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Geochemical Processes of Organic Pollutants in a Typical Subtropical Watershed: A Case Study with Decabromodiphenyl Ether

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Abstract
The Pearl River Delta (PRD) is a subtropical watershed located in South China and noted for its fast-growing economic development. As a result of the high industrial and agricultural productivities, as well as primitive handling of electronic waste (e-waste), organic contamination has become a serious threat to the sustainability of socioeconomic growth in the region. To provide a better understanding of the fate of organic pollutants within the environment of the PRD, this study examined various inter-compartmental fluxes and constructed a mass transport budget using decabromodiphenyl ether (BDE-209), a major constituent in brominated fire retardants abundantly embedded in e-waste, as the target analyte. The results show that atmospheric deposition (9,200 kg/yr annually for dry and wet depositions combined) was the most important vector in cycling of BDE-209; particularly, wet deposition (2,500 kg/yr) was in the same order of magnitude as dry deposition (6,700 kg/yr), which is obviously attributed to the large amounts of precipitations encountered in the PRD subject to the East Asian monsoon system. Riverine runoff, on the other hand, carried an unexpectedly small amount (1,960 kg/yr) of BDE-209 from the PRD; however, most (1,550 kg/yr) of this amount continues to be discharged into deeper oceans. The soil inventory (44,000 kg/yr) of BDE-209 was only several times the amount of annual atmospheric deposition, probably because BDE-209 is still being released from current sources. Nevertheless, imported e-waste (particularly from industrialized countries, mostly from the United States) appears to be a potentially gigantic source of BDE-209 with an annual accumulation rate estimated at 9,400,000 kg/yr. Finally, useful inferences can be drawn for the fate of other organic pollutants from the present study.

Keywords: Organic pollutants; Inter-compartmental flux; Geochemical process; e-Waste; Decabromodiphenyl ether; Pearl River Delta

1. Introduction

The Pearl River Delta (PRD), located in a subtropical region of South China to the north of the South China Sea and with an area of 53,580 km^2 within Guangdong Province (Figure 1), is one of the most economically prosperous areas in China. Rapid economic development and accelerated urbanization in the region over the last three decades have resulted in elevated levels of pollution in various environmental compartments of the PRD. Aside from traditional contaminant inputs such as waste discharges from industrial, agricultural and domestic activities, primitive handling of electronic waste (e-waste), largely derived from importation to the PRD, containing heavy metals and

organic pollutions has drastically added to the magnitude of the problem (Ni and Zeng, 2009). The fact that China has been importing the largest portion of e-waste generated worldwide, with the PRD being the main destiny, has raised great concerns worldwide (Ogunseitan et al., 2009; Stone, 2009).

Polybrominated diphenyl ethers (PBDEs) are major constituents of brominated fire retardants (BFRs) abundantly used in a variety of electronic and electric devices, which become obsolete at the end of their life cycles. Because huge amounts of BFRs have been used in the manufacture of electronics and appliances in the PRD and have been imported to the region in the form of e-waste (a fair amount of BFRs is used locally in manufacturing electronics and appliances, but much larger amounts of BFRs are imported to the region via e-waste), high levels of PBDEs have been detected in various PRD sample matrices including riverine runoff (Guan et al., 2007), soil (Zou et al., 2007), sediment (Mai et al., 2005), air (Chen et al., 2006; Zhang et al., 2009), precipitation (Zhang et al., 2009), aquatic products (Guo et al., 2007; Meng et al., 2007), human breast milk (Bi et al., 2006) and blood plasma (Bi et al., 2006; Qu et al., 2007). In addition, because of their persistency in the environment, ability to accumulate in biota, toxic potency to both the ecological system and human health, and potential for long-range atmospheric transport, tetrabromodiphenyl ethers and pentabromodiphenyl ethers were included in the Stockholm Convention on Organic Persistent Pollutants in May 2009 (http://chm.pops.int/).

