Comments on Policy Relevant Background Ozone

As Discussed In EPA's Draft Integrated Science Assessment for Ozone and Related Photochemical Oxidants

May 5, 2011

by

George T. Wolff, Ph.D.
Principal Scientist
Air Improvement Resource, Inc.
Novi, MI 48734

I. Introduction

In the March 2011 draft "Integrated Science Assessment for Ozone and Related Photochemical Oxidants"[1] EPA is proposing to use the same methodology and values for background ozone as it did in the previous review that led EPA to promulgate in 2008 an 8-hour standard of 75 ppb. The choice of the ozone background values has serious implications for both the amount of perceived risk as well as the degree of precursor controls needed to demonstrate attainment with any chosen ozone National Ambient Air Quality Standard (NAAQS). For reasons discussed below, EPA has understated background ozone levels. However, before I discuss the basis for this conclusion and the implications of this understatement, some discussion of the history of EPA's handling of background ozone is in order.

A. Historical Background

In the 1997 review of the ozone (NAAQS), EPA based the ozone background values on measurements that were designed to determine the ozone concentrations in air masses entering the continental U.S. In their 1996 Staff Paper[2] on page 18, they state:

> Based on a review of the available literature, it is obvious that "natural" O_3 background is a multidimensional and complex concept. Background O_3 concentrations vary by geographic location altitude and season. For the purposes of this document, background ozone is defined as the ozone concentrations that would be observed in the U.S. in the absence of anthropogenic or biogenic emissions of VOCs and NO_x in North America.

[1] U.S. EPA (2011), *Integrated Science Assessment for Ozone and Related Photochemical Oxidants,"* EPA/600/R-10/076A, March 2011.
[2] U.S. EPA (1996), *Review of the National Ambient Air Quality Standards for Ozone Assessment of Scientific and Technical Information OAQPS Staff Paper,* EPA-452/R-96-007, June 1996.

> During the summertime O_3 season in the U.S., daily 1-hr. maximum background is typically between 0.03 to 0.05 ppm. Part of this background is due to the long-range transport of anthropogenic or biogenic emissions.

EPA states that they arrived at this range after a synthesis of the literature, but they relied most heavily on the papers by Altshuller,[3] Kelly et al.,[4,5] and Lefohn and Foley.[6]

B. Policy Relevant Background Ozone

In the subsequent review of the ozone NAAQS, EPA introduced a new term, Policy Relevant Background (PRB), which they define as "the distribution of O_3 concentrations that would be observed in the U.S. in the absence of anthropogenic (man-made) emissions of precursor emissions (e.g., VOC, NOx, and CO) in the U.S., Canada, and Mexico."[7] The differences between this definition and the definition previously used for background are that the current definition refers to a distribution of concentrations rather than a range and it explicitly excluded anthropogenic precursor emissions in Mexico and Canada. EPA does not include emissions from our two neighboring countries because the Agency unrealistically assumes that such emissions can be fixed by international agreements. EPA further states: " As a result of long-range transport of O_3 and its precursors from anthropogenic sources within North America, estimates of PRB O_3 concentrations cannot be derived solely from measurements of O_3, and must be based on modeling." This represented a major departure from the way background was estimated in the past which was based on data. Specifically, EPA used the global photochemical transport model GEOS-CHEM.[8]

As I will show below, based on measurements and new modeling, the estimates of PRB EPA used in the last review were unrealistically low. The consequences of underestimating the background are significant. First, it results in a significant overestimation in EPA's risk estimates. For example, Smith (2011)[9] has shown that 92% to 100% of the mortality risk from ozone disappears if PRB is increased to 40 ppb. Second, it results in an underestimation of the

