Application of enriched stable mercury Isotopes to the Lake 658 watershed for the METAALICUS project, at the Experimental Lakes Area, northwestern Ontario, Canada, 2001-2007

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by

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ABSTRACT

Sandilands, K.A., C. A. Kelly, J.W.M. Rudd, M. T. Tate, H. Hintelmann, B. Dimock, and R. C. Harris. 2008. Application of enriched stable mercury isotopes to the Lake 658 watershed for the METAALICUS project, at the Experimental Lakes Area, northwestern Ontario, Canada, 2001-2007. Can. Tech. Rep. Fish. Aquat. Sci. 2813: vii + 40 p.

Between 2000 and 2007 the Lake 658 watershed at the Experimental Lakes Area in northwestern Ontario was the site of a whole-ecosystem mercury addition experiment designed to study the relationship between atmospheric deposition of mercury and mercury accumulation in fish, and biogeochemical processing of mercury within watersheds. Atmospheric loading was increased to approximately six times the long term wet deposition rate of mercury in this area. A different enriched stable mercury isotope was applied to each part of the watershed (upland, wetland and lake). Additions to the upland and wetland were done via aircraft, loadings to the lake were done by injection into the epilimnion from a boat. The application to the upland, wetland, and lake totalled 111, 107 and 154 µg/m^2 (Averaging 15.8, 17.8 and 22.0 ug/m^2/yr) respectively.

KEY WORDS: Mercury, METAALICUS, ELA, Atmospheric Loading

RÉSUMÉ

Sandilands, K.A., C. A. Kelly, J.W.M. Rudd, M. T. Tate, H. Hintelmann, B. Dimock, and R. C. Harris 2008. Application d'isotopes enrichis stables de mercure dans le bassin versant du lac 658 pour le projet METAALICUS, dans la région des lacs expérimentaux du nord-ouest de l'Ontario, Canada, 2001-2007. Rapp. tech. can. sci. halieut. aquat. 2813 : vii, 40 p.

Entre 2000 et 2007, le bassin versant du lac 658 dans la région des lacs expérimentaux du nord-ouest de l'Ontario a été le site d'une expérience panécosystémique d'ajout de mercure ayant pour objet l'étude du lien entre les dépôts atmosphériques de mercure et l'accumulation de mercure dans les poissons, ainsi que la transformation biogéochimique du mercure au sein des bassins versants. La charge atmosphérique a été augmentée à environ six fois la valeur des retombées humides à long terme de mercure dans cette région. Un isotope enrichi stable de mercure différent a été appliqué dans chaque partie du bassin versant (milieux secs, milieux humides et le lac). Le mercure a été dispersé par avion au-dessus des milieux secs et des milieux humides et injecté à partir d'une embarcation dans l'épilimnion du lac. La charge totale de mercure appliquée dans les milieux secs, les milieux humides et le lac a été respectivement de 111, 107 et 154 µg/m^2 (en moyenne 15,8, 17,8 et 22,0 ug/m^2/an).

MOTS CLÉS : mercure, METAALICUS, RLE, charge atmosphérique

INTRODUCTION

The **M**ercury **E**xperiment **T**o **A**ssess **A**tmospheric **L**oading **I**n **C**anada and the **U**nited **S**tates (METAALICUS) project is a whole ecosystem manipulation experiment that began in 1999 at the Experimental Lakes Area (ELA), northwestern Ontario, Canada. The objectives of METAALICUS are to determine: 1) the relationship between atmospheric mercury (Hg) loading and Hg concentrations in fish and other biota, 2) the relative importance of newly deposited Hg onto wetland, upland, and the lake surface as sources of mercury to fish and other biota, 3) how quickly Hg concentrations in the lake, upland, and wetland respond to changes in Hg deposition rate, and 4) the relative importance of newly deposited Hg as compared to Hg present in the ecosystem from previous years.

The overall design of the experiment involved loading a watershed with an amount of Hg (22 $\mu g/m^2/yr$) equivalent to approximately six times the rate of wet ambient Hg deposition measured from 1990-2006 (3.6 $\mu g/m^2/yr$, Graydon et al. 2008).
To distinguish the relative importance of Hg deposition in different areas of the watershed, the upland, wetland, and the lake, were sprayed with different stable isotopes of mercury, ^{200}Hg, ^{198}Hg, ^{202}Hg respectively.

This report is an update to Sandilands et al. (2005), which detailed mercury applications in 2001, 2002 and 2003. This report summarizes data associated with the application of isotopes for all seven years, with updates to 2001-03 since new data (more accurate mapping) became available. This report also presents an estimate of the spatial distribution of Hg isotopes sprayed on the upland and wetland.

METHODS

APPLICATION OF Hg ISOTOPES TO THE LAKE 658 WATERSHED

DEFINITION OF TERMS

Hg solutions applied in this experiment were enriched to varying degrees with the appropriate isotope. In this report, the term *Spike Hg* is defined as isotopically enriched inorganic Hg(II) applied to the ecosystem. Applications of Spike Hg contained an enriched major isotope and other minor Hg isotopes. For example, the Hg solution applied to the upland was enriched with ^{200}Hg (80.45%), with the remaining 19.55% comprising other Hg isotopes. Therefore, the amount of *Upland Spike Hg* applied is the amount of ^{200}Hg applied divided by 0.8045. For environmental samples, such as lake water, the amount of *Upland Spike Hg* is the amount of ^{200}Hg measured in the sample and calculated to be in excess of the ambient ^{200}Hg, divided by 0.8045. The terms *Wetland Spike Hg* and *Lake Spike Hg* are used in the same way using their respective enrichments. *Ambient Hg* refers to Hg not enriched in any isotope that was deposited under normal circumstances to the watershed.

For the upland and wetland areas, the *release rate* is defined as the amount of Hg that was delivered from the spray boom of the aircraft minus any losses to the aircraft tank or spray delivered outside the spray block or watershed. The *average application rate* ($\mu gHg/m^2$) is the mass of Spike Hg sprayed within the boundary plus the mass of Spike Hg sprayed on the shorelines by hand, divided by the total upland area.

PREPARATION OF ISOTOPE SOLUTIONS

Enriched stable Hg isotopes (^{198}Hg, ^{200}Hg and ^{202}Hg) were purchased from Trace Sciences International in Richmond Hill, Ontario, Canada. The isotopes were produced by the Kurchatov Institute laboratory in Moscow, Russia.

From 2001 to 2004, the Hg was received as mercuric chloride ($HgCl_2$), at the Freshwater Institute, Winnipeg. Spike Hg solutions were made by diluting with 5% nitric acid, with volumes ranging from 100 to 250 mL, and concentrations of 0.5 to 38 g Hg/L. In 2005-07, the Hg isotopes were received as elemental Hg, at Trent University, Peterborough. The elemental Hg was converted to mercuric nitrate. Elemental Hg was transferred into a 50 ml beaker. 10 mL of concentrated HNO_3 was added. Hg was allowed to digest (oxidized) over-night at room temperature. Depending on the amount of Hg, the resulting solution of Hg(II) in nitric acid was yellow to dark green in color. The concentrated solution was poured into a Teflon bottle and diluted by adding approximately 50 ml of deionized water and sent to the Freshwater Institute in Winnipeg.

Hg solutions were then mixed to the volumes and concentrations required for each application. For details of solubilization and blending of spike Hg solutions in 2001-03 see Sandilands et al. (2005).

New batches of isotopes were purchased each year, and the % enrichments differed slightly from year to year. The percentages of each enriched isotope in Table 1 are the guaranteed ratios provided by Trace Sciences (shaded data). Each year, sub-samples of the Hg isotope solutions received from Trace Sciences were also analyzed by Trent University for verification (Table 1, non shaded data).

In 2004-06 Spike Hg solutions also contained remainders of Hg solutions from previous years, and in some cases, the newly acquired Hg from Trace Sciences was more enriched than that used in previous years. As a result the isotopic Hg had to be blended with natural Hg to obtain similar isotope abundances. Since these solutions were more complicated, subsamples were used to determine the final concentration of the solutions used when possible.

DETERMINATION OF RELEASE RATES AND APPLICATION RATES

For the upland and wetland areas, the *release rate* is defined as the amount of HgT that was delivered from the spray boom of the aircraft minus any losses to the aircraft tank or sprayed outside the spray block or watershed (see Figure 1 for a map of the spray blocks). The mass of Spike Hg sprayed inside the watershed boundary was determined by mapping using GIS (see section on mapping of Spike Hg spraying).

The *average application rate* ($\mu gHg/m^2$) is the aerial application of Spike Hg sprayed within the watershed boundary plus the mass of Spike Hg sprayed on the shorelines by hand. The calculation of average application rates assumes that Hg released from the aircraft within the watershed boundary fell straight to the upland or wetland surface, and that no spray drift occurred. Caution should be used in interpreting the application rates, as some spray drift certainly did occur; however we are unable to quantify it here. Therefore application rates should be viewed as the *upper limits* of Hg application. In the case of lake application, there were no similar losses to equipment or spray drift, thus the application rate is the *true* application rate to the lake.

TANK LOSSES

The amount of Spike Hg actually delivered from the aircraft was less than the amount of Spike Hg added to the aircraft tank. Experiments were done to estimate the losses of Hg to the tank surfaces of the aircraft (see Sandilands et al. 2005 for a description of these experiments, and calculation method for estimating tank losses for each spray event.

MAPPING OF SPIKE HG SPRAYING

The upland and wetland were sprayed with Hg isotopes (enriched with ^{200}Hg and ^{198}Hg, respectively) using a Cessna 188 AG truck, commonly used for chemical applications in agriculture and forestry. Upland and wetland Hg isotopes were mixed with acidified lake water immediately prior to spraying (Sandilands et al. 2005). The amounts of Hg used for each spray block and the volumes of lake water used for each spray event are shown in Table 2. The Hg spike solutions were added to the lake water in the airplane tank immediately before each application began, when the pilot was over Lake 658 and ready to commence spraying.

Spatial mapping of the spray events was based on the GPS coordinates obtained from the aircraft during spraying. ArcView GIS and Spatial Analyst were used to calculate the amount of Hg sprayed within the spray block boundaries. Because the aircraft recorded GPS coordinates at a constant rate during spraying, and the spraying rate (and therefore the amount of Hg sprayed) was also constant, the mass of Hg sprayed at each GPS point could be calculated. This made mapping the spraying of Hg using the GPS coordinates possible, and overlapping flight paths could be taken into account. Individual spray events could be mapped, and were summed together to calculate cumulative maps of Spike Hg spraying for each year.

The density of GPS points within a radius of 20 m was calculated using the kernel function in Spatial Analyst. Twenty metres was the distance spray was expected to drift from the flight path of the aircraft under the low wind conditions required for spraying (Art

Robinson, pers. comm.). This calculated density was then multiplied by the amount of Hg sprayed at each GPS point, to determine a spatial map of the amount of Hg sprayed. The total amount of Spike Hg sprayed within a certain area was then calculated by selecting an area (e.g. the watershed polygon), and summarizing zones (which summed up the Hg sprayed for each m^2 within the selected area). Maps of each spray event were then summed together to provide cumulative spray maps over the seven years of the project. The resulting maps can be used to identify areas that were sprayed more frequently over all spray events. GPS data are not available for Area C for 2004 due to an equipment malfunction. The mapping of Spike Hg in 2004 was estimated using the spray tracks from 2003 in Area C, since the target sprayblock to be sprayed was the same in both years.

In the early years of the experiment, the maps that were used to delineate the watershed boundary were the best available at the time, and were based on aerial photography and limited GPS data obtained from walking the watershed boundaries. In 2006, new maps were made using LiDAR (**Li**ght **D**etection **a**nd **R**anging) data collected in July 2005 by LaserMap Inc. The LiDAR data allowed us to better delineate the watershed boundary and resulted in differences between the original boundary, and the new more accurate boundary. This caused some changes to the target areas used in calculations of 2001-2003 application rates. Below we provide revised estimates of the application rates presented by Sandilands et al. (2005).

RESULTS

AERIAL SPRAY EVENTS

For details of individual spray events from 2001 to 2003 see Sandilands et al. (2005).

In 2004, aerial applications to the upland (80.80% ^{200}Hg) and wetland (93.53% ^{198}Hg) areas of Lake 658 took place on August 7 and 8 (Table 3).

A small amount of rain occurred before upland area A&B was sprayed on August 7, 2004. Two small rain events (totalling 0.5 mm) occurred 2.5 and one hour before spraying, enough to wet the canopy. More rain did not occur until a day and a half after spraying (August 9). One hour before upland area C was sprayed, a small rain event occurred (0.23 mm), enough to wet the canopy. One and a half hours afterwards, area E (the wetland) was sprayed. A large rain event started four hours after the end of the spraying, resulting in 27 mm of rain by 9 a.m. on the morning of August 9 (Figure 2). Other conditions during aerial spray events are summarized in Table 4.

Aerial spraying did not occur in 2005 because the pilot was unavailable. Thus only the upland and wetland shorelines had isotope applied in 2005 since this was done by hand (see shoreline spraying section).

In 2006, aerial applications to the upland and wetland occurred on May 24 and 25 (Table 3). The amount of Hg sprayed in 2006 was doubled, because no isotope was applied in 2005. Very little rain was associated with these spray events. Five hours before the wetland was sprayed, there was ~0.2 mm of rain. Because this rain occurred at night and the humidity was 100% before spraying began early in the morning, the canopy was likely still wet. Upland area A&B was sprayed next, ~7 hours after the rain; however the humidity had dropped to ~70 % and the temperature was above the dew point, meaning the canopy may have dried before area A&B was sprayed. Area C was sprayed during a 1.5 mm rain event, however the rain event ended before the entire area was sprayed, and there was no further rain afterwards.

ESTIMATES OF AMOUNTS OF MERCURY LEAVING THE AIRCRAFT

During spraying losses of Hg to the airplane tank were proportional to the time needed for each spray. Times required for each application (Table 5) were used with prior experimental data (Sandilands et al. 2005) to estimate the amounts of mercury leaving the aircraft for each aerial application (Table 6).

FULFILLMENT OF SPRAY CRITERIA

Spray criteria were met fairly well in 2004 (Table 7). The canopy was wet prior to each spray event; however, there was no rainfall during the applications. Sufficient rainfall to wash the applied Hg off the canopy

occurred after spraying of upland area C and the wetland; however this did not occur after upland area A&B were sprayed (Table 7).

In 2006 there was very little rainfall associated with the spray events, both during spraying and afterward, and so it is unlikely that there was enough rain to wash the Hg off the canopy.

SHORELINE SPRAYING

Shoreline spraying of both the wetland and upland was undertaken by USGS as described in Sandilands et al. (2005, See Figure 3 for map of shoreline plots). In 2004, shoreline spraying of the wetland and upland was done on September 6 to 8. The same isotopic solutions that were sprayed from the aircraft in 2004 were used in the shoreline applications (80.80% ^{200}Hg for the upland shoreline and 93.53% ^{198}Hg for the wetland shoreline).

In 2005, shoreline spraying was completed between August 15 and 17. The wetland plots were sprayed with Wetland Spike Hg from 2004 (93.53% ^{198}Hg). The upland plots were sprayed with an aliquot obtained from a composite solution made on May 13, 2005, using remnants from previous years' upland applications. This composite was analyzed by Trent University and found to be 80.38% ^{200}Hg.

In 2006, the upland and wetland shoreline plots were sprayed on August 26 – 28. The isotope used for the wetland was from 2004, and the upland plots were sprayed with leftovers from various batches.

Application rates for the upland and wetland shoreline areas are shown in Tables 8 and 9.

MAPPING OF SPRAY EVENTS

Upland

Once the mapping of each spray event was completed (Figures 4 – 9), the total mass of Spike Hg applied to a spray block was calculated using ArcMap 9.2 and Spatial Analyst. The resulting release rate was then calculated for each spray event (Table 6) as follows:

Upland Release Rate=

$$\frac{\text{mass of Spike Hg released within spray block}}{\text{area of sprayblock}}$$

These rates can be used when examining deposition of Spike Hg within a specific sprayblock. For the total upland watershed area, the average application rate (Table 10) was calculated as follows:

Average Upland Application Rate=

$$\frac{\text{mass of Spike Hg released in the entire upland + mass applied to the shoreline}}{\text{entire upland area}}$$

This average application rate should be used when comparing Spike Hg to ambient Hg in watershed-related measurements, e.g. in runoff.

Wetland

The release rates of Spike Hg applied to the wetland (Table 11) were calculated as follows:

Wetland Release Rate=

$$\frac{\text{mass of Spike Hg released within the wetland}}{\text{wetland area}}$$

This rate is suitable for examining deposition directly to the wetland after a spray event. There was some overspray of Wetland Spike Hg into the upland, along the sides of the wetland (see Figures 10 – 15). However the most likely runoff path for oversprayed Spike Hg is to the wetland before entering the lake. There was also a small amount of overspray of Wetland Spike Hg into the lake. Masses of Wetland Spike Hg oversprayed into the upland and lake are presented in Table 11.

The shoreline area of the wetland was hand sprayed, and so the average application rate for the entire wetland area should be calculated as follows:

Average Wetland Application Rate=

$$\frac{\text{Mass of Wetland Spike Hg leaving aircraft + wetland shoreline spraying}}{\text{wetland area}}$$

This average application rate should be used when concerned with the total amount of Wetland Spike Hg applied (aerial spraying and shoreline spraying). This rate does not include Wetland Spike Hg sprayed alongside the wetland (which may then enter the wetland at a later time). Average application rates for

the wetland are presented in Table 11. Maps of the cumulative loading of Spike Hg are shown in Figures 16 to 27.

