

Appendix 2: Atmospheric Depositions (SI2)

SI2-1. Data Source and Methods

We estimate deposition fluxes of nutrients based on the observational data from 153 stations with ca. 430 data sets across the Changjiang watersheds for the period of 1983-2014 (Figs. SI2-1; Table SI2-1). Because most of observational sites are located in the middle and lower reaches and in order to fill the gap of data in spatial and temporal dimensions, we used measurements from remote and/or national reference stations from Qinghai-Tibetan Plateau in calculating deposition fluxes for the upstream part of the Changjiang watersheds. Also, to calculate the deposition flux and species ratio we used the data from stations outside the Changjiang watersheds to interpolate the concentration and rainfall at the boundary of catchment areas. In the watersheds of the Changjiang, 69% of atmospheric sampling stations are located in the suburb areas of the residence (Group 1), another 31% from countryside and remote areas (Group 2) including forest and grassland (Fig. SI2-1).

In evaluating the significance of atmospheric inputs on the annual scale, we separated wet from dry depositions in data processing. As for wet deposition flux, the volume weighed average concentration (C_i : $\mu\text{mol l}^{-1}$) is used, and the annual deposition flux (F_w^i : $\text{mol km}^{-2} \text{yr}^{-1}$) at a given station is calculated as below:

$$F_w^i = \frac{1}{\sum_{i=1}^m Q_i} \times \sum_{i=1}^m (C_i \times Q_i) \quad (\text{SI2-1})$$

where Q_i is the rainfall (mm). Thus, the annual wet deposition (F_w) of nutrients over the watersheds can be obtained by:

$$F_w = \left(\frac{1}{n} \sum_{i=1}^n F_w^i \right) \times A^i \quad (\text{SI2-2})$$

where n is the number of sampling stations, A^i is the surface area (km^2) represented by Group 1 and/or 2 stations in the watersheds. Then the overall flux of wet deposition

for a given watershed (i.e., tributary) is the integration taking into account the proportion of urban vs remote areas from the GIS data.

With regard to the dry deposition, aerosols and gases species are both considered. As for the aerosols, the deposition flux (F_p) is usually defined as products of aerosol concentration for species of interest ($\mu\text{g m}^{-3}$) and deposition (piston) velocity (m s^{-1}), taking into consideration of element solubility (%) as well.

It should be noted that spatial and temporal coverage is much less for the atmospheric dry deposition samples than rainfall measurements in China. To reduce the bias owing to unbalanced data sets and uneven distribution of observation sites, we estimated the ratio of dry to wet depositions for aerosols ($f_{p/w}^i$) and gas species ($f_{g/w}^i$), respectively, at those stations when both measurements are available:

$$f_{p/w}^i = \frac{F_p^i}{F_w^i} \quad (\text{SI2-3a})$$

and

$$f_{g/w}^i = \frac{F_g^i}{F_w^i} \quad (\text{SI2-3b})$$

Then the dry deposition flux over the watersheds is the integration, taking into account the proportion of urban (Group 1) vs remote (Group 2) areas for a given tributary drainage basin.

SI2-2. Nitrogen

SI2-2.1 Nitrogen deposition flux

With regard to the wet depositions, we focus on NO_3^- and NH_4^+ , since NO_2^- is a minor component and represent only 1%-4% of dissolved inorganic nitrogen (DIN), and data are rather limited in the literature (Zhang et al., 2004; Deng et al., 2009). There is a spatial and temporal variations of nitrogen species across the watersheds of the Changjiang. For example, concentrations of nitrate are 1.69 μM -198 μM

(Average $\pm$ 1sd: 43.4 $\pm$ 32.3 μ M) in rain waters, with the lowest value at stations from the Qinghai-Tibetan Plateau at high elevations and the highest concentration was observed at Nanjing in coastal area. Concentration of NH_4^+ in wet depositions is 8.70 μ M-263 μ M (Average $\pm$ 1sd: 112 $\pm$ 94.3 μ M), with the highest value at Xi'an and lowest concentration from the Lijiang. Overall, concentration of nitrogen in wet depositions is higher in urban areas of middle and lower reaches than the remote region from upper stream of the Changjiang watersheds, with $\text{NO}_3^-/\text{NH}_4^+$ molar ratio of 0.1-2.4 and average of 0.49 $\pm$ 0.38 (1sd), indicating the dominance of NH_4^+ . Dissolved organic nitrogen (DON) concentration and deposition flux in wet depositions are reported from only 31 stations over the whole watersheds of the Changjiang, with DON to DIN ratio (i.e., DON/DIN) of 0.01-1.19, again the highest level of DON was found at Nanjing. Statistically, DON/DIN from Group 1 stations is 0.01-1.19 (Average $\pm$ 1sd: 0.26 $\pm$ 0.34) and 0.01-0.41 (Average $\pm$ 1sd: 0.08 $\pm$ 0.14) from Group 2 stations, respectively.

There are very limited studies on the dry depositions of nitrogen in the watersheds of the Changjiang. Xu et al. (2015) compiled dry deposition data of nitrogen (i.e., NH_3 , NO_2 , HNO_3 , and aerosol- NH_4^+ and NO_3^-) from 43 stations across the country for a study period of 5-years, which showed an average of 1.30 $\pm$ 0.54 (Range: 0.79 to 2.35) of dry to wet deposition ratio of nitrogen for Group 1 and 1.17 $\pm$ 0.93 (Range: 0.30 to 4.34) for Group 2 areas, respectively, shown in Figure SI2-2.

