Atmospheric Arsenic Deposition in Chiayi County in Southern Taiwan

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ABSTRACT

Although arsenic contamination of underground water in southern Taiwan is well known, few studies examine atmospheric arsenic deposition in this area, which might be the major source of such pollution to the soil, water, and even underground water. This research focused on the atmospheric arsenic concentration, dry and wet depositions, and the As distribution around Chiayi County, located in the south of Taiwan. Eight sampling sites are used, both upwind and downwind of an area with heavy industrial and human activities. All samples were collected by a PS-1 high volume sampler at each site, pretreated by a digestion process, and further analyzed with an ICP-MS. The results show that the arsenic deposition flux ranged from 65.0 to 473 μg/m²-month in Chiayi County. This deposition flux has no significant seasonal variation, based on the multiple trend results obtained from pooling the dry and wet deposition data. The average dry deposition flux of As ranged from 34 to 161 μg/m²-month during the sampling year, and had the opposite trend to the wind speed. Additionally, the correlations between atmospheric arsenic concentration and PM levels were significant, supported by the low p-value (< 0.01) at a 99% confidence level. Meanwhile, the highly linear correlation between As and PM concentrations was also evaluated. The amount of precipitation was statistically correlated to the wet deposition flux (11.3–414 μg/m²-month with a p-value = 2 × 10⁻⁷ and r-value = 0.9306. Furthermore, the composition of the As mass concentrations in ambient air were dominated by As (V) in both fall (66.4%) and winter (68.9%). According to the GIS spatial analysis of the As concentration in the dry season, from October to March, the As contribution of local emissions were not significant. Consequently, this study indicates that the atmospheric As around Chiayi County might be sourced from the upwind northern area, and mainly deposited through wet scavenging.

Keywords: Arsenic; Dry deposition; Wet deposition.

INTRODUCTION

The level of atmospheric arsenic (As) level and its deposition are issues of public concern, due their adverse effects on both human health and the environment (Contreras et al., 2009; Kang et al., 2011; Shi et al., 2012). Arsenic compounds have been identified as Group-I carcinogens, which can enter the human body through respiration, ingestion, and skin absorption (Liu et al., 2007). Arsenic poisoning cases have been reported in various areas of the world since they were able to form stable compounds that could be bio-accumulated in living organisms and affect their immune system (Duker et al., 2005; Sanchez-Rodas et al., 2007; Serbula et al., 2010; Bermudez et al., 2012). Arsenics is not only considered as a carcinogen, but has also been shown to have harmful effects on the bladder, liver, kidney, and lung, and cause several non-cancer diseases, such as diabetes mellitus, hypertension and coronal heart disease (Englyst et al., 2001; Cantor and Lubin, 2007; Coronado-Gonzalez et al., 2007; Boamponsem et al., 2010; Fang et al., 2012b).

In general, arsenic compounds are emitted to the atmospheric environment from both natural sources and human activities, and further contained in suspended aerosol in the atmosphere (Lopez-Anton et al., 2006; Kang et al., 2011). The natural sources of arsenic compounds are
mainly released from volcanic activity, since they are widely distributed in the earth’s crust (2 mg/kg) (Wedepohl, 1991). Throughout the related high temperature processes, some arsenic compounds are adsorbed on volcanic ash and deposited to the ground, while the others remain in atmospheric aerosols, and these are then subject to long-range transportation in the troposphere (Thomaidis et al., 2003; Wang et al., 2010; Chen et al., 2012; Fang et al., 2012a). However, human activities account for three times more emissions of arsenic compounds to the environment than volcanic activity, and these mostly come from mining, municipal solid waste incineration, fossil fuel combustion processes, metallurgical facilities, and vehicles (Wang and Mulligan, 2006; Contreras et al., 2009; Boa Monterey et al., 2010; Lee and Nguyen Thi, 2011; Ny and Lee, 2011; Tian et al., 2011; Chen et al., 2012). Nevertheless, the burning of firework during the special festival in Asian culture was reported as one of the main sources of trace metal, including As (Do et al., 2012).