LAKE ADDITIONS

Lake Spike Hg was added to the lake bi-weekly, nine times each year from 2001 to 2007. Table 12 shows the dates and masses of Lake Spike Hg added from 2001 to 2007.

SUMMARY

Over the seven years of Hg loading to the Lake 658 watershed, the Hg Spike applications to the lake were relatively equal (Table 12). However, loadings to the terrestrial parts of the watershed were not equal each year (Tables 10 and 11) primarily due to the necessity for certain conditions to occur before aerial spraying, and pilot availability.

Spike Hg applications to the upland, wetland and lake totalled 111, 107, and 154 ug/m^2 over seven years, averaging 15.8, 17.8, and 22.0 ug/m^2/yr respectively.

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Sandilands, K.A., J.W.M. Rudd, C.A. Kelly, H. Hintelmann, C.C. Gilmour, and M.T. Tate. 2005. Application of enriched stable mercury isotopes to the Lake 658 watershed for the METAALICUS project, at the Experimental Lakes Area, northwestern Ontario, Canada. Can. Tech. Rep. Fish. Aquat. Sci. 2597: viii + 48 p.

Table 1. Percent isotopic enrichment of Spike Hg solutions. Non shaded data are from samples analyzed by Trent Univeristy. Shaded data are the certified purities provided by Trace Sciences.

Upland Spike	^{196}Hg	^{198}Hg	^{199}Hg	**^{200}Hg**	^{201}Hg	^{202}Hg	^{204}Hg
2001	0.08	4.04	3.79	**79.52**	4.22	6.63	1.72
	0.09	3.90	3.62	**80.45**	4.01	6.31	1.62
2002	0.07	3.64	3.28	**79.66**	3.82	8.08	1.45
	0.09	3.90	3.62	**80.45**	4.01	6.31	1.62
2003	0.09	3.06	3.74	**80.61**	3.48	7.42	1.60
	0.11	3.06	3.80	**80.44**	3.54	7.49	1.55
2004	0.09	3.75	3.21	**80.40**	3.60	7.60	1.38
	0.10	3.70	3.10	**80.80**	3.50	7.50	1.30
2006	0.08	3.94	3.52	**79.30**	3.92	7.81	1.43
Wetland Spike	^{196}Hg	**^{198}Hg**	^{199}Hg	^{200}Hg	^{201}Hg	^{202}Hg	^{204}Hg
2001	0.16	**88.98**	1.09	4.25	2.83	2.26	0.43
	0.20	**90.50**	0.80	4.00	2.70	1.50	0.30
2003	0.20	**89.61**	0.91	3.43	3.66	1.79	0.40
	0.23	**90.67**	0.72	3.18	3.55	1.33	0.31
2004	0.01	**92.91**	3.29	1.76	0.70	1.14	0.21
	<0.01	**93.53**	3.19	1.61	0.57	0.93	0.16
2006	0.23	**88.90**	2.19	1.53	5.02	1.60	0.52
Lake Spike	^{196}Hg	^{198}Hg	^{199}Hg	^{200}Hg	^{201}Hg	**^{202}Hg**	^{204}Hg
2001	0.05	0.84	1.67	4.68	1.80	**90.02**	0.94
	0.05	0.70	1.50	4.35	1.65	**90.90**	0.85
2002	0.05	0.94	1.68	4.63	1.81	**89.95**	0.95
	<0.10	0.70	1.50	4.30	1.70	**90.80**	0.90
2003	0.06	1.35	2.08	4.88	1.58	**89.13**	0.92
	0.06	0.96	1.75	4.28	1.33	**90.81**	0.76
2004	0.05	0.51	3.37	2.43	1.77	**89.80**	2.08
	<0.05	0.36	3.21	2.18	1.63	**90.56**	2.01
2005	0.05	0.78	2.97	2.45	2.05	**89.70**	2.00
2006	0.06	1.18	2.59	2.83	2.14	**89.82**	1.39
	0.05	1.15	2.20	2.85	2.15	**90.35**	1.25
2007	0.03	0.80	1.54	3.93	1.80	**89.71**	2.19

Table 2. Areas, amount of Hg put in plane, and spray volumes for each aerial spray application to Lake 658 upland and wetland

Date	Spray block	Hg (mg) added to plane tank	Isotope	Spray volume (L)
13-June-01	A	3410	^{200}Hg	500
18-June-01	B	3390	^{200}Hg	500
27-june-01	E	450	^{198}Hg	250
28-May-02	A&B	2650	^{200}Hg	360
28-May-02	C	8190	^{200}Hg	240
18-May-03	A&B	6620	^{200}Hg	400
18-May-03	C	3620	^{200}Hg	250
18-May-03	E	905	^{198}Hg	250
7-Aug-04	A&B	4300	^{200}Hg	500
8-Aug-04	C	3130	^{200}Hg	250
8-Aug-04	E	104	^{198}Hg	250
24-May-06	A+B	11622	^{200}Hg	500
25-May-06	C	8363	^{200}Hg	250
24-May-06	E	790	^{198}Hg	250

Table 3. Dates and times of upland and wetland aerial applications.

Date	Spray block	Time Began	Time Ended
13-June-01	A	10:44	11:10
18-June-01	B	17:03	17:23
27-june-01	E	19:26	19:38
28-May-02	A	12:03	12:19
28-May-02	B	12:21	12:38
28-May-02	C	13:31	13:55
18-May-03	C	09:56	10:12
18-May-03	A&B	11:10	12:08
18-May-03	E	06:32	06:59
7-Aug-04	A&B	07:15	08:08
8-Aug-04	C	17:15	17:31
8-Aug-04	E	18:24	18:58
24-May-06	E	05:49	06:17
24-May-06	A&B	07:18	08:20
25-May-06	C	09:57	10:43

Table 4. Wind conditions, temperature and relative humidity for each spray event.

Spray Event	Average Temp (ºC)	Avg. Relative Humidity (%)	Average Wind speed (m/s)	Avg. Gust speed (m/s)	Max. Gust speed (m/s)	Avg. Wind Direction (Degrees)
Area A, 13-June-01*	14.5	82	2.7	no data	5.83	no data
Area B, 18-June-01*	19	66	3.4	no data	8	no data
Area E, 27-June-01*	18	92	1.1	no data	3.3	no data
Area A, 28-May-02**	13.5	82.5	0.5	no data	2.5	no data
Area B, 28-May-02**	13	92	0	no data	1.5	no data
Area C, 28-May-02**	12.5	93.5	0.5	no data	2.5	no data
Area A&B, 18-May-03***	13.95	89.34	2.32	2.74	5.57	156.68
Area C, 18-May-03***	11.62	96.19	1.34	1.56	2.78	151.59
Area E, 18-May-03***	11.63	96.56	1.01	1.16	2.41	155.48
Area A&B, 7-Aug-04+	15	83	1.4	3.1	4.5	177
Area C, 8-Aug-04+	17	89	0.8	1.7	2.4	117
Area E, 8-Aug-04+	18	85	1.1	2.4	3.2	132
Area A&B, 24-May-06	19**	67**	1.44***	1.66***	4.08***	156***
Area C, 25-May-06	17**	94**	0.62***	0.73***	2.23***	315***
Area E, 24-May-06	15**	95**	0.82***	0.94***	2.04***	134***

* data are from a meteorological tower located on the ridge to the south of L658, 15 min. averages + 2 min averages, ** data from a meteorological raft station located on L658, data are 15 min. averages, *** data from a meteorological station located on the north side of L658 at the highest point of the upland; data are averages of one minute averages of 10 second interval measurements

Table 5. Estimate of proportion of Hg leaving aircraft during Lake 658 watershed spray events (see Sandilands et al. 2005 for details of method used).

Date	Spray block	Total Length of Spray + 5 mins mixing time (minutes)	Estimate of Proportion Leaving aircraft
13-June-01	A	31	0.90
18-June-01	B	25	0.91
27-june-01	E	17	0.94
28-May-02	A&B	38	0.87
28-May-02	C	29	0.90
18-May-03	A&B	63	0.79
18-May-03	C	21	0.93
18-May-03	E	32	0.89
7-Aug-04	A+B	53	0.81
8-Aug-04	C	26	0.90
8-Aug-04	E	34	0.87
24-May-06	A+B	67	0.78
25-May-06	C	51	0.83
24-May-06	E	33	0.89

Table 6. Masses of Hg leaving aircraft during spraying (mass of Hg added to the aircraft tank – tank losses), mass of Hg released within the spray block (the target area to be sprayed), and the release rates associated with that spray block

Year	Spray block	Mass of Upland Spike Hg leaving aircraft (mg)	Mass of Upland Spike Hg Released within spray block (mg)	Area of Spray block (m^2)	Release Rate (ug/m^2)
2001	A&B	6153.90	5452.57	213220	25.57
2002	A&B	2305.50	2164.15	213220	10.15
2003	A&B	5229.80	4942.50	213220	23.18
2004	A&B	3484.29	3422.35	213220	16.05
2006	A&B	9087.53	8999.64	213220	42.21
2002	C	7371.00	6153.83	178523	34.47
2003	C	3366.60	3026.07	178523	16.95
2004	C	2802.45	2527.10	178523	14.16
2006	C	6961.27	6269.52	178523	35.12

Table 7. Aerial spray criteria, and whether each one was met for each individual spray event (* indicates that criterion was fulfilled adequately), and canopy development.

Spray event	Canopy Wet Prior to spray	Rainfall During Application	Wind light and away from lake	Hg washed off canopy after spray	Deciduous canopy development
Upland A 2001	*		*		full
Upland B 2001			*	*	full
Wetland 2001	*	*	*	*	full
Upland A/B 2002	*	*	*		low
Upland C 2002	*		*		low
Upland A/B 2003	*		*	*	full
Upland C 2003	*	*		*	full
Wetland 2003	*	*	*	*	full
Upland A/B 2004	*		*		full
Upland C 2004	*			*	full
Wetland 2004	*		*	*	full
Upland A/B 2006	Don't know if Canopy was wet		*		full
Upland C 2006	*	*	*		full
Wetland 2006	*		*		full

Table 8. Summary of Hg applications to upland shorelines.

Year	Upland Edge Plot #'s	Individual Plot Area (m^2)	Upland Spike Hg per plot (mg)	Upland Spike Hg (µg /m^2)
2002	112-113,113-114, 115-116, 1-2, 3-4, 4-5	400	23.7	59.3
2003	112-113, 113-114, 115-116, 1-2, 3-4, 5-6	400	6.4	16.0
2003	43 other upland plots	400	30.0	75.0
2004	All 49	400	5.1	12.6
2005	All 49	400	9.4	23.5
2006	All 49	400	9.3	23.1
2002-2006 year totals	**All upland shoreline plots**	**19,600**	**2,507.2 (All 49 plots)**	**127.9**

Table 9. Summary of Hg applications to wetland shorelines.

Year	Wetland Edge Plot #	Area (m^2)	Wetland Spike Hg per plot (mg)	Wetland Spike Hg (µg /m^2)
2002	1	121	6.9	57.03
2002	2	400	19.2	48.00
2002	3	400	19.2	48.00
2003	1+2	521	14.5	27.83
2003	3	400	11	27.50
2004	1	121	0.4	3.31
2004	2	400	5.1	12.75
2004	3	400	5.3	13.25
2005	1	121	2.7	22.31
2005	2	400	8.9	22.25
2005	3	400	8.9	22.25
2006	1	121	2.7	22.31
2006	2	400	8.9	22.25
2006	3	400	8.9	22.25
2003-2006 Year Totals	**1**	**121**	**18.85**	**155.79**
	2	**400**	**46.25**	**115.63**
	3	**400**	**50.2**	**125.50**

Table 10. Mass of Upland Spike Hg released + shoreline spraying, and the average application rates calculated for the entire upland of Lake 658 (regardless of whether it was ever meant to be sprayed or not).

Year	Mass of Upland Spike Hg Released (aerial spraying) + Mass of Hg sprayed (shoreline spraying) within entire upland (mg)	Entire Upland Area (m^2)	Average Application Rate (ug/m^2)
2001	5452.57	410456	13.28
2002	8459.98	410456	20.61
2003	9296.97	410456	22.65
2004	6197.24	410456	15.10
2005	460.00	410456	1.12
2006	15722.73	410456	38.31
		Total	**111.07**
		Mean	**18.51**

Table 11. Mass of Wetland Spike Hg released and the average application rate for the wetland of L. 658 and Mass of Wetland Spike Hg released in upland and lake.

Year	Mass of Wetland Spike Hg leaving aircraft (mg)	Mass of Wetland Spike Hg released within wetland (aerial spraying, mg)	Mass of Wetland Spike Hg sprayed on shoreline (mg)	Total Mass of Spike Hg in wetland (mg)	Average Application rate (ug/m^2)	Mass of Wetland Spike Hg released in upland (mg)	Mass of Wetland Spike Hg released in lake (mg)
2001	423.1	309	0.00	309.00	18.63	109	5.1
2002	0	0	45.30	45.30	2.73	0	0
2003	806	660	25.50	685.50	41.34	133	13
2004	89.8	73.8	10.83	84.63	5.10	16	0
2005	0	0	20.60	20.60	1.24	0	0
2006	703	609	20.46	629.46	37.96	85	9

Table 12. Amount of Lake Spike Hg added to Lake 658 from 2001 to 2007.

2001		2002		2003		2004		2005		2006		2007	
Date	Lake Spike Hg added (mg)	Date	Lake Spike Hg added (mg)	Date	Lake Spike Hg added (mg)	Date	Lake Spike Hg added (mg)	Date	Lake Spike Hg added (mg)	Date	Lake Spike Hg added (mg)	Date	Lake Spike Hg added (mg)
19-Jun	206.13	4-Jun	208.47	10-Jun	208.18	15-Jun	210.94	14-Jun	207.33	13-Jun	179.92	12-Jun	219.33
3-Jul	206.33	18-Jun	207.90	24-Jun	207.83	29-Jun	210.94	28-Jun	206.33	27-Jun	179.92	26-Jun	218.22
17-Jul	205.25	2-Jul	207.30	9-Jul	207.75	13-Jul	210.94	12-Jul	205.88	11-Jul	179.92	12-Jul	217.24
1-Aug	204.95	16-Jul	206.67	22-Jul	207.53	27-Jul	210.94	26-Jul	206.06	25-Jul	179.92	24-Jul	217.10
15-Aug	204.70	30-Jul	206.92	6-Aug	208.38	10-Aug	210.94	9-Aug	206.18	9-Aug	179.92	7-Aug	217.26
28-Aug	206.28	13-Aug	206.80	20-Aug	208.23	24-Aug	210.94	23-Aug	206.51	22-Aug	179.92	22-Aug	217.79
11-Sep	206.13	27-Aug	206.37	3-Sep	208.28	7-Sep	210.94	7-Sep	206.66	6-Sep	179.92	5-Sep	216.79
25-Sep	206.35	10-Sep	206.54	16-Sep	208.18	21-Sep	210.94	21-Sep	206.66	20-Sep	179.92	18-Sep	217.45
9-Oct	206.30	24-Sep	199.08	30-Sep	207.91	5-Oct	210.94	4-Oct	206.39	3-Oct	179.92	2-Oct	218.77
TOTAL	**1852.40**		**1856.05**		**1872.24**		**1898.50**		**1858.00**		**1619.30**		**1959.95**
Area (m^2)	83924		83924		83924		83924		83924		83924		83924
Application Rate (ug/m^2)	**22.07**		**22.12**		**22.31**		**23.03**		**22.14**		**19.29**		**23.35**

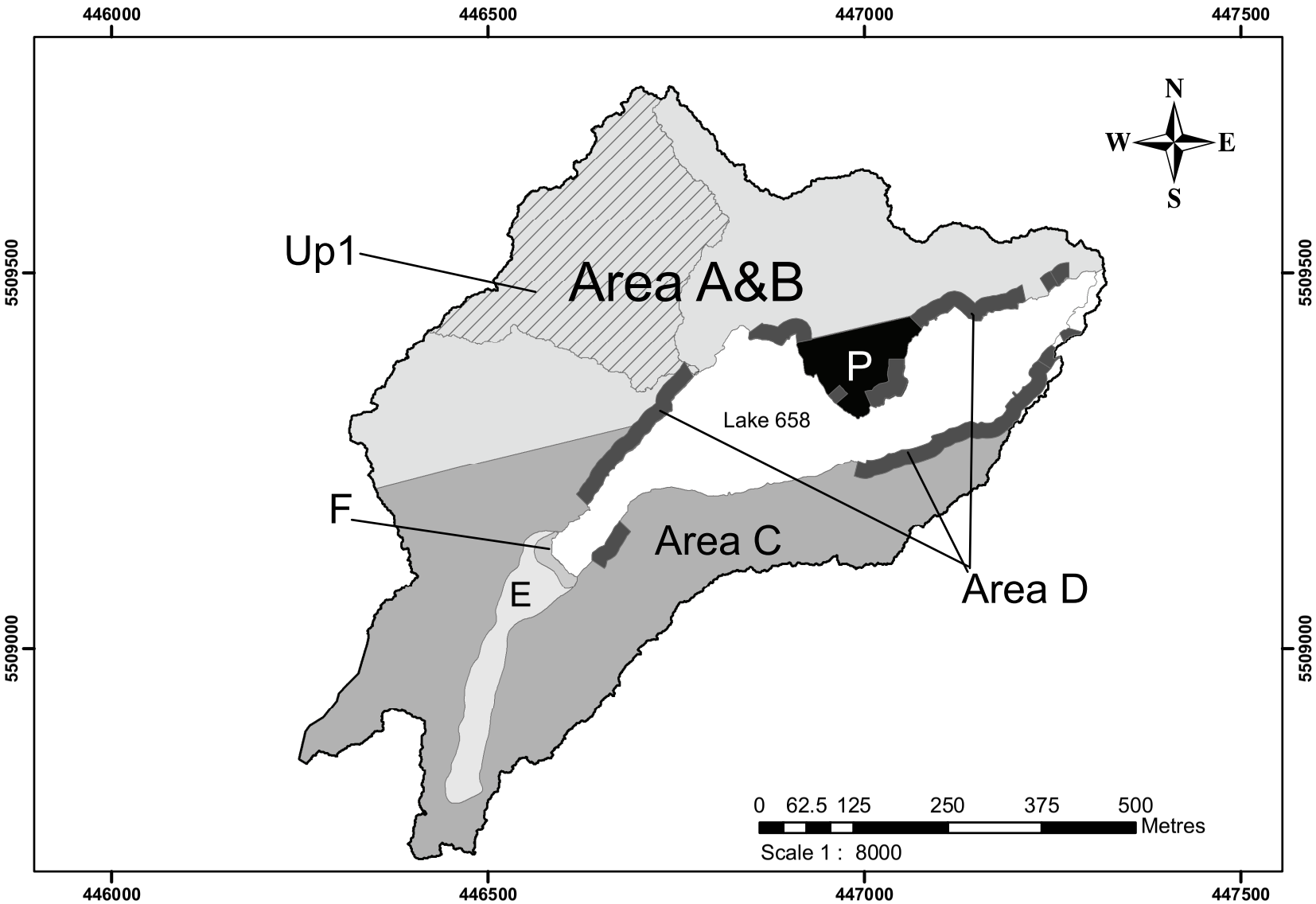

Figure 1. Map of the spray blocks used for aerial and shoreline spraying of Lake 658.