Wet deposition flux of DIN over the Changjiang watersheds has an important spatial structure, i.e., ranging from 14.5 $\times 10^3$ mol N km $^{-2}$ yr $^{-1}$ from remote and upstream stations (e.g., Lijiang) to 535 $\times 10^3$ mol N km $^{-2}$ yr $^{-1}$ at coastal and urban areas (i.e., Tongling). Generally, in the lower part of watersheds, such as provinces of Anhui, Jiangxi and Hunan etc., elevated wet deposition flux from atmospheric pathway has been recorded. In order to constrain the uncertainty in nitrogen depositions over the watersheds of the Changjiang, we compare DIN flux and the proportions of Group 1 vs Group 2 areas for different types of landscape, and then integrate data for the whole drainage basin. Assuming that NO_2^- is 2.5% of $[\text{NO}_3^-]+[\text{NH}_4^+]$ on average, the

results show that DIN deposition amounts to $(257\pm160)\times10^9$ mol N yr⁻¹ (1sd) over the whole watersheds of the Changjiang, with Group 1 being 70% higher than for Group 2 areas.

Furthermore, if we adopt the DON/DIN and the dry to wet deposition ratios mentioned above, a total deposition flux from atmospheric pathways can be obtained, that is $(594\pm367)\times10^9$ mol N yr⁻¹ (1sd) over the whole drainage basin of the Changjiang with 52% from dry depositions.

SI2-2.2 Trend of nitrogen deposition via atmospheric pathways

Owing to the fact that data of nitrogen of atmospheric depositions are still limited to examine the spatial and temporal dimensions from the Changjiang watersheds, we focused on exploring trends of NO₃⁻ and NH₄⁺ in wet depositions in this study. In Group 1 samples concentration of NO₃⁻+NH₄⁺ continues to increase over last 30 years, with a rate of 0.91×10^9 mol N yr⁻¹. However, Group 2 samples do not show any statistical trend for NO₃⁻ and NH₄⁺ over a period of 3 decades, and hence the wet deposition fluxes.

SI2-3. Atmospheric Deposition Flux of Phosphorus and Silicon

When compared to nitrogen, data of dissolved phosphorus (P) and silicon (Si) from atmospheric pathways are even more limited in the literature. Historically, P and Si were not included in atmospheric deposition studies in China, for instance in the national monitoring network for acid rains. In order to estimate the deposition flux of P and Si from atmospheric pathways, we have to compare and summarize the available data in the literature from the whole country (Gan et al., 1995; Bi, 2006; Luo et al., 2007; Yang et al., 2007; Fu et al., 2008; Jiang, 2009; Li et al., 2010; Zhu, 2011; Han, 2013; Qiao et al., 2014; Sun et al., 2014; Zhu et al., 2015; Liao et al., 2015; Wang et al., 2015).

Measurement of P in wet depositions is usually done through oxidation by potassium persulfate according to national protocol GB11893-89 and the results can be considered as the total dissolved phosphorus (TDP_{Wet}), which include both the

inorganic (DIP_{Wet}) and organic (DOP_{Wet}) P species. Dissolved inorganic phosphate in rainwater could be analyzed in photometrical method without digestion. Dry deposition (e.g., aerosols) of dissolvable phosphorus (TDP_{Dry}) was determined based on the measurement of water and/or KCl soluble fractions, and the sum of TDP_{Wet} and TDP_{Dry} represent the total dissolved phosphorus ($\text{TDP}_{\text{Total}}$) via atmospheric pathways. It has been estimated that $\text{DIP}_{\text{Wet}}/\text{DOP}_{\text{Wet}}$ in rainwater is about 1.46 (Bi, 2006; Jiang, 2009), and using this ratio we can estimate that the $\text{DIP}_{\text{Wet}}/\text{TDP}_{\text{Wet}}$ in rainwater is about 0.59. According to the literature reports, $\text{TDP}_{\text{Dry}}/\text{TP}_{\text{Dry}}$ in aerosols is about 0.52, in which TP_{Dry} refers to phosphorus measured in aerosols using potassium persulphate method (Bi, 2006; Jiang, 2009; Zhu, 2011; Han, 2013). For stations where only DIP in aerosols (DIP_{Dry}) were determined, the TDP_{Dry} was estimated using the $\text{TDP}_{\text{Dry}}/\text{DIP}_{\text{Dry}}$ of 1.90 (Bi, 2006; Jiang, 2009).

As shown in Table SI2-2, the estimated TDP_{Wet} ranged from $65 \text{ mol P km}^{-2} \text{ yr}^{-1}$ to $6900 \text{ mol P km}^{-2} \text{ yr}^{-1}$, and TDP_{Dry} ranged from $67 \text{ mol P km}^{-2} \text{ yr}^{-1}$ to $1900 \text{ mol P km}^{-2} \text{ yr}^{-1}$, respectively. The total deposition of dissolved phosphorus via atmospheric pathways was estimated to range from $0.68 \times 10^9 \text{ mol P yr}^{-1}$ to $16.4 \times 10^9 \text{ mol P yr}^{-1}$, with an average of $(3.44 \pm 4.43) \times 10^9 \text{ mol P yr}^{-1}$ (1sd), in which an amount of $0.06 \times 10^9 \text{ mol P yr}^{-1}$ to $2.20 \times 10^9 \text{ mol P yr}^{-1}$ is for Group 1 area with an average of $(0.63 \pm 0.75) \times 10^9 \text{ mol P yr}^{-1}$ (1sd), and $0.62 \times 10^9 \text{ mol P yr}^{-1}$ to $14.2 \times 10^9 \text{ mol P yr}^{-1}$ is for Group 2 region with an average of $(2.80 \pm 3.68) \times 10^9 \text{ mol P yr}^{-1}$ (1sd), respectively.

It should be indicated that gaseous and matrix-bound PH_3 is not included in the flux calculation of phosphorus from atmospheric pathways, because the deposition flux is far much less constrained, with only a few measurements available compared to other P species (cf. Li, 2009).