Anthropogenic emissions of arsenic could accumulate and concentrate on smaller size particulate matters, which cannot be completely removed by air pollutant control devices. These forms of particle-based arsenic have relatively long atmospheric residence times before deposition, and eventually cause pollution several kilometers downwind from the sources (Bencko and Symon, 1977; Saxena et al., 2008; Csavina et al., 2011; Alahmr et al., 2012).

The major removal mechanisms for arsenic suspended in air include gravitational dry deposition and wet scavenging (Andreass et al., 1998; Tsopela et al., 2008; Coskun et al., 2009; Harmens et al., 2010). In dry deposition, arsenic usually presents as a mixture of arsenites As (III) and arsenates As (V) (Mandal and Suzuki, 2002). Both of these can directly cause harmful effects by dermal contact and inhalation, and are predominantly absorbed on particulate matters, especially PM$_{10}$ and PM$_{2.5}$ (Schwartz et al., 1996; Shi et al., 2011; Shi et al., 2012). In addition, the dry deposition of particle-based arsenic on the surfaces of water and soil is considered to be an indirect but important threat to human health (Steinnes et al., 2011; Fang et al., 2012a). The dry deposition flux of total arsenic was reported as 241.39 µg/m$^2$-month in an urban area of Shanghai, which is higher than the amounts reported for suburban (174.33 µg/m$^2$-month) and rural areas (141.89 µg/m$^2$-month) (Shi et al., 2012). For wet deposition, the arsenic adsorbed by aerosols and suspended particles might deposit on water and soil through rain, dew, and snow (Tinggi, 2003). In a coastal area, the arsenic wet deposition flux was reported to be around 20–836 µg/m$^2$-month, varying with the seasonal meteorological conditions. Even in a high altitude atmosphere, the content of Mt. Everest’s ice core has shown to have rising arsenic levels since the 1970s, representing a significant amount of arsenic accumulation (Hong et al., 2009). The impacts of arsenic to these media are a cause for serious concern due to their high toxicity, persistence, and the ability of long-range transportation in the environment (Ng et al., 2003; Kang et al., 2011; De Temmerrman et al., 2012).

In Tainan City, Taiwan, the total amount of particle-based arsenic in the PM$_{2.5}$ and PM$_{10}$ particles has been shown to be 1.09–9.51 ng/m$^2$ and 1.27–13.65 ng/m$^2$, respectively (Tsai et al., 2003). Fang et al. (2012) also reported that the highest concentration of As from dry deposition occurred in winter in central Taiwan for the years 2009 and 2010, and found that it was highly correlated with the steel, electronic and plastic industries, as well as the amount of fossil fuel combustion by a thermal power plant.

Incidents of arsenic contamination and chronic arsenicism have been investigated on the south-west coast of Taiwan, with 7,418 cases of hyperpigmentation, 2,868 of keratosis, and 360 of black-foot disease being observed in a population of 40,421 people in 37 villages, as well as some cases of cancer (Mandal and Suzuki, 2002). However, almost no studies have examined the issue of arsenic deposition, which could directly affect the quality of the soil, water, and underground water around southern Taiwan. Therefore, the current study examines the dry and wet depositions of arsenic in the aerosols and suspended particles in the atmosphere of Chiayi County, which is located in the southern Taiwan. This county contains both agricultural and industrial areas, and the seasonal and regional variations of arsenic concentration and deposition are both considered in this work. The results of this study are expected to be of considerable value with regard to developing better control strategies for arsenic in the atmosphere over Taiwan.

**MATERIALS AND METHODS**

**Sampling Strategy**

All the samples used in this study were obtained from Chiayi County in southern Taiwan, with eight sampling sites chosen upwind and downwind of the potential sources of arsenic emission, as shown in Fig. 1. Five major sources were examined, including Minsyong, Touciao, Jiatai, and Puzi industrial parks and Chiayi City. Sites A (23°35′34.69″N; 120°23′26.13″E) and B (23°33′4.26″N; 120°25′38.73″E) were chosen to provide upwind data, for Minsyong, Touciao, and Jiatai industrial parks and Chiayi City in both cold and warm seasons, while sites C (23°29′24.46″W; 120°17′30.09″E), D (120°19′56.83″W; 23°27′28.82″N) and E (23°25′33.47″N; 120°23′49.82″E) were chose to collect the downwind data. Additionally, sites G and H were located nearby Jiatai and Puzi industrial parks, in order to monitor the arsenic levels in the ambient air that may be related to industrial activity. Finally, site F was located in a rural area that was far from both industrial activity and Chiayi City.

