**Supplementary Material**

Outliers

Ten precipitation events were classified as outliers, and their data are presented in Table SM1.

Five events occurred on Tuesdays and 3 events on Fridays. Hence, it seems possible that an external event could have influenced the local rainwater composition during these events. No additional information was available, but some of these outliers could be related to some periodic event in Barcarena harbor.

**Table SM-1:** Outliers data.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Date | Cations(µeq L-1) | Anions(µeq L-1) | Volume(mL) | Precipitation (mm) | Weekday |
| Jan 24, 2012 | **233** | 132 | 45 | 255 | Tuesday |
| Jan 27, 2012 | 213 | **447** | 340 | 1,925 | Friday |
| Feb 03rd 2012 | **894** | 243 | 480 | 2,718 | Friday |
| May 29, 2012 | 40 | **139** | 142 | 807 | Tuesday |
| Apr 4, 2012 | 96 | **183** | 410 | 2,321 | Wednesday |
| Apr 20, 2012 | 114 | **203** | 305 | 1,727 | Friday |
| Jun 13, 2012 | **185** | 102 | 140 | 793 | Saturday |
| Jun 26, 2012 | **241** | 64 | 400 | 2,265 | Tuesday |
| Jul 03, 2012 | **232** | 128 | 295 | 1,670 | Tuesday |
| Jan 08, 2013 | 110 | **187** | 190 | 1,076 | Tuesday |

Figure SM-1 presents the cation sum versus the anion sum, including outlier results.



**Figure SM-1:** Plot of cation sum versus anion sum, including outlier samples.

Estimation of sulfur deposition due to coal burning

According BARROS (2013), in 2012, Barcarena harbor received 650,000 t of coal. Considering the area of Barcarena is 1,311 km2 and sulfur content in w/w% ranges from low (0.6%) to high (3%) as proposed by YUO, 2010, the sulfur emission in the region was estimated as presented in Table SM-2

**Table SM-2:** Sulfur emission estimate for Barcarena in 2012.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Sulfur content | S(w/w%) | S\*(kg) | S deposition\*\*(g m-2) | SO4-2 deposition(mg m-2) |
| Low | 0.6 | 3,900,000 | 2,975 | 8,900 |
| High | 3.0 | 19,500.000 | 14,874 | 44,600 |

\* present in 650,000 ton of coal.

\*\* over 1,311 km2.

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