---

[3] Altshuller, A.P. (1986), "Review paper: the role of nitrogen oxides in nonurban ozone formation in the planetary boundary layer over N. America, W. Europe and adjacent areas of ocean, *Atmos.Environ.*, 20:245-268.
[4] Kelly, N.A., Wolff, G.T. and Ferman, M.A. (1982), "Background pollutant measurements in air masses affecting the eastern half of the United States- I. air masses arriving from the northwest, " *Atmos.Environ.*, 16:1077-1088.
[5] Kelly, N.A., Wolff, G.T. and Ferman, M.A. (1984), "Sources and sinks of ozone in rural areas," *Atmos.Environ.*, 18:1251-1266.
[6] Lefohn, A.S. and Foley, J.K. (1992) "NCLAN results and their application to the standard-setting process," *J.Air & Waste Mgt. Assoc.* 42: 1046-1052.
[7] U.S. EPA (2007), *Review of the National Ambient Air Quality Standards for Ozone: Policy Assessment of Scientific and Technical Information OAQPS Staff Paper*,EPA-452/R-07-003, January 2007.
[8] Fiore, A., Jacob, D.J., Liu, H., Yantosca, R.M. Farlie, T.D. and Li, Q. (2003), "Variability in surface ozone background over the United States: Implications for air quality policy", *J. Geophys. Res.*, doi:10, 1029/2003JD003855.
[9] http://yosemite.epa.gov/sab/sabproduct.nsf/7401CA2E8A7B7E6C85257831006377AB/$File/API+-+Smith+Comments.pdf.

degree of emission controls needed to achieve the NAAQS. Further by not including Mexican and Canadian anthropogenic precursor emissions, it is penalizing the states which must come up with additional control measures to compensate for the contributions of ozone and precursors from these two countries. It is not logical and it is inconsistent to include worldwide anthropogenic emissions but exclude Canada and Mexico in their definition of PBR. In my opinion, PRB must be redefined to include Mexican and Canadian emissions in PRB because EPA's claim that they merely need an international agreement to fix Canadian and Mexican emissions is not realistic. In addition, because of the way EPA defines PRB, a model performance evaluation of the modeled PRBs is impossible.

II. EPA PRB Estimates

For the last review, the estimates of PRB were based on runs of the GEOS-CHEM model applied to the 2001 warm season (April to November). The model output consisted of hourly gridded ozone estimates in 2^{o} latitude and 2.5^{o} longitude grid cells for the entire U.S. These outputs were used to generate monthly average diurnal profiles for each cell for April through October. Each mean monthly profile was then used in the risk assessment by applying the nearest grid cell's monthly profile to each of the twelve urban areas used in the risk assessment.

The mean profiles for each urban area were plotted in Appendix 2a of the 2007 Staff Paper.[10] An example of these profiles (for Detroit) is reproduced here as Figure 1. As seen from the figure, there is not much hourly variation in the estimated PRB profile. This is the case for the other eleven urban areas as well. In general, April has the highest concentrations and August has the lowest summer concentrations. A broad afternoon/evening peak is observed with relatively constant concentrations from 1300 to 2300. In Table 1, I have estimated from these graphs the afternoon maximum concentration of the PRB for April and August for each urban area. In April, the PRBs range from a low of 27 ppb at a number of Eastern and Midwest cities to a high of 34 ppb in Sacramento while in August they range from a low in Sacramento of 15 ppb to a high of 28 ppb in Houston. In the following sections, these will be compared to actual measurements.

III. A Conceptual Model for PRB in the Continental U.S.

The proposed conceptual model consists of two basic components: 1) background ozone that is transported into the U.S. from any source outside of the country and 2) natural ozone that is formed from either natural sources of precursors in the U.S. or ozone of stratospheric or free tropospheric origin that reaches the surface within the continental U.S. The first component can be both modeled and measured, and the measurements can serve to evaluate the performance of the models including the estimate of PRB as I have redefined it above. Examples of how this

[10] U.S. EPA (2007), *Review of the National Ambient Air Quality Standards for Ozone: Policy Assessment of Scientific and Technical Information OAQPS Staff Paper Appendices to OAQPS Staff Paper,* EPA-452/R-07-003, January 2007.

can be done will follow, but it requires that PRB include all sources out of the U.S. including Canada and Mexico. The second component, which will be important in the interior of the U.S and in the Eastern U.S., needs to be obtained from models. Modeling issues will be addressed in section IV.

