Global Tropospheric OH: Observational Constraints and Model Simulations

Contributed by **Yuhang Wang** (yhw@envsci.rutgers.edu), *Rutgers University, USA*

The abundance of tropospheric hydroxyl radicals (OH) largely defines the oxidizing capacity of the atmosphere. A healthy level of tropospheric OH keeps in check atmospheric concentrations of many potent greenhouse gases such as CH_4 and HCFCs and prevents large amounts of O_3-depleting halogenated hydrocarbons from reaching the stratosphere. Monitoring global tropospheric OH concentrations and understanding the controlling factors, anthropogenic and natural, are necessary to protect our living environment. I will briefly review recent scientific works related to these two global issues.

Introduction

A tutorial of simplified OH chemistry is in order. In the troposphere, OH production is largely initiated by photolysis of O_3 to $O(^1D)$, O atom in an energetically excited state, which reacts with H_2O,

(R1) $$O_3 + h\nu \rightarrow O(^1D) + O_2$$
(R2) $$O(^1D) + H_2O \rightarrow 2OH$$

The reaction sequence leads to primary production of OH since an OH radical (with an unpaired electron) is produced from a much more stable molecule. A large fraction of OH is recycled during oxidation of reduced compounds in the presence of nitrogen oxides ($NO_x = NO + NO_2$),

(R3) $$OH + CO + (O_2) \rightarrow HO_2 + CO_2$$
(R4) $$HO_2 + NO \rightarrow OH + NO_2$$
(R5) $$NO_2 + h\nu + (O_2) \rightarrow NO + O_3$$

In this sequence, a CO molecule is oxidized to CO_2 and an O_3 molecule is produced. Radicals of HO_x ($OH + HO_2$) and NO_x are conserved. Removal of OH is generally through the self reaction of its HO_x-family sibling HO_2,

(R6) $$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$$

H_2O_2 is removed from the atmosphere by deposition to surface and rainout.

Although highly simplified, the above scheme suggests that in order to understand tropospheric OH, it is necessary that we understand the distributions of O_3, NO_x, CO (and hydrocarbons), water vapor, solar radiance (for photolysis), and atmospheric deposition processes. Global simulations of OH, its precursors, and its proxies are generally carried out using 3-D models. In the following section, I will describe atmospheric observations that can be used to constrain tropospheric OH concentrations. In the discussion on global OH modeling, I will examine three categories, constrained respectively by emission sources and atmospheric transport, observed distributions of OH precursors, and observed distributions of OH proxies. The division among the categories is somewhat artificial but it serves the purpose of streamlining the discussion.

Constraints from observations

Dorn and Hofzumahaus [this issue] reviewed instruments employed for in situ measurement of OH near the surface or onboard aircraft and the comparison studies among these observations and model simulations. Surface observations are occasionally lower than model results at continental sites; heterogeneous loss of HO_2 to surfaces or unknown biogenic hydrocarbons, not measured at the sites, may account for the difference [e.g., McKeen et al., 1997]. Jaeglé [2000] reviewed upper tropospheric OH measurements in comparison to model simulations for recent aircraft campaigns; measurements generally support the current understanding of HO_x chemistry. The issues of additional HO_x sources not from reactions R1-2 are also discussed by Brune [this issue]. Although significant for OH concentrations in the upper troposphere, these issues bear far less significance on model predictions of the global mean OH concentration, largely dictated by concentrations in the lower and middle tropical troposphere [Spivakovsky et al., 2000]. The most comprehensive in situ measurements of regional OH concentrations to date were conducted onboard DC-8 and P-3 aircraft over the tropical Pacific during the Pacific Exploratory Mission (PEM)– Tropics B. Good agreement was found between observed and simulated OH concentrations [Tan et al., 2000; Mauldin et al., 2000], landing some confidence in model simulations of tropospheric OH.

