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17

18 **Abstract:**

19 **Spatial patterns and temporal trend of nitrogen (N) and phosphorus (P) deposition are important for**  
20 **quantifying their impact on forest carbon (C) uptake. In a first step, we modeled historical and**  
21 **change in the global distributions of the atmospheric deposition of N and P from the dry and wet**  
22 **deposition of aerosols and gases containing N and P. Future projections were compared between two**  
23 **scenarios with contrasting aerosol emissions. Modeled fields of N and P deposition and P**  
24 **concentration were evaluated using globally distributed *in situ* measurements. N deposition peaked**  
25 **around 1990 in European forests and around 2010 in East Asian forests, and both increased 7-fold**  
26 **relative to 1850. P deposition peaked around 2010 in South Asian forests and increased 3.5-fold**  
27 **relative to 1850. In a second step, we estimated the change in C storage in forests due to the**  
28 **fertilization by deposited N and P ( $\Delta C_{v\ dep}$ ), based on the retention of deposited nutrients, their**  
29 **allocation within plants, and C:N and C:P stoichiometry.  $\Delta C_{v\ dep}$  for 1997-2013 was estimated to be**  
30  **$0.27 \pm 0.13$  Pg C yr<sup>-1</sup> from N and  $0.054 \pm 0.10$  Pg C yr<sup>-1</sup> from P, contributing 9% and 2% of the**  
31 **terrestrial C sink, respectively. Sensitivity tests show that uncertainty of  $\Delta C_{v\ dep}$  was larger from P**  
32 **than from N, mainly due to uncertainty in the fraction of deposited P that is fixed by soil.  $\Delta C_{P\ dep}$  was**  
33 **exceeded by  $\Delta C_{N\ dep}$  over 1960-2007 in a large area of East Asian and West European forests due to a**  
34 **faster growth in N deposition than P. Our results suggest a significant contribution of anthropogenic**  
35 **P deposition to C storage, and additional sources of N are needed to support C storage by P in some**  
36 **Asian tropical forests where the deposition rate increased even faster for P than for N.**

37

## 38 1. Introduction

39 The processes controlling the terrestrial carbon (C) sink are a major source of uncertainty in projections of  
40 the historical and future evolution of atmospheric CO<sub>2</sub> (Matthews, 2007; Ahlstrom *et al.*, 2012).  
41 Observations and models both suggest that terrestrial C sinks are limited by nitrogen (N) and phosphorus (P)  
42 with notable regional differences (Hungate *et al.*, 2004; Reich *et al.*, 2006; Elser *et al.*, 2007; Thornton *et*  
43 *al.*, 2009; Norby *et al.*, 2010; Vitousek, *et al.*, 2010; Goll *et al.*, 2012, Penuelas *et al.*, 2013,  
44 Fernández-Martínez *et al.*, 2014; Wieder *et al.*, 2015). The majority of studies predict a large limitation of  
45 the C sink in the 21<sup>st</sup> century due to limited availabilities of N and P, but the extent of this limitation varies  
46 widely among studies. This effect of limitation implies that terrestrial C sinks are sensitive to the inputs of  
47 N and P from atmospheric deposition in the case of non-cultivated ecosystems. In particular, anthropogenic  
48 emissions of reactive N, including oxidized (*e.g.* NO<sub>x</sub>) and reduced (*e.g.* NH<sub>3</sub>) N have increased  
49 significantly in the past decades (Galloway *et al.*, 2004), causing notable change in N availability in  
50 northern temperate and boreal forests (Magnani *et al.*, 2007) and more recently in tropical forests (Hietz *et*  
51 *al.*, 2011). During 1982-2009, N deposition has contributed 9% to the observed greening of the Earth (Zhu  
52 *et al.*, 2016).

53 Atmospheric deposition increases the availability of N and P, and thus should increase or sustain the  
54 terrestrial C sink (Graham and Duce, 1979; Galloway *et al.*, 2004; Okin *et al.*, 2004; Mahowald, 2011).  
55 Nadelhoffer *et al.* (1999) estimated that N deposition accounts for a C sink of 0.25 Pg C yr<sup>-1</sup> in 1990s based  
56 on <sup>15</sup>N tracer studies in nine forests and a prescribed N input of 5.1 Tg N yr<sup>-1</sup> to the Earth's forests. For the  
57 same period, Liu and Greaver (2009) estimated a somewhat larger contribution of N deposition to the  
58 global terrestrial C sink of 0.35-0.58 Pg C yr<sup>-1</sup> based on a meta-analysis of field observations. Thomas *et al.*  
59 (2010) attributed a C sink of 0.31 Pg C yr<sup>-1</sup> to this factor in forests by comparing forest growth rates for  
60 different exposures of N deposition during the 1980s and 1990s based on data from the national forest  
61 inventory in the USA. Zaehle *et al.* (2010, 2014) estimated that N deposition resulted in a net terrestrial C  
62 sink of 0.2 Pg C yr<sup>-1</sup> during 1996-2005 using an OCN process-based vegetation model. de Vries *et al.*  
63 (2014) estimated a contribution of comparable magnitude (0.2-0.5 Pg C yr<sup>-1</sup>) from N deposition to the  
64 global land C sink using a stoichiometric approach. While the impact of atmospheric N deposition on  
65 terrestrial C sink has been assessed by several studies, the contribution of P deposition has not yet been  
66 quantified. This is because reconstruction of historical changes in P deposition are lacking.

67 Several studies have attempted to simulate the spatial distributions of N deposition (Holland *et al.*, 1997;  
68 Lamarque *et al.*, 2005, 2013; Dentener *et al.*, 2006; Phoenix *et al.*, 2006; Paulot *et al.*, 2013; Hauglustaine  
69 *et al.*, 2014) and P deposition (Mahowald *et al.*, 2008) under current and future conditions driven by  
70 emission inventories. These studies differ by their emission inventories, aerosol chemistry and horizontal  
71 resolutions of chemical transport models (CTM) (Table S1). However, the modeled N and P deposition  
72 rates are subject to high uncertainty, which remains unquantified. For example, the modeled wet deposition  
73 rates of nitrate (NO<sub>3</sub>) and ammonium (NH<sub>4</sub>) as means of 11 CTMs were underestimated by 40-140%  
74 compared to atmospheric station data depending on the region (Lamarque *et al.*, 2013). Hauglustaine *et al.*  
75 (2014) suggested that the underestimation in wet N deposition for Asia in their CTM is of 50-60% due to a

76 bias in the region's N emissions estimates and the model's coarse horizontal resolution. Earlier estimates of  
77 present day total P deposition (Mahowald *et al.*, 2008) were underestimated by almost one order of  
78 magnitude, likely due to an underestimation of the contribution of human activities to P emissions (Wang *et al.*,  
79 2015a). Recent measurements in China underline the anthropogenic component of P deposition  
80 showing that the bulk deposition of P followed a power-law increase with decreasing distance of  
81 monitoring sites to the nearest cities (Du *et al.*, 2016). More measurements over a wide range of  
82 representative stations are necessary to confirm the regional contribution of combustion sources to P in the  
83 atmosphere. High-resolution data sets of modeled N and P deposition over forests supported by global  
84 measurements are critical for understanding the C sink that offsets the increase in fossil-fuel C emissions  
85 and the limitations of primary productivity by N and P. However, such data sets are not available due to  
86 uncertainties in the emission inventories of N and P and limited understanding of N chemistry in aerosols.

87 Elemental stoichiometry has been used to identify major constraints in the change in C storage by terrestrial  
88 ecosystems due to nutrients either available in soils or deposited from the atmosphere, based on  
89 assumptions about the allocation of nutrients and their elemental ratios (e.g., C:N or C:P) in ecosystemic C  
90 pools. This method assumes that N and P limitation of net primary productivity (NPP) is widespread across  
91 global biomes (Elser *et al.*, 2007). For example, Cleveland *et al.* (2013) used this method to show that  
92 external inputs of N and P through atmospheric deposition supports 3.8% and 16% of new NPP, but the  
93 uncertainty has not been considered. De Vries *et al.* (2014) used this method to infer that N deposition  
94 supports a global forest C sink of 0.28-0.45 Pg C yr<sup>-1</sup> by assuming that 15% of deposited N is retained in  
95 forest biomass and 15% in soil in tropical forests, but the impact of P was not considered in this study.

96 Our study aims to fill these gaps by providing the first time-series of N & P deposition and the resulting C  
97 sink taking into account the major sources of uncertainty. To do so, we calculated the temporal evolutions  
98 of N and P deposition in a CTM prescribed by reconstructions of historical and future scenarios of  
99 anthropogenic emissions and evaluated the so derived N wet deposition, P total deposition and surface P  
100 concentrations in aerosols with in situ measurements. We built a modeling framework to estimate the C  
101 sequestration of global forests due to historical and future N and P deposition. We accounted for  
102 uncertainties in the stoichiometric and allocation parameters by employing a Monte Carlo method in a  
103 stoichiometric mass-balance approach, and identify key factors influencing the forest C sequestration.

## 104 **2. Materials and methods**

### 105 **2.1. Atmospheric deposition of N and P for 1850-2100**

106 The global aerosol chemistry climate model LMDZ-INCA couples the LMDz (Laboratoire de  
107 Météorologie Dynamique, version-4) General Circulation Model (Hourdin *et al.*, 2006) and the INCA  
108 (INteraction with Chemistry and Aerosols, version-4) aerosol module (Hauglustaine *et al.*, 2014). A full  
109 description of the model is provided in the **Supporting Information**. To run the model, emissions data  
110 included sea-salt and dust for P, primary biogenic aerosol particles for P, oceanic emissions for N (NH<sub>3</sub>),  
111 vegetation emissions for N (NO), agricultural activities (including fertilizer use and livestock) for N, and  
112 fuel combustion for both N (NO<sub>y</sub> and NH<sub>x</sub>) and P. Regarding N-containing aerosols and gases,

113 LMDZ-INCA was run with a fully interactive atmospheric N cycle (Hauglustaine *et al.*, 2014) at a  
114 horizontal resolution of 1.27° latitude by 2.5° longitude with 39 vertical layers in the atmosphere to  
115 simulate the global dry and wet deposition of NO<sub>y</sub> and NH<sub>x</sub> for 1850, 1960, 1970, 1980, 1990, 1997-2013,  
116 2030, 2050 and 2100. The same resolution is used for the transport and deposition of P aerosols in  
117 LMDZ-INCA with the emission data provided by Wang *et al.* (2015a), which are transported in three size  
118 bins for P emitted by combustion processes with a diagnostic mass median diameter (MMD) of 0.14, 2.5  
119 and 10.0 μm, one size bin for P from primary biogenic aerosol particles (MMD=5.0 μm), one size bin for P  
120 from mineral dust (MMD=2.5 μm), and three size bins for P from marine sea-salt particles (Balkanski *et al.*,  
121 2011). Meteorological fields from a reanalysis of the European Centre for Medium-Range Weather  
122 Forecasts (ECMWF) have been used in the present configuration to nudge the model transport and removal  
123 processes for 1980 and 1990 and for each year during the recent 1997-2013 period. Additional simulations  
124 were performed with emissions for 1850, 1960, 1970 and into the future all using meteorological fields for  
125 2005.

126 The emissions prescribed to LMDZ-INCA were obtained from published data sets or emission inventories.  
127 We focused on generating coherent emission data sets for all species, and the methods are fully described  
128 in the **Supporting Information. Table S2** lists the emission data sets used in our study. In brief, global  
129 0.5°×0.5° emissions of NO<sub>x</sub>, NH<sub>3</sub>, sulfur dioxide (SO<sub>2</sub>), non-methane volatile organic compounds  
130 (NMVOCs), methane (CH<sub>4</sub>), carbon monoxide (CO), organic carbon (OC) and black carbon (BC) for 2005,  
131 2010 and 2030 were obtained from the ECLIPSE.GAINS.4a model (Klimont *et al.*, 2013). SO<sub>2</sub>, CO, CH<sub>4</sub>  
132 and NMVOCs species should be treated consistently with N aerosols, because they influence N chemistry  
133 in the model (Hauglustaine *et al.*, 2014). The ECLIPSE.GAINS.4a emissions of all species were extended  
134 to other years of simulations using historical data from the ACCMIP and MACCity inventories (Lamarque  
135 *et al.*, 2010; Granier *et al.*, 2011) and future data from the RCP4.5 and RCP8.5 scenarios by Lamarque *et al.*  
136 (2011). In addition, natural emissions of NO and NH<sub>3</sub> from soil (Bouwman *et al.*, 1997; Lathière *et al.*,  
137 2006) were assumed to be constant throughout the period; natural oceanic emissions of NH<sub>3</sub> were  
138 calculated in the INCA model following the formulations proposed by Paulot *et al.* (2015), with monthly  
139 2°×2° fields of surface sea-water concentrations of NH<sub>4</sub>, pH and salinity as simulated by the oceanic  
140 biogeochemical model PISCES (Wang *et al.*, 2015b). Emissions of P from fossil fuels and biofuel were  
141 estimated annually for 1960 to 2007 (Wang *et al.*, 2015a), which were extended to other years of  
142 simulations. Natural emissions of P from mineral dust, primary biogenic aerosol particles, sea salt and  
143 volcanoes were assumed to be constant (Wang *et al.*, 2015a). All N and P emissions from fossil fuels,  
144 biofuel and agricultural activities were assumed to be constant throughout each year without seasonal  
145 variation. Monthly 0.5°×0.5° gridded emissions by natural or anthropogenic (deforestation) burning of  
146 biomass were generated for 1997-2013 from the GFED4.1 inventory (Giglio *et al.*, 2013) and for other  
147 years from the ACCMIP inventory (Lamarque *et al.*, 2010). Emissions from biomass burning for 2030,  
148 2050 and 2100 were assumed to be the same as the averages for 2010-2013. We only analyzed the changes  
149 of future N and P deposition from anthropogenic sources due to the lack of estimates of natural emissions,  
150 but both natural and anthropogenic sources were included for evaluating modeled N and P deposition

151 against measurements.

