World Applied Sciences Journal
14 (Exploring Pathways to Sustainable Living in Malaysia: Solving the Current Environmental Issues): 31-38, 2011
ISSN 1818-4952; © IDOSI Publications, 2011

Ozone Pollution and Historical Trends of Surface Background Ozone Level: A Review

Nor Liyana Arif and Ahmad Makmom Abdullah

Department of Environmental Science, Faculty of Environmental Studies,
Universiti Putra Malaysia, Selangor Darul Ehsan, Malaysia

Abstract: A literature survey was conducted to review the historical ozone data from background stations around the world to characterize background ozone levels and trends while also determining the reasons underlying such trends. Ozone pollution is regarded as a great concern nowadays because of its potential effects on human health and the environment especially vegetation. Historical increasing of ozone trends as reported ranged between 0.06 and 2.6% year with some of the largest increasing trend were observed at stations in Europe and Japan. For most stations, the trends were not consistent during the reporting period, but from the year 1970s until the mid 1980s, the trends are generally larger and steeper compared to the year 1990s. Sensitivity studies using chemical transport model were conducted to explain the rising trends in background ozone levels over the past decades.

Key words: Background ozone ・ Ozone trends ・ Tropospheric ozone ・ Ozone pollution

INTRODUCTION

Ozone pollution has been of great concern in the past few years because it is the most frequently occurring secondary pollutant in the atmosphere [1] and produced through photochemical reaction involving primary pollutants such as nitrogen oxides (NO_x), carbon monoxide (CO) and volatile organic compounds (VOCs) [2]. WHO[1] has stated that tropospheric ozone has the potential to be very damaging to human health [3, 4]. It is also associated with 21000 premature deaths per year and other respiratory diseases as mention by WHO[2]. Among the symptoms related to tropospheric ozone exposure include asthma [5], decrease in lung function [6] and breathlessness. The intercontinental transport and hemispheric air pollution by ozone jeopardize agricultural and natural ecosystem [7] worldwide and have a strong effect on climate [8]. Injury to vegetation is the most common effect found on vegetation exposed to ozone pollution [9-14]. This effect on vegetation can have a significant impact on agriculture production, as crop yield may be reduced due to ozone pollution. Past research also shows that ozone has a significant impact on non-biological materials as well. Damage to materials by photochemical oxidants has been studied for many years. Most research has focused on economically important or abundant materials that are susceptible to oxidant damage. These include elastomers, textile fibres, dyes and paints. Ozone results in a reduced breaking strength and increased rate of wear. Ozone damage usually takes the form of cracking leading to brittleness [15]. Damage to paint is similar to that of elastomers, that is embrittlement and cracking due to chain scission and cross-linking. The effects are small in comparison with those of other factors. It has been shown that several pigments faded severely if exposed to ozone level typical of a Los Angeles photochemical smog [16, 17, 18]. Additionally, ozone may also affect photographic materials [19] and damage books [20].

Sources of background ozone precursors come from natural and anthropogenic sources, where urban areas are the major anthropogenic sources of air pollution. The increase in anthropogenic ozone precursor emission (mainly NO_x and VOCs) is due to the rise in energy demands resulting from urbanization and rapid increase in industrialization and motorized transports.

[1] WHO, 2009. *Air Quality and Health* World Health Organization Regional Office for Europe: http://www.euro.who.int/air
[2] WHO, 2005. 'WHO Air Quality Guidelines Global Update 2005', Report of a working group meeting, Bonn, Germany. 18-20 October 2005, World Health Organization, Geneva, www.cepis.ops-oms.org/bvsea/fulltext/guidelines05.pdf

Corresponding Author: Nor Liyana Arif, Department of Environmental Science, Faculty of Environmental Studies,
Universiti Putra Malaysia, Selangor Darul Ehsan, Malaysia.
Tel: +603-89466759, Fax: +603-89466768.

There is also a greater concern of the contribution of biogenic VOC emission, where its reaction products react with NO_x from anthropogenic and natural sources for regional and urban ozone production [21, 22, 23]. In recent years, atmospheric ozone concentration is changing and over the decades, various regions of the globe have shown divergent trends of tropospheric ozone. This paper focuses on the current ozone background levels and trends from different ozone background stations around the world. A survey of past literature was done to further understand the recent surface ozone data from background stations from around the world. Trends of ozone reported at different elevation measurements and period were studied to evaluate the increase trends at the background ozone stations.

