*García de Jalón S., Burgess P.J., Curiel Yuste J., Moreno G., Graves A., Palma J.H.N., Crous-Duran J., Kay S., Chiabai A.* 2019. **Dry deposition of air pollutants on trees at regional scale: A case study in the Basque Country.** AGRICULTURAL AND FOREST METEOROLOGY. 278. DOI (10.1016/j.agrformet.2019.107648).

# Dry deposition of air pollutants on trees at regional scale: a case study in the Basque Country

GARCÍA DE JALÓN S[1*], BURGESS PJ[2], CURIEL YUSTE J[1, 6*], MORENO G[3], GRAVES A[2], PALMA JHN[4], CROUS-DURÁN J[4], KAY S[5], CHIABAI A[1]

(1) Basque Centre for Climate Change (BC3), 48940, Leioa, Basque Country, Spain
(2) Cranfield University, Cranfield, Bedfordshire, MK43 0AL, UK
(3) Forestry Research Group, INDEHESA, Universidad de Extremadura, 10600, Plasencia, Spain
(4) Forest Research Centre, University of Lisbon, Tapada da Ajuda, 1349-017 Lisboa, Portugal
(5) Agroscope. Research Division Agroecology and Environment. Reckenholzstrasse 191, CH-8046 Zürich, Switzerland
(6) IKERBASQUE - Basque Foundation for Science, Maria Diaz de Haro 3, 48013, Bilbao, Basque Country, Spain

(*) Corresponding author. E-mail address: Silvestre.garciadejalon@bc3research.org

## Abstract

There is increased interest in the role of trees to reduce air pollution and thereby improve human health and well-being. This study determined the removal of air pollutants by dry deposition of trees across the Basque Country and estimated its annual economic value. A model that calculates the hourly dry deposition of $NO_2$, $O_3$, $SO_2$, CO and $PM_{10}$ on trees at a 1 km x 1 km resolution at a regional scale was developed. The calculated mean annual rates of removal of air pollution across various land uses were 12.9 kg $O_3$ $ha^{-1}$, 12.7 kg $PM_{10}$ $ha^{-1}$, 3.0 kg $NO_2$ $ha^{-1}$, 0.8 kg $SO_2$ $ha^{-1}$ and 0.2 kg CO $ha^{-1}$. The results were then categorised according to land use in order to determine how much each land use category contributed to reducing air pollution and to determine to what extent trees provided pollution reduction benefits to society. Despite not being located in the areas of highest pollutions, coniferous forests, which cover 25% of the land, were calculated to absorb 21% of the air pollution. Compared to other land uses, coniferous forests were particularly effective in removing air pollution because of their high tree cover density and the duration of leaf life-span. The total economic value provided by the trees in reducing these pollutants in terms of health benefits was estimated to be €60 million $yr^{-1}$ which represented around 0.09% of the Gross Domestic Product of the Basque Country in 2016. Whilst most health impacts from air pollution are in urban areas the results indicate that most air pollution is removed in rural areas.

**Keywords:** vegetation, health, pollutant, deposition velocity, land cover

# 1. Introduction

Exposure to air pollution has been associated with increased mortality and morbidity. The World Health Organization (WHO) reported that in 2012, about 7 million people died as a result of fine-particulate air pollution exposure. In the United States, Pope et al. (2009) found that sustained reductions in air pollution exposure were significantly correlated with an increase in life expectancy.

Negative impacts from anthropogenic air pollution emissions can be reduced either by mitigating their health and environmental effects or by reducing emissions. Whilst much discussion has focused on measures that reduce emissions, such as decreasing traffic rates, phasing out old technologies, and increasing the use of public transport, there is great potential in reducing air pollution concentration through the use of vegetation (Vailshery et al., 2013; Escobedo et al., 2011; Guidolotti et al., 2017).

Vegetation can reduce air pollution concentration by dry deposition, leading to improvements in human health and well-being (Mohan, 2016; Sanderson, 2008; Nowak et al., 2006; Nowak et al., 2013; Janhall, 2015; Litschke and Kuttler, 2008). In the dry deposition process, particles and gases are collected or are deposited on solid surfaces and this decreases the concentration in the air. Atmospheric particles and gases that are intercepted by vegetation can be either absorbed into plant tissues or retained on the surface of leaves, twigs, branches and the trunk. Pollutants absorbed by plant tissues can sometimes be turned into organic compounds stimulating the development of the plant (Sanderson, 2008; Lockwood et al., 2008). However most intercepted particles are retained on the plant surface and often drop to the ground with leaf and twig fall, are washed off by rain, or are resuspended in the atmosphere (Nowak et al., 2013). Thus, the retention of atmospheric particles in trees is usually temporary. This paper focuses on dry deposition on the surface of trees and does not assess the processes after deposition such as pollutant uptake or resuspension.