## 152 **2.2. Observed N and P deposition rates and P concentrations**

153 To evaluate the modeled N and P deposition rates, we used three observational data sets (**Figure S1**),  
154 including (i) a recent global data set of wet N deposition rates measured during 2002-2006 ([Vet et al.,](#)  
155 [2014](#)), (ii) a recent global data set of total P deposition rates measured during 1960-2010 ([Tippling et al.,](#)  
156 [2014](#)), and (iii) a recent global data set of surface P deposition concentrations measured during 1960-2008  
157 ([Mahowald et al., 2008](#)). Dry N deposition was not evaluated due to the lack of data, similar to previous  
158 studies ([Larmarque et al., 2005](#); [Dentener et al., 2006](#)). Before using these observational data for model  
159 evaluation, we have further collected data to increase coverage of wet N deposition data in South America  
160 and Africa, removed some data of total P deposition with a high potential for contamination, and divided  
161 the measured surface P concentrations into short-term and long-term measurements. A full description is  
162 provided in the **Supporting Information**.

## 163 **2.3. Additional C fixation attributable to N and P deposition**

### 164 **2.3.1. A stoichiometric method**

165 A stoichiometric mass balance approach was used to estimate the change in C storage attributable to N and  
166 P deposition over global forests, based on the fraction of deposited nutrients retained in the ecosystems and  
167 incorporated into biomass and soil carbon pools ([Nadelhoffer et al., 1999](#); [Cleveland et al., 2013](#); [de Vries](#)  
168 [et al., 2014](#); [Wieder et al., 2015](#)). We distinguished between four biomass C pools (leaves, stems, fine roots  
169 and coarse roots; each with different stoichiometry) and one soil C pool. Five types of forests are  
170 considered, namely deciduous broadleaf, deciduous needle-leaf, evergreen broadleaf, evergreen needle-leaf  
171 and mixed forests. The global land cover data set at a spatial resolution of 1 km ([Hansen et al., 2000](#)) for  
172 the year 2010, as a product of Moderate Resolution Imaging Spectroradiometer (MODIS), was used to map  
173 these five types of forests. Forest areas have changed significantly since 1850 ([Foley et al., 2005](#); [Houghton](#)  
174 [et al., 2003](#)). Here, we applied the fixed forest cover map by Hansen *et al.* (2000) in the calculation of  $\Delta C_{v, \text{dep}}$   
175 throughout the period of 1850-2100. The uncertainty induced by assuming a fixed forest cover map on  
176  $\Delta C_{v, \text{dep}}$  is quantified in **Section 3.7**. To do this, we re-calculated  $\Delta C_{v, \text{dep}}$  with the variable forest cover map  
177 for 1850-2010 according to the reconstruction of ([Peng et al., 2017](#)) and the fraction of five types of forests  
178 within each  $1^\circ \times 1^\circ$  grid from Hansen *et al.* (2000).

179 We assumed that in 1850 the forest C stocks were in equilibrium with the pre-industrial N and P deposition  
180 levels (i.e.  $\Delta C_{v, \text{dep}} = 0$ ), so that only the change in N and P deposition relative to the background levels can  
181 cause an extra forest C storage, allowing our estimates to cover the additional N and P emissions from  
182 fossil fuels and biofuel burning, atmospheric  $\text{NH}_3$  and  $\text{NO}_x$  emissions from agriculture, and change in N  
183 and P emissions from biomass burning compared to 1850. Hence our analysis included the wind export of  
184 P from croplands and savanna to forests through fire emissions and atmospheric transport. It should be  
185 noted that direct anthropogenic effects on fires (*e.g.*, fire suppression and land use change) and indirect  
186 anthropogenic effects (*e.g.*, changes in climate affecting fire regimes) were not explicitly distinguished in  
187 the historical fire emission inventory we used ([Giglio et al., 2013](#)). In addition, although our calculation of



188  $\Delta C_{v\text{ dep}}$  covers C storage due to N and P from anthropogenic fires, we did not try to estimate the C  
 189 emissions from these anthropogenic fires which are always termed as part of C emissions from land-use  
 190 change in global C budget (Le Quere *et al.*, 2016). However, we did not account for the change of  
 191 atmospheric CO<sub>2</sub> concentrations, climate and forest cover change (Hansen *et al.*, 2013).

192 In each pixel at a spatial resolution of 1°×1°, additional forest C storages supported by anthropogenic N or  
 193 P deposition ( $\Delta C_{v\text{ dep}}$ , where v stands for either N or P) were expressed as:

194

$$\Delta C_{N\text{ dep}_i}(t) = f_{N_{veg}} a_{N_i} s_{N_i} [d_N(t) - d_N(1850)] \quad (1)$$

195

$$\Delta C_{P\text{ dep}_i}(t) = f_{P_{veg}} a_{P_i} s_{P_i} [d_P(t) - d_P(1850)] \quad (2)$$

196 where  $i$  is biomass pool ( $i = 1$  for leaves, 2 for stems, 3 for fine roots and 4 for coarse roots),  $f_{N_{veg}}$  and  $f_{P_{veg}}$   
 197 are the retention fractions of deposited N and P in biomass pools, respectively,  $a_{N_i}$  and  $a_{P_i}$  are the allocation  
 198 fractions of N and P in each pool, respectively,  $s_{N_i}$  and  $s_{P_i}$  are the C:N and C:P stoichiometric ratios,  
 199 respectively, and  $d_N(t)$  and  $d_P(t)$  are the deposition rates of N and P for year  $t$ , respectively. All deposition  
 200 fields and land cover fractions were re-interpolated to the 1°×1° grid for the calculation of  $\Delta C_{v\text{ dep}}$ .

201 Similarly, in each pixel, the  $\Delta C_{v\text{ dep}}$  realized in soil were expressed as:

202

$$\Delta C_{N\text{ dep}_{soil}}(t) = f_{N_{soil}} s_{N_{soil}} [d_N(t) - d_N(1850)] \quad (3)$$

203

$$\Delta C_{P\text{ dep}_{soil}}(t) = f_{P_{soil}} s_{P_{soil}} [d_P(t) - d_P(1850)] \quad (4)$$

204 where  $f_{N_{soil}}$  and  $f_{P_{soil}}$  are the retention fractions of N and P in soil, respectively, and  $s_{N_{soil}}$  and  $s_{P_{soil}}$  are the C:N  
 205 and C:P stoichiometric ratios in soil biota, respectively.

206 There are two limitations in our approach. First, like de Vries *et al.* (2014), we approximate a time scale of  
 207 10-20 years for the  $\Delta C_{v\text{ dep}}$ , because ecosystems take time to sequester C in forest biomass and soil after an  
 208 initial disturbance (Goulden *et al.*, 2011), such as the level of N and P deposition for most of the global  
 209 forests. It should be noted that our stoichiometric mass-balance model cannot resolve processes governing  
 210 the C storage changes attributed to deposition on shorter timescales. It is likely that the instantaneous effect  
 211 at the early stage of forest succession (e.g., less than 5 years) is lower, due to enhancement of soil C  
 212 decomposition by N deposition (Goulden *et al.*, 2011). Second, different turnover times of the plant and  
 213 soil pools should influence the C response to deposition, which is not included in our model. According to  
 214 de Vries *et al.* (2014), the C storage in the woody biomass (stem and coarse root) determines C  
 215 sequestration by forest trees, while the C storage in the non-woody biomass (leaves and fine root), which is  
 216 fast-turnover (Iversen *et al.*, 2017), determines C sequestration in soil. We estimated the sum of them based  
 217 on the allocation of the deposited nutrient in different pools measured at a time scale of around 2-9 years  
 218 (Schlesinger, 2009). At longer time periods, the C sequestration is determined by other disturbances, such  
 219 as forest fires and forest harvesting, which is not considered in our study. More knowledge gained in  
 220 database analyses for nutrient use in the tree growth (e.g., Sardans and Peñuelas, 2013, 2015) would enable  
 221 us to better understand these processes in future process-based ecosystem models.

222 Although  $\Delta C_{v\text{dep}}$  account for C storage supported by deposited nutrients on timescales of 10-20 years (de  
 223 Vries *et al.*, 2014), it should be noted that a fraction of deposited N and P may still contribute to  $\Delta C_{v\text{dep}}$   
 224 beyond the 10-20 year timeframe. This impact is small for  $\Delta C_{N\text{dep}}$ , because a large fraction of N is lost by  
 225 denitrification or leaching. The impact for  $\Delta C_{P\text{dep}}$ , however, is not negligible, because P is mostly fixed by  
 226 the soil and is less prone to loss. We estimated  $\Delta C_{P\text{dep}}$  by fixing an “effective fraction” of the deposited P ( $f_{\text{fix}}$ )  
 227 to account for this impact (see **Section 2.3.2**).

228 All parameters were determined with central values and uncertainty ranges (see **Section 2.3.2, 2.3.3, and**  
 229 **2.3.4**). In brief, the parameters  $f_{N\text{veg}}$ ,  $f_{P\text{veg}}$ ,  $a_i$ ,  $s_{Ni}$ ,  $s_{Pi}$ ,  $f_{N\text{soil}}$  and  $f_{P\text{soil}}$  were derived for deciduous broadleaf,  
 230 deciduous needle-leaf, evergreen broadleaf, evergreen needle-leaf and mixed forests; parameters values and  
 231 their uncertainty ranges are summarized in **Table 1**. Uncertainties in the modeled deposition rates ( $d_N$  and  
 232  $d_P$ ) were derived from a comparison with the available observation data sets (see **Section 3.2**).

233 **Table 1.** Parameters used to estimate carbon fixation due to anthropogenic N and P deposition in forests.  
 234 The 95% confidential intervals adopted as the lower and upper estimates for each parameter applied in our  
 235 Monte Carlo simulations are in parentheses.

Forest type	Evergreen needleleaf forest	Evergreen broadleaf forest	Deciduous needleleaf forest	Deciduous broadleaf forest	Mixed forest
<b>C:N (g C: g N)</b>					
Leaves	42 (34-50)	21 (17-25)	50 (40-60)	21 (17-25)	28 (22-34)
Stems	250 (200-300)	150 (120-180)	250 (200-300)	175 (140-210)	175 (140-210)
Fine roots	78 (62-94)	78 (62-94)	41 (33-49)	41 (33-49)	41 (33-49)
Coarse roots	250 (200-300)	150 (120-180)	250 (200-300)	175 (140-210)	175 (140-210)
Soil	31 (28-35)	16 (14-18)	20 (18-21)	19 (18-20)	19 (18-20)
<b>C:P (g C: g P)</b>					
Leaves	408 (326-490)	400 (320-480)	405 (324-486)	333 (266-400)	278 (222-334)
Stems	3750 (3000-4500)	2250 (1800-2700)	3750 (3000-4500)	2625 (2100-3150)	2625 (-)
Fine roots	1170 (936-1404)	1020 (816-1224)	615 (492-738)	615 (492-738)	615 (492-738)
Coarse roots	3750 (3000-4500)	2250 (1800-2700)	3750 (3000-4500)	2625 (2100-3150)	2625 (2100-3150)
Soil	1030 (459-2312)	169 (134-214)	318 (214-472)	391 (306-500)	254 (214-300)
<b>Retention fractions of N and P in plants</b>					
$f_{N\text{veg}}$	0.23 (0.14-0.30)	0.15 (0.10-0.20)	0.23 (0.14-0.30)	0.23 (0.14-0.30)	0.23 (0.14-0.30)
$f_{P\text{veg}}$	0.05 (0.016-0.22)	0.09 (0.027-0.36)	0.05 (0.016-0.22)	0.05 (0.016-0.22)	0.05 (0.016-0.22)
<b>Retention fractions of N and P in soil</b>					
$f_{N\text{soil}}$	0.52 (0.26-0.78)	0.15 (0.10-0.20)	0.52 (0.26-0.78)	0.52 (0.26-0.78)	0.52 (0.26-0.78)
$f_{P\text{soil}}$	0.13 (0.038-0.50)	0.09 (0.03-0.36)	0.13 (0.038-0.50)	0.13 (0.038-0.50)	0.13 (0.038-0.50)
<b>Allocation fraction of N (<math>a_{Ni}</math>)</b>					
Leaves	0.42	0.56	0.42	0.49	0.46
Stems	0.34	0.22	0.34	0.33	0.34
Fine roots	0.16	0.17	0.16	0.11	0.13

Coarse roots	0.08	0.05	0.08	0.07	0.07
Allocation fraction of P ( $a_{Pi}$ )					
Leaves	0.32	0.63	0.28	0.50	0.35
Stems	0.40	0.19	0.42	0.32	0.40
Fine roots	0.19	0.13	0.20	0.11	0.16
Coarse roots	0.09	0.04	0.10	0.07	0.09

236

237 Sensitivity tests were performed to quantify the influence of the major parameters on the  $\Delta C_{v\text{dep}}$  by N and  
 238 P (see **Section 3.4**). Finally, Monte Carlo simulations were applied to estimate the central values of  $\Delta C_{v\text{dep}}$   
 239 in global forests and to estimate their uncertainties in each pixel at a spatial resolution of  $1^\circ \times 1^\circ$ . In brief,  
 240 the model was run 10000 times by drawing input parameters from uniform or normal uncertainty  
 241 distributions of model parameter and N- and P-deposition rates. The medians and 95% confidence intervals  
 242 (CI) were used to represent the central estimate and the associated uncertainty, respectively, based on the  
 243 Monte Carlo simulations.