Historical Background Ozone Concentration and Trends: The determination of a secular trend of ozone requires continuous or quasi-continuous measurements over a sufficiently long time period. Additionally, there is often uncertainty in the data due to the measurement technique used, artifacts, calibration and siting problems. When considering the trends one should make a distinction between free tropospheric levels, levels in the less polluted background or rural areas and levels in urban areas which are to a great extent influenced by anthropogenic emissions. Tropospheric ozone originates from both downward transport of ozone from stratosphere and in situ photochemical production [24]. Polluted air masses from urban and industrial areas can affect rural areas at considerable distances and even affect ozone concentrations in parts of the free troposphere. It must be clear that the free troposphere and different regimes of the boundary layer may influence one another.

In addition to the recent tropospheric ozone measurements, there are a few historic records of surface ozone measurement. However, most of the historic data is unreliable because of the uncertainties in the measurement method used. It has been established, however, that towards the end of the nineteenth century surface ozone measurement were approximately half of the mean of daily maximum of observations taken during the last 15 years at similar locations in Europe and North America [25]. The most important set of historical data is series is daily means collected at the Montsouris Observatory in Paris between 1876 and 1905 [26, 27]. Disregarding the apparent trend in the Arkona data, the annual average ozone concentration during the 30 year period following 1956 was about 21 ppv. This shows that today's background ozone levels in the central Europe are approximately 100% higher than those 100 years ago. The Montsouris data indicates considerable year to year fluctuations but no discernable trend. The Arkona data show a marked increase in ozone towards the end of the 1970s of 2-3% per year and a tendency for ozone to decrease after 1979 [28]. This decrease is not easily explained from the current knowledge of ozone loss and formation processes [29]. Concern about the positive trend of tropospheric ozone at mid-latitudes of the Northern Hemisphere has focused attention on historical ozone observations made by the Schobein technique one century ago. Reevaluated historical data from South America show that one century ago the surface ozone levels at mid-latitudes of the Southern Hemisphere were comparable to those observed in the Northern Hemisphere. Currently, mid-latitude ozone levels of the Southern Hemisphere are lower than those of the Northern Hemisphere, indicating a change with respect to the past century.

In Table 1, the highest recorded increase of ozone trends is 2.6% per year, while the lowest increase recorded is 0.06%. For stations in Japan and Europe, largest rise in trends can obviously be found, with average recorded increase ranged between 0.07 and 2.6%. At Hohenpeissenberg (elevation 975 m) in Southern Germany, measurements covered a long time period. Continuous ozone measurements commenced in 1971. Over the full period of the records (up to 1988) both stations, which are about 800km apart, show a positive trend of about 1% per year. There are marked differences in the fluctuations over various subperiods at the two locations [30]. A larger increase in mean surface ozone (~2.1% per year) in 1970s compared with that in the 1980s (~0.5% per year) is evident at Hohenpeissenberg. These differences may result from a combination of many factors which include photochemistry, changes in surface deposition, local changes in atmospheric circulation and data quality. Low *et al.* [31] have shown that the uncertainty due to SO2 interference, at Hohenpeissenberg during pre-1976 period, could have a significant effect on the apparent upward trend and possibly lead to its over estimation by a factor of 3.

Through 1970s until mid 1980s, generally larger trends were observed, though during the reporting period, trends at most stations were inconsistent. Direct comparison of ozone trends between those stations may not be possible. However, this compilation of increasing background ozone trends is useful to see

Table 1: Increase of ozone trends based on background stations reporting at the surface, near surface (900mb) or in the lower troposphere (850-700mb, between ~1500 and 3000m)

