Regarding the role of the atmosphere in the formation of the chemical composition of the waters of Lake Baikal

T.V. Khodzher, V.A. Obolkin, and V.L. Potemkin

Limnological Institute, Siberian Branch of the Russian Academy of Sciences, Irkutsk
Received March 3, 1999

Quantitative estimates are made of the influx of macro- and micro-elements from the atmosphere into the surface waters of Lake Baikal from long-term aerosol data and are compared with earlier data. It is concluded that a substantial part of the base-metal contribution to the chemical balance of the lake comes through the atmospheric channel.

Introduction

Quantitative estimates of the contribution of the primary sources of the influx of chemical substances into Lake Baikal are important for monitoring and prediction of long-term changes in the chemical composition of its waters.

Although the quality and volume of initial data for such estimates can still not be considered to be satisfactory, efforts to calculate the main components of the chemical balance of Baikal—river runoff and atmospheric fallout—have already been undertaken.[1,2] As could have been expected, these calculations indicate an extremely small contribution of atmospheric fallout to the influx of most chemical substances to the waters of Lake Baikal in comparison with the contribution of river runoff (in this regard, it has still not been taken into account that river runoff is also part of the atmospheric precipitation into the basin of the lake). So far, estimates of the fluxes of chemical substances from the atmosphere into the waters of the lake have been based, for the most part, on data of the chemical composition of snow cover.[2] However, snow cover on the lake exists for all of three months and is often swept in one direction or another by the wind; understandably, extrapolation of such winter data to the entire year renders annual estimates based on it very approximate. In the last few years, extensive year-round data on the chemistry of atmospheric aerosol in the Baikal region, including above the lake itself,[3–5] have been obtained, which makes it possible to refine the current estimate of the contribution of the atmospheric component of the chemical balance of the lake and to compare results based on independent data: from the accumulation in snow cover (data from 1976–1984) and from the precipitation of aerosol particles from the atmosphere (data from 1991–1997).

Data and methods

As our input data for calculating atmospheric fallout onto the waters of the lake, we used data on the concentrations of chemical elements in atmospheric aerosols both above the southern shore of the lake (over the course of several years and for all seasons of the year) and directly above the lake itself, obtained during summer cruises on research vessels. Data on the concentrations of chemical elements and on methods of analysis can be found in Refs. 4 and 5 (Table 1).

To a first approximation, the fluxes of dry precipitation of chemical elements from the atmosphere onto any surface can be calculated as the product of the concentration of the element in the atmosphere and its rate of precipitation. Although the precipitation rate of an element is a quite indeterminate quantity, depending on the size and density of the particles in which the given element is found and also on the meteorological conditions and the nature of the underlying surface, as a first approximation we can use the mean values of the precipitation rates of elements, obtained in numerous experiments in different regions of the world.[6] We used such mean estimates in our calculations that follow.

However, besides dry precipitations it is also important to estimate the magnitude of moist fallout. Experience shows that even in moderately humid regions the contribution of moist precipitation of elements is, as a rule, not less, and is often greater, than that of dry precipitation. We have attempted to determine the amount of moist fallout in the region under consideration by estimating the washout coefficient of sulfate precipitations from the data of Ref. 5, which presents results of direct measurements of sulfates and nitrates in precipitations (rain and snow) at several Baikal meteorological stations. The following estimates of the washout coefficients were obtained: for 1 cm of rain K = 18,000; for 1 cm of snow K = 1700. For the annual estimates of moist fallout, we used the following mean climatic data:

	Winter	Summer	Year
Amount of atmospheric precipitation (cm)	10	45	55
Number of days with atmospheric precipitations (> 0.1 mm)	33	62	95

0235-6880/99/06 493-04 $02.00

Table 1. Mean concentrations of elements in air (C, μg/m^3) and annual intensity of their atmospheric fallout (P, mg/m^2·yr.) onto the waters of Lake Baikal along basins.

Element	W, cm/sec	C, southern Baikal	C, middle Baikal	C, northern Baikal	P, southern Baikal	P, middle Baikal	P, northern Baikal	P, Dorset, Ontario[6]
Na	2.5	529	290	247	750	370	320	–
Mg	2.5	298	104	–	425	112	–	30.2
Al	2.5	550	322	221	776	357	284	
Ca	2.5	464	244	177	648	268	134	161
Sc	2.5	0.11	0.05	0.04	0.2	0.06	0.05	–
V	1	1.15	0.62	0.42	1.3	0.5	0.4	0.6
Cr	2	1.8	2.0	–	2.3	2.0	–	–
Mn	1.2	8.1	5.0	5.4	9.1	3.1	–	3.0
Fe	2.5	354	201	190	494	223	146	–
Co	2	0.23	0.15	1.0	0.3	0.15	1.2	–
Cu	0.6	54	37	22	53	25	19	–
Zn	0.5	9.3	26	35	19	–	–	7.0
As	0.5	0.48	0.05	0.14	0.4	0.04	0.1	0.4
Se	0.4	–	0.07	0.08	0.1	–	–	–
Br	0.5	2.3	3.26	1.1	2.3	2.2	0.9	7.3
cD	0.5	8.1	1.8	–	2.8	–	1.3	–
In	0.6	–	–	–	4.9*	–	–	–
Sb	0.5	0.2	0.4	0.3	0.2	0.2	0.2	–
I	0.6	0.5	0.2	0.3	0.4	–	–	–
Cs	1.0	0.09	0.02	–	0.1	0.02	–	–
Ba	1.5	11.3	11.0	14.7	13	9.7	16.0	–
Sr	4.0	–	–	–	5.7*	–	–	–
Pb	0.5	–	–	–	5.5*	–	–	17.1
Ce	3.0	0.7	0.3	0.3	1.0	–	–	-
Sm	1.5	0.07	0.09	0.05	0.1	0.08	0.05	–
Th	2.0	0.1	0.04	0.01	0.15	0.04	0.05	–

