

## APPENDIX C

### Sulfur Emissions from Volcanic Activity in 1985 and 1990

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#### C.1. INTRODUCTION

Sulfur from anthropogenic sources is emitted in the form of sulfur dioxide ( $\text{SO}_2$ ), hydrogen sulfide ( $\text{H}_2\text{S}$ ), or sulfate; estimates of the sulfate fraction vary from 1.5 to 5% (Benkovitz *et al.*, 1994). Global estimates of anthropogenic emissions of sulfur for 1985 are approximately  $65 \text{ Tg S y}^{-1}$  (Benkovitz *et al.*, 1996) and for 1990 approximately  $71 \text{ Tg S y}^{-1}$  (see Appendix A titled Global Anthropogenic Sulfur Emissions for 1985 and 1990 in this report). Sulfur from biogenic sources is emitted in the form of  $\text{SO}_2$  and of several reduced sulfur compounds such as hydrogen sulfide ( $\text{H}_2\text{S}$ ), dimethyl sulfide (DMS), carbonyl sulfide (OCS), carbon disulfide ( $\text{CS}_2$ ), and dimethyl disulfide (DMDS). Recent estimates of emissions of sulfur compounds from non-anthropogenic sources (Bates *et al.*, 1992) place oceanic emissions (mostly DMS) at approximately  $15 \text{ Tg S y}^{-1}$ , terrestrial emissions at approximately  $0.4 \text{ Tg S y}^{-1}$ , emissions from biomass burning at approximately  $2 \text{ Tg S y}^{-1}$  and volcanic emissions at approximately  $9 \text{ Tg S y}^{-1}$ , for a total of  $26.4 \text{ Tg S y}^{-1}$ . These estimates for oceanic and terrestrial sulfur emissions are at the low end of the range given by the IPCC (Houghton *et al.*, 1992), where oceanic emissions are estimated to be in the range 10 to  $50 \text{ Tg S y}^{-1}$  and terrestrial emissions in the range  $0.2$  to  $4 \text{ Tg S y}^{-1}$ .

Sulfur emissions from anthropogenic sources are strongly localized in the highly populated and industrialized regions in Eastern North America, and across Europe from the United Kingdom over Central Europe to the Donbas region

in Russia (Benkovitz *et al.*, 1996). Release heights are in the range of surface to over 200 m (Saeger *et al.*, 1989). Biogenic sulfur emissions from oceanic sources follow the distribution of areas of high productivity which are dependent on season and are located preferentially along coastlines. Sulfur emissions from land sources follow the distribution of global vegetation and are highest in the tropical and subtropical areas. All the biogenic emissions are released at the surface (Benkovitz *et al.*, 1994). Volcanic emissions are located in areas of volcanic activity, are extremely variable in time, and can be released anywhere from ground level to the stratosphere.

The main sulfur compound emitted by volcanoes is sulfur  $\text{SO}_2$  (Gerlach and Nordie, 1975). Hydrogen sulfide ( $\text{H}_2\text{S}$ ) may be abundant in plumes of relatively low temperatures and reducing conditions (Ozawa, 1966, Ross, 1968); however, overall this compound is generally less than 1% of the sulfur gas (Stoiber *et al.*, 1987). Other sulfur compounds present in small amounts are sulfur trioxide ( $\text{SO}_3$ ), sulfate, carbonyl sulfide (OCS), and elemental sulfur (S). For example, Belviso *et al.* (1986) estimate the global OCS volcanic yield to be 3.1 to  $46.1 \times 10^9$  g S  $\text{yr}^{-1}$ , compared to an annual flux of  $3.4 \times 10^{12}$  g S  $\text{yr}^{-1}$  for  $\text{SO}_2$  estimated by Stoiber *et al.* (1987). Reduced sulfur compounds can be oxidized as the hot eruption mixes with ambient oxygen, so that the composition of sulfur gases in volcanic activity can vary widely (Cadle *et al.*, 1971).