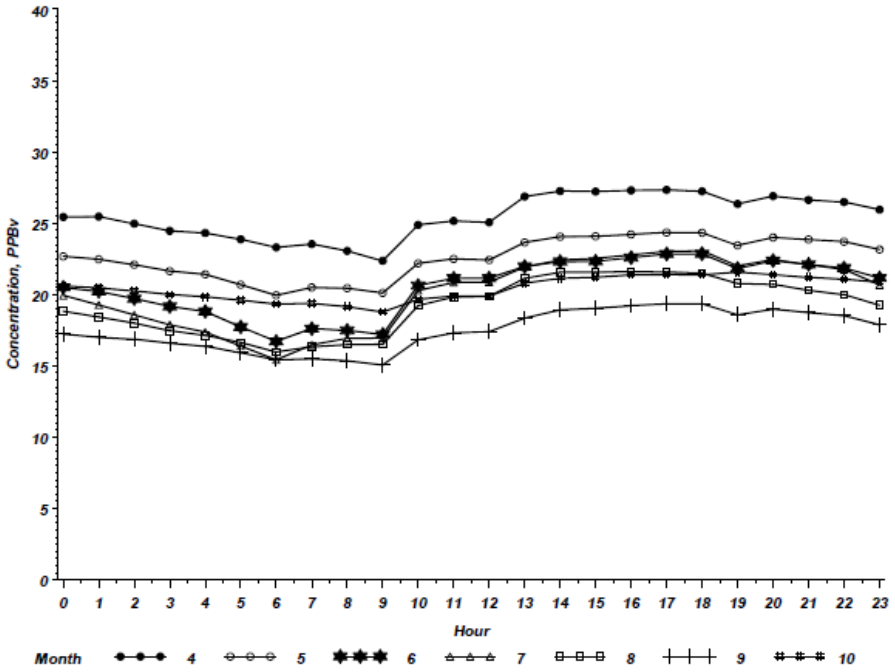

Figure 1: Diurnal Policy Relevant Background (PRB) ozone patterns in the Detroit CSA.[10]

Because of its latitude on the globe, the continental U.S. is in the zone of prevailing westerly winds. That means that the air flow is, on average, from west to east. However, the zone of prevailing westerlies is also where transitory weather systems (high and low pressure areas) continually travel through leading to the alternation of cold air from the north and warm air from the south. As a result, four major types of air masses dominate the U.S. and each one is associated with different levels of background ozone because of where they originate. The four types of air masses are: maritime polar from the Pacific Ocean, continental polar (or arctic) from Canada, continental tropic from Mexico, and maritime tropic from the Gulf of Mexico. The background levels of ozone should be examined for each one of these air masses separately.

CSA	April	August
Atlanta, GA	28 ppb	22 ppb
Boston, MA	30	20
Chicago, IL	32	23
Cleveland, OH	27	22
Detroit, MI	27	22
Houston, TX	31	28
Los Angeles, CA	32	20
New York, NY	27	22
Philadelphia, PA	28	23
Sacramento, CA	34	15
St. Louis, MO	28	26
Washington, DC	27	23

Table 1: Maximum afternoon ozone PRB estimates by month for GEOS-Chem.[10]

A. Maritime Polar

The background concentration of ozone in maritime polar air entering the U.S. from the Pacific Ocean has been the subject of a few recent papers. Oltmans et al., (2008)[11] analyzed a 6-year continuous ozone data base from a coastal site, Trinidad Head, in northern California. They observed frequent Spring excursions of hourly data over 50 ppb with a overall springtime mean of 40 ppb. August typically had the lowest concentrations of the April to August time frame. The yearly maxima in August ranged from 38 to 44 ppb, while the median values ranged from 22 to 27 ppb. They also compared the data from Trinidad Head to data collected at Channel Islands National Park which is about 40 miles off the coast of Southern California. They found a similar seasonal pattern for the April through August time period except that the concentrations were about 5 ppb higher at Channel Islands National Park than at Trinidad Head.

Parrish et al., (2009)[12] also analyzed data from Trinidad Head but set up strict meteorological criteria to insure that they selected data from air parcels that were of marine origin with no contamination from local continental sources. They concluded that in April of 2002, the average ozone concentration in air of marine origin was 42.0 ppb. This very similar to the average Oltmans et al. found using all the data. Applying the same criteria to select air parcels of only marine origin to seven other near coastal sites in California and Washington, they detected an increasing trend in ozone of 0.46 ppb/year over the period from 1990 to 2007.

[11] Oltmans, S.J., Lefohn, A.S., Harris, J.M. and Shadwick, D.S. (2008), "Background ozone levels of air entering the west coast on the US and assessment of longer-term changes," *Atmos. Environ.* 42:6020-6038.
[12] Parrish, D.D., Miller, D.B. and Goldstein, A.H. (2009) "Increasing ozone in marine boundary layer inflow at the west coast of North America and Europe," *Atmos. Chem. Phys.* 9:1303-1323.