Atmospheric particles can be deposited when they pass close to a surface. Compared with manufactured surfaces, trees have a large surface area per unit volume and a high surface roughness, which increases the probability of deposition (Janhall, 2015). Trees directly affect air quality by removing atmospheric particulate concentration, emitting pollen and volatile organic compounds, and through resuspension of particles captured on the plant surface (Nowak et al., 2013; Freer-Smith et al., 2004; Beckett et al., 2000a). Trees can also affect air quality by changing the microclimate by reducing exposure to solar radiation, modifying the wind, and by buffering air temperatures (Beckett et al., 2000b).

Some studies have questioned the effectiveness of the filtration role of plants in reducing pollution concentrations, arguing that the net reduction by vegetation is not always clear (Ries and Eichhorn, 2001; Litschke and Kuttler, 2008; Gromke and Ruck, 2007). One reason can be because vegetated areas are a barrier to air flow which can reduce air circulation in comparison with non-vegetated areas (Ries and Eichhorn, 2001; Gromke and Ruck, 2007). Thus, the volume of air that is exchanged per unit of time can be lower in vegetated areas than in non-vegetated areas. Litschke and Kuttler (2008) claimed that in order to provide a net reduction of air pollution, the particulate emissions of plants and the reduction in near-surface air exchange must be offset against the filtration performance. The authors gave the case of a road with trees on the roadside where a reduction in air exchange would result in an accumulation of dust and the reduction in pollutant concentration through deposition would be offset by reduced air exchange which would increase levels of pollutant concentration. This argument is mostly applicable to local scale assessments such as road trees or urban areas. However, pollutants will still persist and will eventually disperse elsewhere. For this reason, regional scale assessments of the air pollution removal by trees need to also include the impact of forest and agricultural land.

A further complication in the assessment of the filtration capacity of plants is that a number of factors influence dry deposition. Particle size and shape greatly influence deposition on plant surfaces (Janhall, 2015). Meteorological variables such as precipitation, solar radiation, humidity, wind speed, temperature and turbulence affect deposition velocity and thus the filtration performance of plants (Litschke and Kuttler, 2008). Dry deposition is also affected by plant characteristics such as plant species or planting configuration. Since most particles are deposited on leaves, higher deposition can be expected on evergreen species than on deciduous species since leaves remain on the tree throughout the year (Beckett et al., 2000a; Freer-Smith et al., 2004). Furthermore, cuticular, stomatal and mesophyll resistances of leaves, stems, and other organs directly affect deposition and these vary depending on plant species. Hairiness and wax content have also been reported to increase deposition (Janhall, 2015). Other factors that affect deposition and dispersion are vegetation density and distribution as well as the size and shape of the canopy.

There are several models developed to simulate the dry deposition of air pollutants on trees. The European Monitoring and Evaluation Programme (EMEP) was developed to provide governments with scientific information on the evaluation of international protocols for emission reductions (EMEP, 2018). Within EMEP, several models have been developed. The GAINS/RAINS model was developed to explore synergies and trade-offs between the control of local and regional air pollution and the mitigation of greenhouse gas emissions across various scales. The HM and POP models are chemical transport models that assess the regional atmospheric dispersion and deposition of heavy metals and persistent organic pollutants. The MSC-W chemical transport model assesses atmospheric dispersion and deposition of acidifying and eutrophying compounds, ground level ozone and particulate matter. Since 2017 the spatial resolution of these models has been a $0.1^{o}$ x $0.1^{o}$ longitude-latitude grid. In the United States, the USDA Forest Service developed the UFORE-D and i-Tree models which utilize field-surveyed urban forest information, location specific data, weather data, and air pollutant measurements to quantify urban forest structure and forest-related effects such as quantifying dry deposition of air pollution by trees and shrubs (Hirabayashi et al., 2015).

This study develops a regional scale model for evaluating dry deposition on vegetation. The model is based on the UFORE-D and i-Tree models and uses equations from previous studies (e.g. Baldocchi, 1994; Farquhar et al., 1980). In comparison to previous studies, our model presents some advances for regional scale assessments by land cover through the use of new input variables such as satellite data, population density, road density, or land cover.

This work aims to evaluate air pollution removal by dry deposition of trees in the Basque Country in northern Spain (Figure 1) and to assess air pollution removal by individual land uses. Whilst most studies that have assessed dry deposition of air pollution have focused on urban trees (e.g. Nowak et al., 2006; Nowak et al., 2013; Janhall, 2015), this study presents a regional scale approach for simulating the dry deposition of nitrogen dioxide ($NO_2$), ozone ($O_3$), sulphur dioxide ($SO_2$), carbon monoxide (CO) and particulate matter ($PM_{10}$) on trees. The separate assessment by land cover is used to identify regions and land covers where trees provide the highest benefits to society.

< INSERT FIGURE 1 >

# 2. Material and methods

## 2.1. Materials

This study used the Basque Country as a case study. The Basque Country occupies 7,234 km$^2$ in which the population in 2016 was around 2.2 million people. Bilbao and the surroundings is the largest and the most industrialised metropolitan area in Basque Country (see population density map in Figure 1). Apart from Madrid and Barcelona, the metropolitan area of Bilbao is the most affected by air pollution in Spain (Ibarra-Berastegi et al., 2003; 2008; Gómez et al., 2004).