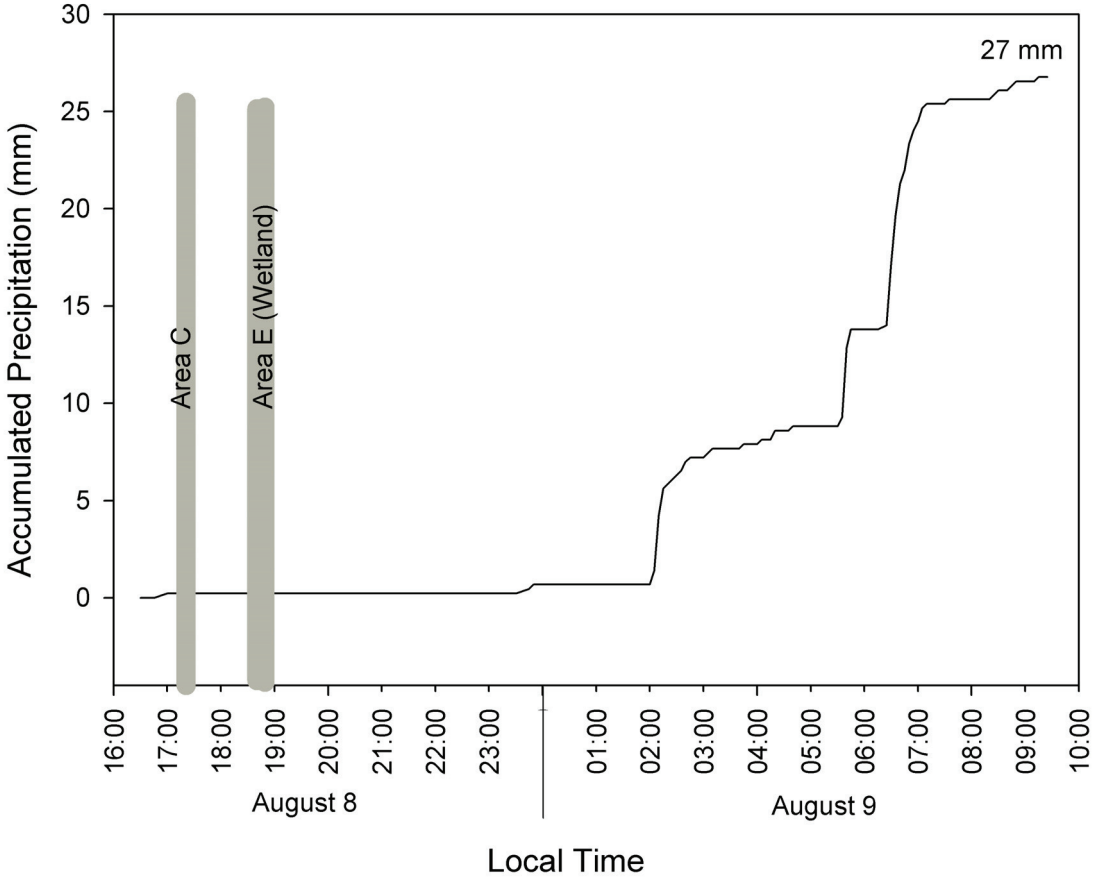

Figure 2. Accumulated precipitation after spraying of Areas C and E in 2004. Data from USGS open rain gauge located at Lake 658.

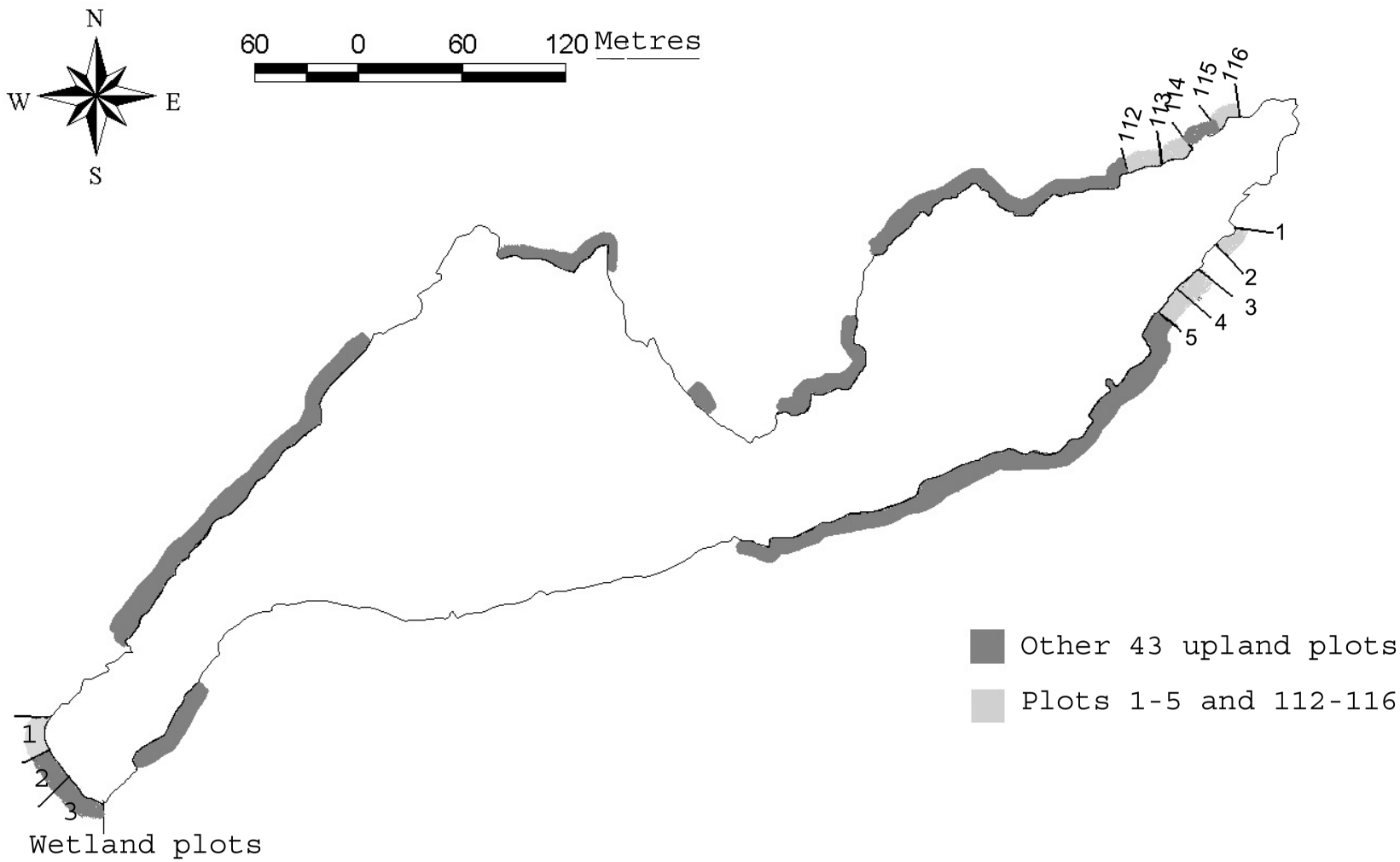

Figure 3. Map of upland and wetland shoreline spraying plots at Lake 658.

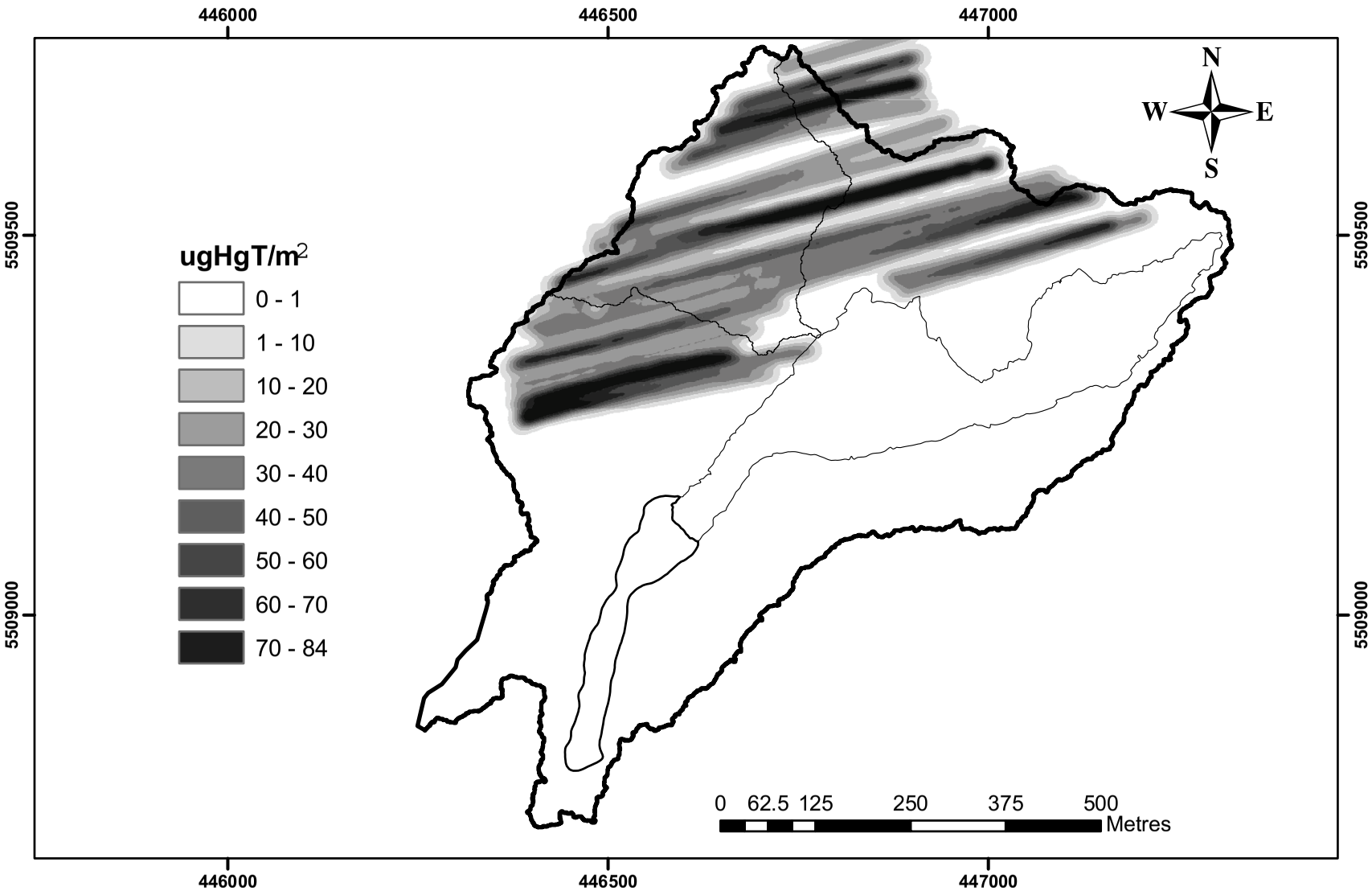

Figure 4. Map of Upland Spike Hg release from aerial spraying in the Upland of Lake 658 in 2001.

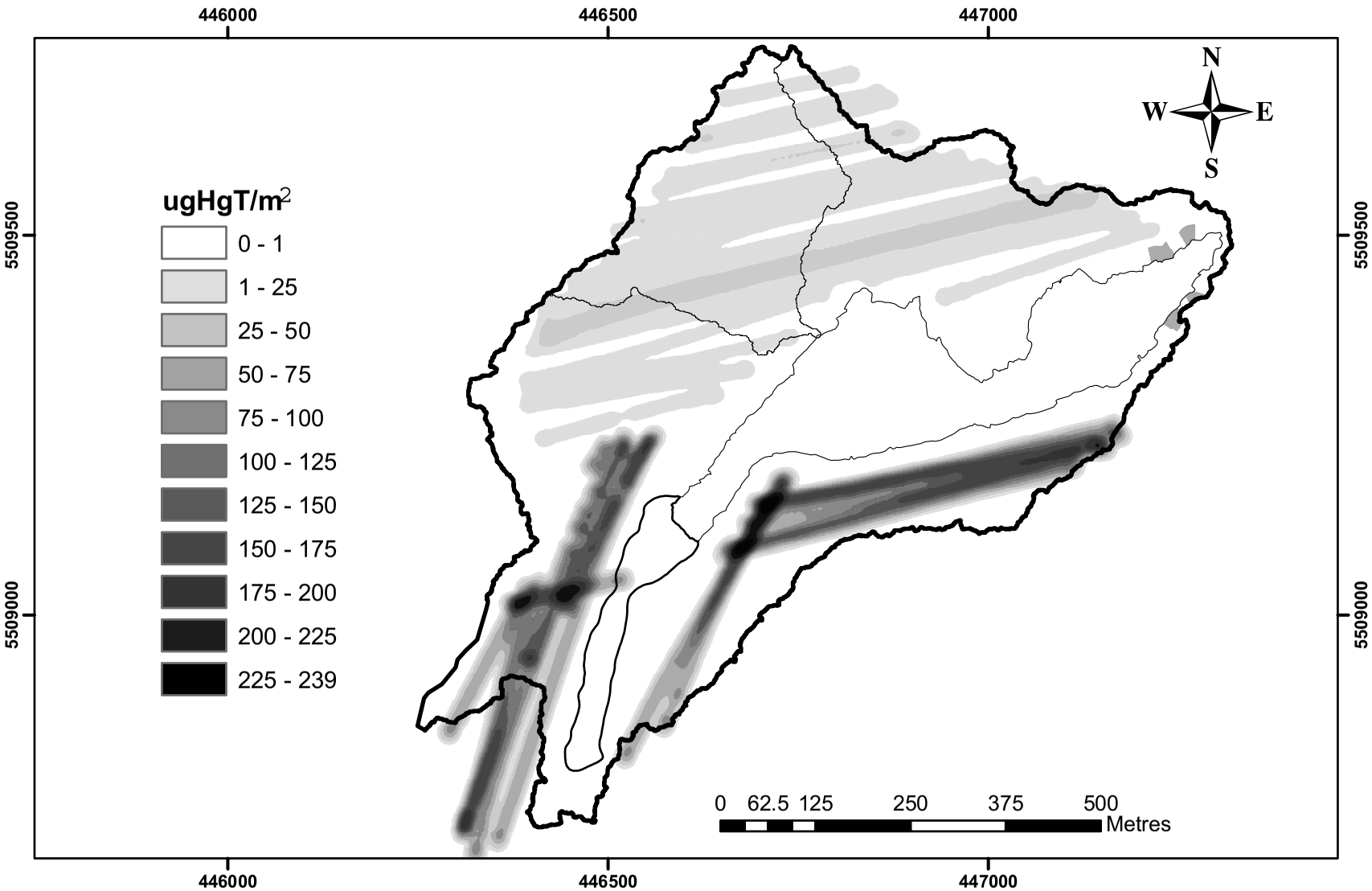

Figure 5. Map of Upland Spike Hg release from aerial spraying in the upland and shoreline spraying of Lake 658 in 2002.