Using an ensemble of aircraft measurements over the northwest Pacific and surface and ozonesonde data from Trinidad Head and Richland, WA collected as part of the NASA/INTEX-B aircraft campaign from April 17 to May 15, 2006, Zhang et al. (2008)[13] assembled an extensive set of vertical ozone profiles. DC-8 aircraft profiles of ozone were reported from 0.5 km to 11.5 km. A C-130 aircraft reported profiles from 0.5 km to 6.6 km. The ozonesondes at the two sites obtained ozone profiles from 0.5 km to 11.5 km. The lowest level (0.5 km) data should be most comparable to the surface marine boundary layer. The means (and the range) of the measurements at the 0.5 km level were: DC-8 42 ppb (40-55), C-130 50 ppb (45-55), Trinidad Head 41 ppb (35-50), and Richland 54 ppb (45-59). These are on the order of the background values reported by Parrish et al and Oltmans et al.

Based on the considerable amount of data contained in these three papers, reasonable average PRB ozone values for maritime Pacific air masses for April are in the 40 to 50 ppb range and for August they are in the 22 to 32 range. This compares to EPA's April PRB for Los Angeles of 32 ppb and Sacramento of 34 ppb and for August 20 in Los Angeles and 15 in Sacramento. It should be noted however, that higher background values have been observed on individual days as is evidenced by the monthly maxima reported above by Oltmans et al. In addition, Oltmans et al. (2010)[14] report even higher concentrations in April 2008 when the west coast was affected by emissions from Eurasian biomass burning.

It should be also noted that all of the vertical ozone profiles presented by Zhang et al show a slight but gradual increase in ozone concentrations with height. At 2 km, the mean concentration over Trinidad Head is 55 ppb and at 4 km it is 60 ppb. As these air masses travel eastward from the coast, they will encounter mountains that will induce mixing and the air parcels containing these higher concentrations will mix to the surface. Jaffe (2011)[15] found evidence that the higher concentrations in the free troposphere control the ozone concentration at 11 rural CASNET sites in the western U.S. Consequently the higher values at the higher altitudes over Trinidad Head are likely better measures of PRB for the inland Western U.S.

B. Other Air Mass Types

Unfortunately the other air mass types entering the U.S. have not received the same degree of attention that the Pacific air masses have received so we cannot rely on existing studies to determine the PRB of these air masses. In fact, even if the same type of measurements were made along the US-Canadian border and US-Mexican border, they would not fit EPA's definition of PRB because they would contain influences of Canadian or Mexican emissions,

[13] Zhang et al. (2008), "Transpacific transport of ozone pollution and the effect of recent Asian emission increases on air quality in North America: an integrated analysis using satellite, aircraft, ozonesonde, and surface observations," *Atmos. Chem. Phys.* 8:6117-6136.
[14] Oltmans, S.J. et al. (2010) "Enhanced ozone over western North America from biomass burning in Eurasia during April 2008 as seen in surface observations," *Atmos. Environ.*44:4497-4509.
[15] Jaffe, D. (2011) "Relationship between surface and free tropospheric ozone in the Western U.S.," *Environ. Sci.Technol.* 45;432-438.

respectively. That is why EPA's definition of PRB is unrealistic and unworkable. It is unrealistic to assume that all EPA merely needs is an international agreement because they have no authority over Mexican and Canadian emissions. It is unworkable because states simply cannot ignore the Canadian and Mexican contributions if they have any chance of developing a workable SIP that achieves compliance with the NAAQS. For these reasons, the definition of PRB must be changed to include all of the sources of ozone which are beyond the influence of the U.S. to control. Using that definition, it is then possible to estimate PRB from monitoring data collected near the U.S. Border.

It should be noted that there has been one attempt[16] to use a model to separate out the Mexican and Canadian contributions. However, I dismiss these results because they were done using unrealistic emission scenarios. To estimate the Mexican and Canadian contribution to the U.S., Wang et al. (2009) ran 3 emission scenarios: 1) all worldwide emissions included, 2) same as 1 but without North American anthropogenic emissions, and 3) same as 1 but without U.S. anthropogenic emissions. The difference between scenarios 1 and 2 gave them the North American contribution, the difference between 1 and 3 gave them the U.S. contribution and the difference between 2 and 3 gave them the Mexican and Canadian contributions. By shutting down U.S. emissions, they have completely altered the chemical environment over the U.S. in an unrealistic manner. The correct approach is to leave the U.S. emissions intact and eliminate the Canadian and Mexican emissions. Unfortunately, they did not run that scenario.