Despite the tremendous efforts to characterize the occurrence of organic pollutants in the environment of the PRD, it remains relatively unclear how organic pollutants such as PBDEs are transported among various environmental compartments of the PRD. Such information is critical for understanding the dynamics of inter-compartmental fluxes of organic pollutants, and would allow a better assessment of the environmental behavior and effects of organic pollutants. To this end, we will use decabromodiphenyl ether (BDE-209), the most abundant component among the BDE congeners found in the environment of the PRD, as the target compound to construct a mass transport budget using data available from the literature.

To accomplish the above-mentioned goal, individual inter-compartmental processes for BDE-209 have to be examined. The main inter-compartmental processes, specifically applicable to the geographical setting of the PRD, include riverine runoff, dry and wet atmospheric depositions, air-soil and air-water gaseous exchange, long-range atmospheric transport in and out of the region, and e-waste emissions. Minor routes include city emission and bioprocess (e.g., uptake and release by terrestrial and aquatic biota). In addition, soil (and sediment) is perhaps the largest reservoir of BDE-209 and may become a buffer mediating the geochemical cycling of BDE-209. Because the Pearl River Estuary (PRE, Figure 1) connects the main portion of the PRD with the northern South China Sea, the geochemical processes (Figure 2) occurring in the PRE play an important role in controlling the outflows of BDE-209 to deep seas and is also considered in the present study.

In the rest of this paper, I will summarize the procedures used to obtain the fluxes of

BDE-209 for riverine runoff, dry and wet atmospheric depositions and e-waste emission, as well as soil inventory of BDE-209, from available literature or current knowledge. Finally, the implications of the mass transport budget will be briefly discussed.

2. Riverine Runoff Transport

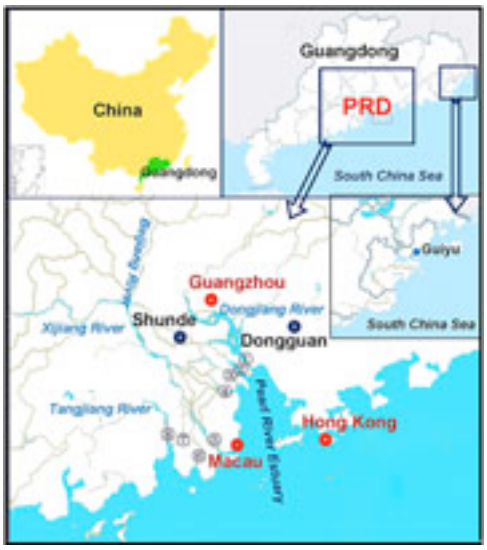

Figure 1
(see appendix for larger image)

Because of the rich river network in the PRD (Figure 1), riverine runoff is an important mode to carry BDE-209, derived from all sources such as industrial, agricultural and residential discharge, surface washoff and precipitation, into the coastal ocean and further to the global oceans. Four main rivers (Xijiang, Beijiang, Dongjiang and Tangjiang Rivers) flow into the PRD and merge into the northern South China Sea through eight outlets, i.e., Humen, Jiaomen, Hongqili, Hengmen, Modaomen, Jitimen, Hutiaomen and Yamen (Figure 1). Monthly flux of BDE-209 through the eight outlets into the coastal ocean was calculated with $F_{i,j} = kC_{i,j} \times D_{i,j}$, where k is a unit conversion factor, $C_{i,j}$ is the concentration of BDE-209 in riverine runoff collected from a specific outlet for a given month, and $D_{i,j}$ stands for the corresponding total water discharge for the same month. The total annual flux (F_i) from the *i*th outlet was then estimated by