Data of dissolved silicon (DSi) in atmospheric depositions are much less than phosphorus, and the information available is essentially based on the studies from coastal areas in China (Bi, 2006; Fu et al., 2008; Jiang, 2009; Zhu, 2011; Han, 2013). If we use the data from coastal area as first approximation for the budgetary calculations, DSi deposition over the Changjiang watersheds can be elaborated. At

this time, atmospheric pathway is $(2.86 \pm 1.75) \times 10^3 \text{ mol Si km}^{-2} \text{ yr}^{-1}$ (1sd), and DSi flux ranges from $0.10 \times 10^3 \text{ mol Si km}^{-2} \text{ yr}^{-1}$ to $7.40 \times 10^3 \text{ mol Si km}^{-2} \text{ yr}^{-1}$ for wet deposition (i.e., water and/or HCl soluble fractions). Dry deposition flux ranges from $0.30 \times 10^3 \text{ mol Si km}^{-2} \text{ yr}^{-1}$ to $8.50 \times 10^3 \text{ mol Si km}^{-2} \text{ yr}^{-1}$, with an average of $(1.82 \pm 3.03) \times 10^3 \text{ mol Si km}^{-2} \text{ yr}^{-1}$ (1sd). The sum of dry and wet depositions integrated over the whole drainage basin of the Changjiang provides an estimate of total atmospheric depositions for soluble Si from $0.82 \times 10^9 \text{ mol yr}^{-1}$ to $32.8 \times 10^9 \text{ mol yr}^{-1}$, with an average of $(9.64 \pm 9.84) \times 10^9 \text{ mol yr}^{-1}$ (1sd) in this study, which represents an amount ca. 10% of DSi flux of the Changjiang exported to the East China Sea.

References

- Bi Y.F. (2006) Atmospheric nutrient deposition at the East China coast and its impact on marine primary production. MSc Dissertation, Ocean University of China, Qingdao, pp 87 (in Chinese).
- Deng J.J., Wang T.J., Li S., Xie M. and Fan J.L. (2009) Study on atmospheric nitrogen oxidant and deposition flux in suburban of Nanjing. *Scientia Meteorologica Sinica*, 29 (1): 25-30 (in Chinese).
- Fu M., Zhao W.H., Wang J.T. and Miao H. (2008) Contribution of atmospheric wet deposition to nutrients in the Yangtze Estuary. *Environmental Science*, 29 (10): 2703-2709 (in Chinese).
- Gan J.M., Xue J.Y. and Zhao H.K. (1995) A preliminary study on change of nutrient import and export in the process of rainfall in the Ailao Mountains region of Yunnan Province. *Journal of Natural Resources*, 10 (1): 43-50 (in Chinese).
- Han L.J. (2013) Nutrient characteristics and source apportionment in atmospheric deposition of the Yellow Sea and East China Sea. MSc Dissertation, Ocean University of China, Qingdao, pp 66 (in Chinese).
- Jiang X.L. (2009) Atmospheric dry and wet deposition of nutrients in East China Sea and Yellow Sea and its impact on marine primary production. MSc Dissertation,

- Ocean University of China, Qingdao, pp 74 (in Chinese).
- Li J.B. (2009) The Biogeochemistry of Phosphine in the Coastal Areas of China. PhD Dissertation, Ocean University of China, Qingdao, pp 120 (in Chinese).
- Li T.Q., Jiao F. and Zheng Y. (2010) Atmospheric nitrogen and phosphorus deposition in spring and summer in Beili Lake in Hangzhou. *Environmental Science*, 23 (6): 66-70 (in Chinese).
- Liao Z.L., Li P. and Shang X. (2015) Difference in the atmospheric nitrogen and phosphorus deposition between the typical urban and suburban areas in Wenzhou. *Journal of Zhejiang Agricultural Sciences*, 56 (1): 123-126 (in Chinese).
- Luo L.C., Qin B.Q., Song Y.Z. and Yang L.Y. (2007) Seasonal and regional variations in precipitation chemistry in the Lake Taihu Basin, China. *Atmospheric Environment*, 41: 2674-2679.
- Qiao X., Jiang L.J., Tang Y., Xiong F., Du J. and Xiao W.Y. (2014) The fluxes and possible aquatic impacts of atmospheric nitrogen, sulfur and phosphorous deposition in Jiuzhaigou. *Mountain Research*, 32 (5): 633-640 (in Chinese).
- Sun L.Y. (2014) Atmospheric Nitrogen and Phosphorus Deposition in Nanjing and Effects of Simulated Nitrogen Deposition on Gaseous Emissions of Soils. PhD Dissertation, Nanjing Agricultural University, Nanjing, pp 109 (in Chinese).
- Wang J.F., Zhou K.J., Wang X.Q., Deng J., Wu L.F., Ma X.Y., Zhong X., Pu F.L., Shi L.L., Chen J. and Jiang C.P. (2015) Atmospheric nitrogen and phosphorous deposition in Hangjiahu area. *China Environmental Science*, 35 (9): 2754-2763 (in Chinese).
- Xu W., Luo S.S., Pan Y.P., Zhang L., Tang A.H., Shen J.L., Zhang Y., Li K.H., Wu Q.H. and Yang D.W. (2015) Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China. *Atmospheric Chemistry and Physics*, 15: 12345-12360.
- Yang L.Y., Qin B.Q., Hu W.P., Luo L.C. and Song Y.Z. (2007) The atmospheric

deposition of nitrogen and phosphorus nutrients in Taihu Lake. *Oceanologia et Limnologia Sinica*, 38 (2): 104-110 (in Chinese).

Zhang D., Shi C.X. and Jia L. (2004) A study of chemical properties of rains on the Tibetan Plateau. *Acta Scientiae Circumstantiae*, 24 (3): 555-557 (in Chinese).

Zhu X., Pan Y., Jiao F. and Gu D.M. (2015) Analysis of the atmospheric deposition of nitrogen and phosphorus—taking with Beili Lake as an example. *Journal of Safety and Environment*, 15 (3): 313-316 (in Chinese).

Zhu Y.M. (2011) Nutrient in Atmospheric Deposition of the Yellow and East China Sea. MSc Dissertation, Ocean University of China, Qingdao, pp 78 (in Chinese).