Stations	Time of record	Measurement elevation	Trend (% per yr unless otherwise) indicated	References
Sapporo, Japan	1969-1982	Near surface	+ 1.3±1.7	[58]
Tateno, Japan	1969-1982	Near surface	+ 2.3±1.0	[58]
Mauna Loa, Hawaii	1974-2001	Surface (3397 m)	+ 0.15±0.06[b]	CMDL (2004)[3]
Cape Grim, Australia	1982-1995	Surface (104 m)	+ 0.18±0.14	[59]
Cape Point, S. Africa	1983-1995	Surface (260 m)	+ 0.53±0.34	[59]
Alert, Nunavut	1987-2001	Surface (62m)	+ 1.2±0.8	[60]
Eureka, Nunavut	1980-2001	Surface (10m)	+ 2.6±2.1	[60]
Resolute, Nunavut	1980-2001	Surface (64m)	+ 0.2±0.6	[60]
Zugspitze, Germany	1978-1995	Surface (2962 m)	+ 1.48±0.51	[59]
Arkona, Germany	1956-1990	Surface (42 m)	+ 1.12±0.38	[61]
			+ 0.98±0.86	[62]
Point Barrow, Alaska	1974-2001	Surface (11 m)	+ 0.07±0.04[b]	CMDL (2004)[3]
Mace Head, Ireland	1987-1995	Surface (25 m)	+ 0.19[b]	[63]
Saturna Island, B.C	1991-2000	Surface (178 m)	+ 0.94±0.74	[64]
Lassen Volcanic Nat.	1988-2002	Surface (1756 m)	+ 0.60±0.30	[65]
Park, California[a] Wallops Island, Virginia	1970-1981	Near surface	+ 0.9±1.2	[58]
	1970-1995	Lower troposphere	+ 0.06±0.24	[59]
Whiteface Mountain, N.Y	1974-1995	Surface (1480 m)	+ 0.45±0.22	[59]
Okinawa, Japan	1989-1997	Near surface	+ 2.5±0.6	[66]
	1989-1997	Surface (76 m)	+ 2.6±2.0	[66]
Tsukuba, Japan	1969-1995	Lower troposphere	+ 0.93±0.26	[59]
Payerne, Switzerland	1968-1977	Near surface	+ 0.6±1.5	[58]
Switzerland, various sites	1991-1999	Surface	+ 0.4-0.9[b]	[67]
Hohenpeissenberg,	1976-1992	Surface (975 m)	+ 0.9±0.3	[62]
Germany		Surface (975 m)		[68]
	1971-1988	Lower	+ 1.02±0.60	[59]
	1967-1995	Troposphere	+ 1.48±0.22	
Dresden	1952-1984		+ 2.6±1.6	[69]
Kalternnordheim	1955-1983		+ 3.1±2.1	[69]
Gr. Inselberg	1972-1983		+ 3.1±2.4	[69]
Fichtelberg	1954-1984		+ 1.1±2.0	[69]

[a]Spring data

[b]Units in ppb yr

the patterns of ozone in different parts of the world, particularly in Europe and Asia. These stations can be grouped into Northern and Southern Hemisphere. Due to limited observations being made in the Southern Hemisphere, it is difficult to distinguish the southern part of the globe and also to make a direct conclusion whether hemispheric difference affected the background ozone concentrations. Barrow and Mauna Loa in the Northern Hemisphere and Cape Point and Cape Grim in the Southern Hemisphere are sited such that ozone concentrations can be considered to be representative of the region at large. Positive trends in the Northern Hemisphere are observed at sites remote from industrial emissions [32, 33]. A significant trend of 0.8 (±0.5)% per year in the first three kilometres of the atmosphere in the Northern Hemisphere is found by averaging over all stations in Table 1. Average trend in the troposphere above this altitude were not significant. At Cape Point, South Africa and Cape Grim, Australia, small increasing trend was observed that may signify African biomass burning [34]. Through careful observation, we can see that ozone trends from year 1980 onwards are either escalated in a smaller scale or have remained invariable [35, 36]. Deterioration of nitrogen oxide emissions in North America and Europe and declination in carbon monoxide concentration [37] are the possible factors that may have influenced the trend of smaller increase rate of background ozone levels.

[3]CMDL (Climate Monitoring and Diagnostics Laboratory), 2004. CMDL Summary Report #26. Climate Monitoring and Diagnostics Laboratory, 325 Broadway R/CMDL, Boulder, Colorado, 80305; http://www.cmdl.noaa.gov/publications/annrpt26/4 1 4.pdf.

Explanation on Probable Causes for Observed Ozone Trends: There are a number of causes that are related to the increasing trends of ozone concentration as reported in past literatures. To investigate these factors, sensitivity studies have been conducted to further analyze and explain the increase in ozone trends such that has been done in Northern Hemisphere using several chemical transport models i.e. NASA GISS[4] and GEOS-CHEM[5].