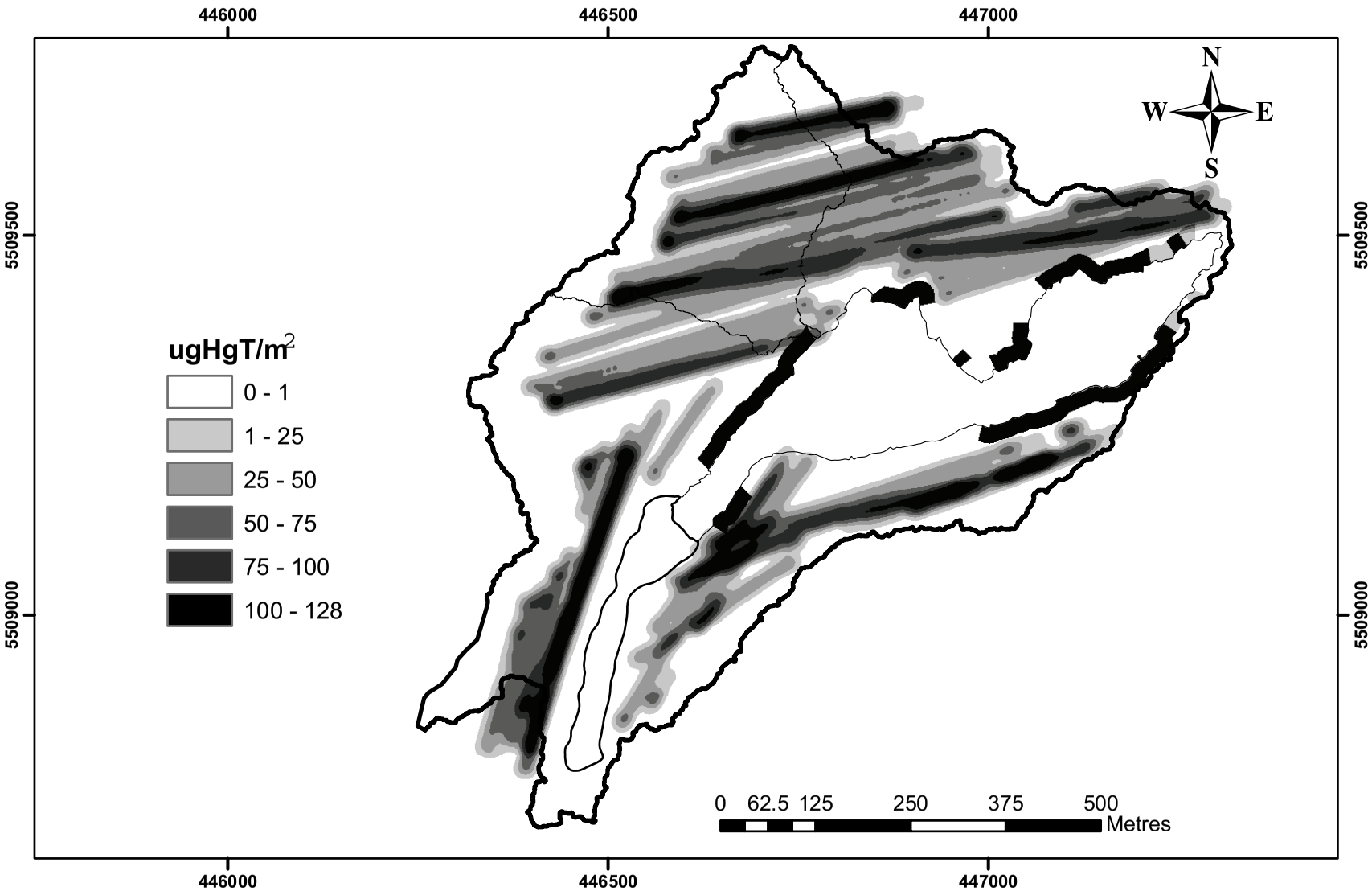

Figure 6. Map of Upland Spike Hg release from aerial spraying in the upland and shoreline spraying of Lake 658 in 2003.

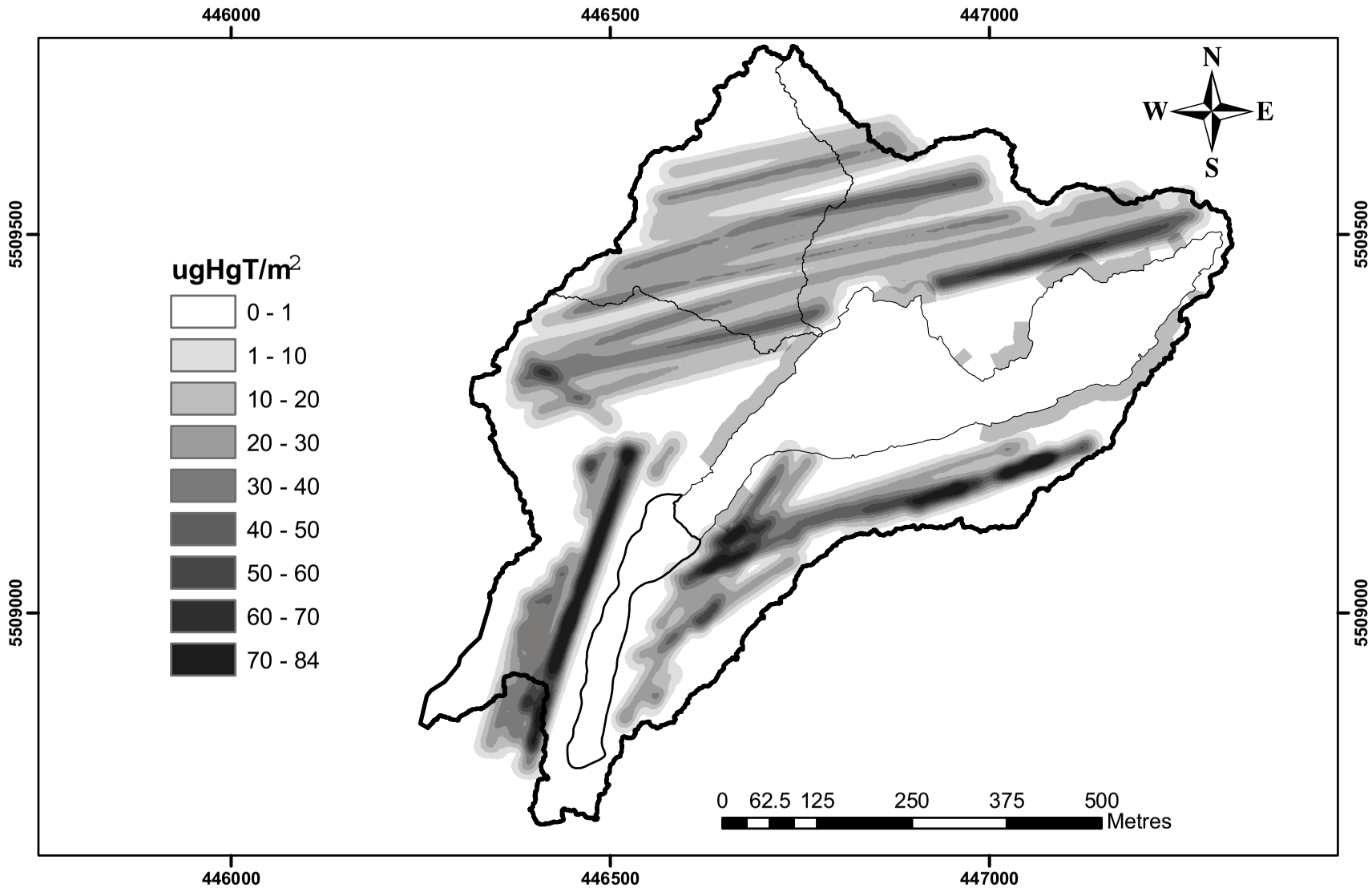

Figure 7. Map of Upland Spike Hg release from aerial spraying in the upland and shoreline spraying of Lake 658 in 2004.

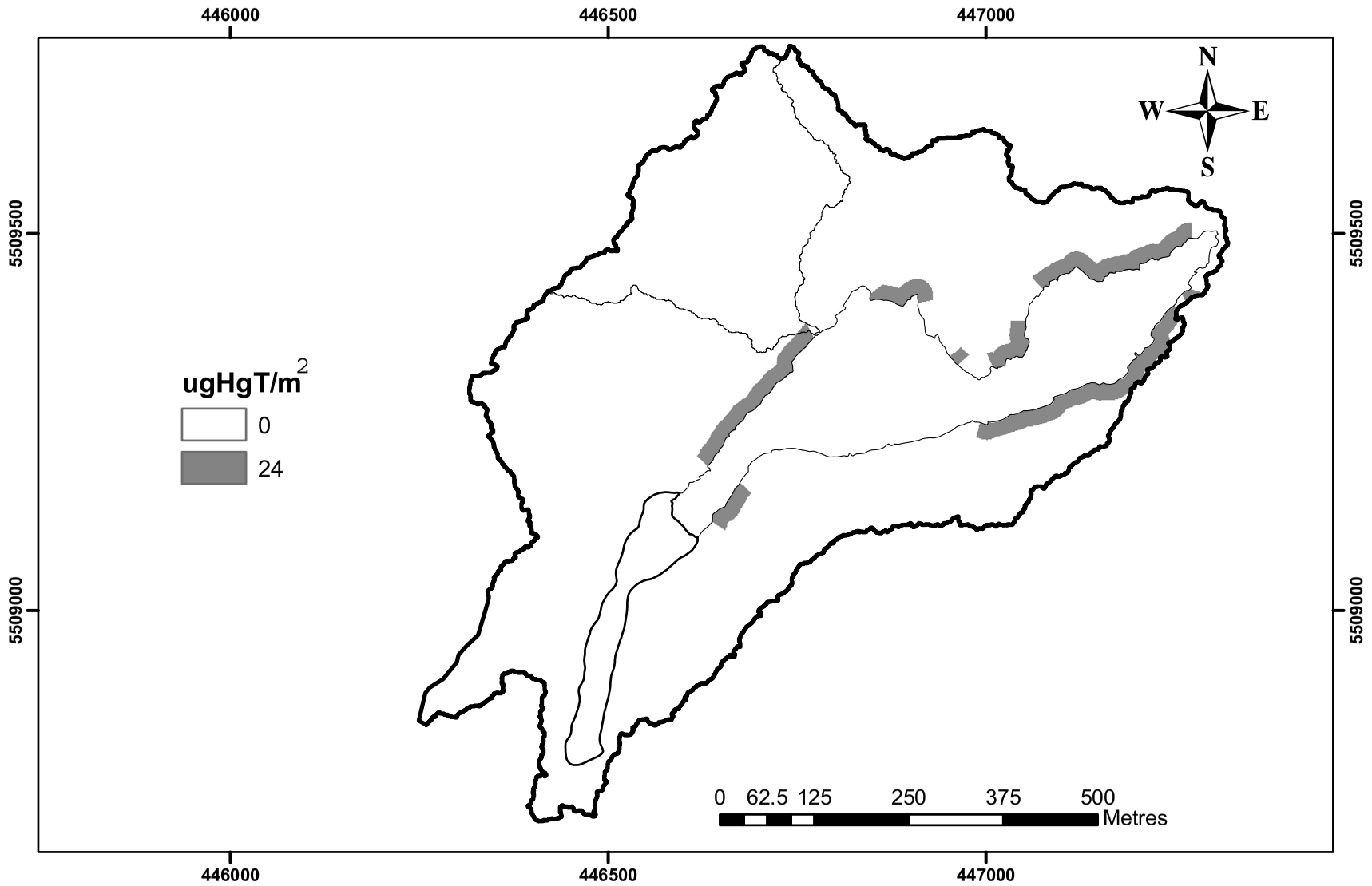

Figure 8. Map of Upland Spike Hg release from shoreline spraying in the upland of Lake 658 in 2005.

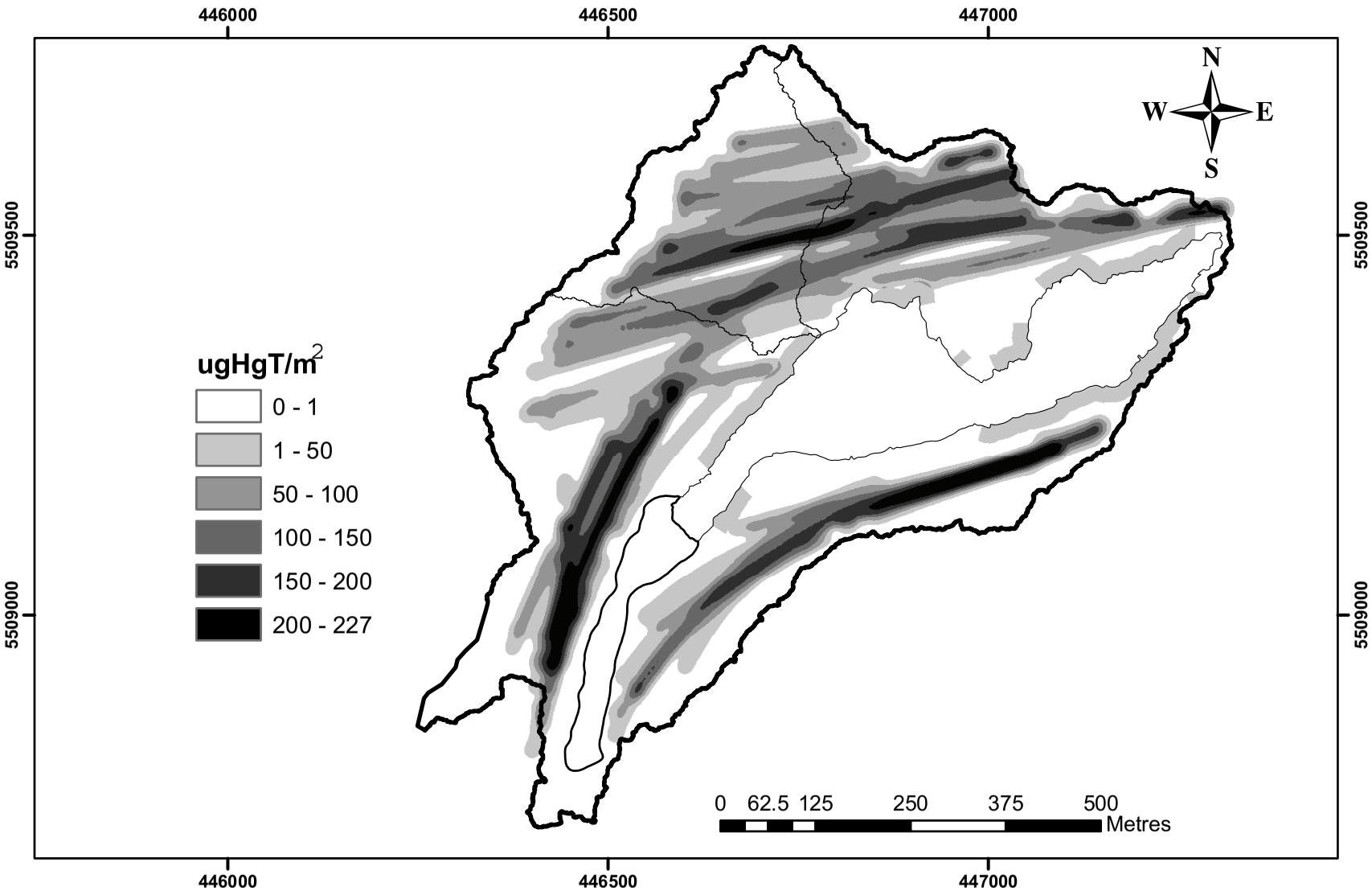

Figure 9. Map of Upland Spike Hg release from aerial spraying in the upland and shoreline spraying of Lake 658 in 2006.