All the meteorological information, PM$_{2.5}$, PM$_{10}$, and total suspended particle (TSP) concentrations for the sampling sites from January 2011 to June 2012 was taken from the Xingang, Puzi, and Chiayi City stations of the Taiwan Air Quality Monitoring Network (Taiwan Environmental Protection Administration, 2012). The average daily temperature of the sampling area, taken from three reference sites, ranged from 15.1°C in January to 29.5°C in August, while the precipitation ranged from 11.3 to 415 mm, and the mean wind speed ranged from 1.86 to 3.22 m/s from January 2011 to June 2012. The precipitation in Chiayi during 2011–2012 showed relatively low values in fall and winter, while
higher values existed in spring and summer. Therefore, the “dry seasons” is defined as October to March, and the “wet season” started from April to September in this study. The atmospheric PM$_{2.5}$, PM$_{10}$, and TSP concentration were illustrated as Fig. 2, comparing with the precipitation and temperature records. The ambient air TSP level in the sampling area varied from the lower values in May, June, July, and August to the higher levels in November, December, January, and February. Generally, the relatively colder atmosphere inhibits the vertical convection of pollutants, while less precipitation reduces the amount of scavenging in fall and winter. Therefore, the atmospheric TSP concentration was higher in the colder and dryer periods (Fig. 2). Notably, the amount of PM$_{2.5}$ tended to be less affected by these two environmental conditions than the amount of coarser particles, since larger particles are mainly formed by surface accumulation, coalescence and agglomeration (Maricq et al., 2002a, 2002b), which might be affected by rainfall and temperature. These environmental conditions will be discussed in more detail later in this work, with regard to their relationships with the level of arsenic in the ambient air.

The atmospheric arsenic samples were collected with a high-volume air sampler (PS-1, Graseby Anderson, GA, USA) and using the standard method (NIEA A102.12A) established by the Environmental Analysis Laboratory, Taiwan. The PS-1 sampling flow rate was controlled at 250–255 L/min for a total of 720 m$^3$ with 48-hour continuous sampling. A quartz fiber filter was used to capture the particulate samples after they had first been conditioned in an electric desiccator operating at 25 ± 5°C and 45 ± 5% humidity for 24 hours. After two days of sampling, the particle-based As samples were then stored in a sealed plastic bag. The chemical analysis of As, As(III), and A(V) was based on NIEA A302.73C and M105.00B, and included an acidic digestion process using HCl and HNO$_3$ in 3:1 (v/v), and further quantification by an inductively coupled plasma mass spectrometer (ICP-MS). The instrument detection limit (IDL) was defined as three times the standard deviation of seven successive blank samples. The IDL of total As, As (III), and As (V) analyses were 0.0233, 0.00145, and 0.0261 ng/m$^3$. The onsite blank samples (n = 2) contained 12.9 pg/m$^3$ As, 0.707 pg/m$^3$ As (III), and 13.4 pg/m$^3$ As (V), which were all determined to be not detectable (ND) and to have analytical purity. The quality assured blank samples were added to the analytical sequence every 10 sample injections, and all showed ND results. The standard solution containing an identified amount of As was used to check the reliability of the calibration line every 10 to 15 samples during the ICP-MS analysis. The calibrated value should be ±10% of the prepared concentration, or the calibration line should be updated to fit the instrument conditions. In this study, the calibration line passed all the reliability tests.
RESULTS AND DISCUSSION