* Calculated on the basis of data from the southern shore of Lake Baikal.

Table 2. Annual dry and moist fallout of chemical elements onto the waters of Lake Baikal, t/yr.

	Fluxes from the aerosol data				From the accumulation in snow	
Element	southern Baikal	middle Baikal	northern Baikal	The entire lake	Published data[2]	The same, but corrected to agree with the aerosol data
Na	500	3400	4400	13300	1700	7800
Mg	3100	1200	–	5800	44200	7100
Al	5700	3700	3900	13300	4000	13900
Ca	4800	2800	1800	9400	15000	10000
Sc	1.5	0.6	0.7	2,8	1.9	2.8
V	9.6	5.2	5.4	22	29	25
Cr	17.0	20.9	20	58	26.7	66
Mn	70	30	30	130	100	220
Fe	3600	2300	2000	7900	3600	7700
Co	2.8	1.6	1.6	6.0	2.0	9.1
Cu	400	260	260	920	15	362
Zn	140	–	–	3008	57	344
aS	2.9	0.4	1.4	4.7	3.3	10.8
sE	0.7	–	–	2.0*	–	–
Br	17.00	23.0	12.3	52.0	7.5	12.5
Cd	20.7	18	17.7	56	–	–
In	36.2	–	–	90*	–	–
Sb	1.5	2.1	2.7	6.3	0.65	7.2
I	2.9	2	2	7	–	–
Cs	0.7	0.2	0.2	1.0	0.5	2
Ba	96	102	220	428	9.8	270
Sr	42	–	–	120*	20	–
Pb	41	–	–	120*	20	–
Ce	7.4	–	–	20*	10.7	34.8
Sm	0.7	0.8	0.7	2.2	–	–
Th	1.1	0.4	0.7	2.2	1.5	3.9

* Rough estimate, made by extrapolating the data from the southern shore of Lake Baikal.

Thus, dry precipitation was defined as the product of the concentration of the element in the atmosphere by its rate of precipitation and the period of time without atmospheric precipitations; moist precipitation was defined as the product of the seasonal mean concentration of the element in the atmosphere and the washout coefficient and the amount of atmospheric precipitation. Results of our calculations are shown in Table 2. For comparison, the second-to-the-last column lists published data obtained mainly from the accumulation of substances in snow cover on lake ice.[2,3]

Discussion

As can be seen from Table 2, there is quite good agreement in the ordering of elements, but there are also substantial differences (especially for the micro-elements); therefore, at the present stage, where the transparency and accuracy of neither method is yet ideal, it is important to find a way of estimating which of the figures is closer to reality. One possible criterion is the ratio of elements in the primary source of atmospheric fallout, namely, atmospheric aerosol. This ratio should be close to the ratio of elements in the Earth's crust. Upon closer inspection, a noncorrespondence in the values of the fallout of some of the macro-elements is apparent in the published data: in particular, such elements as sodium and aluminum should not be present in lesser amounts than magnesium and calcium since in the primary source of aerosol—soil and the Earth's crust the content of the first two is higher. In addition, for the southern part of the lake there is an additional—well-known, in fact, notorious—source of anthropogenic sodium, the Baikal paper mill.

Thus, the new estimates of these elements from the aerosol data look more realistic. The same may be said about such an element as iron. To verify this assertion, we made an attempt to correct the published data by recalculating them in accordance with the ratio of elements in aerosol, taking as our reference point elements having the most similar values for both methods. The results of this recalculation are shown in Table 2 (last column). As can be seen from Table 2, the series of elements on an order-of-magnitude scale has been brought substantially in line with the fluxes calculated from the aerosol data.

However, a series of micro-elements remains whose atmospheric fallout sometimes exceeds the published data by an order of magnitude or more (Cu, Zn, Br, Sb, Ba, Sr, Pb). To analyze the revealed discrepancies, let us first analyze the differences in the composition of the aerosol above the waters of Lake Baikal and above its shore (Fig. 1).