Table SI2-1. Data source list for the nutrient concentrations in wet depositions in the Changjiang drainage basin.

NO	References
1	Aas W.C., Shao M., Jin L. et al. (2007) Air concentrations and wet deposition of major inorganic ions at five non-urban sites in China, 2001–2006. <i>Atmospheric Environment</i> , 41(8): 1706-1716.
2	Bi Y.F. (2006) Atmospheric nutrient deposition at the East China coast and it's impact on marine primary production. Ocean University of China (Master Thesis), Qingdao.
3	Chen J., Meng X.X., Zhang W.D. et al. (2009) Influence of the forest canopy on the precipitation chemical composition in the Simian Mountain in Chongqing. <i>Journal of Anhui Agricultural Sciences</i> , 37(32): 16098-16101.
4	Chen K.J., Chen G.C., Zhang W.D. et al. (2003) Affecting factors on precipitation acidity in chongqing city, China. <i>Bulletin of Mineralogy, Petrology and Geochemistry</i> , 22(2): 163-166.

5	Chen S.J., Tian D.L., Kang W.X. et al. (2004) Chemical properties of the precipitation in the artificial forest of <i>Cinnamomum camphora</i> (L.) presl. <i>Journal of Central South Forestry University</i> , 24(4): 6-10.
6	Chen S.J., Tian D.L., Yan W.D. et al. (2006) Hydrochemical characteristics of through-fall in different layers of <i>Cinnamomum camphora</i> plantation. <i>Chinese Journal of Ecology</i> , 25(7): 747-752.
7	Chen X.Y., Mulder J. (2007) Atmospheric deposition of nitrogen at five subtropical forested sites in South China. <i>Science of the Total Environment</i> , 378: 317-330.
8	Chen X.Y., Mulder J., Wang Y.H. et al. (2004) Atmospheric deposition, mineralization and leaching of nitrogen in subtropical forested catchments, south China. <i>Environmental Geochemistry and Health</i> , 26: 179-186.
9	Chen Y., Tang X., Yang S.M. et al. (2009) Atmospheric N wet deposition in Hangzhou region under rice-wheat-vegetable cropping system. <i>Acta Ecologica Sinica</i> , 29(11): 6102-6109.
10	Cui J., Zhou J., Yang H. (2009) Observations of the input for atmospheric wet-deposition of nitrogen and sulfur into Agro-ecosystem. <i>Ecology and Environmental Sciences</i> , 18(6): 2243-2248.
11	Deng J.J., Wang T.J., Li S. et al. (2009) Study on atmospheric nitrogen oxidant and deposition flux in suburban of Nanjing. <i>Scientia Meteorologica Sinica</i> , 29(1): 25-30.
12	Du C.Y., Zeng G.M., Zhang G. et al. (2008) Input-output budgets for inorganic nitrogen under acid rain in a subtropical evergreen mixed forest in central-south China. <i>Water Air Soil Pollut</i> , 190: 171-181.
13	Du G.Z., Huang X.H., Huang X. et al. (2003) Temporal and spatial

	distribution of acid rain in Hubei province with a discussion on the cause. Resources and environment in the Yangtze basin, 12(4): 377-381.
14	Du M.M., Lu J.X. (1998) Characteristic analysis of acidic precipitation in Nantong area. Environmental Monitoring Management and Technology, 10(5): 12-14.
15	Du X.D., Sun Y. (2003) The characters and strategy of acid rain in Changzhou. Gansu Environmental Research and Monitoring, 16(2): 130-131.
16	Du X.Q. (1999) The present situation of acid precipitation in Zhenjiang city and its countermeasures. Pollution Control Technology, 12(1): 24-26.
17	Duan H., Su Z.X., Li C.Z. et al. (2008) Research on the regional distribution and mechanism of acid rain in Nanchong. Environmental Monitoring in China, 24(3): 79-82.
18	Fan J.L., Hu Z.Y., Wang T.J. et al. (2009) Atmospheric inorganic nitrogen deposition to a typical red soil forestland in southeastern China. Environmental Monitoring & Assessment, 159(1-4): 241-253.
19	Fan J.L., Hu Z.Y., Zhou J. et al. (2013) Comparative study on the observation of atmospheric nitrogen deposition in a forestland. China Environmental Science, 33(5): 786-792.
20	Fan J.L., Hu Z.Y., Zhuang S.Y. et al. (2007) Observation of atmospheric nitrogen deposition into forestland. China Environmental Science, 27(1): 7-9.
21	Fu M., Zhao W.H., Wang J.T. et al. (2008) Contribution of atmospheric wet deposition to nutrients in the Yangtze Estuary. Environmental Science, 29(10): 2703-2709.

22	Fu M.Y., Cao Q.G., Fang M.Y. et al. (1992) Nutrients Cycling in bamboo stands: nutrients input with throughfall and its loss through the run-off of the watershed in pure <i>Phyllostachys pubescens</i> stands. <i>Forest Research</i> , 5(5): 497-505.
23	Fujita S.I., Takahashi A., Weng J.H. et al. (2000) Precipitation chemistry in East Asia. <i>Atmospheric Environment</i> , 34: 525-537.
24	Gan J.M., Xue J.Y., Zhao H.K. (1995) A preliminary study on chance of nutrient import and export in the process of rainfall in the Ailao mountains region of Yunnan province. <i>Journal of natural resources</i> , 10(1): 43-50.
25	Guo W. (2007) Acid rain pollution situation and its chemical components and countermeasures in Xiangtan. <i>Environmental Science Survey</i> , 26(5): 63-66.
26	Han G.L., Liu C.Q. (2006) Strontium isotope and major ion chemistry of the rainwaters from Guiyang, Guizhou Province, China. <i>Science of the Total Environment</i> , 364: 165-174.
27	Han L.J. (2013) Nutrient Characteristics and source apportionment in atmospheric deposition of the Yellow Sea and East China Sea. Ocean University of China (Master Thesis), Qingdao.
28	Hong S.M. (1989) Acidity of the precipitation impacted by the local atmospheric pollution and its chemical composition in the East China area. <i>Meteorological</i> , 15(9): 43-47.
29	Hu C.Y., Hu F.P., Li C.L. (2002) The present situation, characteristics, formative causes and prevention of the acid precipitation in Nanchang city. <i>Acta Agriculhurae Universitatis Jiangxiensis</i> , 24(2): 284-287.
30	Hu Y. (2008) Atmospheric wet deposition chemistry and acid rain control in