Direct monitoring of tropospheric OH on a global scale is unattainable at present. The global mean OH concentration, however, can be estimated indirectly from the concentrations of proxy gases, such as CH_3CCl_3 [Singh, 1977; Lovelock, 1977], through budget calculations. Good proxy gases have known source distributions, relatively long atmospheric lifetimes, significant OH oxidation compared to other loss pathways, and long-term measurements. Spivakovsky et al. [2000] reviewed five commonly used proxies, CH_3CCl_3, HCFC-22, CH_2Cl_2, C_2Cl_4, and ^{14}CO. Observations of ^{14}CO turned out to be most difficult to interpret. The lifetime of ^{14}CO against OH oxidation is 1-2 months in the tropics and midlatitude summer. The short lifetime implies high sensitivities of observed concentrations to transport of cosmically generated ^{14}CO in the stratosphere and high latitudes to the lower latitude troposphere. The lifetimes of CH_2Cl_2 and C_2Cl_4 are also short in the tropics, 2 and 4 months, respectively. However, the industrial proxies have better known source strength and distribution. Particularly since their anthropogenic sources are mostly located at northern midlatitudes, the north-south gradients of their concentrations pose some constraints on the global and hemispheric means of OH concentrations. Observations of CH_2Cl_2 are more useful because of Cl-oxidation of C_2Cl_4. In comparison, CH_3CCl_3 has a lifetime of about 5 years accompanied by decade-long observations around the globe, making it an ideal proxy for the global tropospheric OH [e.g., Prinn et al., 1995]. The atmospheric lifetime of HCFC-22 is about 11 years [Miller et al., 1998]. As the concentrations continue to grow in the atmosphere, the constraints afforded by HCFC-22 observations will be increasingly important. The proxy constraints, although powerful, depend on the accuracy of source magnitudes and distribution, absolute measurement calibration, loss pathways other than OH oxidation, and kinetic data for the reaction with OH. The cumulative uncertainty in derived global mean OH concentration is up to 20-30% [Spivakovsky et al., 2000].

Chemistry and transport modeling

This class of modeling, unlike the other two discussed in following sections, is not specifically designed to simulate tropospheric OH. It simulates the primary components of the natural system, emissions, atmospheric transport, chemistry, and deposition. As reactions R1-6 implied, tropospheric O_3-HO_x-NO_x-CO-hydrocarbon chemistry is tightly coupled. Emissions for NO_x, CO, and nonmethane hydrocarbons (NMHCs) in the models include anthropogenic sources from fossil fuel combustions, industry, and biomass burning, and natural sources from vegetation (isoprene, terpenes, acetone), lightning (NO_x), and soils (NO_x). Concentrations of CH_4, well mixed in the troposphere, are generally specified in the models. The uncertainties in emission estimates, except fossil fuel combustions and industry, are a factor of 2 or more. Transport is generally simulated in the models using meteorological fields from a general circulation model or weather forecast model. Extensive model evaluations for all related species using observations from surface sites, balloon, aircraft, and satellite, are carried out to ensure the quality of model results [e.g., Müller and Brasseur, 1995; Roelofs and Lelieveld, 1995; Berntsen and Isaksen, 1997; Wang et al., 1998a; Hauglustaine, 1998; Lawrence et al., 1999]. The complexity of the models offers the flexibility to investigate various factors affecting the system, including sources of NO_x [Larmaque et al., 1996; Penner et al., 1998; Horowitz and Jacob, 1999; Levy et al., 1999], CO [Holloway et al., 2000], and NMHCs [Houweling et al., 1998; Wang et al., 1998b; Poisson et al., 2000], emissions from aircraft [e.g. Brasseur et al., 1996] and ships [Lawrence and Crutzen, 1999; Kasibhatla et al., 2000], O_3 transport from the stratosphere [Roelofs and Lelieveld, 1997; Wang et al. 1998b; Lelieveld and Dentener, 2000], and heterogeneous chemistry [e.g., Dentener and Crutzen, 1993; Lary et al.,1997].