$$F_i = \sum_{j=1}^{12} F_{i,j} \quad \textbf{(1)}$$

Based on Eq. (1), our previous study obtained an annual riverine input of BDE-209 at 1.96 tons/yr (Guan et al., 2007). For comparison, the annual riverine mass emissions of polycyclic aromatic hydrocarbons (PAHs; sum of 28 components), dichlorodiphenyltrichloroethanes (DDTs; sum of *o,p*'- and *p,p*'-DDT, DDD and DDE), hexachlorocyclohexanes (sum of α-HCH, β-HCH, γ-HCH, and δ-HCH) and PBDEs (sum of 10 congeners including BDE-209) were 33.9 (Wang et al., 2007), 1.3 (Guan et al., 2009b), 1.23 (Guan et al., 2009b) and 2.14 tons/yr (Guan et al., 2007), respectively. Further analyses of the data unveiled considerably linear correlations between the riverine inputs of organic pollutants and water discharge (Guan et al., 2007; Guan et al., 2009b; Wang et al., 2007; Wang et al., 2008), suggesting that water discharge is likely to be the crucial factor in controlling the pollutant riverine inputs in the PRD.

3. Atmospheric Depositional Rates

Atmospheric depositional rates can be contributed from dry and wet depositions, and they are defined as Fdry and Fwet, respectively and estimated with the following equations (Cetin and Odabasi, 2007; Venier and Hites, 2008):

$$F_{dry} = C_p v_d A = (C_{p,u} A_u + C_{p,r} A_r)\, v_d \quad \textbf{(2)}$$

$$F_{wet} = (VWM)pA = (VWM_u A_u + VWM_r A_r)p \quad \textbf{(3)}$$

where C_p is the BDE-209 concentration in the atmospheric particulate phase (kg/m^3), v_d is the particle dry depositional velocity (m/yr), A is the study area (m^2), *VWM* is the volume weighted mean BDE-209 concentration in precipitation (kg/m^3), p is the precipitation rate (m/yr), and u and r denote the urban and rural areas, respectively.

Concentrations of BDE-209 in air (Chen et al., 2006) and rain (Zhang et al., 2009) of Guangzhou, the capital of Guangdong Province (Figure 1), have been measured, and were used as the urban data for Eqs. (2) and (3). In addition, Zhang et al. (2009) measured the concentrations of BDE-209 in air and precipitation from Dongguan and Shunde (Figure 1), which were regarded as the rural concentrations. As a result, the annual dry and wet depositional rates of BDE-209 were estimated at 6,700 and 2,500 kg/yr, respectively, for the entire PRD (Zhang et al., 2009). Apparently, dry deposition is the predominant mode to remove BDE-209 from the atmosphere in the PRD, but wet deposition also contributes substantially to the overall atmospheric fallout of BDE-209. The sizeable contribution from wet deposition can be attributable to the generally large amount of precipitations in the region, e.g., average 1300 to 2284 mm in various parts of Guangzhou Province embracing the PRD in 2008 (Statistical Bureau of Guangdong Province, 2009).

4. Potential Loading from E-Waste

The amount of BDE-209 potentially emitted from e-waste to the PRD (I; tons/yr) was estimated with the following equation:

$$I = kVC \quad \textbf{(4)}$$

where k is a unit conversion constant, V stands for the volume (V does not necessarily represent volume. In the current example, weight is used to designate the amount of e-waste generally annually worldwide. In eq. (4), k is used to transform different units) of e-waste annually generated in and/or imported to the PRD area, and C represents the average concentration of BDE-209 in e-waste. A literature survey indicated that the gross amount of e-waste generated annually worldwide is 20-50 million tons/yr, approximately 70% of which is shipped to China (Ni et al., 2010). In addition, plastics account for ~12-20% of the e-waste weight and the concentration of BDE-209 in plastics is 21,000,000 ng/g (Choi et al., 2009). If 16% is taken as the relative abundance of plastics in e-waste, substituting the above values into Eq. (4) yields the annual potential loading of BDE-209 from e-waste imported to China at 47,000-118,000 tons/yr. If it is assume that 20% of the total amount of e-waste imported to China is shipped to the PRD, the low end of the annual loading of BDE-209 from e-waste to the PRD is ~9,400 tons/yr. This assumption is extremely conservative as Guiyu, located in Guangdong Province (Figure 1), is home to some of the world's largest e-waste dismantling and recycling facilities; as a result, large amounts of e-waste are expected to be processed in the region.