Arsenic Deposition

In this study, the arsenic concentrations in the atmosphere of eight sampling sites were pooled together, and the mean value thus derived represents the average atmospheric arsenic level around Chiayi County in the period of January 2011 to June 2012. The As concentrations ranged from 2.40 to 6.24 ng/m³, close to value found in previous studies (Glooschenko and Arafat, 1988; Boamponsem et al., 2010; Sakata and Asakura, 2011; Fang et al., 2012b). Fig. 3 shows the total (dry + wet) arsenic deposition flux in each month ranged from 65.0 to 473 μg/m²·month, with a mean of 192 μg/m²·month. The seasonal flux variation was not significant, as there were only two very high values, which occurred in November 2011 and June 2012. According to the atmospheric arsenic concentration curve shown in Fig. 3, there were higher values in spring (February to April) and fall (September to November), similar to the trend found in previous studies (Sakata and Asakura, 2011; Shi et al., 2012). However, these concentrations were not closely correlated to the total arsenic deposition fluxes, as seen in Fig. 3. Additionally, the partition ratio of wet deposition (wet deposition flux/total deposition flux × 100%) was higher from May to August (63.6 to 73.2% in mass), which had the similar partition result of the major ionic components reported in the previous study (Budhavant et al., 2012). The two different mechanisms of dry and wet deposition might be the reason why no clear seasonal effect was seen in the pooled deposition fluxes. Therefore, these two pathways are discussed separately in a later section.

Seasonal Variation of Dry Deposition

With regard to the dry deposition flux, the most influential factors were the wind velocity and the particulate concentration in the atmosphere (Sakata and Asakura, 2009). Dry deposition flux levels rose from 34.4 μg/m²·month in January to 152 μg/m²·month in April, then fell again to a low
and PM10 were both 0.00001 (< 0.01) at a 99% confidence. These correlations were carried out using the STATISTICA 10.0 (StatSoft®, 2010) statistical software, with the results shown in Table 1. The results show significant correlations between arsenic concentration and PM levels, while the *p*-values of PM2.5 and PM10 were both 0.00001 (< 0.01) at a 99% confidence level. The highly linear correlations between arsenic and PM concentrations were also evaluated, with *r*-values of 0.9417 and 0.9347 for PM2.5 and PM10, respectively. These high correlations are supported by the results of previous research that focuses on the temporal characteristics of arsenic aerosol in urban areas (Tsai et al., 2003). In addition, the dry deposition fluxes were also significantly correlated with both PM2.5 (*p*-value = 0.0036) and PM10 (*p*-value = 0.0012), although the correlations were weaker than those between arsenic and PM levels. These results might be caused by various factors, such as variations in wind speed and precipitation, which may have affected dry deposition mechanism during the period examined.

The composition of total atmospheric arsenic was also quantified in terms of As(III), As(V), and total arsenic, since they have different toxicities [As(III) > AS(V)]. Table 2 shows that the As(III) level ranged from 0.65 to 1.13 ng/m³ in fall and went up to 1.19 and to 2.80 ng/m³ in winter. Meanwhile, the As(V) concentrations ranged in 1.30–2.20 ng/m³ in fall and increased to 3.25–5.56 ng/m³ in the following winter. The data range found in this study is close to that reported in a previous work that focused on atmospheric arsenic in central Taiwan (Fang et al., 2012a). The results show that the composition of arsenic species in fall was dominated by As(V) in the range of 63.2 to 68.1 wt% (mean = 66.4 wt%), while this increased slightly to 63.0 to 82.4 wt% (mean = 68.9 wt%) in winter. The solubilities of As(V) and As(III) are 167 and 20 g/L, respectively, which supports the higher mass fraction of As(V) being extracted out by wet scavenging. The results of the composition analysis also indicate that As(III) was more abundant in fall 2011 (33.6%) than in winter (31.1%), and thus the level of toxicity was also greater in fall 2011.

**Precipitation and Deposition**

Rainfall can wash out most of the pollutants in the atmosphere (Calvo et al., 2012) and thus also reduce the level of arsenic, and thus can be seen in the wet deposition.
Dry deposition flux of total As ($\mu$g/m$^2$-month) vs. wind speed and As concentration.

Fig. 4. Dry deposition flux of total As vs. wind speed and As concentration.

Total atmospheric As concentration vs. suspended particle levels in the ambient air around Chiayi County.

Fig. 5. Total atmospheric As concentration vs. suspended particle levels in the ambient air around Chiayi County.