One particularly relevant issue is the change of tropospheric composition since preindustrial times. Wang and Jacob [1998] reviewed available observations for preindustrial times and found the best constraints existed for O_3 near surface. Even these (reconstructed) records have seasonal and altitude variations that can hardly be reconciled with our current understanding of tropospheric O_3. They estimated that the sources of NO_x (S_N) and of CO and hydrocarbons (S_C) increased by factors of 4.7 and 2.5, respectively, from preindustrial times to present. Whereas both sources contribute, respectively by 60% and 40%, to the estimated 60% increase in the global O_3 burden, they have offsetting effects on the global mean OH concentration [Thompson, 1992]. Wang and Jacob [1998] showed a model sensitivity of $\Delta[OH] \propto \Delta(S_N / S_C^{3/2})$, resulting in a 9% decrease of the global (mass-weighted) mean OH concentration from preindustrial times in their model. Their results of the much smaller OH change compared to O_3 are consistent with other global 3-D model studies [e.g., Crutzen and Zimmermann, 1991; Martinerie et al., 1995; Levy et al., 1997; Roelofs et al., 1997; Berntsen et al., 1997; Mickley et al., 2000]. Figure 1 shows the zonally-averaged annual mean OH concentrations for the present atmosphere and model simulated changes from preindustrial times. Large increases of 20-60% occur in the lower troposphere of the northern hemisphere while decreases of about 20% take place in the northern upper troposphere and most of the southern hemisphere. The asymmetry reflects the longer range transport of CO and hydrocarbons than that of NO_x. The distribution shift towards warmer lower troposphere from preindustrial times boosts OH oxidation of CH_3CCl_3, the

rate of which increases with temperature; the model estimated the same 5.1 years for the lifetimes of tropospheric CH_3CCl_3 against OH oxidation for preindustrial times and present.

Precursor-constrained modeling

Uncertainties in emissions and atmospheric transport can be minimized to some extent by constraining the model using atmospheric observations. Spivakovsky [2000] carefully compiled climatologies for meteorological variables and OH precursors including O_3, CO, NO_x, and hydrocarbons from available atmospheric observations and applied them to global OH simulations. This work heralds the 3-D chemical data assimilations that will take the center stage in the years to come. Figure 2 shows the simulated annual mean OH distribution. The resulting lifetime of tropospheric CH_3CCl_3 against OH oxidation is 4.8 years. The striking difference compared to Figure 1 is the symmetry in OH distribution across the equator. The two results have about the same mean OH concentrations in the northern hemisphere but Figure 2 is higher by 30% in the southern hemisphere. The interhemspheirc asymmetry with higher OH concentrations in the northern hemisphere, as illustrated in Figure 1, is common among chemistry and transport models. The more up-to-date simulation by Mickley et al. [2000] shows mean OH concentrations 16% higher in the northern than southern hemisphere. The smaller interhemispheric ratio is due solely to less OH in the northern hemisphere compared to Wang et al. [1998a].

The interhemispheric OH asymmetry in chemistry and transport models arises from the asymmetry in anthropogenic emissions, which boost OH in the vicinity but depress OH in remote areas (Figure 1). The latter effect was observed over the tropical Pacific during PEM-Tropics B [Wang et al., 2000]. Presently most models have symmetric (across equator) biomass burning emissions, the only large anthropogenic source in the southern hemisphere. A symmetric OH distribution would imply much larger emissions from biomass burning in the southern than northern tropics.

The observational evidence for a hemisphere-symmetrical OH distribution is not yet affirmative. Observations of CH_3CCl_3 tend to support this distribution [Spivakovsky et al., 2000; Montzka et al., 2000]. The constraint on the inter-hemispheric ratio will improve to ±50% as CH_3CCl_3 emissions are phased out [Spivakovsky et al., 2000]. Montzka et al. [2000] suggested that the air mass boundary is more appropriately defined by the Inter-Tropical Convergence Zone (ITCZ), the annual-mean position of which is north of the equator. They found that CH_3CCl_3 observations implied a higher mean OH concentration in the southern than northern air mass. The observations of [14]CO also support the symmetric distribution or even a higher mean OH concentration in the southern hemisphere [e.g., Brenninkmeijer et al., 1992]; however, the interpretation is marred by large uncertainties in [14]CO transport [Spivakovsky et al., 2000; Quay et al., 2000]. Observations of CH_2Cl_2 suggest, on the other hand, higher OH concentrations by 45-95% in the northern than southern hemisphere [Spivakovsky et al., 2000].