	Changde city. <i>Environmental Science and Management</i> , 33(8): 72-75.
31	Hu Z.Y., Xu C.K., Zhou L.N. et al. (2007) Contribution of atmospheric nitrogen compounds to N deposition in a broadleaf forest of southern China. <i>Pedosphere</i> , 17(3): 360-365.
32	Huang D.Y., Xu Y.G., Zhou B. et al. (2010) Wet deposition of nitrogen and sulfur in Guangzhou, a subtropical area in South China. <i>Environmental Monitoring & Assessment</i> , 171: 429-439.
33	Huang F., Sun Y.J., He L. et al. (2007) Acid rain pollution in Yibin city: status and future trends analysis. <i>Environmental Science and Technology</i> , 30(10): 45-49.
34	Huang K., Zhuang G.S., Xu C. et al. (2008) The chemistry of the severe acidic precipitation in Shanghai, China. <i>Atmospheric Research</i> , 89: 149-160.
35	Jia J.Y., Zhang Y., Cai X.B. et al. (2009) A dynamic changes of wet deposition of nitrogen in southeast Tibet: taking Linzhi experiment station as an example. <i>Acta Ecologica Sinica</i> , 29(4): 1907-1913.
36	Jiang X.L. (2009) Atmospheric dry and wet deposition of nutrient in East China Sea and Yellow Sea and it's impact on marine primary production. Ocean University of China (Master Thesis), Qingdao.
37	Jiang Y.M., Zeng G.M., Zhang G. et al. (2003) Atmospheric acid deposition chemistry and the variation characteristics in Changsha City. <i>Urban Environment and Urban Ecology</i> , 16(supply): 23-25.
38	Jin H., Xu D.C., Ye J. (1998) Study on the origin of acid rain formation in the northwest mountain area of Zhejiang. <i>Chongqing Environmental Sciences</i> , 20(5): 26-29.

39	Jin W.M. (2009) Analysis on acid rain component in Nantong, Jiangsu. Environmental protection Science, 35(1): 4-16.
40	Jin X., Hu X.D., Zhou G.L. et al. (2012) Features and causes of acid rain pollution in Jiaying city. Journal of Green Sciences and Technology, (2): 135-137.
41	Li C.L., Kang S.C., Zhang Q.G. et al. (2007) Major ionic composition of precipitation in the Nam Co region, central Tibetan Plateau. Atmospheric Research, 85: 351-360.
42	Li J.B. (2009) The biogeochemistry of phosphine in the coastal areas of China. Ocean University of China (PhD Thesis), Qingdao.
43	Li L.S. (2005) Features of the precipitation pollution and acid rain formation in Zunyi urban area. Guizhou Environmental Protection Science and Technology, 11(4): 6-9.
44	Li T.Q., Jiao F., Zheng Y. (2010) Atmospheric nitrogen and phosphorus deposition in spring and summer in Beili Lake in Hangzhou. Environmental Science, 23(6): 66-70.
45	Li W., Li J., Hou J.X. et al. (2010) Analysis of acid rain's characteristics and variation tendency in Longli experimental plot, Guizhou. Chinese Environmental Science, 30(2): 155-160.
46	Li Y., Yu X.L., Cheng H.B. et al. (2010) Chemical characteristics of precipitation at three Chinese regional background stations from 2006 to 2007. Atmospheric Research, 96(1): 173-183.
47	Liang T., Tong Y.A., Liu X.J. et al. (2014) Dynamics of atmospheric nitrogen wet deposition fluxes in Guanzhong area, Shanxi. Journal of Agro-Environment Science 33(12): 2389-2394.
48	Liao Z.L., Li P., Shang X. (2015) Difference in the atmospheric nitrogen

	and phosphorus deposition between the typical urban and suburban areas in Wenzhou. Journal of Zhejiang Agricultural Sciences, 56(1): 123-126.
49	Lin W. (2012) Acid precipitation variation in Panzhihua city and its chemical features analysis. Panzhihua Science, Technology and Information, 37(2): 53-57.
50	Liu A.P., Li L.S., Ren X.W. et al. (2013) Fluxes and composition of the atmospheric dry and wet deposition of nitrogen and phosphorus in Zhujiang delta areas. Proceedings of the Annual Conference of the Chinese Academy of Environmental Science , Chapter 5: 4649-4656.
51	Liu D.B., Zhang X.Y., Ba R.X. et al. (2015) Atmospheric nitrogen deposition in Danjiangkou Reservoir area of Northwest Hubei. Acta Ecologica Sinica, 35(10): 3419-3427.
52	Liu J. (2010) Preliminary discussion of the present situation and countermeasures of acid rain pollution in Jian city. Jiangxi Chemical Engineering, (4): 119-122.
53	Liu J.Q., Keene W.C., Wu G.P. (1993) Study of precipitation background value in Lijiang, China. China Environmental Science, 13(4): 246-251.
54	Liu L., Liao B.H., Lei M. et al. (2005) The present situation and formative cause and countermeasure research of acid precipitation in Changsha City. Journal of Hunan Agricultural University, 31(2): 216-218.
55	Liu L., Liao B.H., Lei M. et al. (2008) Features and countermeasures of acid rain in Changsha City. Journal of South China Normal University, (3): 89-94.
56	Liu W.J., Ding A.J., Qi W. et al. (2012) Spatial distribution and temporal variation of acid rain in Tongling city and their causes. Journal of Hefei