#### 244 **2.3.2. Retention of N and P in the ecosystems**

245 Schlesinger (2009) reported a median N retention fraction of 23% (with an interquartile range of 14-30%)  
 246 for biomass pools ( $f_{N\text{veg}}$ ) and 52% (with an interquartile range of 26-78%) for soils ( $f_{N\text{soil}}$ ) in boreal and  
 247 temperate forests. De Vries *et al.* (2007) suggested a lower N-retention fraction of 15% (with a 90%  
 248 uncertainty range of 10-20%) for both  $f_{N\text{veg}}$  and  $f_{N\text{soil}}$  in tropical forests. We adopted these median estimates  
 249 and their uncertainty ranges. The non-retained fraction of deposited N lost by leaching, volatilization and  
 250 denitrification is not explicitly modeled.

251 Evidence suggests that N inputs from atmospheric deposition can be taken up by forest canopies (Sievering  
 252 *et al.*, 2007; Gaige *et al.*, 2007; Sparks, 2009). De Vries *et al.* (2014), however, suggested that the effect of  
 253 uptake of N by canopies on C fixation is likely to be small, because a small fraction of canopy-retained N is  
 254 absorbed and used in leaves. Gaige *et al.* (2007) suggested that denitrification and nitrification do not occur  
 255 in the canopy, despite a high retention fraction of N deposition in the canopy. Sparks (2009) suggested that  
 256 foliar uptake of reactive N should be considered separately from soil-deposited N, but pointed out that it is  
 257 difficult to link canopy uptake of N directly to assimilation. Dail *et al.* (2009) found that only 3-6% of the  
 258 labeled  $^{15}\text{N}$  recoverable in plant biomass was recovered in live foliage and bole wood and that tree twigs,  
 259 branches and bark were the major sinks (50%) after 2 years of  $\text{NH}_4\text{NO}_3$  addition. We therefore did not  
 260 account for this process in our central case, following the suggestion by de Vries *et al.* (2014), but discuss  
 261 the potential impact in a sensitivity test in **Section 4.2**.

262 The fate of deposited P in ecosystems differs from that of N. First, P is less mobile than N in soils (Aerts  
 263 and Chapin, 2000), so the fractional P loss by leaching is smaller than for N. Second, denitrification has no  
 264 counterpart for gaseous P loss to the atmosphere. Third, the physical fixation of P and its eventual  
 265 occlusion in soil reduces the availability of deposited P to leaching and uptake by plants and microbes. The  
 266 fate of deposited P in forests has unfortunately not yet been measured, to the best of our knowledge, and

267 was estimated in our study based on two assumptions. First, we assumed that 10% of the deposited P was  
 268 directly lost by runoff as Sattari *et al.* (2012) assumed for fertilizer P. Second, by assuming that P  
 269 deposition input to soil is much smaller than the actual stocks of labile and stable P in soil and thus does not  
 270 affect the stock size, we approximated the fixation of an “effective fraction” of deposited P ( $f_{P_{fix}}$ ),  
 271 corresponding to the net transfer of P between ‘labile’ and ‘stable’ forms, based on transfer coefficients  
 272 ( $\mu_{SL}$  and  $\mu_{LS}$ ) between stable and labile P:

$$273 \quad f_{P_{fix}} = 1 - \mu_{SL}/\mu_{LS} \quad (5)$$

274 We used  $\mu_{SL}$  and  $\mu_{LS}$  to derive a central estimate of  $f_{P_{fix}}$  (80%) but a wide range of uncertainty (20% to 94%)  
 275 as recommended by Sattari *et al.* (2012). We also assumed that the fraction of ecosystem-retained P taken  
 276 up by plants ( $f_{P_{uptake}}$ ) was the same as that of N, at 30% for boreal and temperate forests and 50% for tropical  
 277 forests (Schlesinger, 2009; de Vries *et al.*, 2007), due to lack of direct measurements of the fraction of  
 278 ecosystem-retained P taken up by plants. We then derived the retention of P in vegetation ( $f_{P_{veg}}$ ) and soil ( $f_{P_{soil}}$ )  
 279 using  $f_{P_{fix}}$ ,  $f_{P_{loss}}$  and  $f_{P_{uptake}}$ :

$$280 \quad f_{P_{veg}} = (1 - f_{P_{loss}})(1 - f_{P_{fix}})f_{P_{uptake}} \quad (6)$$

$$281 \quad f_{P_{soil}} = (1 - f_{P_{loss}})(1 - f_{P_{fix}})(1 - f_{P_{uptake}}) \quad (7)$$

282 We estimated that  $f_{P_{veg}}$  was 5.4% (1.6-22% as the uncertainty range) and  $f_{P_{soil}}$  was 13% (3.8-50% as the  
 283 uncertainty range) in boreal and temperate forests and that  $f_{P_{veg}}$  was 9.0% (2.7-36% as the uncertainty range)  
 284 and  $f_{P_{soil}}$  was 9.0% (2.7-36% as the uncertainty range) in tropical forests (**Table 1**).

### 285 **2.3.3. Allocation of N and P in plants**

286 We used the values of  $a_{Ni}$  diagnosed by de Vries *et al.* (2014) for deciduous broadleaf, evergreen broadleaf  
 287 and needleleaf forests. The  $a_{Ni}$  averaged over deciduous broadleaf and needleleaf forests was applied for  
 288 mixed forests (**Table 1**). The uncertainty associated with  $a_{Ni}$ , however, was not provided and was estimated  
 289 below. The C:N ratio differs for woody (stem and coarse root) and non-woody (leaf and fine root) pools, so  
 290 the assumption that uncertainties in  $a_{Ni}$  are the same as the uncertainties associated with the fraction of N  
 291 allocated to the woody component is reasonable. A meta-analysis of  $^{15}N$  addition experiments suggested  
 292 that 53% of total N uptake is allocated to woody biomass (Templer *et al.*, 2012), compared to 25% reported  
 293 by Nadelhoffer *et al.* (1999). This latter low value is 7-40% lower than that used in our study, and the high  
 294 value is 26-96% higher than the central value used in our study (**Table 1**). We accordingly applied two half  
 295 normal distributions to cover the uncertainty of  $a_{Ni}$ .

296 We are not aware of any experiment measuring the allocation of deposited P to different biomass pools, so  
 297 we derived the allocation fractions of P and their uncertainties from the allocation of N and the  
 298 stoichiometric ratios of these two elements as:

$$299 \quad a_{P_i} = \frac{a_{N_i} \cdot s_{N_i} / s_{P_i}}{\sum_{i=1}^4 (a_{N_i} \cdot s_{N_i} / s_{P_i})} \quad (8)$$

300 where  $s_{N_i}$  and  $s_{P_i}$  are the C:N and C:P stoichiometric ratios, respectively, in each pool of the plant.

### 301 **2.3.4. C:N and C:P stoichiometric ratios in different pools**

302 C:N and C:P stoichiometric ratios in soil and their 95% CI were derived from a global data set of the  
303 nutrient composition of soil (Xu *et al.*, 2013). The median C:N and C:P stoichiometric ratios of leaves,  
304 stems, fine roots and coarse roots were obtained from Cleveland *et al.* (2013). Wang *et al.* (2010) suggested  
305 that the minimum and maximum of C:N and C:P stoichiometric ratios were  $\pm 20\%$  relative to the means,  
306 which we applied as uncertainties for  $s_{N_i}$  and  $s_{P_i}$ .

#### 307 2.4. Maximum and minimum effects of N and P deposition

308 The change in C fixation due to nutrients addition in an ecosystem can be approximated based on the most  
309 limiting element (Vitousek *et al.*, 1984, 2010), but the stoichiometric mass balance approach we used could  
310 not identify the most limiting element. Therefore, we estimated the maximum and minimum effects of  $\Delta C_{v \text{ dep}}$   
311 due to anthropogenic N and P deposition to characterize the degree of limitation by one of the two  
312 elements. Accordingly, the minimum effects of anthropogenic N and P deposition yields a lower estimate  
313 as:

$$314 \Delta C_{v \text{ dep}}^{\min} = \min[\Delta C_{N \text{ dep}}, \Delta C_{P \text{ dep}}] \quad (9)$$

315 and the maximum effects of N and P deposition yields an upper estimate as:

$$316 \Delta C_{v \text{ dep}}^{\max} = \max[\Delta C_{N \text{ dep}}, \Delta C_{P \text{ dep}}] \quad (10)$$

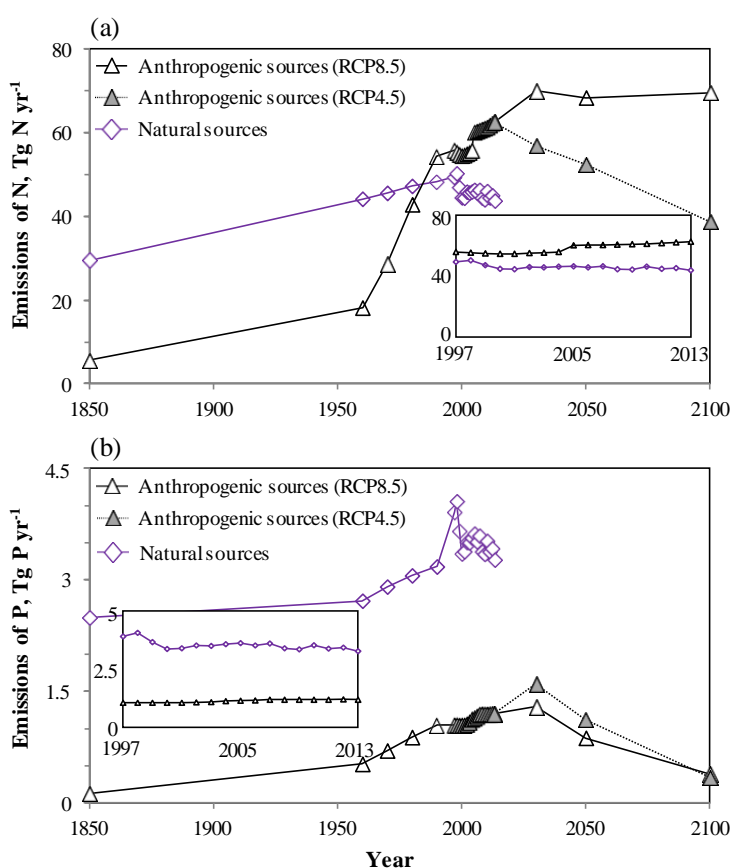
317 The minimum and maximum effects calculated here should be interpreted with caution. First, the minimum  
318 effect is based on a hypothesis that the element with a lower capacity to fix C by deposition is more  
319 limiting and that there is no additional source of this element than atmospheric deposition to support a  
320 larger C storage. The difference between the effect by an element alone and the minimum effect denotes  
321 the limitation by the other element that is enhanced by the deposition. Second, the maximum effect is based  
322 on a hypothesis that the element with a higher capacity to fix C by deposition is more limiting and there is  
323 enough of the other element to allow for the largest C storage. The difference relative to the effect by an  
324 element alone denotes the limitation by the other one that is alleviated by the deposition. Regarding  
325 uncertainties in the limitations by N and P, the combined effect of N and P deposition should fall between  
326 the minimum and maximum effects. Although we cannot rule out that there are synergistic “additive”  
327 effects (Elser *et al.*, 2007) as well as strictly negative effects (Penuelas *et al.*, 2013), such effects are rather  
328 unlikely to occur in wide ranges of ecosystems and deposition loads. In addition, difference between the  
329 maximum and minimum effect quantifies an imbalance between N and P induced by the deposition  
330 providing that other sources of N and P were held constant. However, it should be noted that our  
331 calculation does not account for the interaction between N and P cycles implied by recent studies. For  
332 example, increasing N deposition can affect availability, uptake and using efficiency of P by altering  
333 mycorrhizal activity, phosphatase enzymes and soil properties (Compton and Cole, 1998; Rowe *et al.*, 2008;  
334 Marklein and Houlton, 2012; Lü *et al.*, 2013).