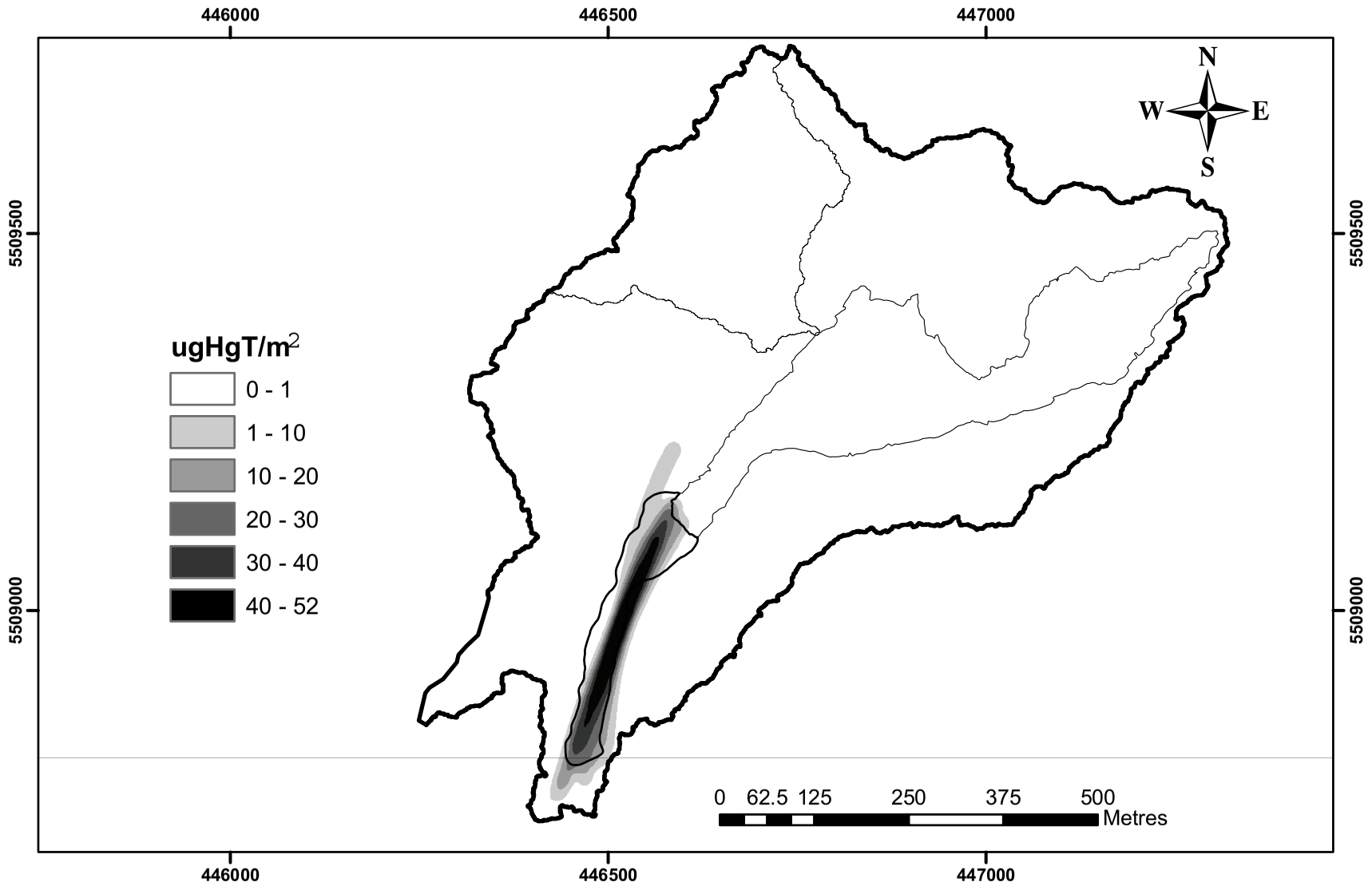

Figure 10. Map of Wetland Spike Hg release from aerial spraying in the wetland of Lake 658 in 2001.

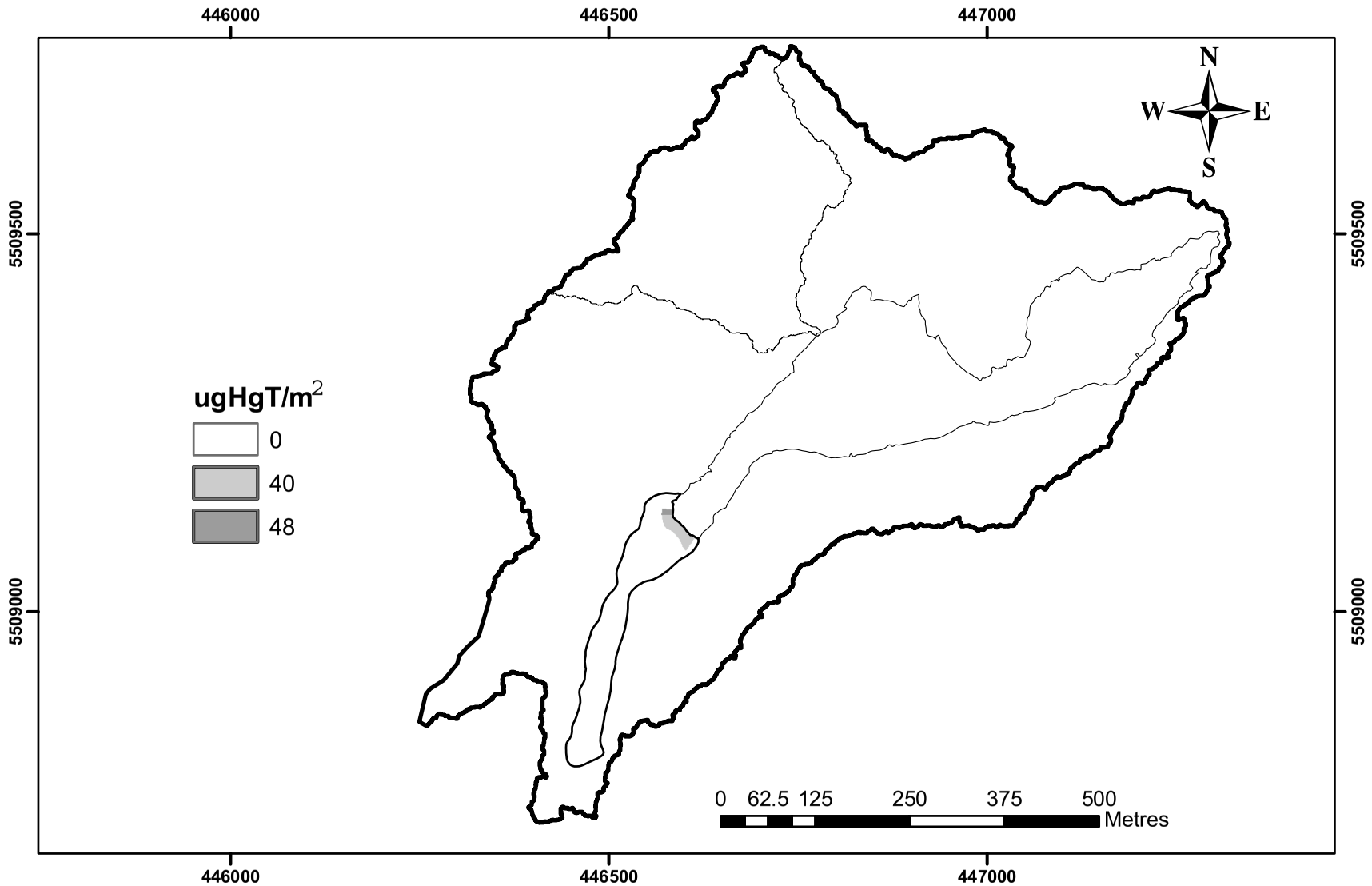

Figure 11. Map of Wetland Spike Hg release from shoreline spraying in the wetland of Lake 658 in 2002.

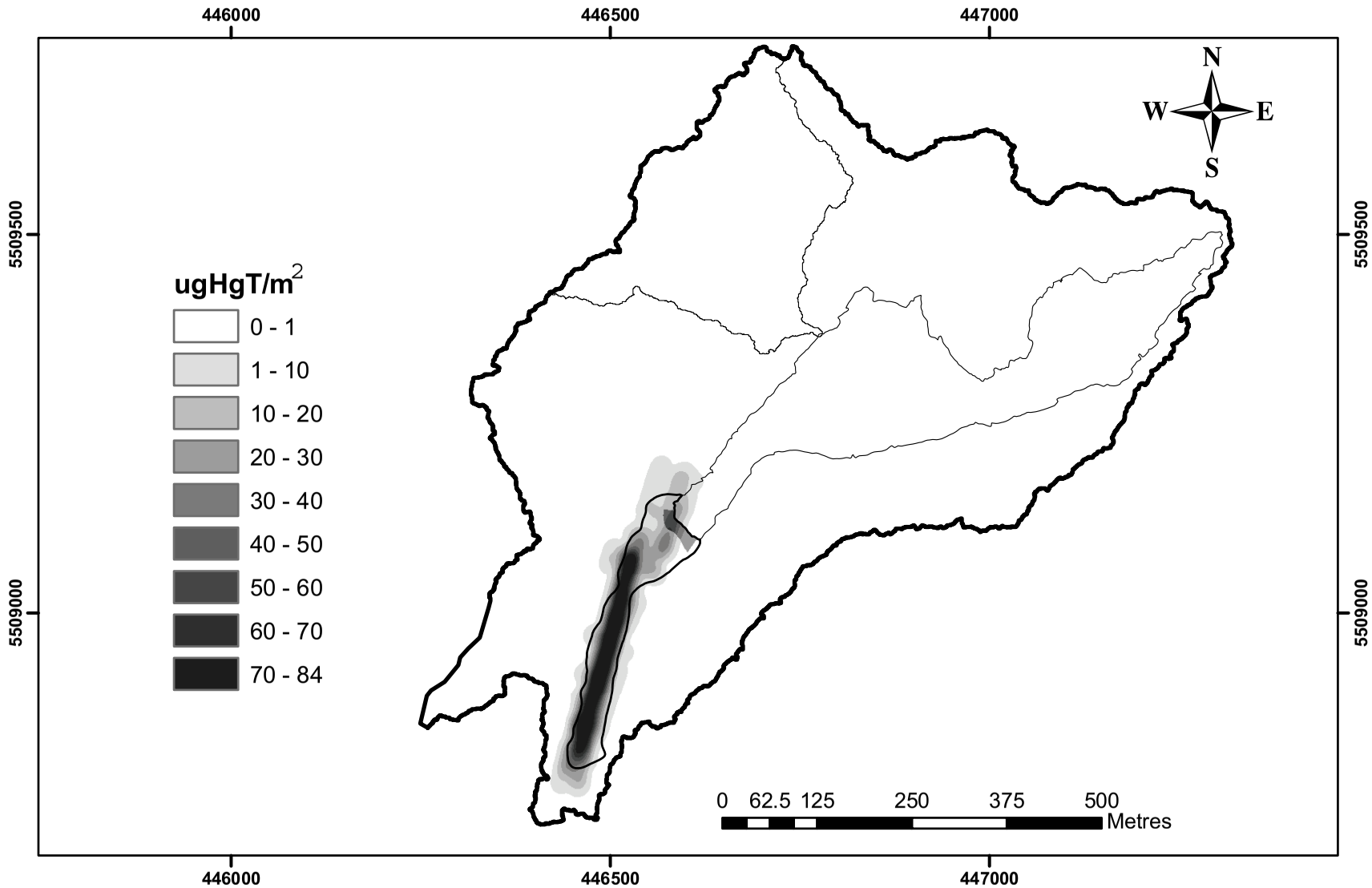

Figure 12. Map of Wetland Spike Hg release from aerial spraying in the wetland and shoreline spraying of Lake 658 in 2003.

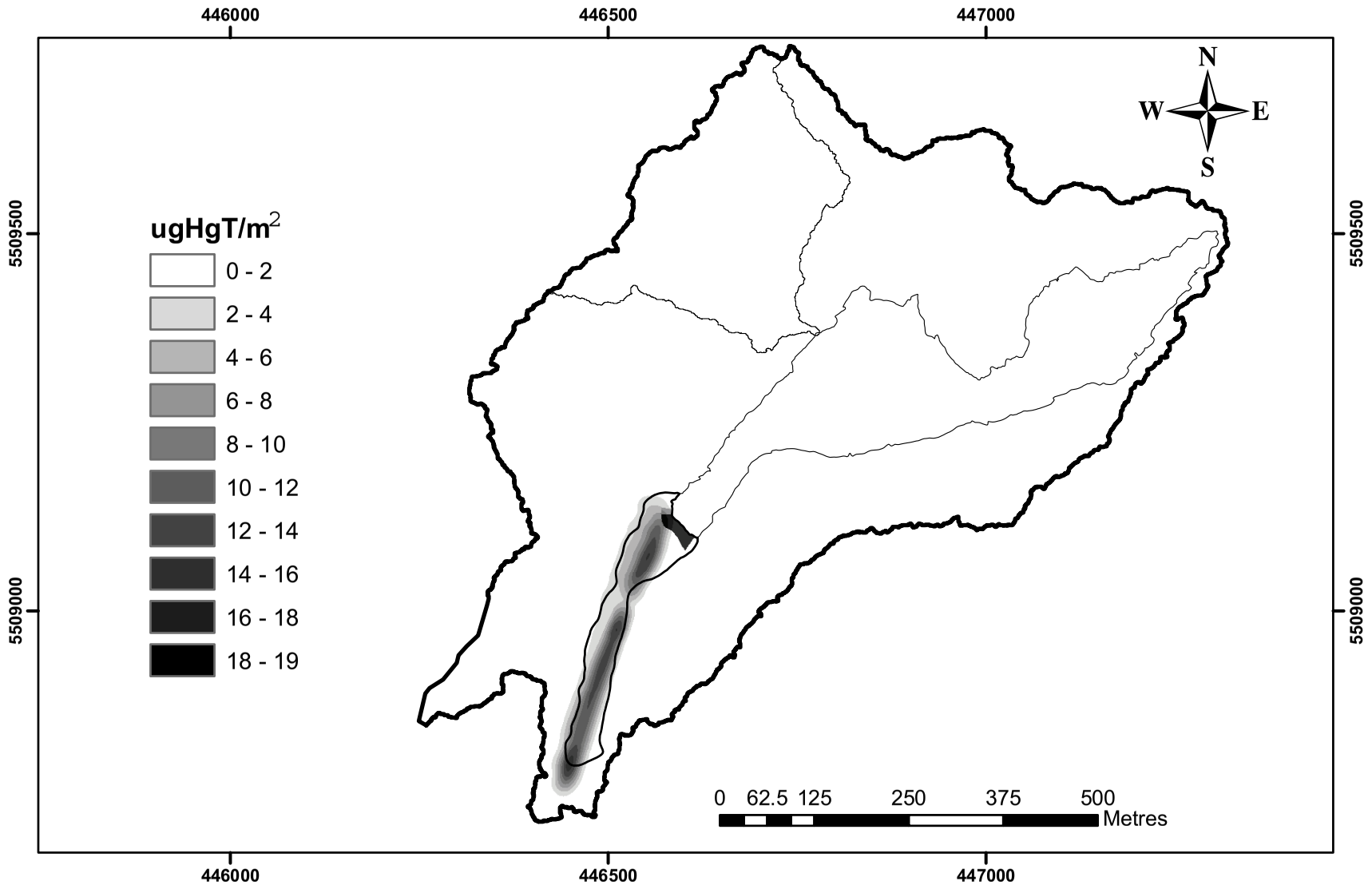

Figure 13. Map of Wetland Spike Hg release from aerial spraying in the wetland and shoreline spraying of Lake 658 in 2004.

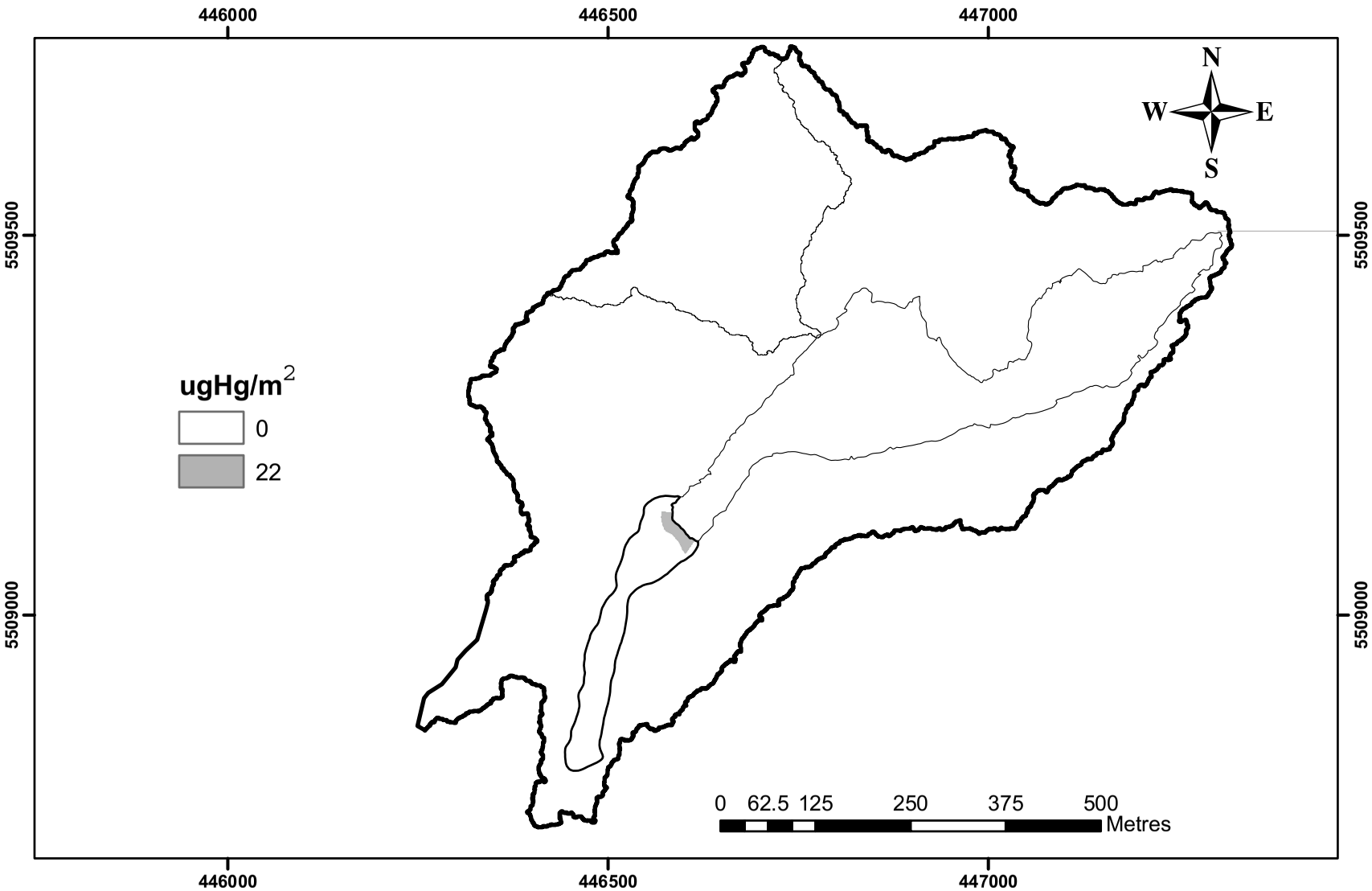

Figure 14. Map of Wetland Spike Hg release from shoreline spraying in the wetland of Lake 658 in 2005.

