Tellus (1983), *35B*, 290–293

LETTERS TO THE EDITOR

Comment on "On the atmospheric input of sulfur into the ocean" by E. Mészáros

By JENNIFER A. LOGAN *Center for Earth and Planetary Physics, Harvard University, Pierce Hall, 29 Oxford Street, Cambridge, MA 02138, USA*

(Manuscript received December 30, 1982)

Mészáros (1982) recently presented an analysis of atmospheric input of sulfur into the ocean. He concluded that wet deposition provides the major sink for sulfur, of magnitude 267 Tg S yr^{-1}, of which 120 Tg S yr^{-1} is derived from sea-salt. Mechanisms which might account for additional input of sulfur to the atmosphere of magnitude 150 Tg S yr^{-1} have not been identified (Granat et al., 1979; Logan et al., 1978; Sze and Ko, 1980; Barnard et al., 1982). Earlier analyses of precipitation data suggested a much smaller removal rate for non-sea-salt, or excess, sulfur, and hence implied a smaller marine source for S (Granat et al., 1976). Analysis of recent precipitation data for island sites indicates that Mészáros overestimated the marine deposition rate of excess S by a factor of between 2.5 and 8.

Table 1. *Concentrations of sulfur in rainfall on Samoa**

Year collected	Rainfall (cm)	No. months data	Total S (μeq/l)	Excess S (μeq/l)
1975	43	6	28	19
1976	63	8	17	6
1977	173	12	45	31
1977†	144	10	18	5
1978	148	12	12	4
1975–77‡	279	26	36	21
1975–77†	250	24	20	8
1975–78†	398	36	17	6

* Volume weighted concentrations are given. Excess sulfur was calculated from measurements of sodium and the mean composition of ocean water, $SO_4^=:Na^+ = 0.25$. Data are from WMO (1977–1981).

† Data for November and December 1977 are omitted.

‡ All data are included.

Mészáros' conclusions relied on monthly precipitation data for $SO_4^=$ and Na^+ collected on Samoa and Hawaii in 1975–1977 (WMO 1977–1980). Average values for Samoa during this period were strongly biased, however, by very high concentrations (150, 230 μeq l^{-1}) reported for two particular months, November and December, 1977. The mean concentration derived for excess S is reduced from 21 μeq l^{-1} to 8 μeq l^{-1} if these two months of data are omitted, and is further reduced to 6 μeq l^{-1} if data for 1978 are included (see Table 1). Pszenny et al. (1982) and Bogen et al. (1979) reported even lower concentrations of excess S in rain on Samoa, ~2 μeq l^{-1}. There is a difference of a factor of 5 between measurements of sulfur in rain at Mauna Loa Observatory, Hawaii, given by WMO (1977–1980), 21 μeq l^{-1}, and the results of Miller for 1981, 4 μeq l^{-1} (private communication). Precipitation samples collected by WMO on a monthly basis appear to give values for excess S which are higher by factors of 2–5 than samples collected at the same locations by other workers employing different protocols for sampling and analysis.

Concentrations of excess S in rain at Amsterdam Island in the southern Indian Ocean, 9 μeq l^{-1} (Galloway et al., 1982), are higher than values reported for the Pacific Islands of Samoa and Hawaii. Concentrations of excess S for the three island sites, 1.5–9 μeq l^{-1}, when combined with annual rainfall at these locations, imply marine deposition rates for excess S of 0.5–1.6 kg S ha^{-1} yr^{-1} (see Table 2). By contrast, the concentration

Table 2. *Measurements of sulfur in rainfall at island locations**

	Location	Rainfall (cm)	Total-S		Excess-S	
			(μeq/l)	(kg/ha/yr)	(μeq/l)	(kg/ha/yr)
Samoa	14° S 170° W	208	27†	9.0	1.5	0.5
			26‡	8.7	2.3	0.8
Amsterdam Island§	38° S 77° E	112	31	5.5	8.8	1.6
Hawaii, MLO¶	19° N 155° W	(80)	4		(4)‖	(0.5)‖

* Volume weighted concentrations are given. Excess sulfur was calculated from measurements of sodium and the mean composition of ocean water, $SO_4^=:Na^+ = 0.25$. Deposition rates were calculated from annual mean precipitation rates at Samoa and Amsterdam Island.

† Results of 33 event samples from February and July, 1981 (Pszenny et al., 1982; Pszenny and Duce, 1981).