### 335 3. Results

#### 336 3.1. N and P emissions

337 Natural and anthropogenic global emissions of N and P were estimated or derived for the past time slices in

338 1850-1990, for each year in 1997-2013 and for the future time slices in 2030-2100 (Figure 1). Total  
 339 reactive-N emissions increased from 35 Tg N yr<sup>-1</sup> in 1850 to an average of 104 Tg N yr<sup>-1</sup> for 1997-2013,  
 340 and were predicted to reach 83 and 114 Tg N yr<sup>-1</sup> by 2100 under the RCP4.5 and RCP8.5 scenarios,  
 341 respectively. N emissions as the oxidized (NO<sub>x</sub>) and reduced (NH<sub>3</sub>) forms are listed in Table S3, which are  
 342 close to previous estimates (Holand *et al.*, 1997; Galloway *et al.*, 2004; Lamarque *et al.*, 2005; Dentener *et al.*,  
 343 2006; Paulot *et al.*, 2013). The main drivers of the increase in historical N emissions were a rapid rise in  
 344 the use of fossil fuels, both the expansion and intensification of agricultural fertilization and an increase in  
 345 the number of livestock (Schlesinger, 2009). N emissions from fossil fuel, biofuel and agricultural activities  
 346 increased from 5.6 Tg N yr<sup>-1</sup> in 1850 (Lamarque *et al.*, 2010) to 60 Tg N yr<sup>-1</sup> in 2005 (Klimont *et al.*, 2013).  
 347 The increase in reactive-N emissions was equivalent to twice the background N emissions in 1850. The  
 348 emissions of P increased from 2.6 to 4.0 Tg P yr<sup>-1</sup> from 1850 to 2013, equivalent to a 57% increase relative  
 349 to the background 1850 emissions. 47%, 25% and 28% of the increase in P emissions were from increases  
 350 in emissions from fossil fuels, biofuel and deforestation fires, respectively. The N:P ratio in the emissions  
 351 nearly doubled from 25 (on a molar basis) in 1850 to 48 in 2013 due to a faster growth of N than P  
 352 emissions as outlined in a previous study (Peñuelas *et al.*, 2013).



353  
 354 **Figure 1.** (a) Global atmospheric N emissions. Anthropogenic sources include fossil fuels, biofuel and  
 355 agricultural activities. Natural sources include NO<sub>x</sub> and NH<sub>3</sub> emissions from biomass burning, NO<sub>x</sub> and  
 356 NH<sub>3</sub> emissions from soil, and NH<sub>3</sub> emissions from oceans. (b) Global atmospheric P emissions.  
 357 Anthropogenic sources include fossil fuels and biofuel. Natural P sources include biomass burning, dust,  
 358 sea salt, volcano particles and primary biogenic aerosol particles. The inset demonstrates the inter-annual  
 359 recent variation over the period 1997-2013.

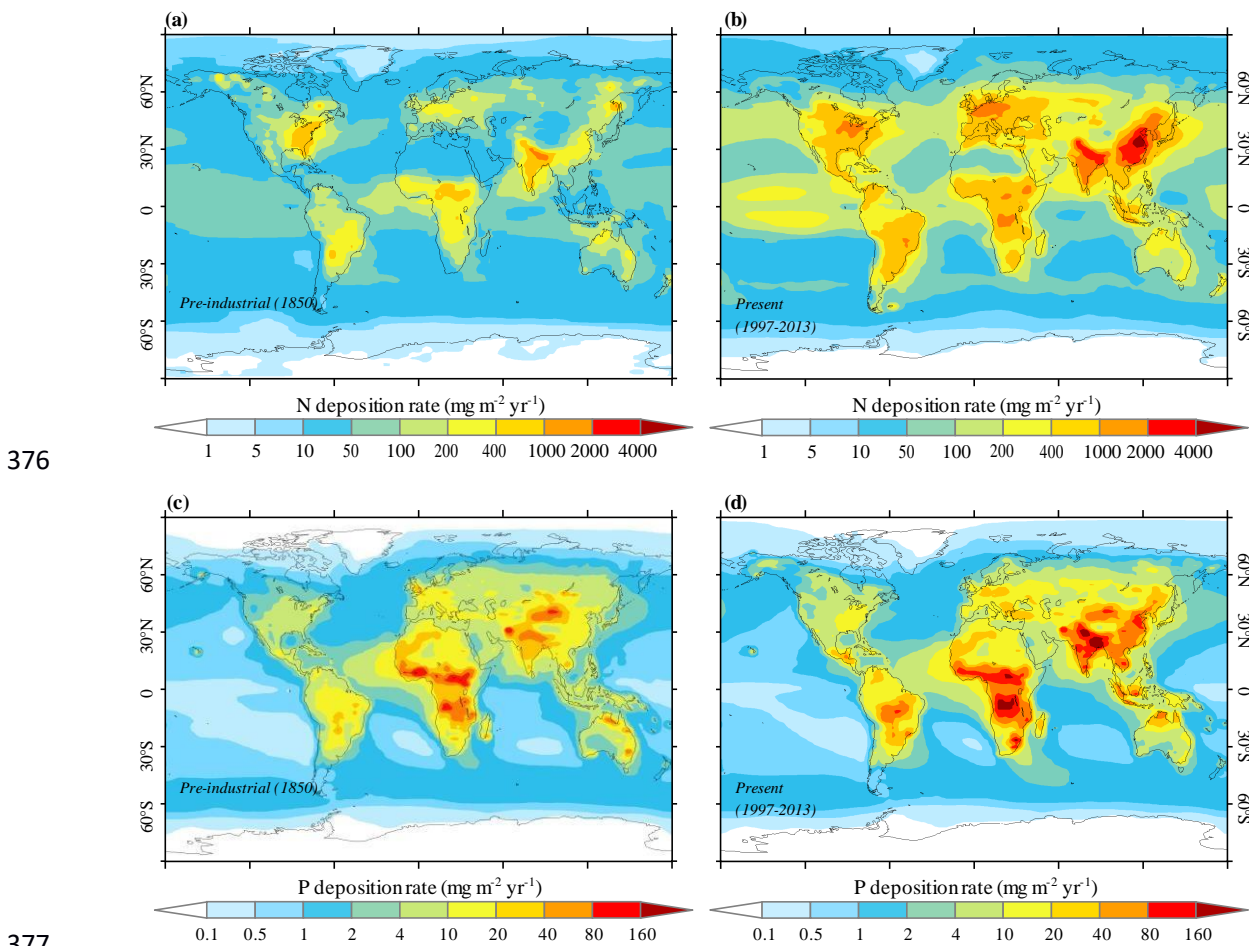


360

361 The future emissions of N differed considerably between the RCP4.5 and RCP8.5 scenarios, but the  
362 emissions of P were similar.  $\text{NH}_3$  emissions in 2100 will be 54% under the RCP8.5 than the RCP4.5  
363 scenario, due to increased agricultural activities needed to meet the food demand of a larger global  
364 population (Lamarque *et al.*, 2011; Riahi *et al.*, 2011). In contrast, 70% of P emissions from fossil fuels and  
365 biofuel were expected to be removed, due to a high penetration rate of clean technology in the industrial  
366 and residential sectors under both the RCP4.5 and RCP8.5 scenarios, leading to a slight difference between  
367 the two scenarios.

### 368 3.2. Spatial distributions of N and P deposition

369 The pre-industrial (1850) and present (1997-2013) spatial distributions of N and P deposition are shown in  
370 **Figure 2**. Dentener *et al.* (2006) recommended  $1000 \text{ mg N m}^{-2} \text{ yr}^{-1}$  as a “critical load” threshold for plant  
371 sustainability, and rates above this threshold may lead to changes in ecosystemic functioning. The  
372 deposition rate of N for 1997-2013 also exceeded  $1000 \text{ mg N m}^{-2} \text{ yr}^{-1}$  in large areas of India (72%), China  
373 (45%) and Europe (26%). The deposition rate of P for 1997-2013 exceeded  $100 \text{ mg P m}^{-2} \text{ yr}^{-1}$  in the  
374 Indo-Gangetic region of India due to a high use of biofuels (*e.g.*, dung cake is widely used for cooking) and  
375 in the Congo Basin in Central Africa due to deforestation (Chen *et al.*, 2010).



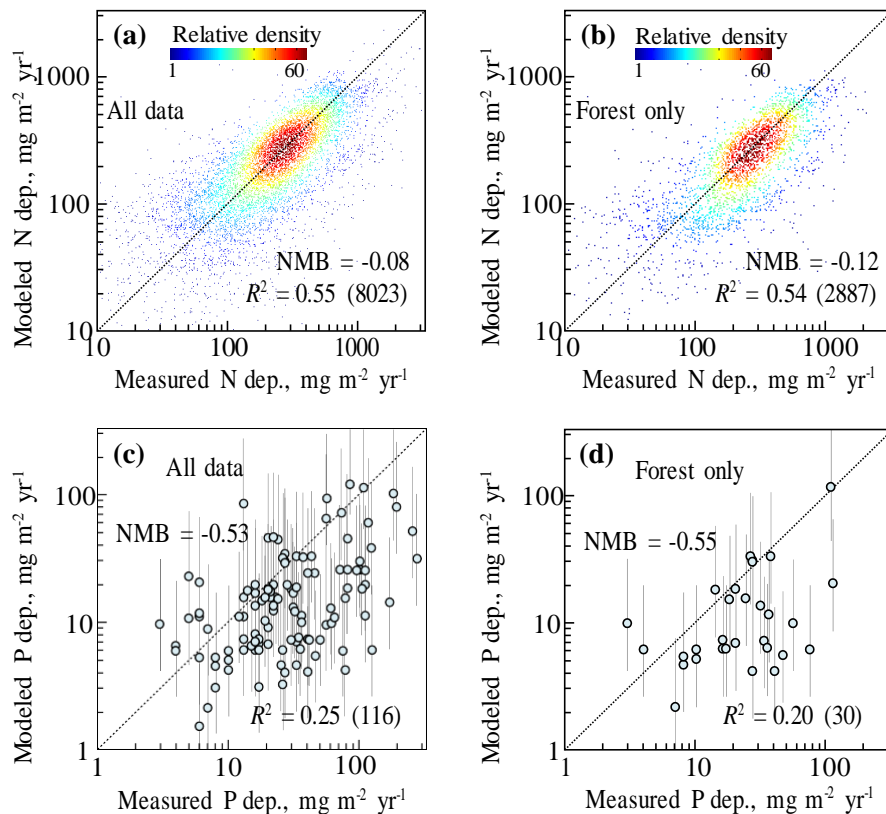
376

377

378 **Figure 2.** (a,b) Spatial distribution of N deposition in 1850 (a) and 1997-2013 (b). (c,d) Spatial distribution  
379 of P deposition in 1850 (c) and 1997-2013 (d).

380

381 The modeled spatial distributions of N wet and P total deposition rates were evaluated by measurements at  
382 the forest sites and at all sites (Figure 3). The comparison indicated that our model broadly captured the  
383 spatial patterns in the observed deposition rates of N and P. A normalized mean bias (difference between  
384 the geometric mean of the model minus the geometric mean of the measurements relative to the latter) was  
385 used to quantify the bias. A statistical analysis show that 50% of the data were subject to a bias of -25% to  
386 50% in the modeled wet N deposition and -4% to 164% in the modeled total P deposition relative to  
387 observations (Figure S2). We addressed the potential uncertainty by scaling the modeled N deposition rate  
388 by a fixed factor that followed a normal distribution, with a mean of 1.2 and a standard deviation of 0.6,  
389 and the modeled P deposition by a fixed factor that followed a normal distribution, with a mean of 1.6 and  
390 a standard deviation of 1.0, which were derived from the frequency distribution of the model bias (Figure  
391 S2). The scaled deposition rates of N and P were used in the calculation of  $\Delta C_{v,dep}$ . The data for observed  
392 N wet deposition enabled us to further evaluate the modeled wet deposition of N in the oxidized ( $NO_3$ ) and  
393 reduced ( $NH_4$ ) forms by region (Figure S3).