To properly estimate PRB as I have redefined it, a series of monitoring sites along the U.S. and Canadian, the U.S. and Mexican border and along the Gulf Coast would need to be established. The data from these sites could then be analyzed in a manner similar to the analyses performed on the Trinidad Head site to establish PRB. Although such networks do not exist today, there are two National Park sites that are part of the CASTNET network that have collected data that can provide a first cut at obtaining PRB information for continental polar and continental tropical air masses. The two sites are Voyageurs NP in Minnesota and Big Bend NP in Texas (see Figure 2). Unfortunately, there are no suitable sites on the Gulf Coast to make PRB estimates for maritime tropical air masses.

The hourly ozone and meteorological data from 1987-2011 were obtained from the EPA CASTNET web site.[17] A computer program was written to process this hourly ozone information from the two sites into daily maximum 8-hour averages. The computer program also computed the corresponding wind vector for each daily maximum 8-hour ozone period. In order for the calculations to be considered valid, 6 or more hours of ozone and wind observations had to be available in each 8-hour period. The wind direction vectors were used to identify 8-hour periods when the wind direction was persistently blowing into the U. S. The wind sectors

[16] Wang, H., Jacob, D.J., Le Sager, P., Streets, D. G., Park, R.J., Giulliland, A.B. and van Donkelaar, A. (2009) "Surface ozone background in the United States: Canadian and Mexican pollution influences," *Atmos. Environ.* 43:1310-1319.

[17] http://java.epa.gov/castnet/

chosen to reflect air being imported into the U.S. are: 123.75^{o} to 281.25^{o} at Big Bend and 303.75^{o} to 56.25^{o} at Voyageurs. The daily 8-hour maximum ozone readings for those days are summarized in Table 2. The mostly recent 3-year periods with valid data were chosen. The months for each year were grouped into two intervals: "April-May" and "June-July-August". The "Period" values represent the total number of days and the weighted average concentrations for each month group. Also shown are the maximum monthly values and the standard deviations.

Figure 2: Locations of CASTNET and National Park ozone monitoring sites. The two sites used in this report are identified in red.

The data in Table 2 indicate that PRB concentrations entering the U.S. from Mexico are on the order of 54 ppb in the spring months and 42 ppb in the summer months. The respective maxima are 70 ppb and 63 ppb. These are significantly higher than the 31 and 28 ppb PRB assigned to Houston by EPA (Table 1). For the air entering the U.S. from Canada, the data from Voyageurs indicate the PRB is on the order of 44 ppb in the spring and 35 ppb in the summer. The maxima are 64 ppb and 51 ppb in the spring and summer, respectively. This compares to the EPA assigned PRB to Chicago, Cleveland and Detroit of 27 to 32 in the spring and 22 to 23 in the summer.

CASTNET "Imported " Average Daily Maximum 8-Hour Ozone Concentrations (ppb)									
Site	Parameter	April-May				June-July-August			
Big Bend NP, TX	Year	2008	2009	2010	Period	2008	2009	2010	Period
	# of Days	32	24	31	87	37	51	44	132
	Maximum O_3	65	70	64	70	60	60	63	63
	Mean O_3	54.7	55.3	53.4	54.4	41.9	44.3	39.8	42.1
	Std. Dev.	6.0	5.3	5.8	5.8	8.8	7.5	10.5	9.1
Voyageurs NP, MN	Year	2004	2005	2006	Period	2004	2005	2006	Period
	Days	31	21	27	79	20	19	20	59
	Maximum O_3	64	55	58	64	51	46	50	51
	Mean O_3	44.6	44.4	42.4	44.2	33.2	33.7	38.0	35.0
	Std. Dev.	6.6	9.0	9.4	8.3	7.7	6.7	7.1	7.4

Table 2: Summary of maximum 8-hour ozone data on days when the air was flowing into the U.S.