5. Mass Inventory in Soil

A hypothetical 20-cm deep layer of soil was used to estimate the mass inventory (I; kg) of BDE-209 in the surface soil of the PRD using

$$I = kCAd\rho \quad \textbf{(5)}$$

where k is a unit conversion factor, C is the area weighted concentration of BDE-209 (ng/g), A is the study area (m^2), d stands for the soil depth (20 cm), and ϱ is the ideal soil density (1.5 g/cm^3). With Eq. (5) Zou et al. (2007) obtained the soil inventory of BDE-209 in the PRD at 44,000 kg. Inevitably, this value should be quite dynamic as the atmospheric deposition estimated above contributes approximately 9,200 kg/yr or ~20% at the current level to the soil inventory annually.

6. Geochemical Dynamics in the Pearl River Estuary

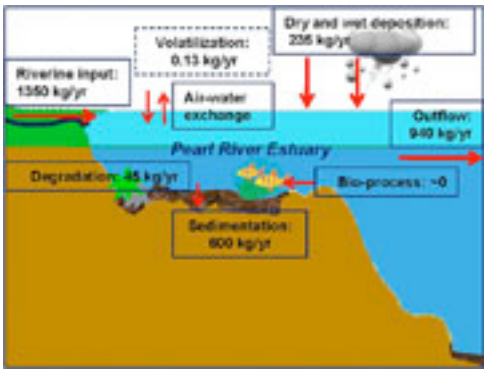

Figure 2
(see appendix for larger image)

As mentioned above, the geochemical dynamics occurring in the PRE (Figure 2) dictates the outflows of BDE-209 into the coastal ocean. The main processes considered include air-water gaseous exchange, dry and wet depositions, sedimentation and degradation and are discussed below.

6.1. Atmospheric Deposition

Dry and wet depositional fluxes can be estimated with Eqs. (2) and (3). For air-water gaseous exchange, the flux (F_g) can be estimated with the following equation:

$$F_g = k_g(C_g - C_w H')A \quad \textbf{(6)}$$

where k_g is the overall gas-phase mass transfer coefficient (m/s), C_w and C_g are the BDE-209 concentrations in the dissolved phase of water and in the gas phase of air (kg/m^3), and H' is the unitless Henry's Law constant (Venier and Hites, 2008). Because no measured data are available, the gas-phase concentration of BDE-209 in the atmosphere of the PRE is assumed to be the same as the average concentration (which was below the detection limit and taken as zero in the present study) of BDE-209 obtained in samples collected from the atmosphere of Guangzhou (Chen et al., 2006). Guan et al. (2009a) determined a value of -0.13 kg/yr for air-water exchange flux of BDE-209 in the PRE with the negative sign indicating vaporization was the dominant transport mode. In addition, the annual combined dry and wet depositional input amounted to 235 kg/yr (Guan et al., 2009a).

6.2. Sedimentation and Degradation

The sedimentation rate of BDE-209 in the PRE (with an area of 2016 km^2) was estimated from a previous study investigating the time trend of BDE-209 in the sediment column of the PRE (Chen et al., 2007). The result indicated that nearly 600 kg/yr of BDE-209 settled into the sediment of the PRE annually in recent years (Chen et al., 2007). Furthermore, a mass attenuation model was used to estimate the degradation rate of BDE-209 in the PRE:

$$M_d = M_o[1 - (1/2)^{t/T}] \quad \textbf{(7)}$$

where M_d is the amount of the degraded BDE-209, M_o is the initial mass, t is the time duration for degradation and T is the degradation half-time of BDE-209. With the hydraulic turnover time (~6 days) regarded as the degradation time (Wong and Cheung, 2008) and the riverine input (1350 kg/yr) of BDE-209 from the four eastern riverine runoff outlets (Figure 1) as the initial mass (Guan et al., 2007), the annual degradation rate of BDE-209 was estimated at 45 kg/yr (Guan et al., 2009a).