394

395 **Figure 3.** Comparison of modelled and observed deposition of wet N (a,b) and total P (c,d). (a,c) show all  
396 data and (b,d) show measurements over forests only. In (a,b), colours show the relative density of data. In  
397 (c,d), the error bars show the uncertainty associated with emissions of P from different sources as estimated  
398 by Wang *et al.* (2015a). Coefficient of correlation ( $R^2$ ) and normalized mean bias (NMB) of log-transferred  
399 deposition rates are given in each panel with the number of data in bracket.

400

401 **Figure S4** shows a comparison of our modeled surface concentrations of P for particles of the same size



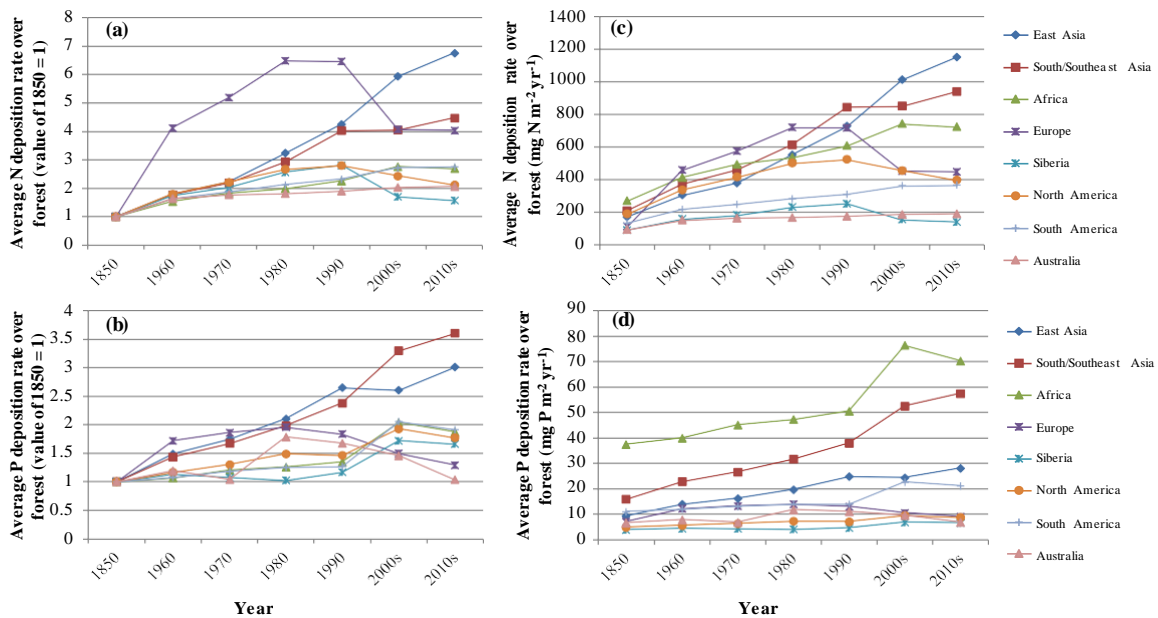
402 with the observational data used by Mahowald *et al.* (2008). These discrete sampling measurements were  
403 temporally variable, so we focused on comparing the modeled P concentrations with long-term  
404 measurements. It shows that including a large contribution of P from combustion sources can reduce the  
405 normalized mean bias from -31% to -11%, while our estimates were subject to uncertainty errors between  
406 -73% to 105% as 95% CIs in the estimation of P emissions from different sources (Wang *et al.*, 2015a).

407

### 408 3.3. Temporal trends of N and P deposition in forests

409 **Figure 4** shows the temporal trends of N and P deposition in forests in various regions from 1850 to the  
410 present. The increase in the rate of N deposition in forests from 1850 to the present ranged from 1.5- to  
411 7-fold by region. The rate of P deposition in forests increased by 1- to 3.5-fold from 1850 to the present,  
412 due to a smaller contribution of anthropogenic source than for N. The deposition of N and P in North  
413 American and European forests both peaked in the 1980s or 1990s. Specifically, the deposition rates of N  
414 and P in European forests peaked in the 1980s and 1990s, respectively. The rate of N deposition in North  
415 American forests peaked in 1990, but the rate of P deposition continued to increase. The increase in the  
416 deposition of P in North American forests was mainly due to an increase in emissions from biomass  
417 burning during the last two decades (Westerling, *et al.* 2006). This increase was captured by the GFED4.1  
418 and ACCMIP inventories (Lamarque *et al.*, 2010; Giglio *et al.*, 2013) used in our study. **Figure S5** shows  
419 that this increasing trend was also confirmed by three other inventories for biomass burning. The rate of N  
420 deposition in Siberian forests had a trend similar to that of European forests due to the atmospheric  
421 transport of N from Europe, but the rate of P deposition was mainly governed by the variability of wildfires.  
422 The rate of N deposition in the forests in Australia and New Zealand increased continuously, but the rate of  
423 P deposition began to decline in Australia after 1980 due to the replacement of P-rich fuels (coal) with  
424 P-poor fuels (petroleum or natural gas) (IEA, 2013).

425 The rate of N deposition in East Asian forests was 7-fold higher in 2010 than 1850 and more than 4-fold in  
426 Southern/Southeastern forests. Total N emissions were highest in China and India, but NO<sub>x</sub> emissions had  
427 increased more in China from 1990 to 2010 by 150%, compared to 80% in India, due to a faster growth in  
428 fleet vehicle in China (Granier *et al.*, 2011).



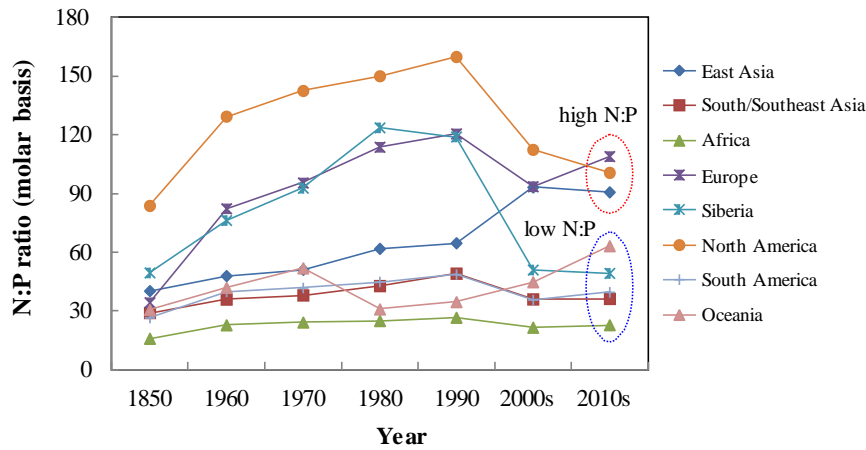
429

430 **Figure 4.** Temporal trends of N (a,c) and P (b,d) deposition rate over forests by region. (a,b) show the  
 431 values relative to 1850, while (c,d) show absolute values. The data for 2000s represent an average over  
 432 1997-2004 and the data for 2010s represent an average over 2005-2013.

433

434 The rate of P deposition in East Asian forests increased 2.5-fold from 1850 to 1990 and stabilized after  
 435 1990. **Figure S6** shows the inter-annual variability of N- and P-deposition rates in the forests in East Asia  
 436 and South/Southeast Asia from 1997 to 2013. The rate of N deposition increased continuously, but the rate  
 437 of P deposition remained more stable in these two regions. The increase in the rate of N deposition was  
 438 driven by the increase of in the use of fossil fuels and in agricultural activities. The rate of P deposition,  
 439 however, was driven by emissions from fossil fuels, biofuel and biomass burning. The increase in fires and  
 440 biomass burnt during the 1997-1998 El Niño led to a peak in the P-deposition rate in the forests in East  
 441 Asia and South/Southeast Asia ([van der Werf \*et al.\*, 2004](#)).

442 **Figure 5** shows the temporal trends of the N:P deposition ratio (on a molar basis) in forests by region. The  
 443 ratio peaked in around 1990 in the forests in Europe, Siberia and North America and then decreased due to  
 444 a decline in the deposition of N and an increase in the deposition of P due to increasing wildfires. The N:P  
 445 deposition ratio in the forests did not vary greatly in the forests in South/Southeast Asia, Africa and South  
 446 America, because the rates of N and P deposition increased at similar rate. In contrast, the N:P deposition  
 447 ratio increased continuously in the forests in East Asia due to rapid increases in the emission and deposition  
 448 of N in this region. The N:P deposition ratio in the 2010s was significantly larger in the forests in Europe,  
 449 North America and East Asia than in the other regions.

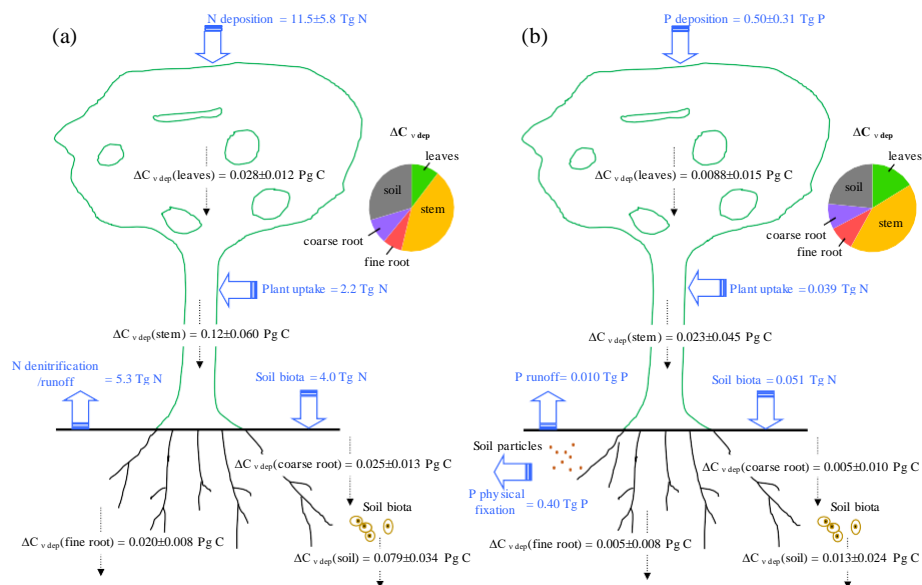


450  
 451 **Figure 5.** Temporal evolution of N:P ratio in the deposition over forests by region. The data for 2000s  
 452 represent an average over 1997-2004 and the data for 2010s represent an average over 2005-2013.

453

### 454 3.4. Forest C fixation due to anthropogenic N and P deposition

455 Based on the maps of N and P deposition, we calculated the global forest  $\Delta C_{v,dep}$  using the stoichiometric  
 456 mass balance approach. **Figure 6** shows that, for control values of the parameters, anthropogenic deposition  
 457 of N and P for 1997-2013 led to global  $\Delta C_{v,dep}$  (median  $\pm$  90% CI from Monte Carlo simulations) of  $0.27 \pm$   
 458  $0.13$  and  $0.054 \pm 0.10$  Pg C yr<sup>-1</sup>, respectively. A total of 10 000 Monte Carlo simulations were run by  
 459 randomly entering parameters from a prior uniform or normal distribution of parameters. The frequency  
 460 distributions of global  $\Delta C_{v,dep}$  by anthropogenic N and P deposition averaged for 1997-2013 are shown in  
 461 **Figure S7**. Some parameters were uniformly or normally distributed, but the output was nearly normally  
 462 distributed for  $\Delta C_{N,dep}$  ( $P > 0.1$ ) and log-normally distributed for  $\Delta C_{P,dep}$  ( $P > 0.2$ ). For a global terrestrial C  
 463 sink of  $3.1 \pm 0.9$  Pg C yr<sup>-1</sup> averaged for 2006-2015 (Le Quere *et al.*, 2016),  $\Delta C_{v,dep}$  by anthropogenic N and  
 464 P deposition contributed 8.7% and 1.7% to the terrestrial C sink, respectively.



465  
 466 **Figure 6.** Anthropogenic N and P deposition over forest and  $\Delta C_{v,dep}$  due to anthropogenic N and P  
 467 deposition per year as an average over 1997-2013. (a) for N and (b) for P. Uncertainty of the modeled N

468 and P deposition as standard deviations is derived from a comparison with observations. Uncertainty of the  
 469  $\Delta C_{v\text{ dep}}$  as 90% CI is derived from Monte Carlo simulations. The pie charts show the distribution of  $\Delta C_{v\text{ dep}}$   
 470 among five pools.