IV. Modeling Results

A. Modeling Used In 2006 Review

As mentioned earlier, EPA derives their PRB values using the modeling results of Fiore et al. (2003).[8] To derive PRBs, Fiore el al**.** ran three emission scenarios using a protocol that differs from that used by Wang et al. (2009).[15] They ran 1) a base case using worldwide anthropogenic and natural emissions, 2) they repeat 1 but set North American anthropogenic emissions to zero, and 3) they repeat 1 but this time set global anthropogenic emissions to zero. They estimate the stratospheric contribution by tagging stratospheric ozone in the base case scenario. Scenario 1 gives them the total ozone concentrations. Scenario 2 gives them the North American background concentrations in the absence of North American anthropogenic emissions which EPA defines as PRB and scenario 3 gives them their estimate of natural background. Even though this protocol is somewhat different than Wang et al.'s, they make the same mistake. The contribution of natural sources and other PRB sources to North American cannot be realistically

assessed in the absence of U.S. anthropogenic emissions. To realistically estimate the contribution of PRB sources, the PRB sources should be shut down in the presence of U.S. sources.

Another issue is that GEOS-CHEM is known to underestimate the stratospheric contribution.[12,18] Zhang et al. (2008)[12] point out that "GEOS-CHEM underestimated observed ozone concentrations from the ITCT 2K2 campaign over California in April-May 2002 by up to 10 ppb due to its failure to reproduce high-ozone layers of stratospheric origin." Referring to the ozonesondes over Richland, Zhang et al state: "At Richland where stratospheric influences are more pronounced, the model is 10 ppb too low in the free troposphere." Langford et al. (2009)[19] shows that the stratospheric contribution to surface ozone is significant, and can lead to exceedances of the 2008 NAAQS ozone standard in a major U.S. metropolitan area. Langford et al. point out that the conclusions of Fiore et al. (2003) that EPA relies on are at odds with many other studies which have presented evidence for significant stratospheric contributions to surface ozone at both high altitude sites and near sea level. The assumption that EPA makes that stratospheric intrusions, if present, can be easily identified and handled as exceptional events has not been verified. Furthermore, Koumoutsaris, et al. (2008)[20] used the GEOS-CHEM model to evaluate the interannual variability of tropospheric ozone and CO. They report that the model reproduced the observed variability in CO well but that the model was less successful in the case of ozone. They attribute the poorer performance for ozone to the model's poor representation of stratospheric chemistry and dynamics.

A number of other recent papers have underscored the importance of adequately characterizing the importance of the stratospheric contribution to PRB. Tarasick et al. (2007),[21] Lightener et al. (2009),[22] Terao et al. (2008)[23] and, Trickl et al. (2009)[24] report additional evidence of stratospheric input. Tarasick et al. point out that while stratospheric intrusion events sometimes

[18] Yang, Q. et al. (2010) "A study of tropospheric ozone column enhancements over North America using satellite data and a global chemical transport model", *J. Geophys. Res.* doi:10.1029/2009JD012616.
[19] Langford, A.O., Aikin, K.C., Eubank, C.S. and Williams, E.J. (2009) "Stratospheric contribution to high surface ozone in Colorado during springtime," *Geophys. Res. Let.*, L12801, doi:10.1029/2009GL038367.
[20] Koumoutsaris, S., I. Bey, S. Generoso, and Thouret V. (2008) "Influence of El Nino–Southern Oscillation on the interannual variability of tropospheric ozone in the northern midlatitudes," *J. Geophys. Res.* 113, D19301, doi:10.1029/2007JD009753.
[21] Tarasick, D. W., et al. (2007) "Comparison of Canadian air quality forecast models with tropospheric ozone profile measurements above midlatitude North America during the IONS/ICARTT campaign: Evidence for stratospheric input," *J. Geophys. Res.* 112, D12S22, doi:10.1029/2006JD007782.
[22] Lightner, K. J., McMillan, W.W., McCann, K.J., Hoff R.M., Newchurch, M.J., Hintsa, E.J. and Barnet, C.D. (2009) "Detection of a tropospheric ozone anomaly using a newly developed ozone retrieval algorithm for an uplooking infrared interferometer," *J. Geophys. Res.* 114, D06304, doi:10.1029/2008JD010270.
[23] Terao, Y., Logan, J.A., Douglass, A.R. and Stolarski, R.S. (2008) "Contribution of stratospheric ozone to the interannual variability of tropospheric ozone in the northern extratropics," *J. Geophys. Res.* 113, D18309, doi:10.1029/2008JD009854.
[24] Trickl, T., Feldmann, H., Kanter, H.-J., Scheel, H.-E. Sprenger, M., Stohl, A. and Wernli. H. (2009) "Forecasted deep stratospheric intrusions over Central Europe: case studies and climatologies," *Atmos. Chem. Phys. Discuss.* 9, 2223–2288.