This study used a range of time dependent and time independent data from diverse sources. Time dependent data included air pollution concentration, weather and leaf area index (LAI). For air pollution concentration and weather variables, this study used data from the monitoring stations of the General Administration of the Autonomous Community of the Basque Country on an hourly basis (Gobierno Vasco, 2017). The air pollution data included atmospheric concentration levels of $NO_2$, $O_3$, $SO_2$, CO and $PM_{10}$ in 2016. Hourly weather data including wind, precipitation, humidity, pressure, solar radiation and temperature were also collected for this period. The data were collected from the online portal of the Basque Country (http://www.euskadi.eus) which had 53 air quality monitoring stations of which 47 stations had hourly data for the studied period (from 1 January 2016 at 00:00 to 31 December 2016 at 23:59). The locations of the monitoring stations are shown in Figure 1. The LAI data were obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS) payload imaging sensor with a 500-m resolution (Myneni et al., 2015). As MODIS provides data on an eight-day basis, 45 maps (raster layers) of LAI in the Basque Country in 2016 were downloaded. The values in each cell of the map were converted to hourly data assuming constant values during eight-day periods.

The time independent data included population density, road density, tree cover, land cover, roughness length and data relating to plant characteristics. Population density data were obtained from the Center for International Earth Science Information Network (CIESIN) (2017) and road density data from OpenStreetMap (2015). Tree cover data were obtained from the raster layer Tree Cover Density 2012 with 20 m resolution from the Copernicus Land Monitoring Service (see Figure 2). Land cover data were obtained from the raster layer CORINE Land Cover (CLC 2012) with 100 m resolution from the Copernicus Land Monitoring Service (European Environment Agency, 2017). The roughness length values used in this study varied according to land use and season and were obtained from previous studies (Brook et al., 1999; EANET, 2010). They are shown in Table S.3. Data relating to the plant characteristics necessary for the calculation of deposition velocity were obtained from previous studies (see Table S.2).

< INSERT FIGURE 2 >

## 2.2. Analytical methods

The method developed for this study aimed to measure the air pollution removal ($NO_2$, $O_3$, $SO_2$, CO and $PM_{10}$) by dry deposition on trees at regional scale. The computational analysis was done using R software (R Development Core Team, 2017).

### *2.2.1. Calculating the downward pollutant flux*

Material deposited per unit ground area

The first step was to calculate the material deposited per unit ground area and time. Typically, this is calculated as the product of deposition velocity and pollutant concentration (Equation 1) (Hicks et al., 1985; Pederson et al., 1995).

$$D_{p,t} = Vd_{p,t} * C_{p,t}, \qquad (1)$$

where $D_{p,t}$ is the deposited amount of pollutant *p* per unit ground area and time instant $t$ (g m$^{-2}$ s$^{-1}$). $Vd_{p,t}$ is the deposition velocity (m s$^{-1}$). As the calculations were made on an hourly basis each time instant represented an hour (3600 s h$^{-1}$). $C_{p,t}$ is the concentration of pollutant *p* (g m$^{-3}$) in every hour.

Deposition velocity

Deposition velocity ($Vd_{p,t}$) is the pollutant removal efficiency due to dry deposition. For $NO_2$, $O_3$, $SO_2$ and CO, deposition velocity was calculated as a function of three main resistances (Hicks et al., 1987; Pederson et al., 1995) (Equation 2):

$$Vd_{p,t} = \frac{1}{Ra_t + Rb_{p,t} + Rc_{p,t}}, \qquad (2)$$

where $Ra_t$ is the aerodynamic resistance, $Rb_{p,t}$ is the quasi-laminar boundary layer resistance and $Rc_{p,t}$ is the canopy resistance (s m$^{-1}$). To limit deposition estimates to periods of dry deposition, $Vd_{p,t}$ was set to zero during periods (hours) of rain (Nowak et al., 2006).

Following Nowak et al. (2006) and Hirabayashi et al. (2015), deposition velocity for $PM_{10}$ ($Vd_{PM_{10},t}$) was set to a constant value during the in-leaf period which could be considered a rough estimate, since deposition velocity depends on the particle size.

Aerodynamic resistance

The aerodynamic resistance ($Ra_t$) is the force exerted by the air on the surface of the plant that is parallel and opposite to the direction of flow relative to the plant. It affects the transport of the pollutant in the atmospheric surface layer towards the surface of the plant (Pederson et al., 1995). Aerodynamic resistance was calculated using a relationship described by Killus et al., (1984) (Equation 3):

$$Ra_t = \frac{u_{t,z}}{{u^*}_t^2}, \qquad (3)$$

where $u_{t,z}$ (m s$^{-1}$) is the mean wind speed at height z at time instant *t* measured on an hourly basis at each monitoring station. When there were no measurements at a selected monitoring station, the data from the closest station were used. The value $u^*{}_t$ is the friction velocity (m s$^{-1}$) at time instant $t$ (see subsection “Friction velocity” below).