Changes in the emissions of ozone precursors significantly influence the background ozone level. Tropospheric ozone level is largely affected by the decrease of NO_x emission at point source and mobile source, while reduction of ozone concentration over large area is due to removal of area and non-area sources of NO_x, eventhough the magnitude is not that significant compared to the cases of point source and mobile source [38]. NOx, CO, CH4 and non-methane hydrocarbons are known to have increased, mainly due to the anthropogenic emissions, in the Northern Hemisphere [39, 40]. Modeled result suggested that the increase of surface emissions of NO_x originated from fossil-fuel combustion have had the largest effect on ozone in the lower troposphere since 1970. Evaluation of the study have indicated that increase in emissions of ozone precursors, particularly NO_x could be the primary cause of more than 10% increase in year round ozone over Canada, Europe and Japan, while also causing another 20% increase over Europe and Japan during summer. Model studies have also indicated that a significant portion of the increase in ozone may be connected to the increase in global anthopogenic NOx emissions while the contribution of CH4 to the increase is small [41]. Further increases in NO_x will contribute to a continued rise in free tropospheric ozone, while the occurrence of ozone episodes in the boundary layer may not change much [42]. However, the same cannot be said for hydrocarbons emissions. As opposed to NO_x emissions, the rise of hydrocarbon emissions from combustion of fossil fuel has been moderate. Although the strength of regional high ozone episodes is been affected by the long-term changes of hydrocarbon emissions, they do not significantly contributed to the mean tropospheric ozone trends, based on the model results presented [43, 44].

Intercontinental transport is another factor worth considering to explain the observed ozone trends. Several studies [45-48] have indicated that intercontinental transport of ozone have a significant impact on North America, where the region is affected by trans-Pacific transport of Asian pollution. Other studies [49, 50, 51] have suggested that North American pollution have affected Europe via trans-Atlantic transport, while Asian region is affected by the trans-European transport of European pollution [52]. This is a critical problem that hampered the regions because the main sources of the pollution cannot be identified (thus making the regional air pollution control measures ineffective), while there are no specific mitigating measures that can be taken to curb this intercontinental transport of pollutants. It is important to note that western United States is also affected by pollution coming from Asia, where the effect of trans-Pacific transport of Asian pollution directly influenced the background ozone levels in that area by 3-10 ppb. This effect is most prevalent in springtime because the storm and frontal activity in Asia is most frequent and the strongest westerly transport of Asian air across North Pacific is most pronounced in this period [53, 54].

If intercontinental transport is considered as a factor that impacted the background ozone level trends, it is highly likely that trends in emissions of ozone precursors from source areas will greatly influence the trend of background ozone levels, since ozone formation is dependent on its precursor's emissions. The increase of NO_x emissions from East Asia, particularly from China has led to the deterioration in air quality in many regions of the country. This is due to the increase motorization in China where transportation sector contributes to 37% production of NO_x, in which NO_x is an essential precursor that make up ozone formation [55]. If developing countries continue produce emission according to their projected increasing trends, emission contribution from Asian countries will rise.

Another possible reason that leads to the changes in background ozone level is the influence of stratospheric-tropospheric ozone exchange. Sources of stratospheric ozone are evaluated to put forth a small but significant influence on ozone levels in the lower troposphere during the winter and spring at northern midlatitudes, where ozone of stratospheric origin is estimated to be approximately 10% of total ozone concentration. However, stratospheric ozone influence is somehow negligible during the summer [56]. Evaluation of the ozone trends has estimated that the stratospheric flux of ozone into the troposphere may have lowered by as much as 30% from early 1970s right through mid-1990s.

[4]NASA, 2004. Goddard Institute for Space Studies; http://www.giss.nasa.gov/research/modeling/gcms.html.
[5]Harvard University, 2004. Atmospheric Chemistry Modeling Group; http://www.as.harvard.edu/chemistry/trop/geos/.

It is believed that the decline of stratospheric flux of ozone may be caused by ozone depletion in the lowermost stratosphere during spring and winter.

Apart from the influence of stratospheric-tropospheric ozone exchange effect, another important effect that consequently related to the possible cause of changes in background ozone level as mentioned above is the increase in UV radiation that reach the lower troposphere due to the decline in stratospheric ozone level, in which photochemical production and ozone loss are expected to occur as the consequences to this effect. Modeled results have implicated that UV radiation's increase (due to stratospheric ozone depletion) do not significantly reduce tropospheric ozone, except at midlatitudes in the Southern Hemisphere, following the break up of the ozone hole.