directly impact the surface, much more frequently, intrusion events are observed to reach the middle troposphere, where they mix in and contribute to background ozone. The Trickl et al. (2009) study of stratospheric-tropospheric exchange using lidar is important because it documents that the available models are, still, far from being quantitative in describing the intrusion phenomena. They conclude that high spatial resolution models are needed to capture the events and that the data they collected contradicts the frequently heard statement that stratospheric-tropospheric transport has a minor influence on the tropospheric ozone budget.

There are also several recent papers that indicate natural NO_x emissions were underestimated in the 2003 GEOS-CHEM modeling. Since nonurban atmospheres are deficient in NO_x, any underestimation of the natural NO_x emissions will result in an underestimation of the PRB ozone. Jaegle et al. (2005)[25] using satellite observations of NO_2 columns, found that soil NO_x emissions were twice as high as assumed in the GEOS-CHEM model. Cooper et al. (2007)[26] documents that lightning NO_x is a significant source of tropospheric ozone, particularly over the Southeastern U.S. This is important because, even though the ozone is formed in the upper troposphere, it will influence surface ozone at a later time. Hudman et al. (2007)[27] found that the GEOS-CHEM estimates of lightning NO_x emissions had to be increased by a factor of four to match recent upper tropospheric NO_x measurements. Finally, Singh et al. (2007)[28] evaluated four global models including GEOS-CHEM against tropospheric measurements of reactive nitrogen species. The authors show that the four models deviate from each other and from the observations at all altitudes, demonstrating that there is still substantial uncertainty in the modeling of ozone precursors that was ascribed to uncertainty in sources, meteorology, and mechanisms in the models. If the modeling of ozone precursors is substantially uncertain, the modeling of the ozone produced from those precursors is also substantially uncertain.

The GEOS-CHEM performance evaluation conducted by Fiore et al. was done for total ozone against CASTNET observations. Unfortunately, a reality check on the estimates of PRB cannot be made as EPA has currently defined PRB. If my definition was used, a reality check could be made by comparing the border measurements with modeling results. Since there were not contributions from Mexican or Canadian sources in the data presented above in the determination of PRB ozone in maritime polar air masses, those data can be used to perform a simple reality check. That was done in the comparison of the EPA assigned PRB for Los Angeles and Sacramento and the PRB estimated from the Trinidad Head which showed that the

[25] Jaegle, L., Steinberger, L., Martin R. and Chance, K. (2005) "Global partitioning of NOx sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning, and soil emissions," *Faraday Discussions*, 130: 407-423.
[26] Cooper, O.R., et al. (2007) "Evidence for a recurring eastern North America upper tropospheric ozone maximum during summer," *J. Geophys. Res.*112, D23304,doi:10.1029/2007JD008710.
[27] Hudman, R. C., et al. (2007) "Surface and lightning sources of nitrogen oxides over the United States: magnitudes, chemical evolution, and outflow," *J. Geophys. Res.* 112, D12S05, doi:10.1029/2006JD007912.
[28] Singh, H. B., et al. (2007) "Reactive nitrogen distribution and partitioning in the North American troposphere and lowermost stratosphere," *J. Geophys. Res.* 112, D12S04, doi:10.1029/2006JD007664.

model significantly under predicted the PRB. If it was under predicted on the West Coast, there are no reasons to expect better performance elsewhere in the U.S.

B. New Modeling Results

Since the original Fiore et al. results were published in 2003, GEOS-CHEM has undergone significant upgrades. The model now includes improved chemistry and physics and stratospheric ozone chemistry, updated meteorology and emissions and a much higher resolution grid of 0.5 x 0.6 km over North America. Recently, BP has initiated a study of PRB with Jacob and Zhang at Harvard using the updated GEOS-CHEM and 2006 meteorology. Preliminary results of these analyses have been presented to the CASAC Ozone Panel that is involved in the reconsideration of the 2008 ozone NAAQS during a February 18, 2011 teleconference.[29,30] Figure 3 is a reproduction of one of the figures presented by BP at that teleconference. This figure clearly shows the occurrence of PRB concentrations of 50 ppb or greater in the West during both spring and summer and in the Midwest in the spring. However, it should be kept in mind that this work is based on EPA's definition of PRB so Mexican and Canadian contributions are not counted. Nevertheless, this provides additional evidence that the PRBs generated by Fiore et al. (2003) and used by EPA in the last ozone review are significantly underestimated.