### Quasi-laminar boundary layer resistance

The quasi-laminar boundary layer resistance ($Rb_{p,t}$) affects the process of the transport by molecular diffusion across an (intermittently present) thin laminar layer (Hicks et al., 1985) (Equation 4):

$$Rb_{p,t} = 2 * \left(Sc_p\right)^{\frac{2}{3}} * (Pr)^{-\frac{2}{3}} * (k * u^*{}_t)^{-1}, \tag{4}$$

where *Sc* is the Schmidt number, *Pr* is the Prandtl number and $k$ is the von Karman constant.

### Canopy or surface resistance

The canopy or surface resistance ($Rc_{p,t}$) is the net resistance corresponding to the entire surface of the plant and affects the physical capture and chemical reactions. The canopy resistance usually dominates and controls the rate of deposition. (Pederson et al., 1995) (Equation 5):

$$Rc_{p,t} = \frac{1}{\frac{1}{r\_s_t + r\_m_p} + \frac{1}{r\_soil_t} + \frac{1}{r\_t_p}}, \tag{5}$$

where $r\_s_t$ is the stomatal resistance at time *t,* $r\_m_p$ is the mesophyll resistance of each pollutant, $r\_soil_t$ is the soil resistance at time $t$ , and $r\_t_p$ is the cuticular resistance of each pollutant.

The calculation of the stomatal resistance used the analytical solution for coupled leaf photosynthesis developed by Baldocchi (1994). The analytical solution is based on four equations with four unknowns. Despite calculating stomatal resistance the model does not estimate stomata uptake. Fares et al. (2008) found a significant relationship between stomatal conductance and stomata uptake.

Firstly, stomatal conductance, which is the inverse of stomatal resistance, is calculated on an hourly basis using the equation of Ball (1989) (Equation 6).

$$Gs_t = \frac{m * A_t * rh_t}{Cs_t} + b', \tag{6}$$

where $Gs_t$ is the stomatal conductance (µmol m$^{-2}$ s$^{-1}$), $A_t$ is the leaf photosynthesis (µmol m$^{-2}$ s$^{-1}$), $rh_t$ is relative humidity (%), and $Cs_t$ is hourly $CO_2$ concentration at the leaf surface (ppm). The coefficient $m$ is a dimensionless slope and $b'$ is the zero intercept when $A_t$ is equal to or less than zero. Units from µmol m$^{-2}$ s$^{-1}$ were converted to m s$^{-1}$.

The value $A_t$ is calculated using Farquhar et al. (1980) as a function of the carboxylation ($V_c$), oxygenation ($V_o$) and dark respiration ($R_d$) rates of $CO_2$ exchange between the leaf and the atmosphere (Equation 7):

$$A_t = V_c - 0.5 * V_o - R_d \tag{7}$$

Finally, to obtain an analytical solution for leaf photosynthesis, two conductance equations were employed (Equations 8 and 9). See Baldocchi (1994) for more details in the calculation.

$$Ci_t = Cs_t - \frac{A_t}{Gs_t} \tag{8}$$

$$Cs_t = Ca - \frac{A_t}{Gb_t}, \tag{9}$$

where $Ci_t$ is the leaf internal $CO_2$ concentration, $Cs_t$ is the leaf surface $CO_2$ concentration, *Ca* is the atmosphere's $CO_2$ concentration (410 ppm) and $Gb_t$ is the conductance across the laminar boundary layer of the leaf (μmol $m^{-2}$ $s^{-1}$) for $CO_2$ exchange.

In the case of CO, it was considered that pollutant removal by vegetation was not directly related to transpiration (Bidwell and Fraser, 1972). Using Hirabayashi et al. (2015), canopy resistance was set to a constant value depending on the in-leaf and out-of-leaf periods.

### Friction velocity

Friction velocity is a scaling parameter that describes the shear stress and atmospheric turbulence in the boundary layer which affects pollutant dispersion. Friction velocity is needed in order to obtain a vertical wind profile for given atmospheric conditions. Since friction velocity was estimated from hourly-averaged horizontal wind measurements within the roughness sublayer the following approximation was used (Equation 10, Prandtl, 1925):

$$u^*_t = \sqrt{0.2} * u_{t,z}, \tag{10}$$

where $u *_t$ is the friction velocity and $u_{t,z}$ the mean wind speed at height z at the at time *t*.

### Material deposited per unit tree-covered area and time

Finally, the amount of pollutant deposited per unit tree-covered area and time (*DT*) was calculated using Equation 11 (Janhall, 2015):

$$DT_{p,t} = D_{p,t} * LAI_{t,sp} * TC, \tag{11}$$

where $DT_{p,t}$ is the deposited amount per unit tree-covered area of pollutant *p* and time instant *t* (g $m^{-2}$ $s^{-1}$). $D_{p,t}$ was calculated following Equation 1. $LAI_{t,sp}$ is the leaf area index under tree canopy in each time instant ($m^2$ of leaf area per $m^2$ of ground area under the tree canopy). $TC$ is the proportional tree cover between 0 and 1.

