ozone [Mattingly, 1977; Poulida et al., 1996]. However, the histogram of derived 100-200mb ozone above clouds is consistent with that of measured SHADOZ ozone (Figure 5).
- Chemical and dynamical effects cannot account for the cloudy ozone excess.



Calibration Errors?

- The cloud/clear total ozone difference EP TOMS data is expected to be positive, but is smaller by 6-7 DU than that in N7 TOMS data. The assumption of same stratospheric ozone above clouds is more valid in EP TOMS data.
- We ascribe the N7/EP bias of 6-7 DU to the nonlinear calibration error in N7 TOMS data.

Summary of the sources of cloudy excess ozone

Table 1

Region	ΔO_3 before ΔP correction	ΔO_3 after ΔP correction	Calibration error*	Incorrect climatology	Unknown sources
Western Pacific Ocean	2(3)	15(3)	-6	-5**	-4
Equatorial Africa	1(5)	11(3)	-7	-5***	-9

** The annual average tropospheric ozone is assumed to be 22 DU in the western Pacific Ocean.
 *** The annual average tropospheric ozone is assumed to be 40 DU in the Equatorial Africa.

3

Ozone absorption enhancement in the clouds

- The unknown sources in Table 1 result mainly from the treatment of clouds as opaque Lambertian surfaces that neglects in-cloud ozone absorption.
- Although the backscattered photons penetrates limitedly high-reflectivity clouds, the multiple scattering enhances the ozone absorption where photons reach. Theoretical calculation shows that TOMS can see most of the ozone in clouds at nadir view even for optically thick clouds.
- The estimated enhanced ozone is typically 5-13 DU over EA and 1-7 DU over WPO.

Comments on the CCD and CCP methods to derive tropospheric ozone

- A basic assumption in CCD and CCP: the same cloudy/clear stratospheric ozone (ozone above cloud-top). However, the cloudy ozone excess due to calibration errors (N7 TOMS only) and unknown sources together tends to underestimate the derived tropospheric ozone in N7 TOMS data.
- The clouds with minimum ozone above clouds used in the CCP and CCD methods are clouds with less in-cloud ozone or has extremely low values within in a period, cancelling the cloudy ozone excess caused by other retrieval errors.
- Although the assumption of the same cloudy/clear stratospheric ozone is more correct during EP period, using minimum ozone above clouds overestimates the tropospheric ozone.

Conclusions

- The incorrect cloud heights lead to three errors: radiation interpolation errors, errors in the added ozone below clouds, and errors in the retrieved ozone above clouds. Errors in the total ozone for high-reflectivity clouds are dominated by the mainly negative ozone retrieval errors below clouds.
- Correcting cloud height errors leads to consistent excess ozone over cloudy areas of 10-15 DU. The overall error in the total ozone is small due to the cancellation of cloudy excess ozone and negative ozone retrieval below clouds.
- We investigated the sources for cloudy ozone excess. After accounting for the inappropriate ozone climatology of ~5 DU over the Pacific Ocean and ~5 DU over Equatorial Africa, and possible calibration errors of ~6 DU in N7 TOMS, we still observe about ~4 DU more ozone over high cloudy areas in the Western Pacific Ocean and ~9 DU over Equatorial Africa. The dynamical and chemical effects are estimated to be small. This remaining ozone excess are caused mainly by the treatment of clouds as Lambertian surfaces that neglects the in-cloud ozone absorption.
- The cloudy ozone excess resulting from calibration errors and ozone absorption enhancement in the clouds tends to underestimate the derived tropospheric ozone. However, using minimum ozone above clouds cancels or partly cancels the cloudy ozone excess.

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