	University of Technology, 35(5): 687-693.
57	Liu X.H. (2005) Present situation, formative causes, detriment and countermeasures of the acid rain in Liuyang urban area. China Science and Technology Information, (11): 43.
58	Liu Y.K., Hao H., Dai H. (2011) The distribution and characteristic and causes of rainfall in Huaian. Instruments Apparatuses and Analysis Detection, (2): 37-39.
59	Luo L.C., Qin B.Q., Song Y.Z. et al. (2007) Seasonal and regional variations in precipitation chemistry in the Lake Taihu Basin, China. Atmospheric Environment, 41: 2674-2679.
60	Ma L.Y., Wang B., Yang J.G. (2008) Spatial-temporal distribution of acid rain in Sichuan Province. Environmental science and management, 33(4): 26-29.
61	Mei Z.L., Liu Z.Q., Liu L. et al. (2005) Analysis on the variation of acidity and chemical compositions of rainwater in Chengdu urban area. Sichuan Environment, 24(3): 52-55.
62	Meng X.X., Chen J., Zhang D. (2009) Comparison of rainfall composition in 3 different types of monitoring point in Chongqing and analysis on their trend. Journal of Anhui Agricultural Sciences, 37(32): 16135-16138.
63	Qiao X., Jiang L.J., Tang Y. et al. (2014) The fluxes and possible aquatic impacts of atmospheric nitrogen, sulfur and phosphorous deposition in Jiuzhaigou. Mountain Research, 32(5): 633-640.
64	Qiao X., Xiao W.Y., Jaffe D. et al. (2015) Atmospheric wet deposition of sulfur and nitrogen in Jiuzhaigou National Nature Reserve, Sichuan Province, China. Science of the Total Environment, 511: 28-36.

65	Sha C.Y., He W.S., Tong C.F. et al. (2007) Analysis on the variation of acidity and chemical compositions of rainwater in Shanghai. <i>Research of Environmental Science</i> , 20(5): 31-34.
66	Sun L.Y. (2014) Atmospheric nitrogen and phosphorus deposition in Nanjing and effects of simulated nitrogen deposition on gaseous emissions of soils. Nanjing Agricultural University (PhD Thesis), Nanjing.
67	Tu J., Wan H.S., Zhang Z.F. et al. (2005) Trends in chemical composition of precipitation in Nanjing, China, during 1992–2003. <i>Atmospheric Research</i> , 73: 283-298.
68	Wang H. (2004) The spatial-temporal variability of precipitation acidity and its driving factors in Ma'anshan City. <i>Journal of Anhui Normal University</i> , 27(2): 208-211.
69	Wang H., Han G.L. (2011) Chemical composition of rainwater and anthropogenic influences in Chengdu, Southwest China. <i>Atmospheric Research</i> , 99: 190-196.
70	Wang J. (2012) Chemical Characteristics of Ions in Atmospheric Precipitation and their Sources Analysis. <i>Environmental Sciences and Management</i> , 37(3):73-92.
71	Wang J.F., Zhou K.J., Wang X.Q. et al. (2015) Atmospheric nitrogen and phosphorous deposition in Hangjiahu area. <i>China Environmental Science</i> , 35(9): 2754-2763.
72	Wang J.J., Zhang K.R., Wu C. et al. (2014) Wet deposition of atmospheric nitrogen of the Jinshui Watershed in the upper Hanjiang River. <i>Environmental Science</i> , 35(1): 66-72.
73	Wang L. (2007) The research about the chemical composition change rule

	of precipitation in Jiangxi. Jiangxi Science, 25(6): 781-783.
74	Wang X.Y., Zhang S.G. (2000) Elementary research of characteristics and trend of chemical composition of precipitation in Wuhu. Journal of Anhui Normal University, 23(3): 276-279.
75	Wang X.Z., Zhu J.G., Gao R. et al. (2004) Dynamics and ecological significance of nitrogen wet-deposition in Taihu Lake region- taking Changshu Agroecological Experiment Station as an example. Chinese Journal of Applied Ecology, 15(9): 1616-1620.
76	Wei H., Wang J.L., Li X.G. (2005) Analysis on Seasonal Variation of Chemical Composition of Rainfall in Jinyun Mountain, Chongqing China. Journal of Southwest China Normal University (Natural Science), 30(4): 725-729.
77	Wu J.Y., Deng S.H., Zheng R.H. et al. (2006) Formation mechanism research of acid rain in Yaan urban area. Journal of Sichuan Agricultural University, 24(4): 432-434.
78	Wu Q.X., Han G.L. (2012) Chemical characteristics of rainwater collected in the head region of Three Gorges Reservoir. China Environmental Science, 32(3): 385-390.
79	Wu Q.X., Han G.L., Tao F.X. et al. (2011) Chemical characterization of rainwater in a karst rural site: a case study of Puding, China. Environmental Sciences, 32(1): 26-32.
80	Wu Q.Z. (2011) Analysis of pollution characteristics and variation tendency of precipitation in Jingzhou City. Journal of Yangtze University, 8(10): 243-248.
81	Wu S.X. (2011) Analysis on characteristics of chemical composition of precipitation in Yingtan. Jiangxi Chemical Engineering, (3): 62-66.