With the above considerations, a mass balance diagram can be constructed for BDE-209 in the PRE (Figure 2). It is clear that the majority (70%) of BDE-209 discharged from the PRD into the PRE eventually flows to the adjacent ocean, which indicates that riverine runoff from the PRD significantly contributes to the mass loadings of BDE-209 (and other organic pollutants) to the global oceans.

7. Mass Transport Budget: Implications and Perspectives

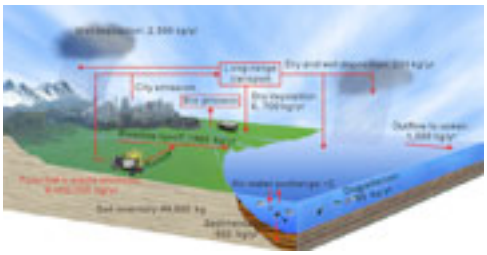

Figure 3
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After all the above processes are taken into account, a mass transport budget can be constructed (Figure 3). It should be noted that the riverine runoff input in the budget is 610 kg/yr higher than that entering the PRE (Figure 2); this additional amount was discharged from the four western outlets (Modaomen, Jitimen, Hutiaomen and Yamen) and directly to the coastal ocean (Figure 1). Figure 3 shows that dry deposition is an important mode to recycle BDE-209 from the atmosphere to the ground, and soil remains the largest sink. On the other hand, fluxes associated with air-soil and air-water gaseous exchange within the PRD should be noticeably less significant compared to dry and wet depositional fluxes as indicted by the small amount of air-water exchange relative to that of dry/wet deposition in the PRE (Figure 2). Terrestrial sediment inventories (e.g., those in rivers, lakes, reservoir and ponds etc.) may be important because of the possibility of receiving point-source inputs from manufacturing plants or e-waste recycling centers, but their impacts on the overall geochemical cycling are largely accounted for by riverine runoff or air-water exchange. The amounts of BDE-209 flowing in and out of the PRD via long-range transport are also unknown; however, the net effect may be small and largely reflected in dry and wet atmospheric depositions. Biological processes that impact BDE-209 cycling are also unknown but are expected to be negligible because the amount of biota (or equivalently biomass) available for accumulation of BDE-209 is expected to be substantially smaller than those of soil/sediment and water. Finally, it should be emphasized that the amount of BDE-209 for a specific inter-compartmental flux or within an environmental compartment in the PRD is no more than a drop in the bucket compared to the potential emission of BDE-209 from e-waste (Figure 3). Apparently, vast amounts of BDE-209 remain in e-waste and commercial products embedded with BFRs, that can explode in the future if no appropriate mitigation measures are immediately installed.

Although this study has examined the geochemical process of BDE-209 only, useful inferences can be derived for other organic pollutants. For example, PAHs are similar to BDE-209 in that both are still being released from current sources and considerably persistent in the environment. It can be deduced that the amount of annual atmospheric PAH depositions would be substantial compared to the current soil inventory as in the case of BDE-209 (9.2 tons/yr for dry and wet depositions versus 44 tons for soil inventory; Figure 3). Based on a field measurement of atmospheric depositions of PAHs (12 components) in an urban lake in Guangzhou (Li et al., 2009), we obtained the annual dry and wet depositional rate of PAHs at 106 tons/yr for the entire PRD. In addition, the soil inventory of the same PAHs in the PRD was estimated at 4,600 tons by extrapolating the results from the study of Ma et al. (2009). On the other hand, the amount of annual atmospheric deposition of DDTs is expected to be fairly small relative to its soil inventory because DDTs in the environment are mostly residues from history uses. In fact, our preliminary assessment indicates that the soil DDT inventory in the PRD is three orders of magnitude higher than the annual dry and wet atmospheric deposition of DDTs (unpublished data). Nevertheless, more refined studies are needed to verify this hypothesis, and more importantly to gain better understanding of the governing mechanisms for the geochemical processes of organic pollutants in general.