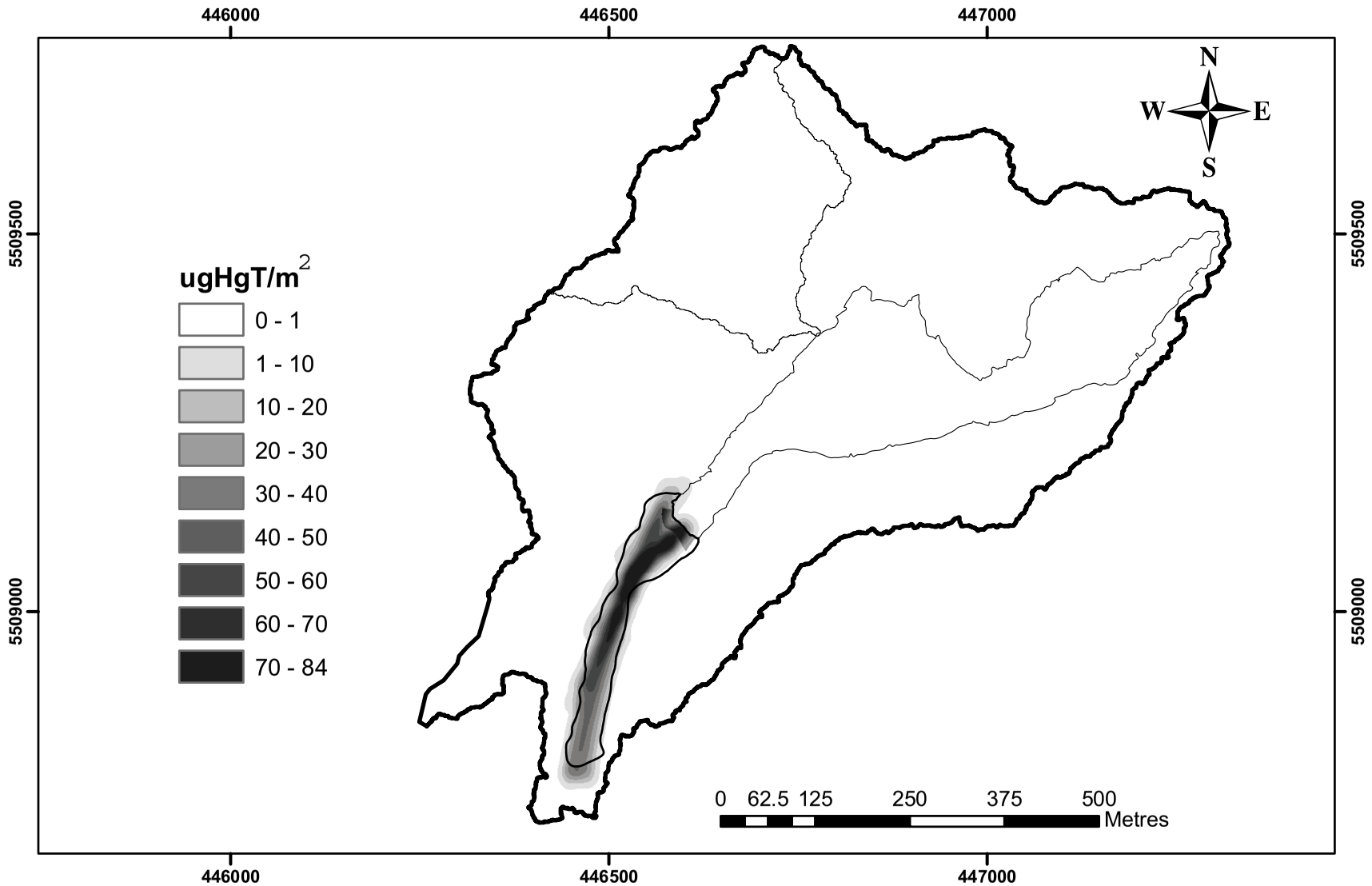

Figure 15. Map of Wetland Spike Hg release from aerial spraying in the wetland and shoreline spraying of Lake 658 in 2006.

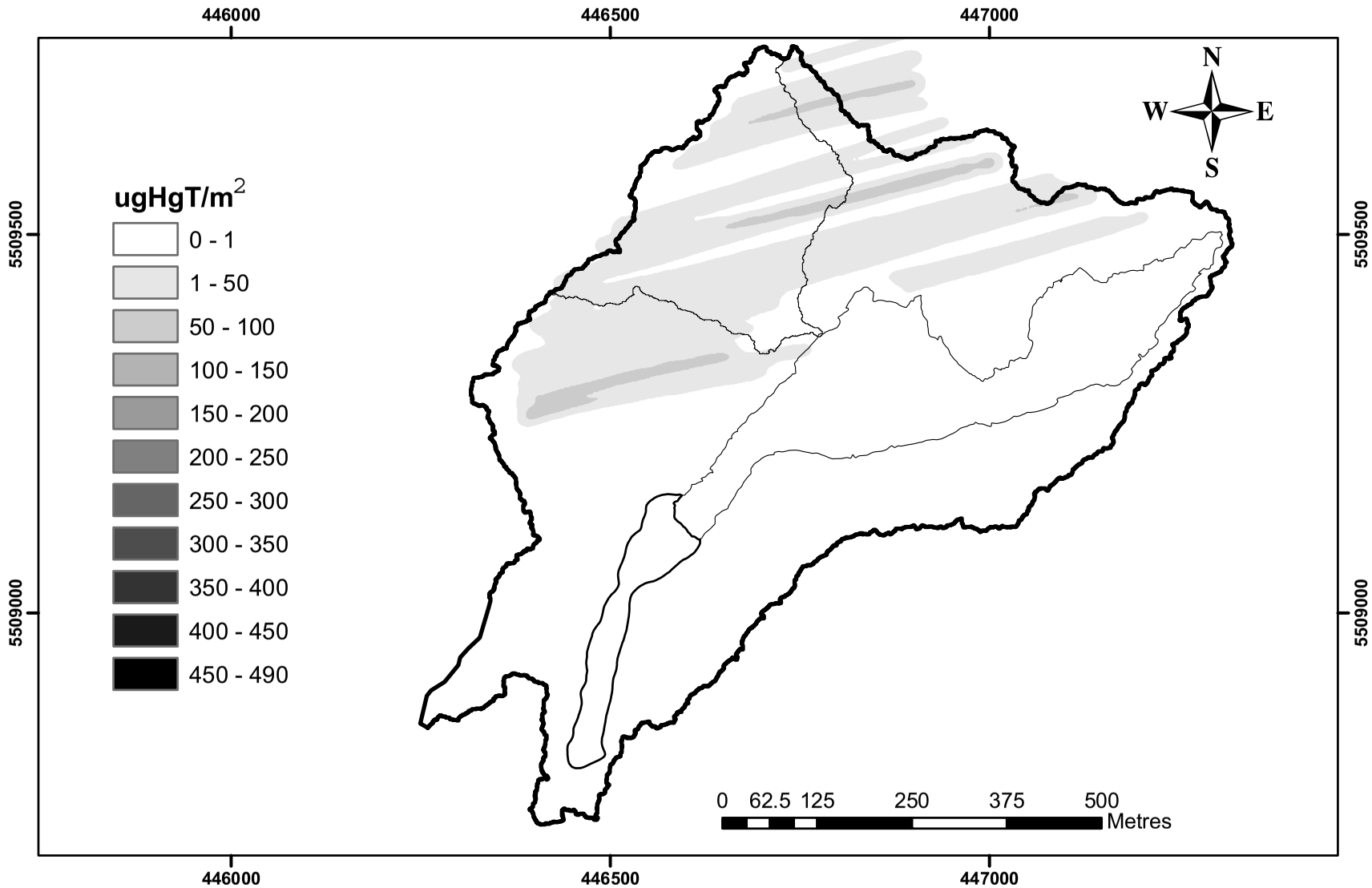

Figure 16. Map of cumulative Upland Spike Hg release from aerial spraying in the upland of Lake 658 in 2001.

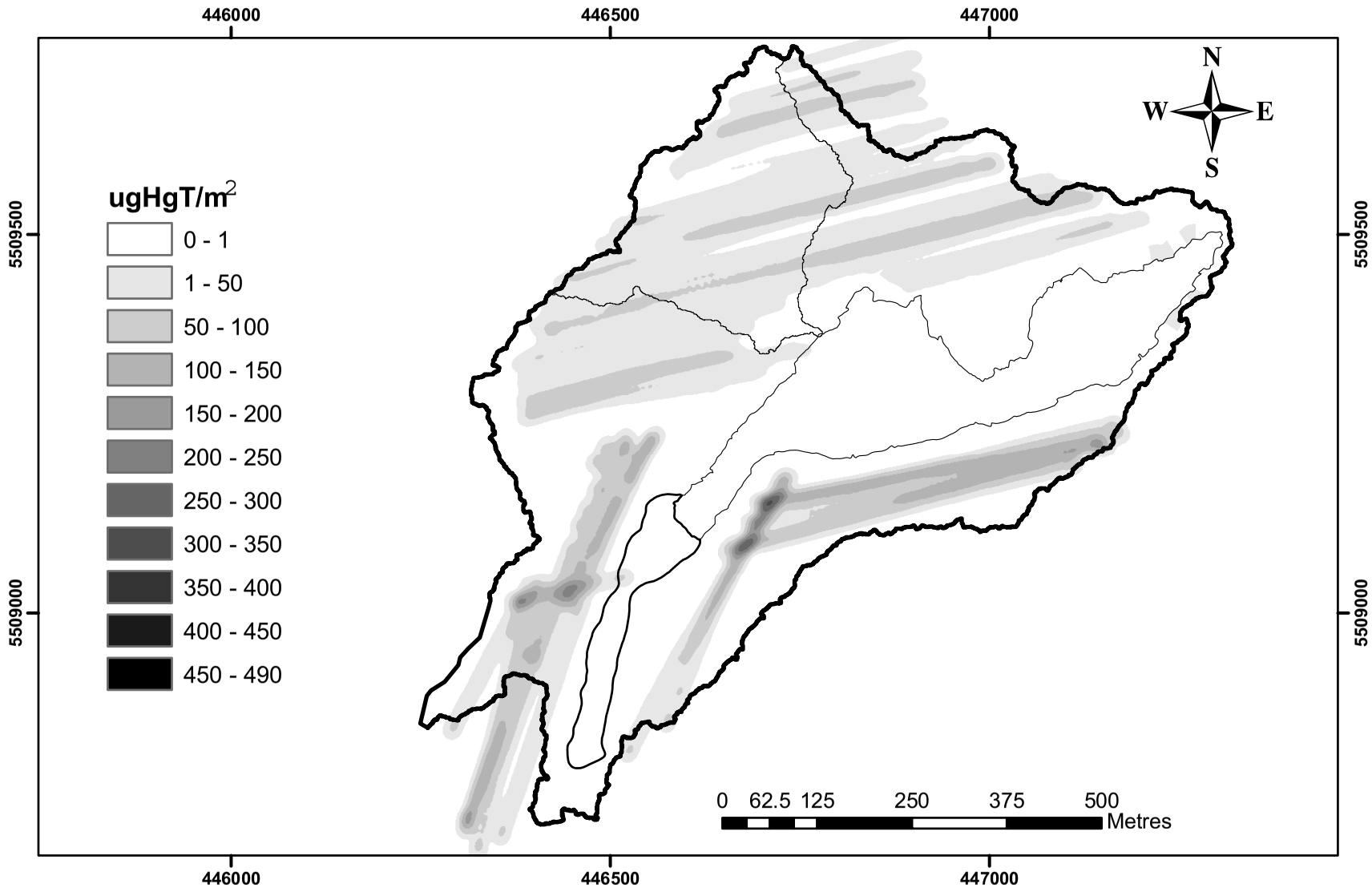

Figure 17. Map of cumulative Upland Spike Hg release from aerial and shoreline spraying of Lake 658 upland in 2002.

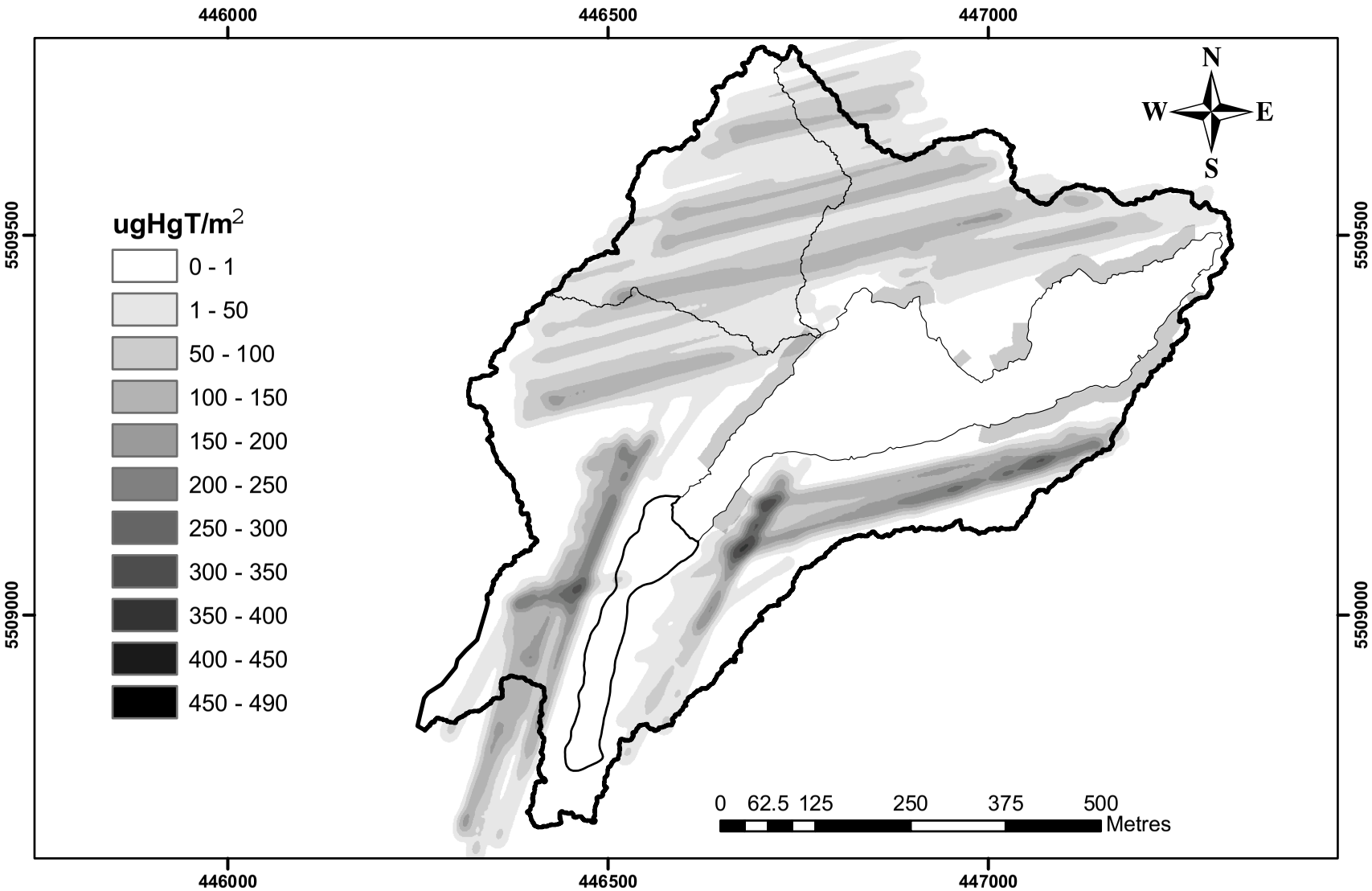

Figure 18. Map of cumulative Upland Spike Hg release from aerial and shoreline spraying of Lake 658 upland in 2003.