As Fig. 1 shows, the concentration of copper above the lake, in contrast to the other elements, exceeds its value above the shore by a factor of 10, which clearly points to contamination of the samples onboard the ship (this could be the result of the brushes of the ship's electric engines, etc.). Therefore, the copper fluxes from the atmosphere should be reduced by a factor of 10 or excluded from consideration (in what follows we exclude this element from the discussion). The remaining elements, as it should be, have a somewhat reduced concentration in the aerosol above the lake itself (in comparison with the shore) and no effects of ship-based sources for them were detected. The only exception to this is sodium, whose concentrations above the lake and above dry land were similar in the southern part of the lake and linked with the effect of the Baikal paper mill. Thus, it is still not possible to give a convincing explanation of such substantial discrepancies as do exist in the fluxes of several of the above-indicated micro-elements calculated by us and taken from Ref. 2.

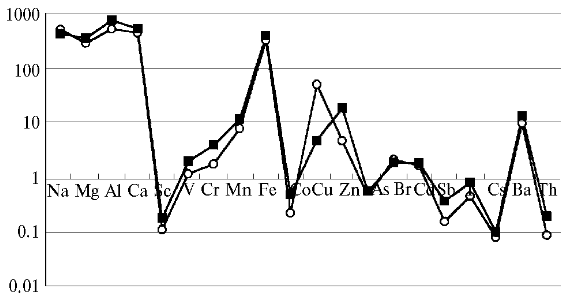

Fig. 1. Concentrations of elements in atmospheric aerosol above the shore and waters of the southern part of Lake Baikal: lake (–○–), shore (-■-).

It can be seen from the data of Table 3, how much the contribution of the atmosphere to the total influx of chemical elements to Lake Baikal changed when the new data were taken into account (the influx of elements from river runoff was taken from Refs. 7 and 8). Note that the accuracy of the estimates of the influx of substances brought into the lake by river runoff also comes under question (especially for the insoluble elements) and is not higher than the accuracy of the estimates of atmospheric fallout.

Table 3. Fraction of chemical elements from atmospheric fallout in the total influx into Lake Baikal, %.

Element	Published data (on accumulation in snow)[7]	From the aerosol data
Na	0.7	5
Mg	1.7	2
Al	2.1	6
Ca	1.4	1
Sc	7.0	11
V	7.6	5
Cr	17.3	31
Mn	2.0	2
Fe	2.2	5
Co	5.0	12
Cu	12.0	–
Zn	12.1	42
Sa	3.0	5
Br	3.0	17
Sb	1.4	12
Cs	2.2	4
Ba	0.3	10
Pb	25.3	67
Ce	2.7	4
Th	4.5	6

Conclusions

A comparison of the values of atmospheric fallout of chemical elements onto the waters of Lake Baikal obtained by two independent methods (from the accumulation of these elements in snow and from their fallout from aerosols) has revealed the following.

Discrepancies in the influx of macro-elements into Lake Baikal are as high as two to threefold while calculations of the fluxes of these elements from the atmosphere based on the snow data underestimate their values as a rule and do not correspond to the actual distribution of elements in the atmosphere. However, the contribution of the atmosphere to the macro-element fluxes based on the aerosol data remains insignificant—with a maximum value of 5–6% for iron and aluminum.

The discrepancies in the micro-element and base-metal fluxes are significantly greater: for such elements as zinc, strontium, bromine, and lead the fluxes according to the aerosol measurements are 5–6 times higher, and for antimony and barium, 10–30 times higher. Correspondingly, the fraction of the atmospheric component grows substantially: for base metals (chromium, zinc, lead) by 30–60%, and for the remaining micro-elements, by 5–15%. Most of the enumerated elements can also have an anthropogenic origin; therefore, a closer examination of their fluxes (both from the atmosphere and from river runoff) is required to iron out the substantial differences in the estimates of their balance.

The fluxes of many of the micro-elements from the atmosphere, calculated from the aerosol data, can be taken as maximum estimates since they were obtained for the southern (more polluted) part of the lake and were then extrapolated to the entire lake.

References

1. *Monitoring of the State of Lake Baikal* (Gidrometeoizdat, Leningrad, 1991), 261 pp.
2. V.A. Vetrov and A.I. Kuznetsova, *Micro-Elements in Natural Media of the Region of Lake Baikal* (Izdat. SO RAN NITs OIGGM, Novosibirsk, 1997), 230 pp.
3. V.A. Obolkin, V.L. Potemkin, and T.V. Khodzher, Geograf. Prirodn. Resursy, No. 4, 41–48 (1996).
4. T.V. Khodzher, V.A. Obolkin, V.L. Potemkin, U. Tomza, and K. Ran, Khimiya v Interesakh Ustoichivogo Razvitiya, No. 5, 547–551 (1997).
5. V.A. Obolkin and T.V. Khodzher, Meteorol. Gidrol., No. 7, 71–76 (1990).
6. L.A. Barrie, J. Geophys. Res. **93D**(4), 3773–3788 (1988).
7. E. Callender and L. Granina, Limnol. Oceanogr. **42**(1), 148–155 (1997).
8. L. Granina, U. Tomza, and R. Arimoto, Nucl. Instrum. Methods Phys. Res. (1999) (in press).