Although increases in surface and tropospheric ozone over the past century seems to be well established, its magnitude and possible causes are still a controversial issue due mainly to a lack of enough reliable data. Experimental techniques and observational frequencies used have differed in different places and even at different times in the same place. Some stations have been operative for over a decade, whereas others have had a transitory existence. Typical tropospheric ozone variability and instrumental precision means that the current ozonesonde network and presumably surface ozone sites, are insufficient to detect a trend in tropospheric ozone of about 1% per year, even at stations with the records a decade in length. At least a doubling of the current network is necessary to detect an ozone trend of 1% per year on a decadal time frame [57].

SUMMARY AND CONCLUSION

Ozone pollution poses several significant threats to human and environment as well. The historical trends of background ozone level presented in this study has indicated that the trend of background ozone level is increasing over the past century, though there are prevalent uncertainties that need to be addressed clearly if comparison among locations is to be done, such as few numbers of background stations, different reporting periods and difficulties in identifying stations which are representative of background conditions. Through this study, it is important to acquire more data from monitoring stations, especially in Asia and Southern Hemisphere as it can greatly facilitate in filling in some of the present gaps in analyzing background ozone distribution and trends globally. Characterization of background ozone is possible if global network exists. This network can also play an important role in the evaluation of the impact of emissions changes and intercontinental transport on background levels. A good understanding of the anthropogenic development to the background ozone has become a vital point with the recent narrowing gap between air quality standards and objectives and background levels. Despite all the uncertainties, the ozone background level data from past literatures provides us with an overview on the trends of ozone background level over the centuries. Much work needs to be done to observe the ozone background level at global, regional and hemispheric level. Continuous data collection from background stations is especially important in this work to better analyze the trends for further studies in the future, especially in projecting the distribution of background ozone globally in the future.

REFERENCES

1. Lee, C.T. and S.C. Chang, 2006. Ozone variations through vehicle emissions reductions based on air quality monitoring data in Taipei City, Taiwan, from 1994 to 2003. Atmospheric Environ., 40: 3513-3526.
2. Yamaji, K., T. Ohara, I. Uno, H. Tanimoto, J. Kurokawa and H. Akimoto, 2006. Analysis of the seasonal variation of ozone in the boundary layer in East Asia using the Community Multi-scale Air Quality model: What controls surface ozone levels over Japan? Atmospheric Environ., 40: 1856-1868.
3. Lee, D.S., M.K. Holland and N. Falla, 1996. The potential impact of ozone on materials in the UK. Atmospheric Environ., 30: 1053-1065.
4. Mazzeo, N.A., L.E. Venegas and H. Choren, 2005. Analysis of NO, NO_2, O_3 and NO_x concentrations measured at a green area of Buenos Aires City during wintertime. Atmospheric Environ., 39: 3055-3068.
5. Cody, R.P., C.P. Weisel, G. Birnbaum and P.J. Lioy, 1992. The effect of ozone associated with summertime photochemical smog on the frequency of asthma visits to hospital emergency departments. Environmental Res., 58: 184-194.
6. Schwartz, J. and S. Zeger, 1990. Passive smoking, air pollution and acute respiratory symptoms in a diary of student nurses. American Review of Respiratory Disease, 141: 62-67.
7. Englert, N., 2004. Fine particles and human health-a review of epidemiological studies. Toxicology Letters, 149: 235-242.