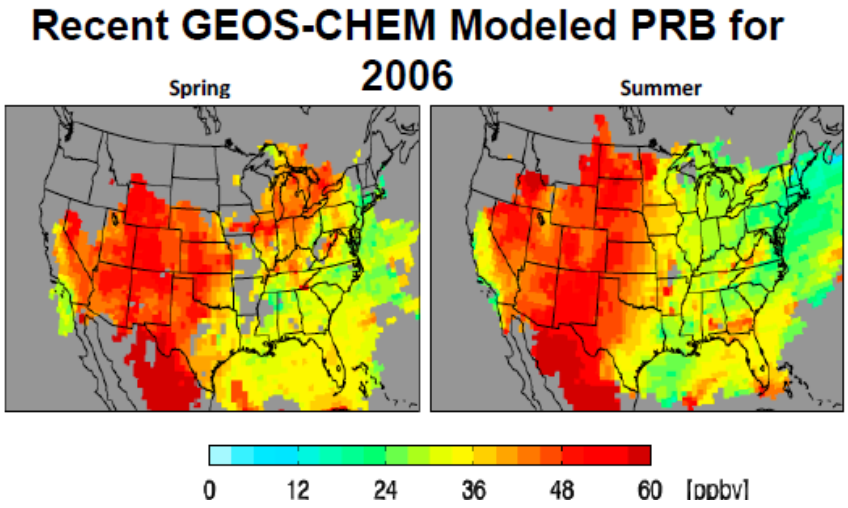

Figure 3: Recent GEOS-CHEM modeling results for 2006 showing EPA's PRB on days when the maximum 8-hour ozone values are ≥ 60 ppb.[29,30]

[29] http://yosemite.epa.gov/sab/sabproduct.nsf/F023F6EA5B41FDBF85257831006354AA/$File/BP+America+Production+Company+Comments+Submitted+by+N.+Downey.pdf
[30] http://yosemite.epa.gov/sab/sabproduct.nsf/9199C7054B4E5E8E85257834006CCCAF/$File/British+Petroleum+-+Doug+Blewit.pdf

V. Summary

EPA's definition of PRB is unrealistic and inconsistent. It is unrealistic in that it assumes EPA has control over Mexican and Canadian anthropogenic emissions and all they need to do is sign an international agreement with the two countries. It is inconsistent in that it includes all other anthropogenic emissions from around the world outside of the U.S. but excludes our two closest neighbors whose impacts are the greatest. It is only because of this unrealistic and inconsistent definition that PRB cannot be measured. If PRB was correctly defined to include all sources of ozone that the U.S. has no control over, PRB could be measured by measuring the flux of ozone in air parcel entering the U.S. EPA needs to change their definition of PRB to include Canadian and Mexican sources.

Since the 2005 deadline for inclusion of papers in the last ozone Criteria Document, numerous papers have been published that contain data showing concentrations of ozone at rural locations in the Western U.S. well above EPA's PRB range of 15 to 35 ppb in the absence of U.S. anthropogenic influences. Very recent modeling results with the GEOS-CHEM appear to be telling the same story. EPA must adjust their PRB upwards to be consistent with the latest monitoring and modeling results.

It is of paramount importance that EPA use the best information possible to develop PRB levels. As mentioned earlier, 92% to 100% of the estimated mortality risks disappear if a PRB level of 40 ppb is used. A more accurate depiction of the risks may warrant a different conclusion as to what level of the NAAQS is requisite to protect public health with an adequate margin of safety. In addition, a more accurate knowledge of PRB levels will allow the states to better evaluate the effectiveness of various emission control strategies.

Acknowledgements

Dennis Kahlbaum of AIR, Inc. created Figure 2 and Table 2 and wrote the programs and conducted the analysis to obtain the data in Table 2. Jon Heuss of AIR provided a review and valuable comments on this report.

This work was supported by the Utility Air Regulatory Group (UARG).