Proxy-gas constrained modeling

This class of modeling takes full advantage of long-term observations of OH proxy gases using sophisticated statistical analysis. The best data series to date was gathered by the Atmospheric Lifetime Experiment / Global Atmospheric Gas Experiment (ALE/GAGE). Prinn et al. [1995] and Krol et al. [1998] applied different statistical methods to analyze the data series from 1978 to 1993. Global HCFC-22 measurements have been collected since 1992 by the NOAA Climate Monitoring & Diagnostics Laboratory (CMDL); the program also measures CH_3CCl_3 (beginning in 1988). Miller et al. [1998] demonstrated the utility of HCFC-22 observations using the method similar to Prinn et al. [1995] and found results consistent with the latter work.

The gist of the modeling is trying to deduce global OH concentrations from proxy observations; I will discuss the case for CH_3CCl_3. The deduction is an inverse problem since the observed atmospheric accumulation of CH_3CCl_3 results from industrial emissions, OH oxidation, and minor losses to the oceans and in the stratosphere. Starting with *a priori* global OH distribution, the inverse modeling aims to find the global mean OH concentration and its linear trend (with respect to time) that best fit observed CH_3CCl_3. A long-term data series is essential for the inversion. The *a priori* global OH distribution is taken from a global chemistry and transport model like the ones previously discussed. Prinn et al. [1995] used a 2-D model and Krol et al. [1998] relied on the 3-D model by Crutzen and Zimmermann [1991]. Prinn et al. [1995] applied a recursive weighted least squares (Kalman) filter to the inversion. With a more complicated 3-D OH distribution, Krol et al. [1998] employed a Monte-Carlo ensemble method for the fitting optimization.

Prinn et al. [1995] derived an atmospheric lifetime of CH_3CCl_3 of 4.6 with no apparent trend from 1978-1994. Krol et al. [1998] found a similar CH_3CCl_3 lifetime of 4.5-4.7 years but an increasing trend of 7% in 15 years. The estimated global mean (mass-weighted) OH concentration from the two studies is 9.7-10.7x10^5 molecules cm^{-3} in the troposphere. Prinn and Huang [2000] and Krol et al. [2000] examined the causes for the trend discrepancy. The different treatment of CH_3CCl_3 concentrations at the beginning of the observation series appears to be a significant factor. Prinn et al. [1995] computed CH_3CCl_3 concentrations from 1951 and had only two free parameters (mean OH and trend) in their inverse model. Krol et al. [1998], on the other hand, computed CH_3CCl_3 concentrations from 1978 and needed a third optimizing parameter to adjust model initial CH_3CCl_3 concentrations.

 The 7% OH increase from 1978 to 1993 estimated by Krol et al. [1997] is significant in light of ±10% change from preindustrial times to present found in most global 3-D simulations [Wang and Jacob, 1998]. One factor not considered in the other global 3-D studies is the decrease of stratospheric O_3 concentrations since the late 1970s, which, Krol et al. [1998] estimated, contributes to a 2% increase of the global mean OH concentration. They suggested that increasing water vapor concentrations in the tropics and a decoupling of NO_x and CO emissions (with increasing NO_x but decreasing CO emissions) by about 10% each could explain the rest of the OH increase. A shift in the

global mean OH concentration by this magnitude would therefore signify substantial changes taking place in anthropogenic emissions or global climate. Resolving the difference between the two studies and obtaining additional independent estimates are critical.

Acknowledgements

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Figure Captions

Figure 1. Zonally-averaged annual mean concentrations of OH for the present atmosphere and percentage changes from preindustrial times to present. Adopted from Wang and Jacob [1998].

Figure 2. Zonally-averaged annual mean concentrations of OH for the present atmosphere computed by Spivakovsky et al. [2000]. (Courtesy of C. M. Spivakovsky.)

OH [10⁵ molecules cm⁻³]

OH Change (%)

OH [10^5 molecules cm^{-3}]