Sulfur emission rates of volcanoes have recently begun to be estimated based on surface measurements (e.g., Stoiber *et al.*, 1983; Allard *et al.*, 1994, Caltabiano *et al.*, 1994) and derived from satellite measurements (Bluth *et al.*, 1993, Mouginis-Mark *et al.*, 1989). Lacking direct measurements,

various methodologies to estimate sulfur emissions have been developed and applied to obtain estimates of the contribution of volcanoes to the global atmospheric sulfur budget; for example, see Newhall and Self (1982), Berresheim and Jaeschke (1983), Devine *et al.* (1984b), Stoiber *et al.*, (1987), and Pinto *et al.* (1989). Several of these methodologies and estimates have been included as a component of global inventories of sulfur emissions from all sources by various authors; for example, see Cullis and Hirschler (1980), Möller (1984), Bates *et al.* (1992), and Spiro *et al.* (1992). In general, global estimates of total sulfur emissions are given as global totals for an "average" year, except in the Spiro *et al.* inventory, where the data reflect the year 1980.

Much more attention has been given to sulfur emissions from erupting volcanoes since these were considered a more abundant source of sulfur compounds; for example, Stoiber *et al.* (1987) estimate that the annual sulfur flux from degassing, non-erupting volcanoes is approximately 53% of the flux from eruptive events. For their estimate, these authors used average, normalized data for the previous 20 years based on the compendium of Simkin *et al.* (1981). In contrast, Bluth *et al.* (1993) used data from the Total Ozone Mapping Spectrometer aboard NIMBUS 7 to estimate that from November 1978 to May 1993 the annual flux from explosive volcanism is less than half of the non-explosive output. These results give an indication of the interannual variability of the volcano sulfur source, so that applying averages to obtain emissions estimates would not yield the correct picture of these emissions for particular years.

The purpose of this project is to compile estimates of sulfur emissions from volcanic activity for years 1985 and 1990.

## C.2. EMISSIONS OF SULFUR FROM VOLCANOES FOR 1985 AND 1990.

### C.2.1. Estimation of sulfur emissions from volcanoes.

Petrologic estimates and direct measurements are the two prevalent methodologies used to estimate sulfur emissions from volcanoes. In addition the algorithms used to estimate various parameters of large eruption plumes from satellite measurements have been developed and are being refined.

The petrologic method assumes that the volatile contents of melt inclusions trapped in crystals represent those of the pre-eruption melt, and that the volatile contents of coexisting degassed matrix glass represent those of the melt after eruption (Gerlach *et al.*, 1994); the difference is taken as a measure of volatile degassing which, scaled up to the mass of erupted melt, yields estimates of volcanic emissions of SO<sub>2</sub> and other volatiles (Cl, F) during eruptions (Devine *et al.*, 1984a, Johnson, 1980, Palais and Sigurdsson, 1989, Sigurdsson, 1990, Sigurdsson *et al.*, 1985). Petrologic estimates of volatile emissions during eruptions are taken to be minimum estimates, since the assumptions inherent in the method may not be fully satisfied and lead to low results (Devine *et al.*, 1984a, Palais and Sigurdsson, 1989).

Direct measurement techniques using the correlation spectrometer (COSPEC) have been used since 1973 and the satellite-based Total Ozone Mapping Spectrometer (TOMS) has been used since 1978 to measure SO<sub>2</sub> emissions by remote sensing during volcanic eruptions (Gerlach *et al.*, 1994). The correlation spectrometer method uses the solar ultraviolet light transmitted and scattered by the earth's atmosphere as an illumination source. With the instrument directed towards the sky, the COSPEC measures the amount of ultraviolet light absorbed by SO<sub>2</sub> molecules present along the optical path in units of concentration-path length (ppm-m). Calibration is performed by using

a pair of internal standards with known concentrations of SO<sub>2</sub>. A profile of SO<sub>2</sub> concentration across the plume is measured at a right angle to the axis of the plume. The product of the concentration-path length (ppm-m), the width of the plume (m), and the plume speed (m/s) gives the emission rate of SO<sub>2</sub> which is reported in metric tons (10<sup>3</sup> kg) per day (t/d).

In addition to actual variation in the discharge rate, measured SO<sub>2</sub> emission rates may vary due to fluctuations in the wind speed and direction, change in conditions of cloud cover, change of sun angle and amount of solar ultraviolet radiation, and variation in plume opacity due to suspended ash particles. Uncertainty in COSPEC measurements arises from several operational factors including instrument calibration ( $\pm 2\%$ ) and chart reading error ( $\pm 4\%$ ), variation in aircraft speed during the measurement traverse ( $\pm 5\%$ ), operator variance affecting instrument operation and reduction of the data ( $\pm 5\%$ ), and measurement of wind speed and direction ( $\pm 10-40\%$ ) (Casadevall et al., 1981, Stoiber et al., 1983).