Figure 3 shows an example of the calculated air pollutants deposited per unit broadleaf deciduous tree-covered area in 2016 at the "Algorta (Bbizi2)" monitoring station (*Station code = 4*). As shown in the upper graphs, most of the dry deposition was produced during the in-leaf period between April and November. In the lower graphs, we see that deposition is usually higher during early morning.

< INSERT FIGURE 3 >

### *2.2.2. Regional air pollution removal*

In order to estimate air pollution removal at regional scale the material deposited per unit tree-covered area (see Equation 17) was calculated in each cell of a raster layer of the Basque Country with a resolution of 1 km × 1 km. For doing this, each cell needed hourly data (8,784 values in 2016) of the pollutant concentration, weather and LAI variables.

As pollutant concentration and weather data were only collected in selected monitoring stations, the data were spatially interpolated for the rest of the Basque territory. In the case of air pollution concentration ($NO_2$, $O_3$, $SO_2$, CO and $PM_{10}$), the spatial interpolation included two steps. The first step was to identify the closest monitoring station for each raster cell. Previously, the location of each station that had data available was identified separately for each pollutant. Then the distance from each cell to each station was measured. This allowed us to assign the closest station to each cell in the 1 km × 1 km raster layer (see the assignation of the stations to each raster cell in Figure S.1 in the supplementary material). In this way, each monitoring station represented a geographical area in which cells would use the same air pollutant concentration data. The second step was to add to each raster cell an increment of pollutant concentration (positive or negative) based on the density of roads and population of each raster cell. In order to estimate the increment of pollutant concentration, the influence of density of roads and population on the atmospheric concentration of each pollutant was determined by linear regressions.

Equation 12 shows the calculation of the increment of concentration ($\Delta C_{p,(i-st)}$) of pollutant *p* in cell *i* respect to the closest station *st*:

$$C_{p,i} = C_{p,st} + \Delta C_{p,(i-st)}, \tag{12}$$

where $C_{p,st}$ is the concentration of pollutant *p* in station *st* and $C_{p,i}$ in cell *i*.

Equation 13 shows the linear regression (ordinary least squares, OLS) used to measure the effect of density of roads and population on pollutant concentration in the cells where the stations are located (*st*). Equation 14 shows the regression for the raster cells where there were no stations (*i*):

$$C_{p,st} = \beta_0 + \beta_1 * RD_{st} + \beta_2 * PD_{st} + \varepsilon \tag{13}$$

$$C_{p,i} = \beta_0 + \beta_1 * RD_i + \beta_2 * PD_i + \varepsilon \tag{14}$$

where *RD* and *PD* indicate road and population density in each cell.

The increment of pollutant concentration between cell *i* and station *st* ($\Delta_{p,(i-st)}$) was calculated as the difference between the two equations (Equation 15).

$$\Delta C_{p,(i-st)} = C_{p,i} - C_{p,st} = \beta_1 * (RD_i - RD_{st}) + \beta_2 * (PD_i - RD_{st}) \tag{15}$$

As all parameters in Equation 13 are known ($C_{p,st}, RD_{st}$ and $PD_{st}$) the coefficients ($\beta_0, \beta_1$ and $\beta_2$) can be estimated through the OLS regression. Since $RD_i$ and $PD_i$ are also known, $\Delta C_{p,(i-st)}$ can be calculated for each cell in respect to its closest monitoring station through Equation 15. In each cell, this increment was added to the 8,784 hourly values of pollutant concentration.

In the case of weather data (hourly wind, precipitation, humidity, pressure, radiation and temperature), the same principle of using the closest weather station to each raster cell was used for each weather variable.

These steps allowed the calculation of the quantity of pollutant deposited per unit ground area and per unit time ($D_{p,t}$) in each cell of the 1 km × 1 km raster layer (see Equation 1). The hourly data for LAI and the tree cover map were used to calculate the deposited quantity of pollution per unit tree-covered area ($DT_{p,t}$) in each cell of the raster layer (see Equation 11).

### *2.2.3. Assessment by land cover and sensitivity analysis*

The last step was to assess air pollutant removal by land use and to undertake a sensitivity analysis. The raster layer CORINE Land Cover raster layer was used to locate the different land uses in the Basque Country. The CORINE Land Cover map was overlain with the generated maps of air pollutant removal. This then allowed identification of those areas where dry deposition on trees could be important and provided especially high levels of benefits to society.

A sensitivity analysis was conducted to assess the model robustness and to increase the reliability of the model performance. The sensitivity analysis focused on the main sources of uncertainty within the input factors. One source of uncertainty was the representativeness of measurement stations. For example, the southern part of the Basque Country had very few stations, and consequently, there were some raster cells located far away from the stations. The other main source of uncertainty was the fact that meteorological measurements were point measurements and registered at a certain height above the surface which was not the same for all stations. Thus, the aerodynamic resistance due to vegetation was not determined at a constant height. The sensitivity analysis assessed the uncertainty of these two sources.