471

472 **Figure 6** also shows that 43% of  $\Delta C_{N\text{ dep}}$  was stored in stems, followed by soil (29%), leaves (11%), coarse  
 473 roots (9%) and fine roots (7%). Similarly, stems (42%) were the most important storage pool for  $\Delta C_{P\text{ dep}}$ . In  
 474 addition, we performed sensitivity tests using the lower or upper bounds of the parameters to investigate the  
 475 influences on our estimated  $\Delta C_{N\text{ dep}}$  (**Table 2**). It shows that the major parameters influencing  $\Delta C_{v\text{ dep}}$  by N  
 476 or P include the retention ratios of N in vegetation and soils, the physical fixation of P by soil, the fraction  
 477 of N and P allocated to woody biomass, and the C:N and C:P stoichiometric ratios in each pool.  $\Delta C_{P\text{ dep}}$  was  
 478 generally associated with a far higher uncertainty than  $\Delta C_{N\text{ dep}}$ . Most importantly, we assumed that a large  
 479 fraction of P (80%) was fixed by the soil as inorganic P, which is unavailable for C storage ([Walker and](#)  
 480 [Syres, 1976](#)). Assuming a weaker or stronger fixation of P by soils, which influences estimates of  $\Delta C_{v\text{ dep}}$   
 481 by P the most, however, led to a difference in  $\Delta C_{P\text{ dep}}$  from -70% to +300%.

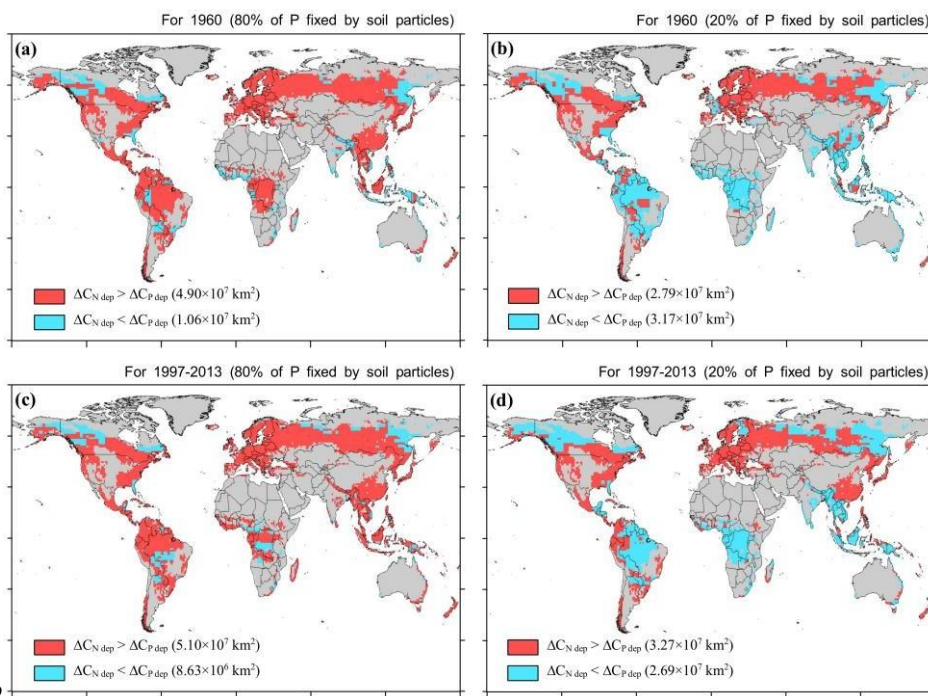
482 **Table 2. Global forest  $\Delta C_{v\text{ dep}}$  due to anthropogenic N and P deposition over 1997-2013.** Sensitivity  
 483 tests were run to compare with a standard run with central values of parameter. The sensitivity tests include  
 484 high or low retention fraction of N in vegetation and soil; weak or strong fixation of P by soil particles;  
 485 high or low fraction of N and P allocated to the woody part; high or low C:N and C:P stoichiometric ratios.  
 486 Values in brackets show the percentage changes relative to the standard run.

	Leaf	Stem	Fine root	Coarse root	Soil	Woody	Total
<b>Forest <math>\Delta C_{v\text{ dep}}</math> by N deposition over 1997-2013 (Pg C yr<sup>-1</sup>)</b>							
Standard run	0.028	0.117	0.020	0.025	0.079	0.142	0.269
High retention fraction	0.037	0.153	0.026	0.033	0.107	0.186 (+31%)	0.356 (+32%)
Low retention fraction	0.018	0.073	0.013	0.016	0.042	0.088 (-38%)	0.160 (-40%)
High woody fraction	0.021	0.168	0.014	0.037	0.079	0.205 (+45%)	0.320 (+19%)
Low woody fraction	0.034	0.079	0.022	0.017	0.079	0.097 (-32%)	0.232 (-14%)
High C:N ratio	0.034	0.140	0.024	0.030	0.079	0.170 (+20%)	0.307 (+15%)
Low C:N ratio	0.023	0.093	0.016	0.020	0.079	0.113 (-20%)	0.231 (-15%)
<b>Forest <math>\Delta C_{v\text{ dep}}</math> by P deposition over 1997-2013 (Pg C yr<sup>-1</sup>)</b>							
Standard run	0.009	0.023	0.005	0.005	0.013	0.028	0.054
Weak fixation by soil	0.035	0.091	0.020	0.020	0.051	0.111 (+300%)	0.218 (+300%)
Strong fixation by soil	0.003	0.007	0.002	0.002	0.004	0.008 (-70%)	0.016 (-70%)
High woody fraction	0.006	0.039	0.003	0.009	0.013	0.048 (+73%)	0.070 (+29%)
Low woody fraction	0.009	0.018	0.005	0.004	0.013	0.022 (-19%)	0.050 (-8%)
High C:P ratio	0.011	0.027	0.006	0.006	0.013	0.033 (+20%)	0.063 (+15%)
Low C:P ratio	0.007	0.018	0.004	0.004	0.013	0.022 (-20%)	0.046 (-15%)

487

### 488 3.5. Spatial patterns of N and P limitation

489 Increasing N from atmospheric deposition has been postulated to lead to a progressive emerging limitation  
 490 of P (Vitousek *et al.*, 1984, 2010; Penuelas *et al.*, 2013). **Figure 7** compares the forest area where  $\Delta C_{N\text{ dep}}$  is  
 491 larger than  $\Delta C_{P\text{ dep}}$  between 1960 and the present (1997-2013) under different assumptions of the physical  
 492 fixation of P by the soil. A faster increase in the deposition of N than P in forests close to industrialized  
 493 regions had led to a growth in  $\Delta C_{v\text{ dep}}$  that is higher for N than for P. It should be noted that, as we did not  
 494 account for other sources of N and P (*e.g.* weathering for P and nitrification for N), lower  $\Delta C_{v\text{ dep}}$  from N  
 495 than P deposition is only one factor contributing to a limitation by P (Vitousek *et al.*, 1984, 2010; Penuelas  
 496 *et al.*, 2012, 2013). Globally, the forests where  $\Delta C_{N\text{ dep}}$  is larger than  $\Delta C_{P\text{ dep}}$  in 1960 covered an area  
 497 between  $2.79$  and  $4.90 \times 10^7$  km<sup>2</sup>, depending on the strength of soil P fixation. In these forests, additional  
 498 supply of P from other sources (*e.g.*, by mineralization) is needed to support  $\Delta C_{v\text{ dep}}$  by N. The forest area  
 499 where  $\Delta C_{N\text{ dep}}$  is larger than  $\Delta C_{P\text{ dep}}$  had extended by 4% or 18% from 1960 to the present, which also  
 500 depends on the strength of soil P fixation, mainly located in forests in South/Southeast Asia, East Asia,  
 501 Europe and North America.



502 **Figure 7.** Spatial distribution of forest area where  $\Delta C_{v\text{ dep}}$  by anthropogenic N deposition is larger or  
 503 smaller than  $\Delta C_{v\text{ dep}}$  by anthropogenic P deposition in 1960 (**a,b**) and as an average over 1997-2013 (**c,d**).  
 504 In (**a,c**), the model assumes that 80% of the deposited P is fixed by soil particles, while (**b,d**) shows results  
 505 in a sensitivity case where 20% of P is fixed by soil particles. The total areas where  $\Delta C_{v\text{ dep}}$  by  
 506 anthropogenic N deposition is larger or smaller than  $\Delta C_{v\text{ dep}}$  by anthropogenic P deposition are in  
 507 parentheses. Non-forest land areas are shown as grey.

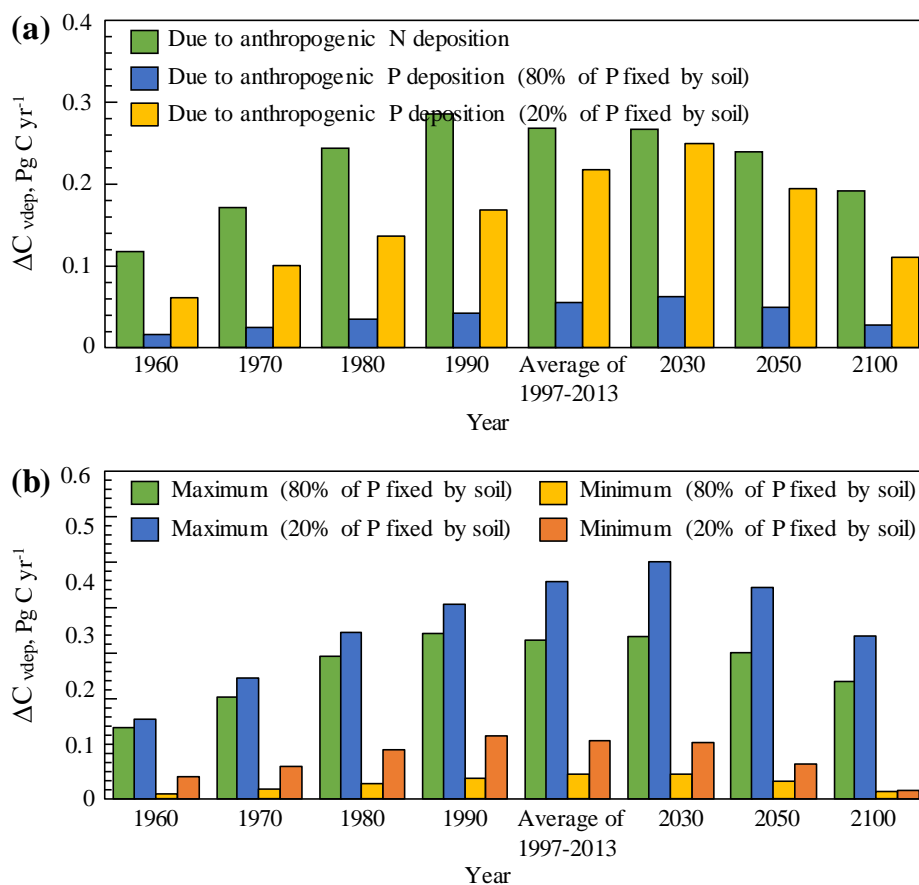
508

### 509 3.6. Temporal trends of $\Delta C_{v\text{ dep}}$ by N and P

510 **Figure 8a** shows the temporal trends of  $\Delta C_{v\text{ dep}}$  by N and P deposition alone for 1960-2100 under different  
 511 physical fraction of P fixed in soil.  $\Delta C_{v\text{ dep}}$  by N peaked in 1990 and then declined due to reductions of N  
 512 emissions and deposition in the forests in Europe and North America, but  $\Delta C_{v\text{ dep}}$  by P was projected to  
 513

514 increase by 2030 due to continuous increases in P emissions and deposition in South and Southeast Asia,  
 515 Africa and South America under the RCP4.5 scenario. We also found that  $\Delta C_{v\text{dep}}$  by N was much higher  
 516 than  $\Delta C_{v\text{dep}}$  by P in our central case where 80% of the P was fixed by soil, but they were very similar in a  
 517 sensitivity test where only 20% of P was fixed by soil.  $\Delta C_{v\text{dep}}$  by N for 2030, 2050 and 2100 was expected  
 518 to be 28-60% higher under the RCP8.5 than the RCP4.5 scenario due to higher  $\text{NH}_3$  emissions from  
 519 agriculture. Difference in  $\Delta C_{v\text{dep}}$  by P would be less than 15% due to a high control rate of industrial  
 520 particulate emissions under both scenarios (Lamarque *et al.*, 2011) (Figure S8).