#### C.2.2. Development of Sulfur Emission Estimates for 1985 and 1990

The basic information on volcanoes was taken from the computerized data file of Simkin and Siebert (1994). This information includes the location and summit height of the volcano, activity, 'continuity' or 'discontinuity' of the eruptions, and the Volcanic Explosive Index (VEI). The monthly bulletins of the Scientific Event Alert Network (SEAN) which report seismic and volcanic activity and other such events were used as the primary source of information for the years of interest. Detailed information on volcanic activity such as height of the ash/gas column, SO<sub>2</sub> emission rates, start and stop date and emission type, were taken from the SEAN bulletins and

from the literature, for example, Caltabiano (1994), Casadevall (1994), Andres (1991), and Allard (1994). If available, minimum, average and maximum emission estimates were recorded. Ash/gas columns, lava flow, and other activity were reported from some volcanoes which were not listed to be active for the years 1985 and 1990 by Simkin and Siebert (1994); we have classified these cases as non-eruptive, degassing activities.

The TOMS estimates of the sulfur emissions from major volcanic activity in 1985 and 1990 were received from G. Bluth (personal communication, 1994). These data have been discussed in Bluth *et al.* (1993), Krueger *et al.* (1990), Shannon and Bluth (1994), and Schnetzler *et al.* (1994). The algorithms used to retrieve the volcanic emissions information from satellite measurements are currently under review.

Sulfur emissions were estimated for all volcanic activity in both years using the methodology of Stoiber *et al.* (1987) which bases the estimates on the type of eruption (continuous or episodic), the volcano location (at a convergent plate margin or other) and the VEI. We assumed that all volcanoes listed as active in the two years of interest are located at convergent plate margins. This assumption is based on the fact that only 5% of all volcanoes listed by Simkin and Siebert (1994) are of the divergent margin type (rift volcanism). Moreover, the 'spreading apart' of major plates is characterized by the relatively nonexplosive outpouring of fluid lava and *commonly* takes place one or more kilometers below the surface of the ocean (Simkin and Siebert, 1994). In addition, such volcanism is dominant in regions like East Africa and Iceland, where the spreading apart of the plates takes place above sea level (Simkin and Siebert, 1994). None of the volcanoes located in East Africa or Iceland are shown to be active for the years 1985 or 1990 in the

sources consulted.

### C.3.3. Initial Results

Table C.3.3.1 presents information on the volcanoes that were active in 1985 and the sulfur emissions that have been estimated. Table C.3.3.2 presents the same information for volcanoes active in 1990. Columns labeled 'Literature' present emissions estimates obtained from the SEAN bulletins and/or the literature (column labeled 'Mid'), with estimated ranges if available (columns labeled 'Low' and 'High'). Column labeled 'Simkin/Stoiber' presents the emissions estimates obtained by using the data from Simkin and Siebert (1994) and the methodology of Stoiber *et al.* (1987). Stoiber's methodology consistently overestimates sulfur emissions, often by an order of magnitude or more, when compared to the literature values which are volcano-specific and generally based on some measurement data. In addition to the inherent uncertainty in the correlation between the VEI and the sulfur emissions, there is also some uncertainty in the 'continuity/discontinuity' classification for the eruptions of some volcanoes. These uncertainties are significant for volcanoes of higher VEI. For example with a VEI of 3, a *non-continuous eruption* as per Stoiber would have  $2.87 \text{ Tg yr}^{-1}$  total sulfur flux as against  $0.075 \text{ Tg yr}^{-1}$  for a *continuous eruption*. Therefore we consider estimates based on the Stoiber methodology to be an upper value for sulfur emissions.