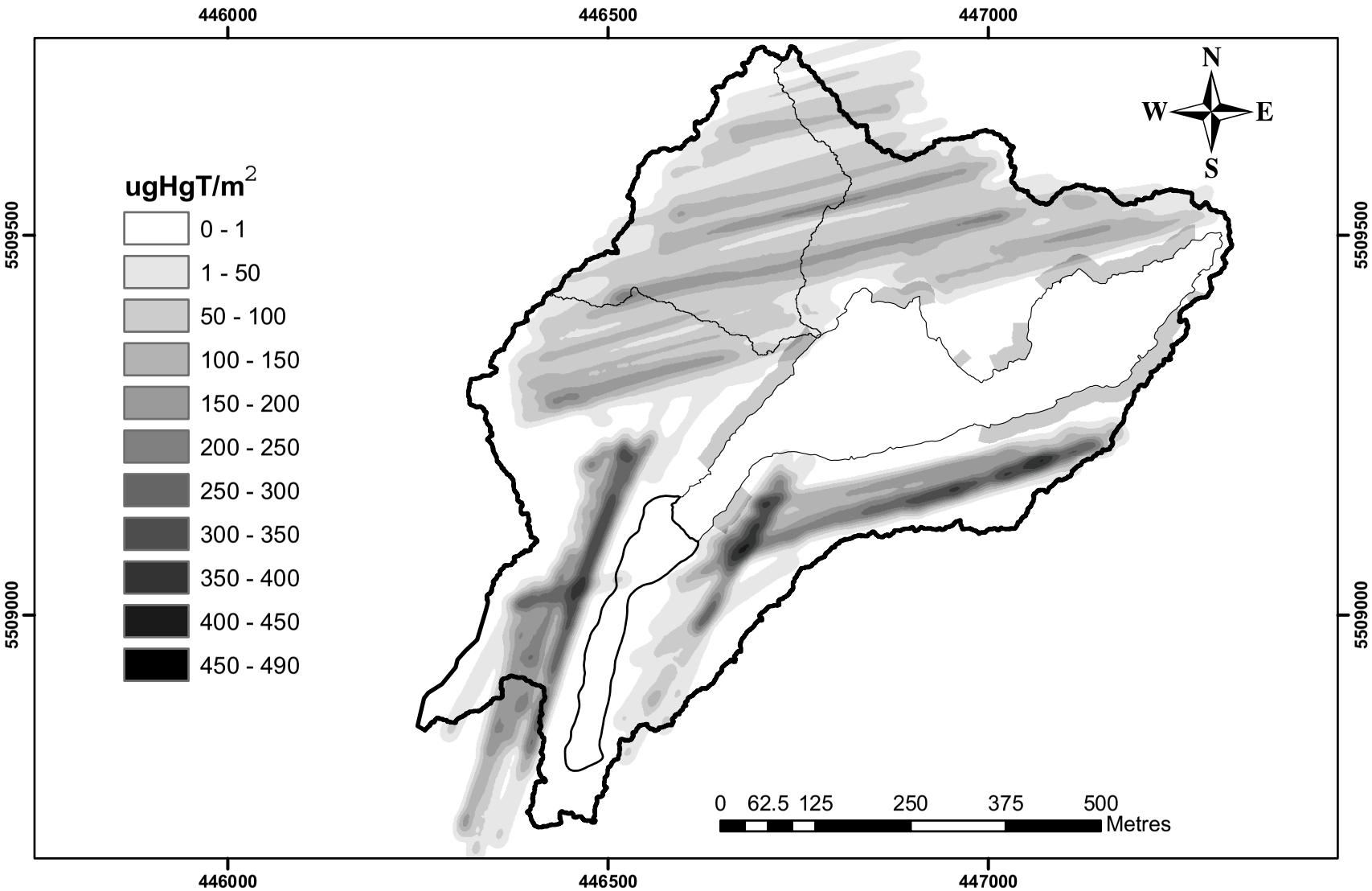

Figure 19. Map of cumulative Upland Spike Hg release from aerial and shoreline spraying of Lake 658 upland in 2004.

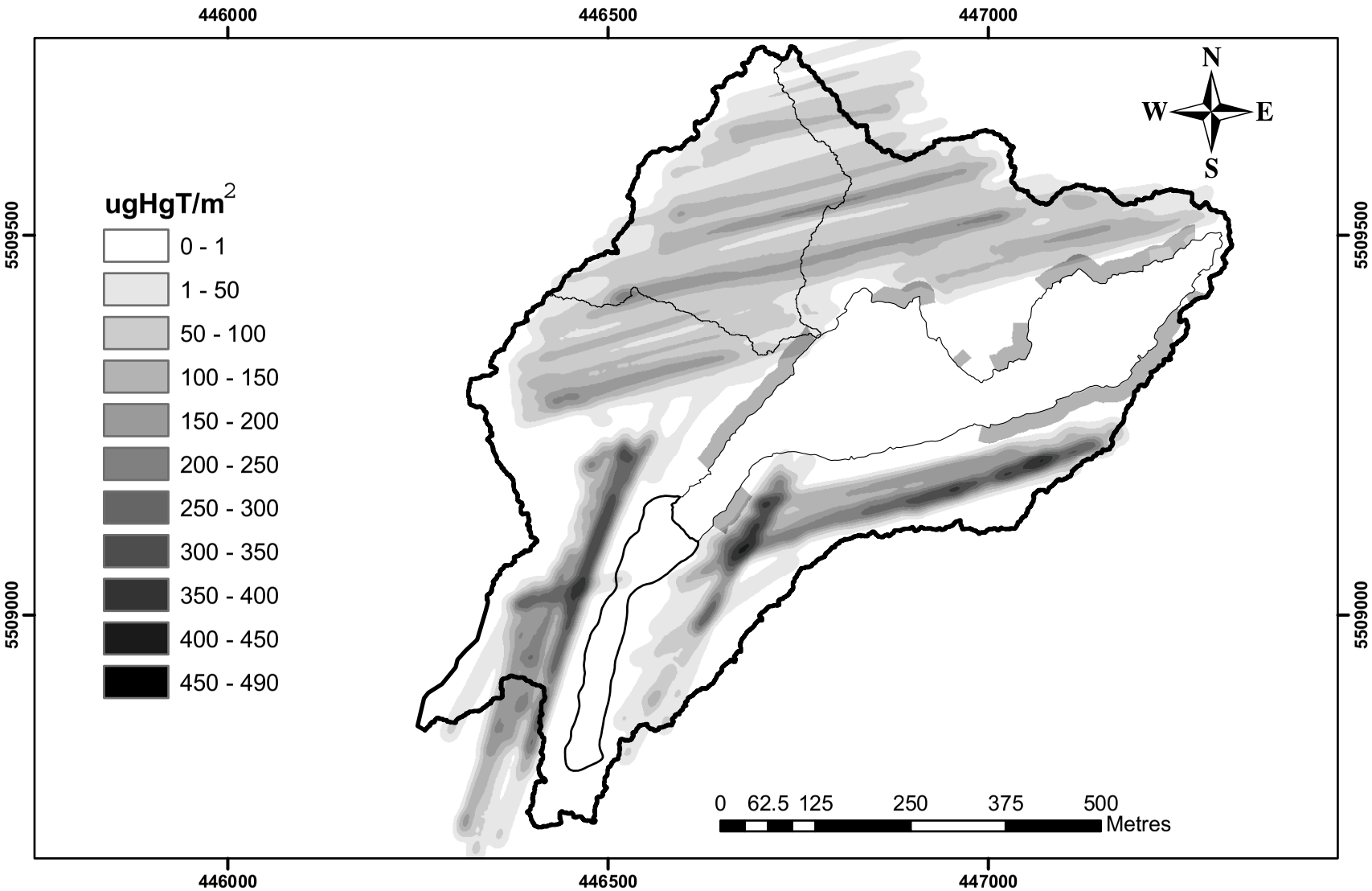

Figure 20. Map of cumulative Upland Spike Hg release from aerial and shoreline spraying of Lake 658 upland in 2005.

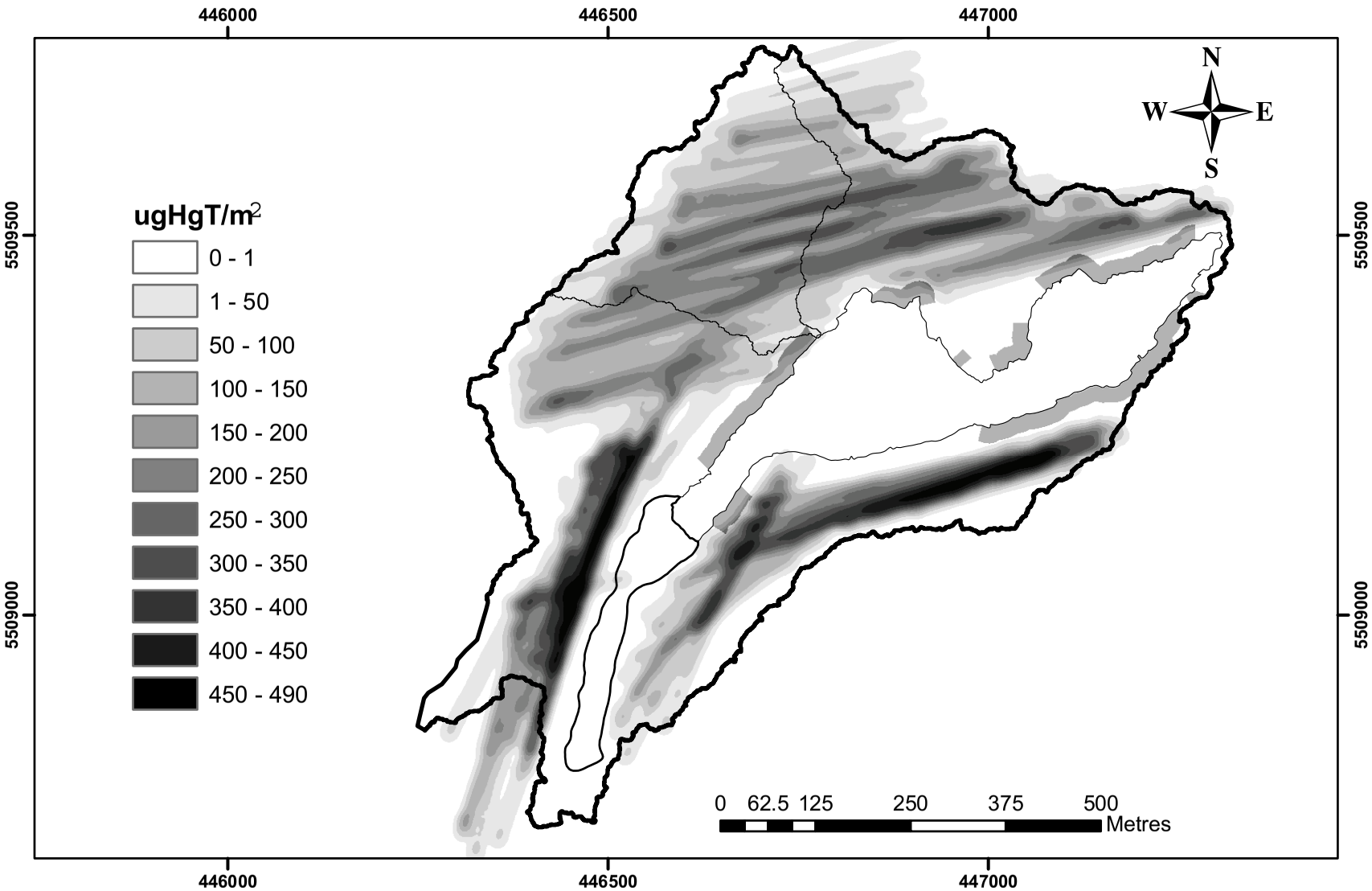

Figure 21. Map of cumulative Upland Spike Hg release from aerial and shoreline spraying of Lake 658 upland in 2006.

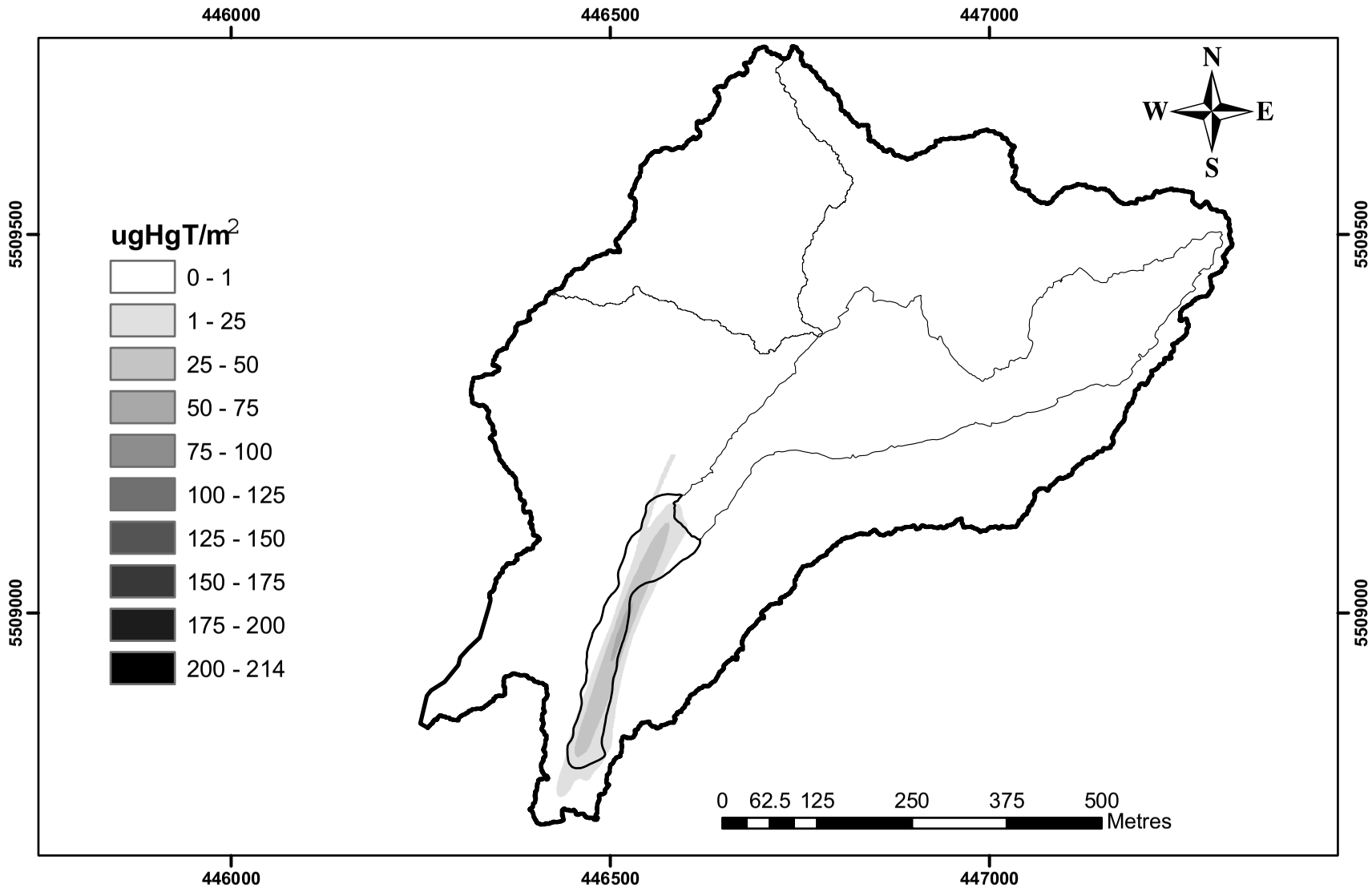

Figure 22. Map of cumulative Wetland Spike Hg release from aerial spraying of Lake 658 wetland in 2001.

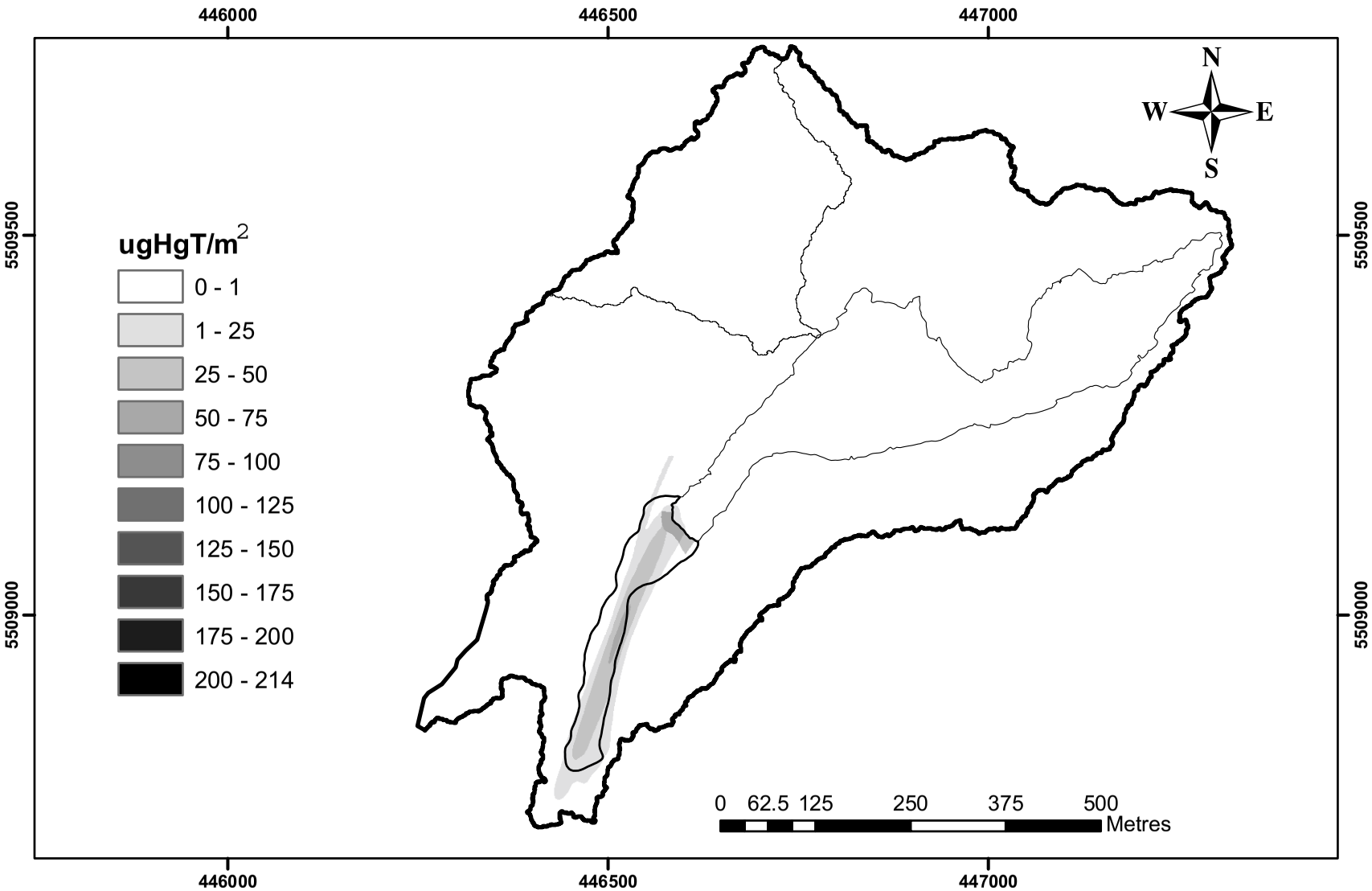

Figure 23. Map of cumulative Wetland Spike Hg release from aerial and shoreline spraying of Lake 658 wetland in 2002.