8. Akimoto, 2003. Science 5 December 2003: 302(5651): 1716-1719.
9. Laurence, J.A. and L.H. Weinstein, 1981. Effects of air pollutants on plant productivity. Annual Review of Phytopathol., 19: 257-271.
10. Jacobson, J.S., 1982. Ozone and the growth and productivity of agricultural crops. Effects of Gaseous Air Pollution in Agriculture and Horticulture. M.H. Unsworth and D.P. Ormrod (Eds.), Butterworth, London, pp: 293-304.
11. Taylor, O.C., 1984. Organismal responses of higher plants to atmospheric pollutants: photochemical and other. Air Pollution and Plant Life. M. Treshow, (Ed.), John Wiley and Sons, NY., pp: 215-238.
12. Guderian, R., (Ed.), 1985. Air Pollution by Photochemical Oxidants: Formation, Transport, Control and Effects on Plants, Springer-Verlag, Berlin.
13. Prinz, B. and C.J. Brandt, 1985. Effects of air pollution on vegetation. Pollutants and their Ecotoxicological Significance. H.W. Nurnberg (Ed.), John Wiley and Sons, NY., pp: 67-84.
14. Ashmore, M., N. Bell and J. Rutter, 1985. The role of ozone in forest damage in West Germany. Ambio, 14: 81-87.
15. Colbeck, I. and A.R. Mackenzie, 1994. Air Pollution by Photochemical Oxidants. Elsevier Science B.V.
16. Shaver, C.L., G.R. Cass and J.R. Druzik, 1983. Ozone and deterioration of works of art. Environmental Science and Technol., 17: 748-752.
17. Whitmore, P.M. and G.R. Cass, 1987. The ozone fading of traditional natural organic colorants on paper. Journal of American Institute of Conservatory, 26: 45-58.
18. Whitmore, P.M. and G.R. Cass, 1988. The ozone fading of traditional Japanese colorants. Studies in Conservatory, 33: 29-40.
19. Swan, A., 1981. Conservation of photographic print collections. Library Trends, 30: 267-296.
20. Shahani, C.J. and W.K. Wilson, 1987. The preservation of library and archives. American Scientist, 75: 240-251.
21. Cardelino, C.A. and W.L. Chameides, 1990. Natural hydrocarbons, urbanization and urban ozone. J. Geophysical Res., 95: 3971-3979.
22. Chameides, W.L., R.W. Lindsay, J. Richardson and C.S. Kiang, 1988. The role of biogenic hydrocarbons in urban photochemical smog: Atlanta as case study. Sci., 241: 1473-1475.
23. Trainer, M., E.T. Williams, D.D. Parrish, M.P. Buhr, E.J. Allwine, H.H. Westberg, F.C. Fehsenfeld and S.C. Liu, 1987. Models and observations of the impact of natural hydrocarbons on rural ozone. Nature, 329: 705-707.
24. Colbeck, I. and A.R. Mackenzie, 1994. Air Pollution by Photochemical Oxidants. Elsevier Science B.V.
25. Bojkov, R.D., 1986. Surface ozone during the second half of the nineteenth century. J. Climate Applied Meteorol., 25: 343-352.
26. Kley, D., A. Volz and F. Mulheins, 1988. Ozone measurements in historic perspective, In: Tropospheric Ozone, Isaksen I.S.A. (Ed.) D. Reidel, Dodrecht, pp: 63-72.
27. Volz, A. and D. Kley, 1988. Evaluation of the Montsouris series of ozone measurements made in the nineteenth century, Nature, 332: 240-242.
28. Feister, U. and W. Warmbt, 1987. Long-term measurements of surface ozone in German Democratic Republic, J. Atmospheric Chemistry, 5: 1-21.
29. Isaksen, I.S.A., 1988. Is the oxidizing capacity of the atmosphere changing? The Changing Atmosphere, F.S. Rowland and I.S.A. Isaksen, (Eds.) John Wiley, Chichester, pp: 141-157.
30. Low, P.S., T.D. Davies, P.M. Kelly and G. Farmer, 1990. Trends in surface ozone at Hohenpeissenberg and Arkona. J. Geophysical Res., 95: 22441-22453.
31. Low, P.S., T.D. Davies, P.M. Kelly and R. Reiter, 1991. Uncertainties in surface ozone at Hohenpeissenberg. Atmospheric Environment, 25A: 511-515.
32. Oltmans, S.J. and W.D. Komhyr, 1986. Surface ozone distributions and variations from 1973-1984 measurements at the NOAA geophysical monitoring for climatic change baseline observations. J. Geophysical Res., 91: 5229-5236.
33. Bodhaine, B.A. and R.M. Rosson, 1988. Geophysical monitoring for climate change. Summary Report 1987, NOAA Air Resources Lab, Boulder.
34. Fishman, J., K. Fakhruzzaman, B. Cros and D. Nganga, 1991. Identification of widespread pollution in the Southern Hemisphere deduced from satellite analyses. Sci., 252: 1693-1696.
35. Logan, J.A., 1994. Trends in the vertical distribution of ozone: an analysis of ozonesonde data. J. Geophysical Res., 99: 25553-25585.