82	Xiao H.W., Xiao H.Y., Wang Y.L. (2010) Chemical characteristics and source apportionment of precipitation in Guiyang. Chinese Environmental Science, 30(12): 1590-1596.
83	Xie Y.X., Xiong Z.Q., Xing G.X. et al. (2008) Source of nitrogen in wet deposition to a rice agroecosystem at Tai lake region. Atmospheric Environment, 42: 5182–5192.
84	Xiong C.J. (2006) Preliminary study of the present situation, characteristics and formative causes of the acid rain in Yingtian city. Acta Agriculturae Jiangxi, 18(4): 165-166.
85	Xu J., Zhang X.L., Xu X.B. et al. (2008) Variations and source identification of chemical compositions in wet deposition at Shangdianzi background station. Acta Scientiae Circumstantiae, 28(5); 1001-1006.
86	Xu W., Luo S.S., Pan Y.P. et al. (2015) Quantifying atmospheric nitrogen deposition through a nationwide monitoring network across China. Atmospheric Chemistry and Physics, 15:12345-12360.
87	Yang L.Y., Huang L., Sun Y. (2009) Acidity and Ionic Characteristics of Precipitation in Changzhou, 2007. Environmental Chemistry, 28(6): 946-947.
88	Yang L.Y., Qin B.Q., Hu W.P. et al. (2007) The atmospheric deposition of nitrogen and phosphorus nutrients in Taihu Lake. Oceanologia et Limnologia Sinica, 38(2): 104-110.
89	Yang L.Y., Yu X.N. (2010) Chemical characteristics of acidic precipitation and its relation to atmospheric pollutants in Changzhou city. Urban Environment and Urban Ecology, 23(5): 30-33.
90	Yang R., Hayashi K., Zhu B. et al. (2010) Atmospheric NH ₃ and NO ₂

	concentration and nitrogen deposition in an agricultural catchment of eastern China. <i>Science of the Total Environment</i> , 408: 4624-4632.
91	Yuan L., Zhou X.B., Gu X.R. et al. (2009) Variation in wet deposition of nitrogen from atmosphere in typical areas of Chongqing. <i>Acta Ecologica Sinica</i> , 29(11): 6098-6101.
92	Zeng K., Ju W.M., Tu L.Y. et al. (2010) Characteristics of acid rain in the suburb and urban areas of Nanchang during 2006 and 2007. <i>Journal of Agro-Environment Science</i> , 29(3): 609-612.
93	Zhang D., Shi C.X., Jia L. (2004) A study of chemical properties of rains on the Tibetan Plateau. <i>Acta Scientiae Circumstantiae</i> , 24(3): 555-557.
94	Zhang F.Z., Zhang J.Y., Zhang H.R. et al. (1995) Chemical Composition of Precipitation in a Forest Area of Chongqing, Southwest of China. <i>Water Air Soil Pollut</i> , 90: 407-415.
95	Zhang G., Zeng G.M., Jiang Y.M. et al. (2003) Study on the atmospheric chemistry of wet deposition in Hunan Province. <i>Environmental monitoring in China</i> , 19(5): 7-11.
96	Zhang N.N., He Y.Q., Cao J.L. et al. (2012) Long-term trends in chemical composition of precipitation at Lijiang, southeast Tibetan Plateau, southwestern China. <i>Atmospheric Research</i> , 106: 50-60.
97	Zhang Q., Yu J., Yu Y.Y. (2009) Analysis on the characteristics and developing trend of the acid rain in Nanjing. <i>Environmental Science and Management</i> , 34(12): 108-126.
98	Zhang Y., Song L., Liu X.J. et al. (2012) Atmospheric organic nitrogen deposition in China. <i>Atmospheric Environment</i> , 46: 195-204.
99	Zhang Y., Zheng L.X., Liu X.J. et al. (2008) Evidence for organic N deposition and its anthropogenic sources in China. <i>Atmospheric</i>

	Environment, 42(5): 1035-1041.
100	Zhao D., Xiong J., Xu Y. et al. (1988) Acid rain in Southwestern China. Atmospheric Environment, 22(2): 349-358.
101	Zhao L., Lu Q.M., Li L. et al. (2013) Chemical characteristics of atmospheric precipitation at Wanzhou District of Chongqing. Environmental and Ecology in Three Gorges, 35(2): 9-15.
102	Zhao X., Yan X.Y., Xiong Z.Q. et al. (2009) Spatial and temporal variation of inorganic nitrogen wet Deposition to the Yangtze River delta region, China. Water Air Soil Pollut, 203: 277-289.
103	Zhong L.L. (2003) The pollution and prevention of acid rain in Jingdezhen. China Environmental Management, 22(2): 43-44.
104	Zhou J.Y., Liu G., Ma X.M. et al. (2004) The chracterristic of acid deposition, disciplinarian and its policy of prevention and cure in the area of Leshan. Journal of Geological Hazards and Environment Preservation, 15(3): 31-34.
105	Zhu J.P. (2003) Analysis of characteristics of acid rain pollution in Mianyang city. Journal of Southwest University of Science and Technology, 18(2): 59-62.
106	Zhu J.X., He N.P., Wang Q.F. et al. (2015) The composition, spatial patterns, and influencing factors of atmospheric wet nitrogen deposition in Chinese terrestrial ecosystems. Science of the Total Environment, 511: 777-785.
107	Zhu X., Pan Y., Jiao F. et al. (2015) Analysis of the atmospheric deposition of nitrogen and phosphorus—taking with Beili Lake as an example. Journal of Safety and Environment, 15(3): 313-316.
108	Zhu Y.F., Yang W.W. (2004) A preliminary study of the features and

	formative causes of the acid precipitation in Taizhou city. Environmental Monitoring Management and Technology, 16(2): 20-22.
109	Zhu Y.M. (2011) Nutrient in atmospheric deposition of the Yellow and East China Sea. Ocean University of China (Master Thesis), Qingdao.
110	Zhu Z.Y., Chen D.R., Yin J. et al. (2003) Analysis of the chemical characteristics of the precipitation in Chongqing city. Chongqing Environmental Science, 25(11): 112-114.

Table SI2-2. Deposition flux of phosphorus via atmospheric pathways in China (Unit: mol P km⁻² yr⁻¹).