Table 1. The correlations among As concentration, dry deposition flux and PM levels in the atmosphere.

<table>
<thead>
<tr>
<th>Arsenic levels</th>
<th>$r$-value</th>
<th>$p$-value</th>
<th>$r$-value</th>
<th>$p$-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>As conc.</td>
<td>0.9417</td>
<td>0.00001</td>
<td>0.9347</td>
<td>0.00001</td>
</tr>
<tr>
<td>As dry deposition flux</td>
<td>0.6489</td>
<td>0.0036</td>
<td>0.7008</td>
<td>0.0012</td>
</tr>
</tbody>
</table>

Fluxes calculated in the current study. Fig. 6 shows the highest wet deposition flux of arsenic was in June 2012, with the value of 414 $\mu$g/m$^2$-month, while the amount of precipitation at this time 415 mm. Additionally, the lowest value was recorded in April 2011, with the arsenic wet deposition flux value and the precipitation being 11.3 $\mu$g/m$^2$-month and 11.3 mm, respectively. The average value for wet deposition over the whole period studied was 108 $\mu$g/m$^2$-month. Additionally, seasonal changes in wet deposition flux were not observed in this study, in contrast to the findings of a previous work (Sakata and Asakura, 2009). According to the calculation of wet deposition, as shown in Eq. (2), the flux was affected by the scavenging ratio, precipitation, and concentration, with the scavenging ratio in both warm and cold seasons set as constants in this study. The seasonal trends of the other two factors are also shown in Fig. 6, indicating very similar
Table 2. Chemical composition of the atmospheric dry arsenic concentration in fall and winter.

<table>
<thead>
<tr>
<th>Sample site</th>
<th>October (Fall) in 2011</th>
<th>February (Winter) in 2012</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>As (III) (ng/m³)</td>
<td>As (V) (ng/m³)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>1.13</td>
<td>2.20</td>
</tr>
<tr>
<td>B</td>
<td>1.01</td>
<td>1.73</td>
</tr>
<tr>
<td>C</td>
<td>0.79</td>
<td>1.69</td>
</tr>
<tr>
<td>D</td>
<td>0.79</td>
<td>1.63</td>
</tr>
<tr>
<td>E</td>
<td>0.77</td>
<td>1.53</td>
</tr>
<tr>
<td>F</td>
<td>0.65</td>
<td>1.30</td>
</tr>
<tr>
<td>G</td>
<td>0.69</td>
<td>1.43</td>
</tr>
<tr>
<td>H</td>
<td>0.86</td>
<td>1.72</td>
</tr>
<tr>
<td>Mean</td>
<td>0.84</td>
<td>1.65</td>
</tr>
<tr>
<td>RSD</td>
<td>19.3</td>
<td>16.2</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample site</th>
<th>October (Fall) in 2011</th>
<th>February (Winter) in 2012</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>As (III) (ng/m³)</td>
<td>As (V) (ng/m³)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>2.80</td>
<td>4.76</td>
</tr>
<tr>
<td>B</td>
<td>1.19</td>
<td>5.56</td>
</tr>
<tr>
<td>C</td>
<td>2.41</td>
<td>5.14</td>
</tr>
<tr>
<td>D</td>
<td>1.88</td>
<td>3.97</td>
</tr>
<tr>
<td>E</td>
<td>1.54</td>
<td>3.28</td>
</tr>
<tr>
<td>F</td>
<td>1.69</td>
<td>3.25</td>
</tr>
<tr>
<td>G</td>
<td>1.67</td>
<td>3.41</td>
</tr>
<tr>
<td>H</td>
<td>2.05</td>
<td>4.26</td>
</tr>
<tr>
<td>Mean</td>
<td>1.90</td>
<td>4.21</td>
</tr>
<tr>
<td>RSD</td>
<td>26.9</td>
<td>21.0</td>
</tr>
</tbody>
</table>

Fig. 6. Wet deposition flux of total As vs. precipitation and As concentration.