521 Based on  $\Delta C_{v\text{dep}}$  by N and P in each  $1^\circ \times 1^\circ$  grid, we calculated  $\Delta C_{v\text{dep}}$  by N and P deposition together in  
 522 the case of maximum or minimum effects from 1960 to 2100 (Figure 8b).  $\Delta C_{v\text{dep}}$  was about 6-fold higher  
 523 for maximum (0.28  $\text{Pg C yr}^{-1}$  averaged for 1997-2013) than minimum (0.044  $\text{Pg C yr}^{-1}$ ) effects,  
 524 highlighting the imbalance between N and P in the deposition.  $\Delta C_{v\text{dep}}$  for maximum effects peaked at 0.29  
 525  $\text{Pg C yr}^{-1}$  in 1990 and then decreased due to reductions of N emissions and deposition rates in the forests in  
 526 Europe and North America. In a sensitivity test where 20% of P was fixed by soil, however,  $\Delta C_{v\text{dep}}$  for  
 527 maximum effect was predicted to increase continuously from 0.14  $\text{Pg C yr}^{-1}$  in 1960 to 0.42  $\text{Pg C yr}^{-1}$  in  
 528 2030 due to increases in both N and P deposition rates before 1990 and an increase in P deposition rate  
 529 alone after 1990. Figure S8 shows  $\Delta C_{v\text{dep}}$  due to N and P together under the RCP8.5 scenario, where  $\Delta C_{v\text{dep}}$   
 530 for maximum effect was expected to be 23-54% higher than under the RCP4.5 scenario.



531

532

533 **Figure 8.** Temporal trends of global  $\Delta C_{v\text{dep}}$  due to anthropogenic N and P deposition alone (a) and together  
 534 as maximum or minimum effects (b) from 1960 to 2100.  $\Delta C_{v\text{dep}}$  in a case where 80% of P is fixed by soil

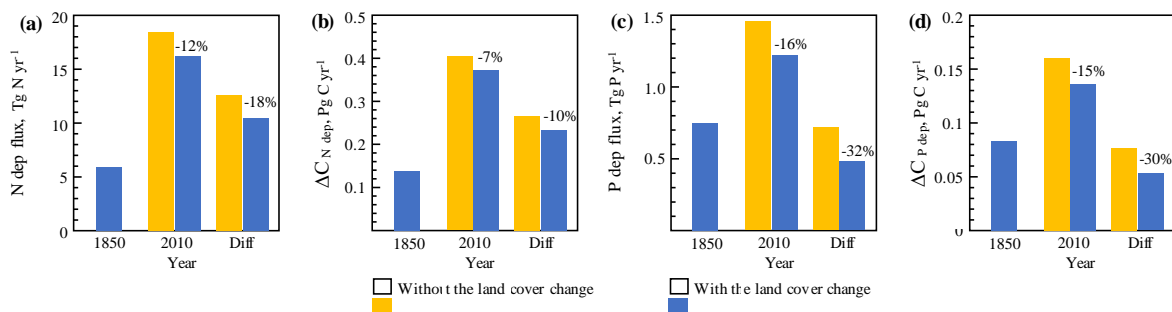


535 is compared to a sensitivity case where 20% of P is fixed by soil. The  $\Delta C_{v\text{ dep}}$  for 2030, 2050 and 2100 are  
 536 estimated based on the anthropogenic N and P deposition under the RCP 4.5 storyline.

537

### 538 3.7. Impact of forest cover change on $\Delta C_{v\text{ dep}}$

539 In addition to the change of N and P deposition rates, the change of forest areas due to deforestation and  
 540 afforestation influences  $\Delta C_{v\text{ dep}}$ . **Table S4** shows the change of total forest areas, average N and P  
 541 deposition fluxes over forests and  $\Delta C_{v\text{ dep}}$  estimated for the five types of forests using a reconstructed  
 542 global data set of forest cover change from 1850 to 2010 (Peng *et al.*, 2017). **Figure 9** compares the  
 543 deposition fluxes and the  $\Delta C_{v\text{ dep}}$  for 1850 and 2010 with or without forest cover change by taking 1850 as  
 544 a reference, whereas the results for 2010 are calculated using the forest cover map in 1850 or 2010. It  
 545 shows that, if accounting for forest cover change,  $\Delta C_{v\text{ dep}}$  due to change in N and P deposition from 1850 to  
 546 2010 would decrease by 10% and 30%, respectively. Likewise, the deposition fluxes and  $\Delta C_{v\text{ dep}}$  for 1850  
 547 and 2010 are compared with or without the forest cover change by taking 2010 as a reference, whereas the  
 548 results for 1850 are calculated using the forest cover map in 1850 or 2010 (**Figure S9**). Similarly, it shows  
 549 that the forest cover change leads to a difference in  $\Delta C_{v\text{ dep}}$  by -6% and -19% for N and P, respectively.



550

551 **Figure 9.** Comparison of global N deposition fluxes over forests (a),  $\Delta C_{v\text{ dep}}$  due to N deposition (b), P  
 552 deposition fluxes over forests (c) and  $\Delta C_{v\text{ dep}}$  due to P deposition (d) in 1850 and 2010 and the difference  
 553 between 1850 and 2010 (Diff) without or with the forest cover change by taking year 1850 as a reference.  
 554 The yellow bars show the  $\Delta C_{v\text{ dep}}$  calculated based on the forest cover map in 1850, and the blue bars show  
 555 the  $\Delta C_{v\text{ dep}}$  calculated based on the forest cover map for 1850 and 2010 respectively. In all cases,  $\Delta C_{v\text{ dep}}$  is  
 556 estimated using the central values of parameters. Relative difference as a percentage by accounting for the  
 557 forest cover change is given over each bar.

558

559 There are three reasons for the difference. First, the global total forest area had declined by 12% from  $29.5$   
 560  $\times 10^6$  km<sup>2</sup> in 1850 to  $25.9 \times 10^6$  km<sup>2</sup> in 2010. Second, the C response to N or P deposition varies across the  
 561 five types of forest and change in the relative cover of each forest type influences the estimation of  $\Delta C_{v\text{ dep}}$ .  
 562 For example, the C response to N deposition is the highest for the evergreen needle-leaf forests (47 kg C kg  
 563 N<sup>-1</sup>) and the lowest for the evergreen broadleaf forests (12 kg C kg N<sup>-1</sup>). From 1850 to 2010, the cover  
 564 fraction of evergreen needle-leaf forests had increased from 19% to 21%, while the cover of evergreen  
 565 broadleaf forests had decreased from 49% to 44%, leading to less reduction in  $\Delta C_{N\text{ dep}}$  than N deposition  
 566 fluxes after accounting for the land cover change (**Figure 9**). At last, change in the spatial distribution of  
 567 forests also leads to some changes in the average N or P deposition rates, but our data sets show that this  
 568 influence is relatively small. The average N and P deposition rates over global forests in 2010 were 628 mg

569 N m<sup>-2</sup> yr<sup>-1</sup> and 47 mg P m<sup>-2</sup> yr<sup>-1</sup> using the forest cover map in 2010, compared with 626 mg N m<sup>-2</sup> yr<sup>-1</sup> and  
570 49 mg P m<sup>-2</sup> yr<sup>-1</sup> using the forest cover map in 1850.

571

## 572 4. Discussion

### 573 4.1. Global data sets of N and P deposition

574 Our study provides the first global gridded data sets for both N and P deposition from pre-industrial (1850),  
575 historical periods (1960, 1970, 1980 and 1990), to present (1997-2013), and into the future (2030, 2050 and  
576 2100) from a global climate-chemistry model driven by complete bottom-up emission inventories. **Table**  
577 **S1** provides a comparison of species represented by the model, horizontal resolution, and observational  
578 data used to evaluate the model with previous studies. In the simulation of N, we employed state-of-the-art  
579 inventories for reactive N and other tracers that influence the atmospheric chemistry of N (Klimont *et al.*,  
580 2013). The chemistry of N in the model is advanced in the treatments of the ammonia cycle and the nitrate  
581 particle formation (Hauglustaine *et al.*, 2014), which are only represented in few global models (Dentener  
582 *et al.*, 2006). Our model also provides a horizontal resolution of 1.2°×2.5° that is finer than most of  
583 previous models (Dentener *et al.*, 2006). For P, we used a new emission inventory from combustion  
584 sources with the uncertainties quantified (Wang *et al.*, 2015a), which reduced the underestimation in the  
585 modeled P deposition by covering P in all sizes of particles which are carried to the measurement sites.  
586 Prior to our work, there was only one global data set of present day P deposition available (Mahowald *et al.*,  
587 2008). In addition to its lack of temporal changes, this previous data set covered P in particles with  
588 diameter smaller than 10 μm. Wang *et al.* (2015a) showed that it led to a discrepancy between the modeled  
589 and observed P deposition rates at globally distributed measurement stations if P in particles with diameter  
590 larger than 10 μm was neglected even if uncertainties in P emissions from other sources were accounted  
591 for.

592 The modeled NO<sub>3</sub> and NH<sub>4</sub> wet deposition rates were evaluated by comparing with 8023 observations from  
593 globally distributed measurement stations. The modeled P deposition rates were compared with 116  
594 observations. The normalized mean bias (NMB) is -8% for the NO<sub>3</sub> and NH<sub>4</sub> wet deposition rates, and  
595 -53% for the P deposition rates. It also shows that 50% of the data were associated with a bias between  
596 -25% and 50% for the NO<sub>3</sub> and NH<sub>4</sub> wet deposition and between -4% and 164% for the P deposition. The  
597 correlation coefficient of the log-transformed deposition rate is higher for N ( $R=0.74$ ) than for P ( $R=0.50$ ).  
598 The underestimation of P deposition was likely due to errors in the transport model or ignored local  
599 biogenic aerosols in the measurement samples. The distributions of these model errors were treated as  
600 uncertainties in our data sets. It is not a surprise that there is a larger uncertainty associated with the  
601 modeled deposition rates of P than N. For P, in addition to the errors associated with atmospheric transport  
602 and removal, emissions of P from wind erosion of soil dusts, biogenic aerosols, marine sea-salt particles,  
603 volcanoes and other sources (*e.g.*, phosphine from freshwater wetlands and rice paddies) are all subject to  
604 high uncertainties (Graham and Duce, 1979; Mahowald *et al.*, 2008; Wang *et al.*, 2015a). A limitation of  
605 our P deposition simulations set is that the long-term variations of these emissions are not resolved, because



606 no information on evolution of these sources is currently available at the global scale. More measurements  
607 of P deposition rates in contrasting environments are useful to reduce the uncertainty in the modeled P  
608 deposition rates (e.g., Du *et al.*, 2016), while more long-term measurements of the surface concentrations of  
609 P with additional source information are useful to constrain the model and emissions of P from difference  
610 sources (Mahowald *et al.*, 2008).

611 For N, we had lower model biases in some regions when compared with previous studies. For example,  
612 Hauglustaine *et al.* (2014) simulated N deposition using the same climate-chemistry model and evaluated  
613 the model using 4036 observations from global monitoring stations. They reported that NMB is  
614 respectively -32%, -4.5% and -60% in North America, Europe and Asia for the modeled NH<sub>4</sub> wet  
615 deposition, and -28%, 13% and -54% for the modeled NO<sub>3</sub> wet deposition. In contrast, NMB in our study is  
616 -12% in North America, -42% in Europe and -28% in East Asia respectively for NH<sub>4</sub> wet deposition, and  
617 29%, -29% and -11% respectively for NO<sub>3</sub> wet deposition (Figure S3). Thus, our results are better for Asia  
618 but worse for Europe. Hauglustaine *et al.* (2014) attributed the underestimation in Asia to a coarse  
619 horizontal resolution of the model and an underestimation of reactive-N emissions in the region (Lamarque  
620 *et al.*, 2010). We updated reactive-N emissions by using a more recent ECLIPSE GAINS.4a inventory  
621 (Klimont *et al.*, 2013) and used a higher horizontal resolution version of the atmospheric model, from  
622 1.9°×3.75° previously to 1.2°×2.5°. These together reduced the model bias in Asia. Despite a larger NMB  
623 in Europe in our model, the correlation coefficient (*R*) is 0.63 and 0.79 for wet NH<sub>4</sub> and NO<sub>3</sub> deposition in  
624 this region, respectively, higher than 0.33 and 0.49 by Hauglustaine *et al.* (2014), indicating that our model  
625 better captures the spatial pattern of N deposition.

626 Overall, our study provides consistent gridded simulations of both N and P deposition at a high horizontal  
627 resolution, which agrees better with observations from a large number of globally distributed measurement  
628 stations than previous studies (Mahowald *et al.*, 2008; Hauglustaine *et al.*, 2014). A statistical analysis of  
629 the model-observation comparison generates a probabilistic distribution of the model bias, which enabled  
630 us to address the uncertainty in our data sets. Whereas previous studies (de Vries *et al.*, 2014) have only  
631 estimated the C sequestration by current N deposition without spatial and temporal variations, we estimated  
632 C sink changes from changes in N and P deposition maps and included an uncertainty analysis to both N, P  
633 deposition and C sink responses.