‡ Results of monthly samples obtained in wet–dry collectors from April 1976–March 1978 (Bogen et al., 1979).

§ Results from 27 event samples in 1980–1981 (Galloway et al., 1982).

¶ Results from samples collected every three days in 1981 (Miller, private communication).

‖ There should be little contribution from sea-salt sulfur at the altitude of Mauna Loa Observatory, 3400 m. The deposition rate of excess S in the vicinity of Hawaii is based on a concentration of ~4 μeq l^{-1} and marine rainfall of ~80 cm yr^{-1} in this region (Dorman and Bouke, 1979; Korzoun, 1977).

of excess S adopted by Mészáros, 21 μeq l^{-1}, when combined with average marine rainfall, 127 cm yr^{-1}, implies a deposition rate of 4.1 kg S ha^{-1} yr^{-1}.

Deposition rates in the range 0.5–1.6 kg S ha^{-1} yr^{-1} correspond to global removal rates for excess S of 18–58 Tg S yr^{-1} over the oceans, considerably smaller than the rate derived by Mészáros, 150 Tg S yr^{-1}. Biogenic emissions of reduced S compounds may provide a marine source for S of about 30 Tg yr^{-1} (Nguyen et al., 1978; Barnard et al., 1982). Higher concentrations of excess sulfur in rain on Amsterdam Island compared to Samoa may result from proximity to more productive ocean waters (Koblentz-Mishke et al., 1970), and hence larger biogenic sources of S. Data for S in marine rain and for emission rates of gaseous precursors are very sparse. Estimates of marine sources and sinks of sulfur based on these fragmentary data are not, however, inconsistent as was implied by the analysis of Mészáros.

REFERENCES

Barnard, W. R., Andreae, M. O., Watkins, W. E., Bingemer, H. and Georgii, H. W. 1982. The flux of dimethyl sulfide from the oceans to the atmosphere. *J. Geophys. Res. 87*, 8787–8794.

Bogen, D. C., Nagourney, S. J. and Torquato, C. 1979. Measurements of total, wet and dry deposition at American Samoa. Report EML-361, Environmental Measurements Laboratory, U.S. Department of Energy, New York.

Dorman, C. E. and Bourke, R. H. 1979. Precipitation over the Pacific Ocean, 30°S to 60°N. *Mon. Wea. Rev. 107*, 896–910.

Galloway, J. N., Likens, G. E., Keene, W. C. and Miller, J. M. 1982. The composition of precipitation in remote areas of the world. *J. Geophys. Res. 87*, 8771–8786.

Granat, L., Hallberg, R. O. and Rodhe, H. 1976. The global sulfur cycle. *Ecol. Bull. 22*, 89–134.

Koblentz-Mishke, O. J., Volvovinsky, V. V. and Kabanova, J. G. 1970. Plankton primary production of the world ocean. In *Scientific exploration of the South Pacific* (ed. W. S. Wooster). National Academy of Sciences, Washington, D.C., 183–193.

Korzoun, V. I. 1977. *Atlas of World Water Balance.* Hydrometeorological Publishing House, the UNESCO Press, Paris.

Logan, J. A., McElroy, M. B., Wofsy, S. C. and Prather, M. J. 1979. Oxidation of CS_2 and COS: sources for atmospheric SO_2. *Nature 281*, 185–188.

Mészáros, E. 1982. On the atmospheric input of sulfur into the ocean. *Tellus 34*, 277–282.

Nguyen, B. C., Gaudry, A., Bonsang, B. and Lambert, G. 1978. Re-evaluation of the role of dimethyl sulfide in the sulfur budget. *Nature 275*, 637–639.

Pszenny, A. and Duce, R. 1981. Ionic composition of Samoan rain: preliminary results. *SEAREX Newsletter 4*, 19–26.

Pszenny, A. A. P., MacIntyre, F. and Duce, R. A. 1982. Sea-salt and the acidity of marine rain on the

windward coast of Samoa. *Geophys. Res. Lett. 9*, 751–754.

Sze, N. D. and Ko, M. K. W. 1981. Photochemistry of COS, CS_2, CH_3SCH_3 and H_2S: implications for the atmospheric sulfur cycle. *Atmos. Environ. 14*, 1223–1239.

World Meteorological Organization. 1977–1981. Global monitoring of the environment for selected environmental parameters. BAPMoN data for 1975–1978, available from National Climate Centre, Ashville, N.C. 28801.