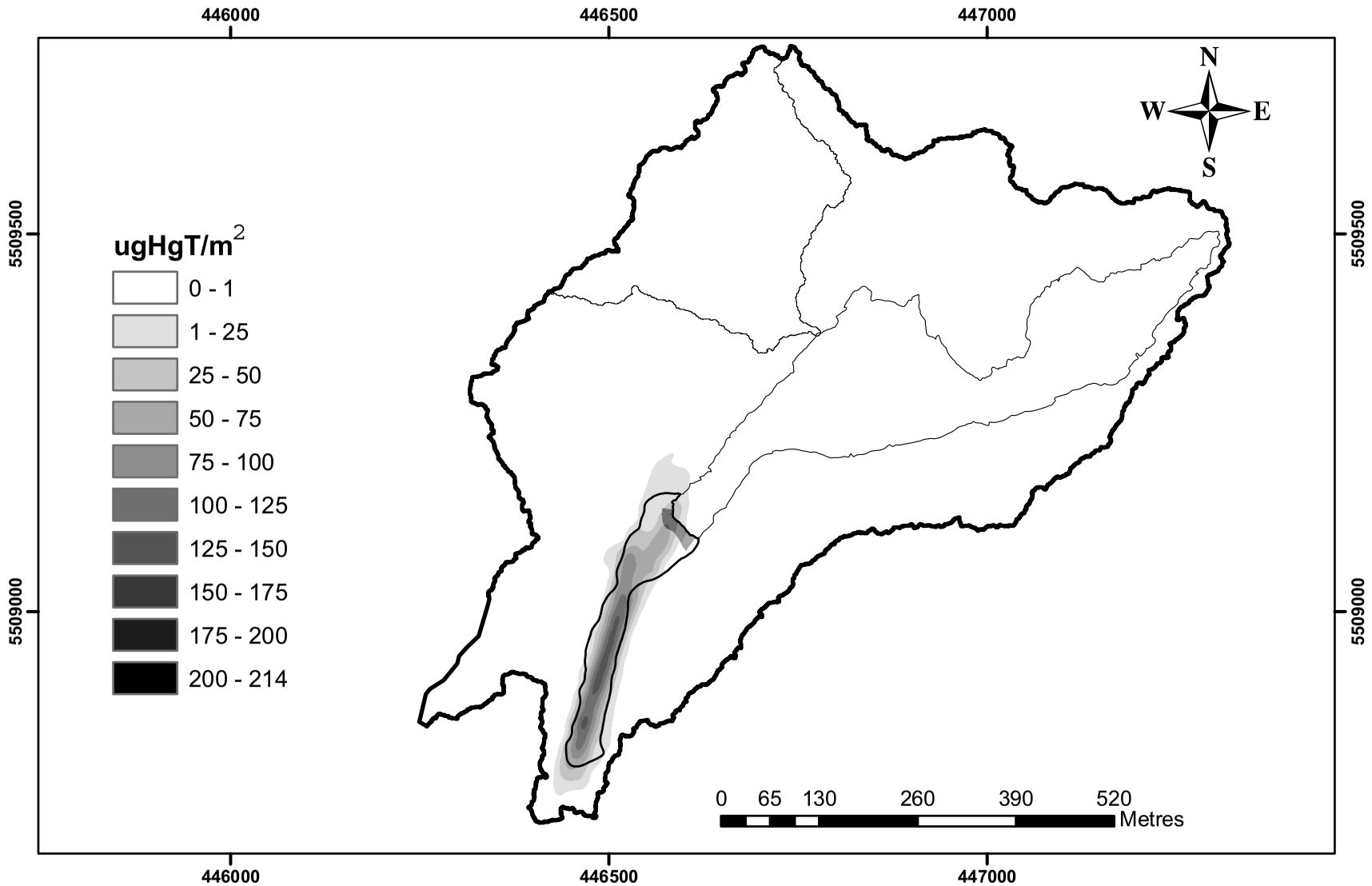

Figure 24. Map of cumulative Wetland Spike Hg release from aerial and shoreline spraying of Lake 658 wetland in 2003.

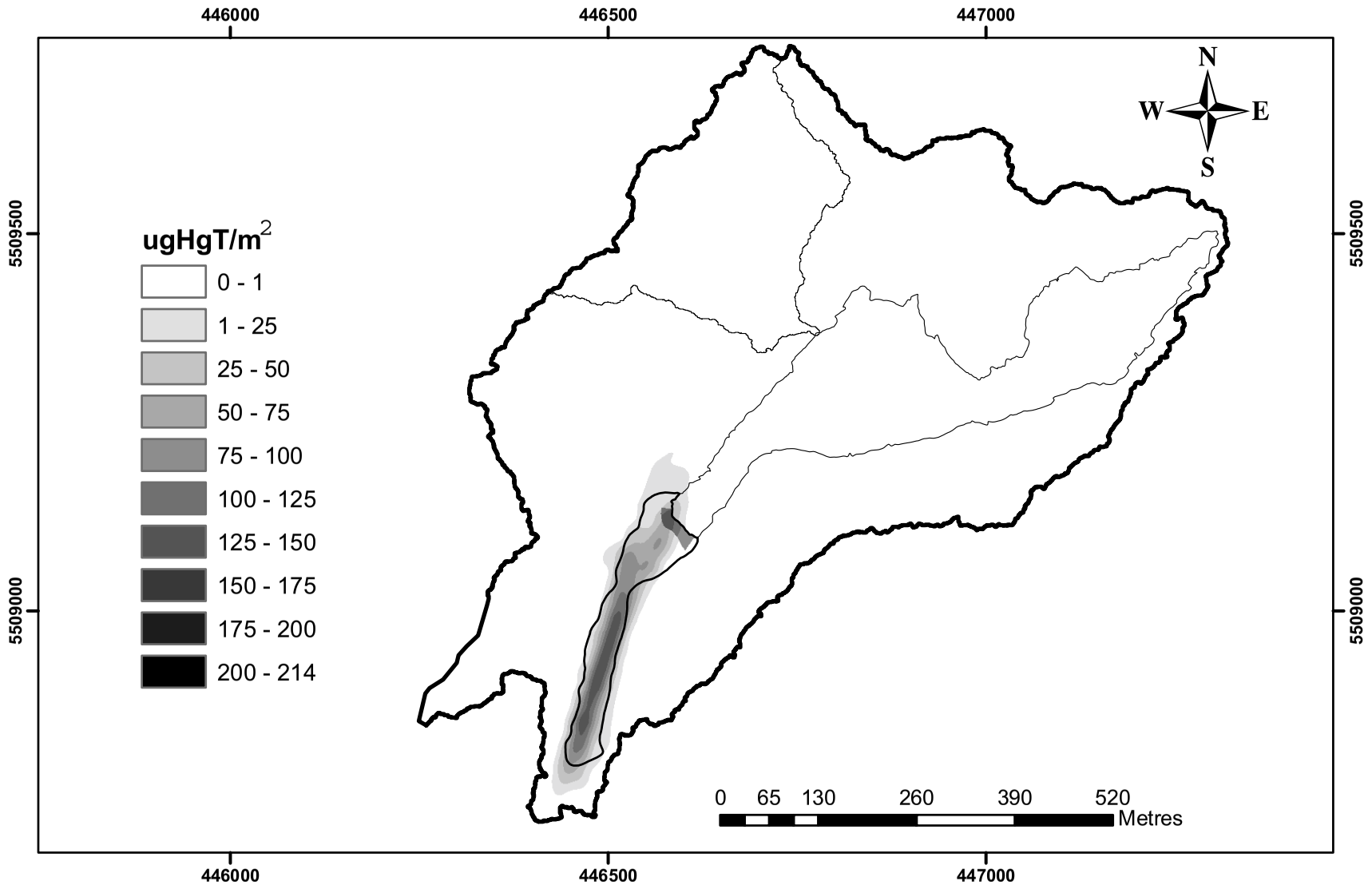

Figure 25. Map of cumulative Wetland Spike Hg release from aerial and shoreline spraying of Lake 658 wetland in 2004.

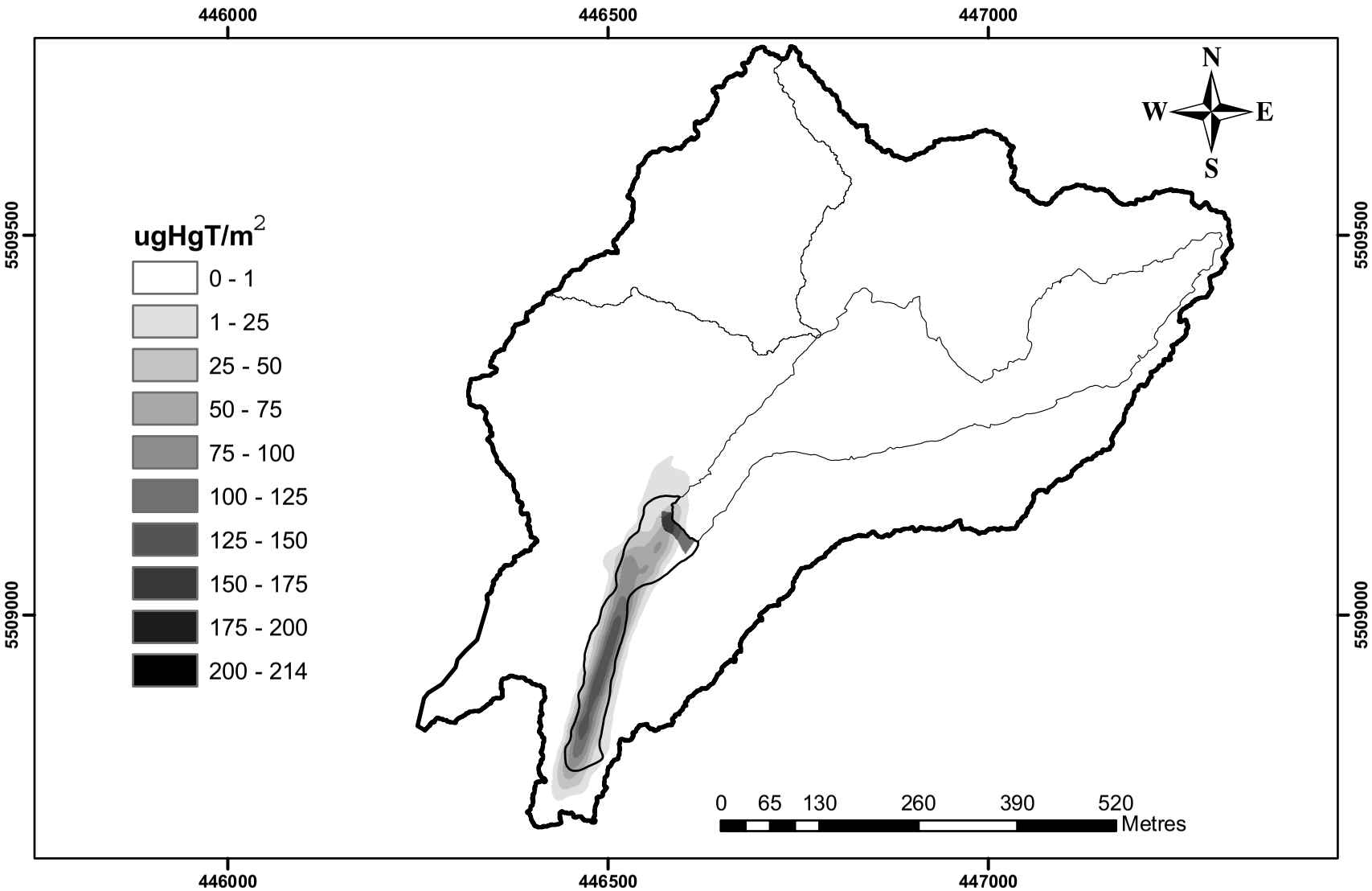

Figure 26. Map of cumulative Wetland Spike Hg release from aerial and shoreline spraying of Lake 658 wetland in 2005.

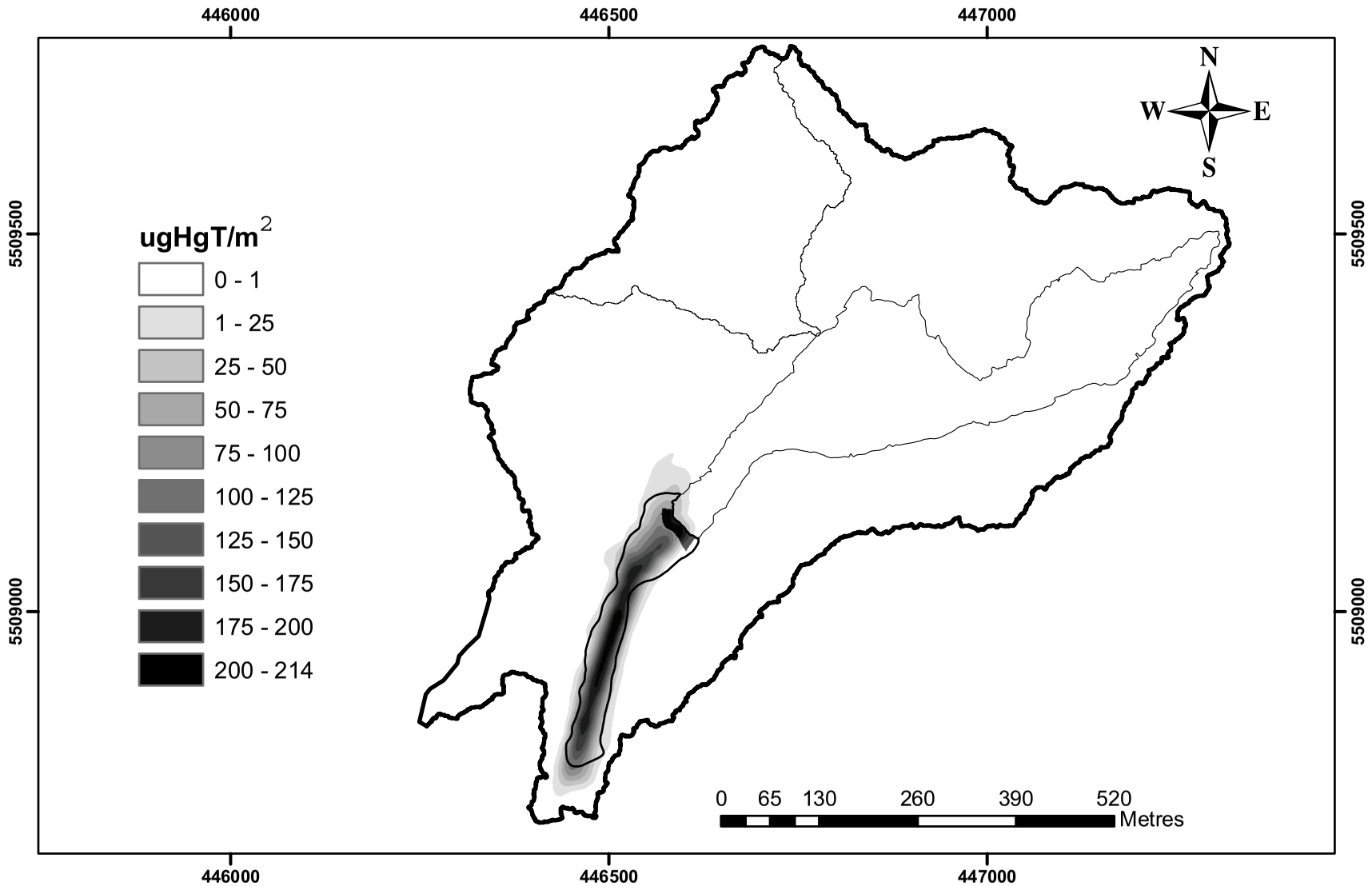

Figure 27. Map of cumulative Wetland Spike Hg release from aerial and shoreline spraying of Lake 658 wetland in 2006.