Trends for both precipitation and wet deposition flux. With regard to the statistical analyses results shown in Figs. 7(a) and 7(b), a significant linear correlation was found between precipitation and wet deposition flux (p-value = 0.0000002; r-value = 0.9306). Sakata and Asakura (2009) reported that the As wet deposition flux had the same trend as its atmospheric concentration for the period 2003 to 2005, along with significant seasonal variations; however, the As concentration was not a dominant factor with regard to wet deposition (p-value = 0.1077) in the current study. Comparing the amount of precipitation in the sampling period examined in this study and the one in Sakata and Asakura (2009), a relatively high value was found (521 mm) in this work compared to the earlier one (< 450 mm). The wet deposition trends found in the current study were thus also significantly affected by the precipitation, rather than only by the arsenic concentration.

GIS Spatial Distribution of Atmospheric Arsenic Concentration

To analyze the level of arsenic in the ambient air around Chiayi County, a geographical information system, Arc View GIS 3.3, was used to obtain the As distributions. Data for the arsenic concentrations at eight sampling sites in October, 2011 were obtained, because higher levels of air pollutants are generally found in dry and cold seasons, and this was also true in the current work. In Fig. 8, the main wind direction in northern Chiayi was from the northwest (recorded at the Xingang site), and arsenic levels were also monitored in the north-east (in Puzi site) and west north-west (at the Chiayi City site) downwind of areas with heavy industrial and human activity. For the first transport pathway from sites A (3.33 ng/m³) to H (2.58 ng/m³), C (2.48 ng/m³) and G (2.12 ng/m³), the As concentration reduced along with the wind direction. This indicates that the Jiatai and Puzi industrial parks did not make significant contributions to the As concentration, even in their local areas, because the major industries in these parks are metal processing, polymer production, and food production, instead the ones that produce a lot of As, such as copper smelting and solid waste incineration. The same trend was found in another pollutant transport path, from sites A (3.33 ng/m³) to B (2.74 ng/m³) and E (2.30 ng/m³), indicating that Minsyong and Touciao industrial parks and Chiayi City did not have any
significant effects on the As concentration. Based on the As concentration distribution in the dry season, the main contribution to atmospheric As was mid-term transport from the north of Chiayi County, instead of local emissions.

**CONCLUSIONS**

1. The total (dry + wet) arsenic deposition flux of each month ranged from 65.0 to 473 μg/m²-month, with a mean of 192 μg/m²-month in Chiayi County. The seasonal variation of total As deposition flux was not significant, and this was because two different mechanisms (dry deposition and wet scavenging) were pooled together in this work.

2. The average dry deposition flux of arsenic was 84.5 μg/m²-month in Chiayi County, while the seasonal variation ranged from 34 μg/m²-month (wet season) to 161 μg/m²-month (dry season). Additionally, the dry deposition flux had the opposite trend to the wind speed.

3. The correlations between the atmospheric arsenic concentration and PM levels were significant, while the \( p \)-values of PM\(_{2.5} \) and PM\(_{10} \) were both 0.00001 (<0.01) at a 99% confidence level. Highly linear correlation between arsenic and PM concentrations were also evaluated by \( r \)-values equal to 0.9417 and 0.9347 for PM\(_{2.5} \) and PM\(_{10} \) respectively.

4. The composition of arsenic species in fall were dominated by As(V), with the ranged of 63.2 to 68.1 wt% (mean = 66.4%), while this slightly increased to a ranger of 63.0 to 82.4% (mean = 68.9%) in winter.

5. The average value for wet deposition was 108 μg/m²-month, while the highest value of 414 μg/m²-month was found in the wet season, and the lowest value of 11.3 μg/m²-month in the dry season. A significant linear correlation between precipitation and wet As deposition flux was found (\( p \)-value = 0.00000002; \( r \)-value = 0.9306), and the As concentration was not a dominant factor with regard to the wet deposition (\( p \)-value = 0.1077).
6. According to the results of GIS spatial analysis of the As concentration in the dry season, the contribution of atmospheric As due to mid-term transport from the north of Chiayi County, instead of local emissions. The results of the current study indicate that the atmospheric As around Chiayi County might be sourced from the upwind northern area and mainly deposited through wet scavenging, which has not been reported in previous studies. These findings could provide useful information for the development of local control strategies for arsenic emissions.

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