In order to evaluate the uncertainty of the representativeness of measurement stations, an increment of -50%, -20%, 0%, 20% and 50% of pollutant concentration values were added in those cells that were far away from the stations. To test the uncertainty of point measurements an increment of -50%, -20%, 0%, 20% and 50% of aerodynamic resistance was considered in the analysis.

# 3. Results

## 3.1. Air pollution removal by dry deposition on trees

Figure 4 shows the spatial distribution of annual air pollution removal by dry deposition for each air pollutant in 2016. Mean $NO_2$ deposition in the Basque Country was around 3 kg ha$^{-1}$ yr$^{-1}$ ranging from 0 to 17.1 kg ha$^{-1}$ yr$^{-1}$. Deposition values above the 95 percentile were measured during daylight. A large amount of $NO_2$ deposition was determined for the extensive forests to the south of the metropolitan areas of Bilbao and San Sebastian. This can be explained by the displacement and deposition of $NO_x$ gases generated from roads and densely populated areas in nearby areas. For $O_3$, the highest values were obtained between June and August in areas far away from large cities. Mean $O_3$ deposition was around 12.9 kg ha$^{-1}$ yr$^{-1}$ ranging from 0 to 42.8 kg ha$^{-1}$ yr$^{-1}$. Most $SO_2$ was deposited in the forests surrounding the Bilbao metropolitan area. Mean $SO_2$ deposition was around 0.8 kg ha$^{-1}$ yr$^{-1}$ ranging from 0 to 3.7 kg ha$^{-1}$ yr$^{-1}$. For CO, the highest hourly values were obtained from dusk until dawn between June and November. Mean CO deposition was around 0.2 kg ha$^{-1}$ yr$^{-1}$ ranging from 0 to 0.8 kg ha$^{-1}$ yr$^{-1}$. Similar to $NO_2$, the highest $PM_{10}$ deposition was calculated to occur in the extensive forests to the south of the metropolitan areas of Bilbao and San Sebastian. Mean $PM_{10}$ deposition was around 12.7 kg $ha^{-1}$ $yr^{-1}$ ranging from 0 to 38.5 kg $ha^{-1}$ $yr^{-1}$.

Nowak et al. (2014) calculated the removal of $NO_2$, $O_3$, $SO_2$ and $PM_{2.5}$ in each state of the conterminous United States. In their study, the sum of $NO_2$, $O_3$, $SO_2$ and $PM_{2.5}$ removed by trees ranged from 49.9 kg $ha^{-1}$ $yr^{-1}$ in Maine to 1.2 kg $ha^{-1}$ $yr^{-1}$ in North Dakota. In our study, the calculated mean combined removal of $NO_2$, $O_3$, $SO_2$ and $PM_{10}$ was 29.4 kg $ha^{-1}$ $yr^{-1}$.

The air pollution removal maps show that dry deposition was very low in areas with low tree cover density such as southern Basque Country which is mainly occupied by arable crops and vineyards (see tree cover map in Figure 2). Apart from tree cover density, concentration was the other main driver of air pollution removal. Overall, with the exception of $O_3$, air pollution removal was found to be greater in the surroundings of areas with high industry development and population density which are usually associated with high pollutant concentration levels (Ilan Levy and Broday, 2017; Hao et al., 2018).

< INSERT FIGURE 4 >

Amongst the studied pollutants, $O_3$ showed the greatest reduction through dry deposition on trees (9,325 t of $O_3$ $yr^{-1}$, see Table 1). Regarding the other pollutants, 9,158 t of $PM_{10}$ $yr^{-1}$, 2,192 t of $NO_2$ $yr^{-1}$, 608 t of $SO_2$ $yr^{-1}$ and 174 t of CO $yr^{-1}$ were also removed. Assuming constant externality values from the literature across the Basque Country, the economic value of reducing the concentration of each pollutant was estimated. The externality values transferred in this study were based on the damage cost approach, typically used for evaluating air pollution effects. This approach focuses on the quantification of the explicit impact that the emissions have on human health, environment and economic activity (Ricardo-AEA, 2014). Removal of $PM_{10}$ showed the greatest economic benefit at approximately €34 million $yr^{-1}$. The total economic value of reducing all the pollutants was approximately €60 million $yr^{-1}$, which was about 0.09% of the Gross Domestic Product of Basque Country in 2016.

< INSERT TABLE 1 >

## 3.2. Air pollution removal by land cover

The last step was to analyse air pollution removal by the different land covers in the Basque Country (Table 2). Air pollution removal in the land-cover group “Forest and semi-natural areas” provided about 93.7% of the total removal of all air pollutants which occupies 65.4% of the total area in the Basque Country with a 70.1% mean tree cover. Within the land-use group “Forest and semi-natural areas”, coniferous forest was the land cover that reduced air pollution the most. Whilst coniferous forest on average removed 6.47 kg $NO_2$ $ha^{-1}$ $yr^{-1}$, deciduous forest removed 3.49 kg $NO_2$ $ha^{-1}$ $yr^{-1}$. This was due to all of the main coniferous trees in the Basque Country are evergreen with a significant LAI throughout the year.