36. Oltmans, S.J., A.S. Lefohn, H.E. Scheel, J.M. Harris, H. Levy, I.E. Galbally, E.G. Brunke, C.P. Meyer, J.A. Lathrop, B.J. Johnson, D.S. Shadwick, E. Cuevas, F.J. Schmidlin, D.W. Tarasick, H. Claude, J.B. Kerr, O. Uchino and V. Mohnen, 1998. Trends of ozone in the troposphere. Geophysical Research Letters, 25: 139-142.
37. Novelli, P.C., K.A. Masarie and P.M. Lang, 1998. Distributions and recent changes of carbon monoxide in the lower troposphere. J. Geophysical Res., 103: 19015.
38. Lin, C.J., T.C. Ho, H.W. Chu, H. Yang, S. Chandru, N. Krishnarjanagar, P. Chiou and J.R. Hopper, 2005. Sensitivity analysis of ground-level ozone concentration to emission changes in two urban regions of southeast Texas. J. Environmental Management, 75: 315-323.
39. Gammon, R., S.C. Wofsy, R.J. Cicerone, A.C. Delany, R.T. Harris, M.A.K. Khalil, J.A. Logan, P. Midgley and M. Prather, 1986. Trends in source species. Atmospheric Ozone 1985 1: 57-116. WMO Global Ozone Research and Monitoring Project, Report No, 16.
40. Ehhalt, D.H., P.J. Fraser, D. Albritton, Y. Makide, R.J. Cicerone, F.S. Rowland, M.A.K. Khalil, L.P. Steele, M. Legrand and R. Zander, 1989. Trends in source gases. Report of the International Ozone Trends Panel-1988. WMO Report No. 18, Geneva.
41. Dignon, J. and S. Hameed, 1985. A model investigation of the impact of increases in anthropogenic NOx emissions between 1967 and 1980 on tropospheric ozone. J. Atmospheric Chemistry, 3: 491-506.
42. Hov, O., 1990. The role of nitrogen oxides in the long-range transport of photochemical oxidants. Science of Total Environment, 96: 101.
43. Lefohn, A.S., D.S. Shadwick and S.D. Ziman, 1998. The difficult challenge of attaining EPA's new ozone standard. Environmental Science and Technol., 276A-282A.
44. Fiore, A.M., D.J. Jacob, I. Bey, R.M. Yantosca, B.D. Field and J.G. Wilkinson, 2002. Background ozone over the United States in summer: origin and contribution to pollution episodes. Journal of Geophysical Res., 107(15): ACH 11-1-ACH 11-25.
45. Jaffe, D.A., T. Anderson, D. Covert, R. Kotchenruther, B. Trost, J. Danielson, W. Simpson, T. Bertsen, S. Karlsdottir, D. Blake, J. Harris, G. Carmichael and I. Uno, 1999. Transport of Asian air pollution to North America. Geophysical Research Letters, 26: 711-714.
46. Jacob, D.J., J.A. Logan and P.P. Murti, 1999. Effect of rising Asian emissions on surface ozone in the United States. Geophysical Research Letters, 26: 2175-2178.
47. Yienger, J.J., M. Galanter, T.A. Holloway, M.J. Phadinx, S.K. Guttikunda, G.R. Carmichael, W.J. Moxim and H. Levy, 2000. The episodic nature of air pollution transport from Asia to North America. J. Geophysical Res., 105(26): 931-945.
48. Wilkening, K.E., L.A. Barrie and M. Engle, 2000. Transpacific air pollution. Sci., 290: 65-67.
49. Ryall, D.B., R.H. Maryon, R.G. Derwent and P.G. Simmonds, 1998. Modeling long range transport of CFCs to Mace Head, Ireland. Quarterly J. the Royal Meteorological Society, 124: 417-446.
50. Derwent, R.G., P.G. Simmonds, S. Scuring and C. Dimmer, 1998. Observation and interpretatio n of seasonal cycles in the surface concentrations of ozone and carbon monoxide at Mace Head, Ireland, from 1990 to 1994. Atmospheric Environ., 32: 145-157.
51. Stohl, A. and T. Trickl, 1999. A textbook example of long-range transport: simultaneous observation of ozone maxima of stratospheric and North American origin in the free troposphere over Europe. J. Geophysical Res., 104(30): 445-462.
52. Liu, H., D.J. Jacob, L.Y. Chan, S.J. Oltmans, I. Bey, R.M. Yantosca, J.M. Harris, B.N. Duncan and R.V. Martin, 2002. Sources of tropospheric ozone along the Asian Pacific Rim: an analysis of ozonesonde observations. J. Geophysical Research 107 (D21), 4573 (doi: 10.1029/ 2001JD002005).
53. Savoie, D.L., J.M. Prospero and E.S. Salzman, 1989. Nitrate, non-sea salt sulfate and methane sulfonate over the Pacific Ocean. Chemical Oceanography, 10: 220-251.
54. Merril, J.T., 1989. Atmospheric long-range transport to the Pacific Ocean. Chemical Oceanography, 10: 15-50.
55. Schwela, D., G. Haq, C. Huizenga, J.H. Wha, H. Fabian and M. Ajero, 2006. Urban Air Pollution in Asian Cities: Status, Challenges and Management, pp: 10-11.
56. Vingarzan, R., 2004. A review of surface ozone background levels and trends. Atmospheric Environ., 38: 3431-3442.
57. Prinn, R.G., 1988. Toward an improved global network for determination of tropospheric ozone climatology and trends. J. Atmospheric Chemistry, 6: 281-298.
58. Logan, J.A., 1985. Tropospheric ozone: seasonal behavior, trends and anthropogenic influence. J. Geophysical Res., 90(10): 463-482.