The columns labeled 'Current Estimate' present what we consider to be best estimates from the data sources accessed. These estimates are a combination of the literature values when available, defaulting to the Simkin/Stoiber estimate when no other data are available. Our current

estimate of the total sulfur emitted by volcanic activity in 1985 is 29.6 Tg SO<sub>2</sub> from 41 active volcanoes; literature estimates are available for only 12 (29%) of these volcanoes. Emissions based on literature values total 1.4 Tg SO<sub>2</sub>; the corresponding Simkin/Stoiber estimates total 10.8 Tg SO<sub>2</sub>. Our current estimate of the total sulfur emitted by volcanic activity in 1990 is 23.3 Tg SO<sub>2</sub> from 44 active volcanoes; literature estimates are available for only 15 (34%) of these volcanoes. Emissions based on literature values total 7.2 Tg SO<sub>2</sub>; the corresponding Simkin/Stoiber estimates total 12.5 Tg SO<sub>2</sub>. The much smaller difference in the totals for 1990 is the result of estimates of the activity of Mt. Etna during this year. Literature estimates, based on monthly COSPEC measurements of the plume, are higher (by a factor of 3) than the Simkin/Stoiber estimates, compensating for the greater difference between the estimates for the other volcanoes. The major difference in volcanic activity between the two years is provided by the eruptions of Mt. Etna and Redoubt; Mt. Etna showed only non-eruptive activity in 1985 and Redoubt showed no activity for this year. These results, although preliminary, provide graphic illustration of the variability of the volcanic source of sulfur emissions.

Work on this project continues. The on-going literature search is expected to provide volcano-specific estimates of the sulfur emissions from additional volcanoes active in these two years. Other data sources being consulted include the Smithsonian Institution Global Volcanism Program and World Data center A for Solid Physics in Boulder, CO.

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## List of Acronyms in Appendix C

**COSPEC:** COrelation SPECTrometer.  
**IPCC:** Intergovernmental Panel on Climate Change.  
**SEAN:** Scientific Event Alert Network.  
**TOMS:** Total Ozone Mapping Spectrometer.  
**VEI:** Volcanic Explosivity Index.

Table C.3.3.1 Estimate of Sulfur Emissions from Volcanic Activity in 1985

Volcano Name	Region	Sub region	Latitude	Longitude	Estimated Sulfur Emissions (Tg SO <sub>2</sub> )				
					Literature Low	Mid	High	Simkin/ Stoiber	Current Estimate
ETNA	Europe	Italy	37.73 N	15.00 E				1.640	1.640
STROMBOLI	(1)	Italy	38.79 N	15.21 E	0.201	0.243	0.285	2.090	0.243
FOURNAISE, PITON DE	AustralAsia	Indian O.-W	21.23 S	55.71 E				0.329	0.329
HEARD	(4)	Indian O.-S	53.11 S	73.51 E		0.010		2.090	0.010
RUAPEHU		New Zealand	39.28 S	175.57 E				0.329	0.329
NIUAFO'OU	Pacific	Tonga-SW Pacific	15.60 S	175.63 W				0.020	0.020
MANAM		New Guinea	4.10 S	145.06 E				1.640	1.640
LANGILA		New Britain	5.53 S	148.42 E				0.075	0.075
ULAWUN	(4)	New Britain	5.05 S	151.33 E		0.075		0.329	0.075
BAGANA		Bougainville	6.14 S	155.20 E				0.329	0.329
KAVACHI		Solomon Islands	9.02 S	157.95 E				0.170	0.170
PAGAN, MT.		Mariana Is land	18.13 N	145.80 E				1.640	1.640
TINAKULA		Santa Cruz Island	10.38 S	165.80 E				1.640	1.640
TANGKUBANPARAHU	South East &	Java	6.77 S	107.60 E				0.170	0.170
SEMERU	Far East Asia	Java	8.11 S	112.92 E				1.640	1.640
RAUNG		Java	8.13 S	114.04 E				2.090	2.090
SANGEANG API		Lesser Sunda Is	8.18 S	119.06 E				2.870	2.870
SOPUTAN		Sulawesi-Indonesia	1.11 N	124.73 E				2.090	2.090
API SIAU		Sangihe Is-Indones	2.78 N	125.48 E				1.640	1.640
CANLAON	(4)	Philippines-C	10.41 N	123.13 E		0.010		0.170	0.010
SUWANOSE-JIMA		Ryukyu Is	29.53 N	129.72 E				1.640	1.640
SAKURA-JIMA	(5)	Kyushu-Japan	31.58 N	130.67 E		0.262		1.640	0.262
ASO	(5)	Kyushu-Japan	32.88 N	131.10 E		0.016		0.329	0.016
FUKUTOKU-OKA-NO-BA		Bonin Is-Japan	24.28 N	141.52 E				0.084	0.084
TOKACHI		Hokkaido-Japan	43.42 N	142.68 E				0.170	0.170
GORELY		Kamchatka	52.56 N	158.03 E				1.640	1.640
BEZYMIANNY	(4)	Kamchatka	55.97 N	160.60 E		0.010		0.075	0.010
KLIUCHEVSKOI	(4)	Kamchatka	56.06 N	160.64 E		0.010		1.640	0.010