After “Forest and semi-natural areas”, the next most effective land cover for removal of air pollution was “Agricultural areas” which removed around 5.8% of the total. The land cover group “Artificial surfaces" then removed around 0.5% of the total. Trees near shorelines and marine coasts (Water bodies group) removed only 0.02% of the total value.

< INSERT TABLE 2 >

The results of the sensitivity analysis (Tables S.5 – S.14 in Supplementary Material) show that the model is sensitive to variations in pollutant concentration values. The spatial distribution of monitoring stations across the Basque Country was not homogenous as there were more stations in densely populated areas. Hence, the representativeness of measurement stations is a limitation that should be considered when interpreting the results of this study. The furthest distance from a station was about 15 km. The results also showed that estimated annual deposition was not strongly affected by the aerodynamic resistance.

# 4. Discussion

There are a number of factors that could explain why air pollution removal due to trees in the Basque Country was high. Firstly, the percentage of tree cover is a key determinant. In the Basque Country, the tree cover density within "forest and semi-natural areas" at around 49% is relatively high (Hansen et al., 2013a; Hansen et al., 2013b; Schuck et al., 2002). Secondly, Basque Country has large industrial areas and the population density of 303 inhabitants km$^{-2}$ is the second highest in the country (Gómez et al., 2004). High population densities, with associated high levels of transportation, and industrialisation lead to high pollution concentrations (Ilan Levy and Broday, 2017; Hao et al., 2018). In turn, high air pollution concentrations can accentuate the deposition of pollutants by trees which is calculated as the product of concentration and deposition velocity (Janhall, 2015). The deposition velocity is mainly determined by meteorological variables and plant parameters. Litschke and Kuttler (2008) suggested that the main meteorological determinants of deposition velocity and the filtration performance of plants were precipitation, wind speed and radiation. Precipitation in the Basque Country is relatively high (around 1,000 mm yr$^{-1}$) which would have an inverse relationship with air pollution removal as dry deposition velocity was set to zero during rain events. Although precipitation contributes to air pollution removal it was considered to be wet deposition and hence, excluded from dry deposition.

The results of this study seem to indicate that the main driver of the air pollution removal by dry deposition and its economic value was the tree cover density and the duration of the LAI. One reason for this is that the effect of tree species on stomata conductance was only considered through the temporal LAI in each raster cell. Overall, the land cover that provided the highest economic benefit was coniferous forests (tree cover density = 83.9%) which is largely occupied by the introduced species *Pinus radiata* D. Don. Coniferous and broad-leaved forests occupy almost half of the total area in the Basque Country but they provide about 83% of the total economic value of air pollution removal. This finding is in line with Nowak et al. (2014) who found that North Dakota (tree cover density = 3%) and Nebraska (tree cover density = 4%) were the states where tree cover density and air pollution removal were lowest, whilst New Hampshire (tree cover density = 89%) and Maine (tree cover density = 83%) had the highest tree cover and level of air pollution removal.

About 94% of the dry deposition on trees in the Basque Country occurred in forest and natural areas, primarily in rural areas. However, as in many other regions, most of the population in the Basque Country is concentrated in urban areas. Therefore, it could be argued that in terms of reducing air pollution concentration by dry deposition, trees in urban areas are likely to be more important than rural trees due to their proximity to people (Nowak et al., 2014). In this respect, the largest benefits could be in areas with the highest population density as the impact on human health would be greater, and it is recommended that the implications of this should be integrated in future research. In line with this finding but at considerably smaller spatial scale, the review on deposition on urban vegetation by Janhall (2015) suggested that vegetation should be close to the pollution source, e.g. low bushes between traffic lanes since proximity to the source increases pollutant concentration and thus deposition. This is supported by our study at regional scale since in most cases, the highest deposition rates were found in extensive forests close to the metropolitan area of Bilbao and to a lesser extent, in San Sebastian (Figure 4). However, it is worth noting that sometimes trees in urban areas can also have negative effects on urban air quality because they can act as a barrier to air flow and emit organic compounds with harmful effects on human health (Ries and Eichhorn, 2001; Gromke and Ruck, 2007; Litschke and Kuttler, 2008).