Location /City	Wet Depositions			Dry Depositions				References	Groups
	DIP _{-Wet}	DOP _{-Wet}	TDP _{-Wet}	DIP _{-Dry}	DOP _{-Dry}	TP _{-Dry}	TDP _{-Dry}		
Wuxi			516					Luo et al., 2007	1
Suzhou			2000					Luo et al., 2007	1
Changzhou			581					Luo et al., 2007	1
Jintan			1065					Luo et al., 2007	1
Jingdong			3548					Gan et al., 1995	2
Hangzhou			65			146	76 ^c	Wang et al., 2015	1

Jiaxing			73		152	79 ^c	Wang et al., 2015	1
Huzhou			88		129	67 ^c	Wang et al., 2015	1
Nanjing			3548			1290	Xie et al., 2008	1
Zhoushan	150	170	320 ^a	180		342 ^d	Sun, 2014	2
Zhoushan	110	110	220 ^a				Jiang,2009	2
Zhoushan	80		136 ^b				Zhu, 2011	1
Zhoushan	110		186 ^b				Han, 2013	1
Fulongshan	570	290	860 ^a	1000		1900 ^d	Sun, 2014	1
Fulongshan	70		119 ^b	600	160	760 ^a	Zhu, 2011	1
Fulongshan	3700	3200	6900 ^a	400		760 ^d	Jiang,2009	1
Qianliyan	450	130	580 ^a	220		418 ^d	Sun, 2014	1
Qianliyan	280		475 ^b	240		456 ^d	Jiang,2009	1
Qianliyan	790		1339 ^b	240	490	730 ^a	Zhu, 2011	1
Qianliyan	400		678 ^b	310	360	670 ^a	Han, 2013	1
Taihu			281		713	371 ^c	Yang et al., 2007	2
Rize			484		516	268 ^c	Qiao et al. 2014	1
Goukou			419		581	302 ^c	Qiao et al., 2014	2

Hangzhou		581	2645	1375 ^c	Zhu et al., 2015b	1
Chongming	170	288 ^b			Fu et al., 2008	1
Guangzhou		1181	368	191 ^c	Liu et al., 2013	1

Note: Normal letters show the data from the literature, and the italic letters show the values from calculation. In the table, “a” is based on $TDP = DIP + DOP$; “b” is based on $DIP_{-Wet}/TDP_{-Wet}=0.59$; “c” is based on $TDP_{-Dry}/TP_{-Dry}=0.52$; and “d” is based on $DIP_{-Dry}/TDP_{-Dry}=1.90$.

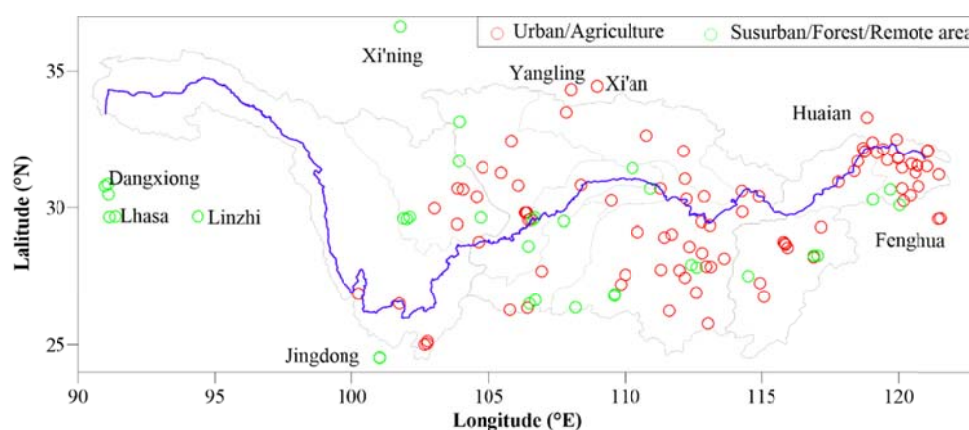


Figure SI2-1. Sample locations of atmospheric wet and dry deposition studies over the watersheds of the Changjiang. Due to very limited sampling coverage in the upstream area, observations from station outside the drainage area but with similar landscape and/or land-use pattern are shown for data compilation, such as Jingdong, Linzhi, Lhasa, Dangxiong, Xi'ning, Yangling and Xi'an. In the figure, samples are classified into Group 1: urban/agriculture areas and Group 2: suburban/forest/remote areas, respectively.

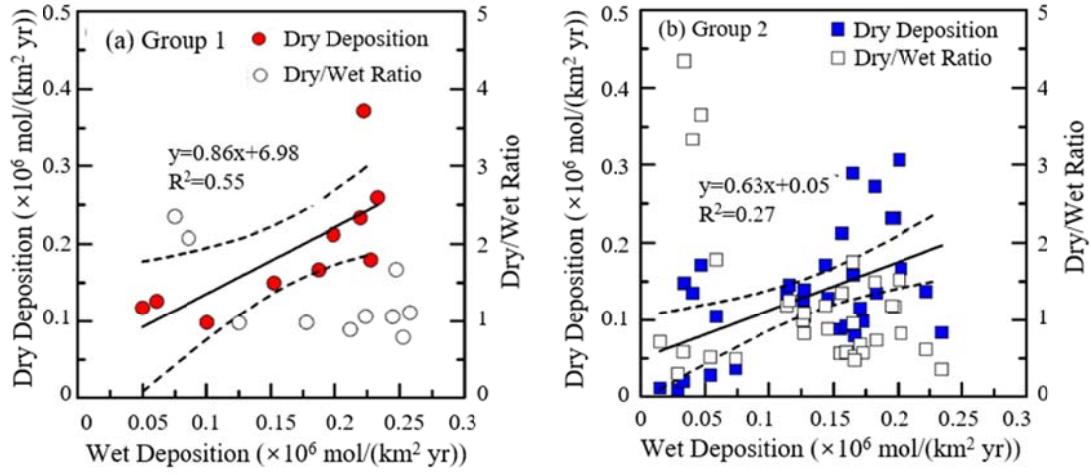


Figure SI2-2. Relationship between the dry and wet depositions from 43 stations over the watersheds of the Changjiang, data are from Xu et al., 2015. For the purpose of data comparison samples are classified into Group 1 (a) and Group 2 (b), respectively. The black solid lines and dashed curves refer to the linear regression and 95% confidence levels.