Table C.3.3.1 Estimate of Sulfur Emissions from Volcanic Activity in 1985

Volcano Name	Region	Sub region	Latitude	Longitude	Estimated Sulfur Emissions (Tg SO <sub>2</sub> )				
					Literature Low	Mid	High	Simkin/ Stoiber	Current Estimate
CLEVELAND		Aleutian Is	52.81 N	169.95 W				0.005	0.005
ST. HELENS, MT. (3)	America	US-Washington	46.20 N	122.18 W	0.014	0.016	0.019	0.170	0.016
KILAUEA		Hawaiian Is	19.43 N	155.29 W				0.329	0.329
LOIHI SEAMONT		Hawaii	18.92 N	155.25 W				0.020	0.020
PACAYA		Guatemala	14.38 N	90.60 W				0.329	0.329
SAN CRISTOBAL		Nicaragua	12.70 N	87.00 W				0.005	0.005
MASAYA		Nicaragua	11.98 N	86.16 W				0.329	0.329
CONCEPCION (4)		Nicaragua	11.54 N	85.62 W			0.010	0.329	0.010
ARENAL		Costa Rica	10.46 N	84.70 W				1.640	1.640
RUIZ (4)		Colombia	4.90 N	75.32 W			0.750	1.640	0.750
VILLARRICA		Chile-C	39.42 S	71.95 W				1.640	1.640
BEERENBERG JAN MAYEN		Atl-N-Jan Mayen	71.08 N	8.17 W				2.090	2.090
EREBUS, MT. (3)	Antarctica	Antarctica	77.53 S	167.17 E			0.011	0.329	0.011
<b>Totals</b>							<b>1.423</b>	<b>39.054</b>	<b>29.646</b>

Table C.3.3.1 Estimate of Sulfur Emissions from Volcanic Activity in 1985

**References**

Simkin/Stoiber (Simkin and Siebert, 1994; Stoiber *et al.*, 1987).

- (1) (Allard *et al.*, 1994).
- (2) (Caltabiano *et al.*, 1994).
- (3) SEAN bulletin.
- (4) TOMS data.
- (5) (Ohta 1988).
- (6) (Hirabayashi *et al.*, 1995).

Table C.3.3.2 Estimate of Sulfur Emissions from Volcanic Activity in 1990

Volcano Name	Region	Sub region	Latitude	Longitude	Estimated Sulfur Emissions (Tg SO <sub>2</sub> )				
					Low	Literature Mid	High	Simkin/ Stoiber	Current Estimate
STROMBOLI (1)	Europe	Italy	38.79 N	15.21 E	0.428	0.445	0.461	0.329	0.445
ETNA (2)		Italy	37.73 N	15.00 E		2.659		0.329	2.658
AKUTAN		Aleutian Is	54.13 N	165.97 W				0.170	0.17
FOURNAISE, PITON DE	AustraliaAsia	Indian O.-W	21.23 S	55.71 E				0.084	0.084
WHITE ISLAND		New Zealand	37.52 S	177.18 E				0.170	0.17
RUAPEHU		New Zealand	39.28 S	175.57 E				0.329	0.329
MANAM	Pacific	New Guinea-NE of	4.10 S	145.06 E				1.640	1.64
LANGILA		New Britain-SW Pac	5.53 S	148.42 E				1.640	1.64
ULAWUN		New Britain-SW Pac	5.05 S	151.33 E				0.005	0.005
RABAU CALDERA		New Britain-SW Pac	4.27 S	152.20 E				0.005	0.005
BAGANA		Bougainville-S W Pac	6.14 S	155.20 E				0.329	0.329
YASUR (3)		Vanuatu-SW Pacific	19.52 S	169.43 E	0.219	0.438	0.657	0.158	0.438
PAGAN, MT.		Mariana Is-C Pac	18.13 N	145.80 E				[1]	
AGRIGAN		Mariana Is-C Pac	18.77 N	145.67 E				0.005	0.005
MERAPI	South East and Java		7.54 S	110.44 E				0.329	0.329