59. Oltmans, S.J., A.S. Lefohn, H.E. Scheel, J.M. Harris, H. Levy, I.E. Galbally, E.G. Brunke, C.P. Meyer, J.A. Lathrop, B.J. Johnson, D.S. Shadwick, E. Cuevas, F.J. Schmidlin, D.W. Tarasick, H. Claude, J.B. Kerr, O. Uchino and V. Mohnen, 1998. Trends of ozone in the troposphere. Geophysical Research Letters, 25: 139-142.
60. Tarasick, D.W., V.E. Fioletov, D.I. Wardle, J.B. Kerr and J. Davies, 2005. Changes in the vertical distribution of ozone over Canada from ozonesondes: 1980-2001, Journal of Geophysical Research, 110, D02304, doi:10.1029/2004JD004643.
61. Lefohn, A.S., D.S. Shadwick, U. Feister and V.A. Mohnen, 1992. Surface-level ozone: climate change and evidence for trends. J. the Air and Waste Management Association, 42(2): 136-144.
62. Low, P.S., T.D. Davies, P.M. Kelly and G. Farmer, 1990. Trends in surface ozone at Hohenpeissenberg and Arkona. J. Geophysical Res., 95(D13) 22: 441-453.
63. Simmonds, P.G., S. Seuring, G. Nickless and R.G. Derwent, 1997. Segregation and Interpretatio n of Ozone and Carbon Monoxide Measurements by Air Mass Origin at the TOR Station Mace Head, Ireland from 1987-1995. J. Atmospheric Chemistry, 28: 45-59.
64. Vingarzan, R. and B. Thomson, 2004. Temporal variation in daily concentrations of ozone and acid related substance s at Saturna Island, British Columbia, Canada. Journal of the Air and Waste Management Association, 54: 459-472.
65. Jaffe, D.A., D. Parrish, A. Goldstein, H. Price and J. Harris, 2003. Increasing backgroundozone during spring on the west coast of North America. J. Geophysical Res., 30(12): 1613 (doi:10.1029/2003GL017024).
66. Lee, S.H., H. Akimoto, H. Nakane, S. Kurnosenko and Y. Kinjo, 1998. Lower tropospheric ozone trendobserved in 1989-1997 at Okinawa, Japan. Geophysical Research Letters, 25: 1637-1640.
67. Bronnimann, S., B. Buchmann and H. Wanner, 2002. Trends in near-surface ozone concentration in Switzerland: the 1990s. Atmospheric Environ., 36(17): 2841-2852.
68. Logan, J.A., 1994. Trends in the vertical distribution of ozone: an analysis of ozonesonde data. J. Geophysical Res., 99: 25553-25585.
69. Scientific Assessment of Stratospheric Ozone: 1989. Volume 1, WMO, (1989). Global Ozone Research and Monitoring Project, Report No. 20.