#### 634 **4.2. P deposition contributes significantly to forest C storage**

635 Increasing atmospheric CO<sub>2</sub> concentrations, longer growing season and N fertilization have been suggested  
636 to lead to limitation by P of productivity in temperate forests (Vitousek *et al.*, 1984, 2010; Penuelas *et al.*,  
637 2012, 2013). Our long-term data of P-deposition rates, combined with data of N-deposition rates, suggested  
638 that  $\Delta C_{P\text{ dep}}$  would be close to  $\Delta C_{N\text{ dep}}$  if 20% of the P is fixed by soil, and C fixation due to P deposition is  
639 higher than previously estimated (Cleveland *et al.*, 2013; de Vries *et al.*, 2014) due to higher P deposition  
640 rates and a less fraction of P loss (Sattari *et al.*, 2012). Our simple stoichiometric mass balance approach  
641 indicated that the strength of P fixation by soil particles exerted the strongest influence on the response of C  
642 fixation to P deposition. Consequently, we adopted a global-constant fraction of deposited P being fixed by

643 soil ranging from 20% to 94%, based on the transfer coefficients between stable and labile P from Sattari *et*  
644 *al.* (2012). However, it should be noted that this fraction was not uniform in space and was probably soil  
645 type dependent (Compton and Cole, 1998). More field experiments measuring fixation of P by soil particles  
646 are needed to reduce this uncertainty (Johnson *et al.*, 2003).

647 Nevertheless, our stoichiometric method found that  $\Delta C_{P\text{ dep}}$  was equivalent to 50-90% of  $\Delta C_{N\text{ dep}}$  when the  
648 fixation of P by soil was weak, implying that deposited P can contribute to forest C storage providing that  
649 other sources of N and P were held constant. Spatially,  $\Delta C_{P\text{ dep}}$  could even exceed  $\Delta C_{N\text{ dep}}$  in some areas of  
650 forests in East Asia, South and Southeast Asia, Africa and South America (Figure 7). In particular, we find  
651 that  $\Delta C_{P\text{ dep}}$  can be significantly higher than  $\Delta C_{N\text{ dep}}$  in tropical forests in the Congo and Amazon Basins and  
652 Indonesia, due to both rapid losses of N from ecosystems through denitrification (Bai *et al.*, 2012) and high  
653 deposition rates of P from deforestation fire emissions (Giglio *et al.*, 2013). The C sink in tropical forests is  
654 sensitive to additional P inputs, because P availability in the soil of tropical forests was found to be far  
655 lower than the global average (Yang *et al.*, 2013) but P use efficiency was higher than that in boreal forests  
656 (Gill and Finzi, 2016). We suggest that more attentions should be paid to estimate the C sink due to  
657 additional P inputs from deposition in tropical forests.

658 However, our current knowledge of nutrient cycling is more limited for P than for N, leading to high  
659 uncertainties global ecosystem-level models (Wang *et al.*, 2010; Goll *et al.*, 2012; Yang *et al.*, 2014). It is  
660 known that most of P is neither directly taken up by plant nor lost by leaching, but a large fraction of P is  
661 fixed by soils before being slowly transferred into a labile pool that can be used by plant (Aerts and Chapin,  
662 2000; Sattari *et al.*, 2012). Different from N, there is no atmospheric loss pathway for P, while P is less  
663 mobile in soil and hence less prone to leaching than for N (Aerts and Chapin, 2000; Goll *et al.*, 2012; Goll,  
664 2016), but it remains unknown whether and how fast the fixed P in soil can be mineralized and used by  
665 plants (Lü *et al.*, 2013). A meta-analysis of P and N plus P fertilization experiments suggested that  
666 increasing N availability, *e.g.* from increasing N deposition, tends to increase P cycling rate and thus  
667 contributes to use of P by plants (Marklein and Houlton, 2012). Measurements of the rate of P uptake in the  
668 seasons with both high N and P-deposition rates are useful to understand the contribution of N and P  
669 deposition together to the C fixation in forests.

670 The cycles of C, N and P differ in their respect to the residence time in terrestrial ecosystem (Walker and  
671 Syres, 1976). Our stoichiometric method attributes the C storage due to N and P deposition at a timescale  
672 of 10-20 years, following de Vries *et al.* (2014). Nonetheless, P would turn over much more slowly than N  
673 (Walker and Syres, 1976), so it is worth highlighting that the effects of P would last longer than those of N.  
674 We expect that the use by plants is much slower for deposited P than deposited N due to the strong physical  
675 fixation of P in soil (Aerts and Chapin, 2000; Goodale *et al.*, 2002), and our estimated  $\Delta C_{P\text{ dep}}$  under a weak  
676 fixation should include part of the long-term effects of P deposition. Under a high CO<sub>2</sub> concentration in the  
677 near term, plants are likely increasing their efficiency in accessing and utilizing these not readily available  
678 P (Buendia *et al.*, 2014, Goll, 2016). Such effects should be better represented in the process-based models  
679 when studying the impact of N and P deposition on P limitation (Cleveland *et al.*, 2013; Wieder *et al.*,

680 2015).

681 While we cannot quantify all sources of N and P for plants, calculation of maximum and minimum  $\Delta C_{v\text{ dep}}$   
682 by N and P provides a preliminary estimation of the C storage supported by anthropogenic P deposition.  
683 Under a weak fixation for P by soil, maximum  $\Delta C_{v\text{ dep}}$  by N and P ( $0.38 \text{ Pg C yr}^{-1}$ ) is 43% higher than  $\Delta C_{v\text{ dep}}$   
684 by N alone ( $0.27 \text{ Pg C yr}^{-1}$ ). Meanwhile, minimum  $\Delta C_{v\text{ dep}}$  by N and P ( $0.10 \text{ Pg C yr}^{-1}$ ) is 53% lower  
685 than  $\Delta C_{v\text{ dep}}$  by P alone ( $0.22 \text{ Pg C yr}^{-1}$ ). They both suggest a potential contribution of anthropogenic P  
686 deposition to additional forest C sink. The map of  $\Delta C_{v\text{ dep}}$  by N and P also implies that other sources of N  
687 are needed to support the C storage by P deposition in some Asian tropical forests where the deposition rate  
688 increased faster for P than for N (Figure 7). However, it should be noted that N fertilization can also exert a  
689 significant effect on the capacity of plant to use the P (Marklein and Houlton, 2012; Lü *et al.*, 2013), and  
690 the maximal co-fertilized  $\Delta C_{v\text{ dep}}$  is likely even larger than maximum  $\Delta C_{v\text{ dep}}$  estimated by our method.

#### 691 4.3. Uncertainty due to N retention by the canopy

692 We aimed at providing a preliminary estimation of forest C response to N and P deposition and investigated  
693 the imbalance of N and P in the deposition at a mid-term scale of 10-20 years when other sources were held  
694 constant. We find that anthropogenic P deposition can contribute to C storage, supplementing  $\Delta C_{v\text{ dep}}$   
695 supported by anthropogenic N deposition by 43%. It is worth highlighting that the impact of N deposition  
696 on C storage also remains uncertain, despite more measurements of N than P retention in ecosystems, due  
697 largely to the uncertain impact of canopy N uptake. It was noticed that ~40% of N deposited is retained by  
698 the canopy in Europe and North America (Lovett and Lindberg, 1993), but the impact on C storage is not  
699 well constrained (Sparks, 2009). Sievering *et al.* (2007) found that 80% of the growing-season N deposition  
700 was retained in canopy foliage and branches and that ~20% of daytime net ecosystem exchange may be  
701 attributed to canopy N uptake. Dezi *et al.* (2010) found that the net ecosystem production would be  
702 increased by 58% under a hypothesis that canopy N uptake can directly stimulate photosynthesis relative to  
703 without canopy N uptake. Nair *et al.* (2016) suggested that accounting for canopy N uptake could lead to an  
704 increased C response to N deposition ( $\text{oC/oN}$ ) from 43 to  $114 \text{ kg C kg N}^{-1}$  (by 2.6-fold) through a  
705 well-designed mesocosm experiment. It should be noted that the experiment by Nair *et al.* studied the  
706 response to N deposition at a time scale of one year and it is likely that N in the aboveground biomass can  
707 be re-allocated in plants at a longer time scale. This is likely the reason why most of applied N was retained  
708 in the plants at mid-long term (Gaije *et al.*, 2007), but most of the N in the plant was recovered in the bark  
709 rather than in the canopy (Dail *et al.*, 2009).

710 Here we calculated the C response to N deposition based on the fate of deposited N at a timescale of 10-20  
711 years following de Vries *et al.* (2014). We estimated that  $\text{oC/oN}$  was  $24 \text{ kg C kg N}^{-1}$ , which is lower than  
712 the estimate by Nair *et al.* (2016), but close to the estimate ( $11.5\text{-}39.8 \text{ kg C kg N}^{-1}$ ) by de Vries *et al.* (2014),  
713 because Nair *et al.* (2016) did not account for loss of N in the ecosystem via denitrification and leaching.  
714 We increased our estimate of  $\Delta C_{N\text{ dep}}$  by 2.6-fold as indicated by Nair *et al.*'s experiment to yield an upper  
715 estimate of the effect by N. Consequently, maximum  $\Delta C_{v\text{ dep}}$  by N and P increased by 2-fold from  $0.38 \text{ Pg}$   
716  $\text{C yr}^{-1}$  to  $0.76 \text{ Pg C yr}^{-1}$ , compared to  $0.70 \text{ Pg C yr}^{-1}$  by anthropogenic N deposition, while minimum  $\Delta C_{v\text{ dep}}$

717 increased by 1.6-fold from 0.10 to 0.16 Pg C yr<sup>-1</sup>, compared to 0.22 Pg C yr<sup>-1</sup> by anthropogenic P  
718 deposition. This suggests that the effect of P will be less significant if the high C response to N deposition  
719 due to direct use of N by the canopy can be confirmed by more evidences.

#### 720 4.4. Ecological implications of N and P deposition

721 The spatial patterns and temporal trends of N and P emissions and subsequent deposition are important for  
722 understanding the variation of nutrient limitation in forests far away from agricultural activities (Reay *et al.*,  
723 2008). Manipulation experiments across global biomes found that many ecosystems are co-limited by the  
724 availability of N and P (Elser *et al.*, 2007; LeBauer and Treseder, 2008; Penuelas *et al.*, 2013). Fossil fuel,  
725 biofuel and deforestation fires provide additional N and P to forests beyond the background level and these  
726 emitted from the populated regions can reach remote forests by atmospheric transport. Our long-term data  
727 of the deposition for both N and P help to quantify the deposition of N and P as anthropogenic components  
728 and to estimate the additional C storage in forests.

729 Global change in N and P deposition rates arising from N and P emissions by human activities and biomass  
730 burning is only one of factors that influence carbon storage by forests. Our study made a preliminary  
731 attempt to assess the contributions of forest cover change (Peng *et al.*, 2017) and the changes in N and P  
732 deposition rates to the  $\Delta C_{v\ dep}$ . It is not a surprise that forest cover change has offset 10% and 30% of  
733 increase in the  $\Delta C_{v\ dep}$  from 1850 to 2010 due to increase in N and P deposition rates, due to decline in  
734 forest areas mainly in the tropics. In addition to forest cover change, other environmental drivers perturbing  
735 the forest C sequestration from nutrient deposition, such as rising CO<sub>2</sub> levels (Field *et al.*, 1995),  
736 emergency of large-scale drought (Phillips *et al.*, 2009), spring and autumn warming (Piao *et al.*, 2008) and  
737 change in forest water-use efficiency (Keenan *et al.*, 2013), are not considered in this study. A combination  
738 of our global N and P deposition data sets associated with their uncertainties with process-based vegetation  
739 models including both N and P interactions with terrestrial C cycling (*e.g.*, Wang *et al.*, 2010; Goll *et al.*,  
740 2012) would permit a more comprehensive understanding of the impact of atmospheric deposition on the  
741 forest carbon sink.

742 Using a conceptual stoichiometric mass balance approach, we showed that anthropogenic N deposition for  
743 1997-2013 contributed ~9% to the global terrestrial C sink, which is close to a previous estimate of 10% for  
744 2030 (Reay *et al.*, 2008). We emphasized that physical P fixation is an important factor that can lead to an  
745 imbalance between N and P in atmospheric deposition. Anthropogenic P deposition (0.50 Tg P yr<sup>-1</sup>) was  
746 23-fold lower than N deposition (11.5 Tg N yr<sup>-1</sup>) for 1997-2013, but most of the P was stored in soils on a  
747 shorter term and less P than N was prone to loss (5.3 Tg N yr<sup>-1</sup> was lost for N by denitrification or leaching,  
748 and only 0.010 Tg P yr<sup>-1</sup> was lost for P by leaching). Our analysis suggested that historical P deposition  
749 would likely exert a significant cumulative effect on the terrestrial C sink by releasing soil-fixed P in the  
750 long term. Nonetheless, our stoichiometric approach cannot account for the effects of N and P deposition  
751 under elevated CO<sub>2</sub> concentration and varying climate (Gruber and Galloway, 2008; Vitousek *et al.*, 2010).  
752 Comprehensive process-based Earth System ecosystem models representing the biogeochemical cycles of  
753 C, N and P are useful for understanding the ecological effects of historical, present and future depositions

754 of N and P on the C cycle (Reed *et al.*, 2015).

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