Table C.3.3.2 Estimate of Sulfur Emissions from Volcanic Activity in 1990

Volcano Name	Region	Sub region	Latitude	Longitude	Estimated Sulfur Emissions (Tg SO <sub>2</sub> )					
					Low	Literature Mid	High	Simkin/ Stoiber	Current Estimate	
KELUT	Far East Asia	Java	7.93 S	112.31 E	0.150	0.150	0.150	0.910	0.15	
RAUNG		Java	8.13 S	114.04 E				1.640	1.64	
GAMALAMA		Halmahera-Indonesia	0.80 N	127.33 E				0.075	0.075	
SAKURA-JIMA		Kyushu-Japan	31.58 N	130.67 E		0.660		1.640	0.66	
UNZEN		Kyushu-Japan	32.75 N	130.30 E		0.008		0.170	0.008	
ASO		Kyushu-Japan	32.88 N	131.10 E		0.016		0.020	0.016	
ASAMA		Honshu-Japan	36.40 N	138.53 E		0.037		2.090	0.036	
OSHIMA		Izu Is-Japan	34.73 N	139.38 E				2.090	2.09	
KLIUCHEVSKOI		Kamchatka	56.06 N	160.64 E				0.075	0.075	
REDOUBT		America	Alaska-SW	60.48 N	152.75 W		1.127		2.870	1.127
ST. HELENS, MT.			US-Washington	46.20 N	122.18 W				1.640	1.64
KILAUEA			Hawaiian Is	19.43 N	155.29 W				0.084	0.084
COLIMA VOLC COMPLEX	Mexico		19.51 N	103.62 W				0.084	0.084	
SANTIAGUITO DOME	Guatemala		14.76 N	91.55 W	0.012	0.018	0.023	0.075	0.018	
FUEGO	Guatemala		14.47 N	90.88 W	0.059	0.070	0.082	[1]	0.07	
PACAYA	Guatemala		14.38 N	90.60 W		0.011		1.640	0.011	
ARENAL	Costa Rica		10.46 N	84.70 W				1.640	1.64	

Table C.3.3.2 Estimate of Sulfur Emissions from Volcanic Activity in 1990

Volcano Name	Region	Sub region	Latitude	Longitude	Estimated Sulfur Emissions (Tg SO <sub>2</sub> )			Simkin/ Stoiber	Current Estimate
					Low	Mid	High		
POAS		Costa Rica	10.20 N	84.23 W				0.329	0.329
RUIZ	(3)	Colombia	4.90 N	75.32 W		0.699		1.640	0.699
GALERAS	(3)	Colombia	1.22 N	77.37 W		0.873		0.329	0.873
REVENTADOR		Ecuador	0.08 S	77.66 W				0.005	0.005
GUAGUA PICHINCHA		Ecuador	0.17 S	78.60 W				0.170	0.17
SABANCAYA		Peru	15.78 S	71.85 W				1.640	1.64
LASCAR		Chile-N	23.37 S	67.73 W				0.093	0.093
PULAR		Chile-N	24.18 S	68.05 W				0.093	0.093
LONQUIMAY		Chile-C	38.37 S	71.58 W				1.640	1.64
LLAIMA		Chile-C	38.70 S	71.70 W				0.093	0.093
KICK-'EM-JENNY		W Indies	12.30 N	61.63 W				0.020	0.02
EREBUS, MT.	(3)	Antarctica	77.53 S	167.17 E		0.037		0.329	0.037
<b>Totals</b>						<b>7.245</b>		<b>28.631</b>	<b>23.348</b>

**Notes**

[1] VEI information not given in Simkin and Siebert (1994).

**References**

Simkin/Stoiber (Simkin and Siebert, 1994, Stoiber *et al.*, 1987).

Table C.3.3.2 Estimate of Sulfur Emissions from Volcanic Activity in 1990

- (1) (Allard *et al.*, 1994).
- (2) (Caltabiano *et al.*, 1994).
- (3) Scientific Event Alert Network (SEAN) bulletin.
- (4) TOMS data.
- (5) (Ohta, 1988).
- (6) (Hirabayashi *et al.*, 1995)