Acknowledgements

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Appendix – Figure 1

Figure 1. General locality of the study region in the Pearl River Delta (PRD) and Pearl River Estuary (PRE) within Guangdong Province of South China. Upper left-hand quadrant: Locality of Guangdong province. Upper right-hand quadrant: Locality of the PRD within Guangdong Province; the smaller box on the right indicates the location of Guiyu, one of the largest e-waste recycling centers in the world (also enlarged directly below). Lower quadrant: Detailed map showing the various sampling sites within the PRD with the numbers designating the eight major riverine runoff outlets: (1) - Humen; (2) - Jiaomen; (3) - Hongqili; (4) - Hengmen; (5) - Modaomen; (6) - Jitimen; (7) - Hutiaomen; and (8) - Yamen.

In the upper right hand quadrant - One box has PRD in it and is blown up on the left below. Guiyu's location is shown in the smaller box on the right.

Appendix – Figure 2

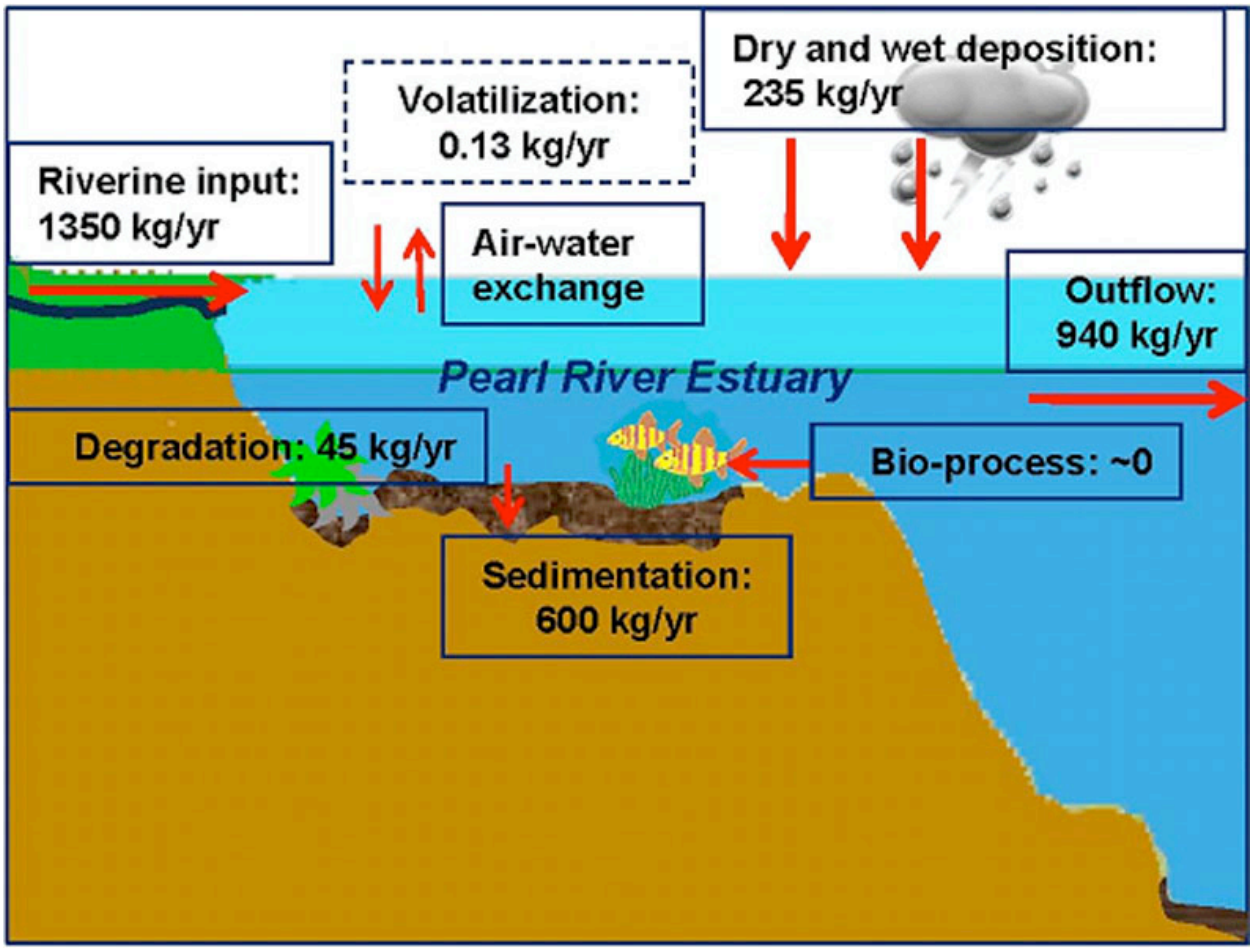

Figure 2. Schematic showing the geochemical process of BDE-209 in the Pearl River Estuary (Figure 1).

Appendix – Figure 3

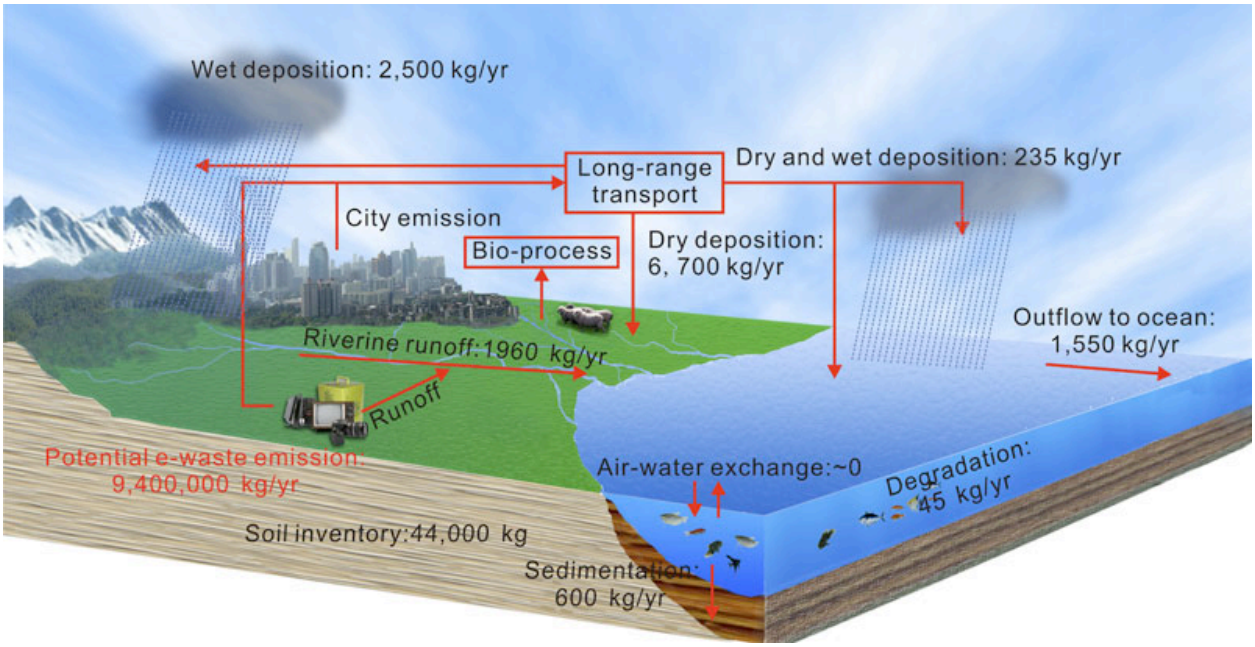

Figure 3. Mass transport budget of BDE-209 within the Pearl River Delta, South China (Figure 1).

About the Author

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