There are a number of ways in which future research using this method could be developed. Firstly, the calculation of the deposition velocity depends on many parameters obtained from the literature, such as plant resistance and those related to the calculation of deposition velocity derived from experimental and modelling studies. In the literature, there are many discrepancies between these values (Litschke and Kuttler, 2008; Petroff et al., 2008). The use of these different values from the literature can lead to contrasting modelling results, which in turn can have a significant impact on regional estimations. Therefore, there is a need for further research of combining experimental analysis with modelling studies. Secondly, the effect of forest edges (transition zones between an area of woodland and fields or other open spaces) was not considered in the analysis. However, dry deposition is greater at the edge of forests than at the centre and consequently, large forests could be less efficient that forests occurring in patches (Templer et al 2015). Likewise, trees in agroforestry systems could provide greater benefits than in extensive forests since the marginal importance of tree cover seems to decrease as tree cover increases. Thirdly, the parameter values used for the calculation of canopy resistance, for example, for mesophyll or cuticular resistance can differ according to the tree species, individuals and even between leaves on the same tree (Lockwood et al., 2008). However, this study has demonstrated a systematic and transparent method to estimate at regional scale the extent by which trees can reduce pollutant concentration and thereby provide beneficial effects for human health. However, it should be noted that trees can also contribute to air pollution by emitting volatile organic compounds that can contribute to $O_3$ and CO formation (Nowak and Heisler, 2010). In addition to this, some limitations could be associated to the use of plant physiological variables as input data in a regional scale model. Since dry deposition was calculated per raster cell (1 km x 1 km) instead of per tree, the effect of tree species on the calculation of some variables such as stomata resistance was not fully considered. Conversely, our model calculates stomatal conductance using temporal LAI data from the MODIS satellite which is affected by tree species. For instance, in a determined raster cell and time instant, if LAI of a particular species equals zero, then there is no stomata opening activity in that particular cell and time instant. Despite these limitations, the methodology allows the impact of regional air pollution removal by dry deposition of trees in different land covers to be calculated using an approach that could be replicated in other areas.

# 5. Conclusion

This study calculated the spatial distribution of air pollution removal by dry deposition of trees in the Basque Country and estimated its economic value. In doing so, a regional scale model calculating the hourly deposited amount of $NO_2$, $O_3$, $SO_2$, CO and $PM_{10}$ on trees was presented. Although the methodology has limitations, it provides an estimate of the removal by trees of different forms of air pollution in the Basque Country. The proposed methodology could be used in different regions or contexts as long as representative hourly weather and air pollution concentration data are available. The estimated annual dry deposition of pollutants by trees in the Basque Country in 2016 was calculated to be 9,325 t $O_3$, 9,158 t $PM_{10}$, 2,192 t $NO_2$, 608 t $SO_2$ and 174 t CO. The estimated total economic benefit of reducing these pollutants was around €60 million $yr^{-1}$ which represented around 0.09% of the Gross Domestic Product of the Basque Country in 2016. Coniferous forests, which occupy 25% of the area, were found to provide the most of the economic benefit from dry deposition as tree cover density and the duration of leaf life-span were important determinants of the amount of the deposited material.  Although the greatest health impacts from air pollution occur in urban areas where population density is highest, most air pollution is removed in rural areas. To this end, the hourly modelling approach presented here, using air pollution, weather, and leaf area index data collected in monitoring stations and by satellites provides an objective and transparent means of estimating air pollution benefits by trees.

# Acknowledgements

Silvestre García de Jalón, Jorge Curiel Yuste and Aline Chiabai acknowledge support from the Basque Government through the BERC 2018-2021 program and from the Spanish Ministry of Science, Innovation and Universities through the BC3 María de Maeztu excellence accreditation (MDM-2017-0714).

# References

Baldocchi, D., 1994. An analytical solution for coupled leaf photosynthesis and stomata conductance models. Tree Physiol. 14, 1069-1079.

Ball, J.T., 1988. An analysis of stomata1 conductance. Ph.D. Thesis, Stanford University, CA, 89 p.

Beckett, K.P., Freer, P.H., Taylor, G., 2000a. Effective tree species for local air-quality management. Journal of Arboriculture 26 (1), 12-19.

Beckett, K.P., Freer-Smith, P.H., Taylor, G., 2000b. Particulate pollution capture by urban trees: effect of species and windspeed. Glob. Change Biol. 6, 995-1003.

Bidwell, R.G.S., Fraser, D.D.E., 1972. Carbon monoxide uptake and metabolism by leaves. Can. J. Bot. 50, 1435-1439.

Brook, J., Zhang, L., Digiovanni, F., Padro, J. (1999) Modelling of deposition velocities for routine estimates of dry deposition across N.A. Part I. Model development. Atmospheric Environment, 33, 5037–5051.

Center for International Earth Science Information Network, (2017). Gridded Population of the World, Version 4 (GPWv4): Population Density, Revision 10. Palisades, NY: NASA Socioeconomic Data and Applications Center (SEDAC). Columbia University, USA. https://doi.org/10.7927/H4DZ068D.

EANET (2010). Technical Manual on Dry Deposition Flux Estimation in East Asia. Acid Deposition Monitoring Network in East Asia. Available at: http://www.eanet.asia/product/manual/techdry.pdf.

EMEP, 2018. EMEP Status Report 1/2018 Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. Joint MSC-W & CCC & CEIP Report. Available at: http://emep.int/publ/reports/2018/EMEP_Status_Report_1_2018.pdf.

Escobedo, F.J., Kroeger, T., Wagner, J.E., 2011. Urban forests and pollution mitigation: analyzing ecosystem services and disservices. Environ. Pollut. 159, 2078–2087.

European Environment Agency, 2017. Copernicus Land Service - Pan-European Component: CORINE Land Cover. Available at: http://land.copernicus.eu/pan-european/corine-land-cover/clc-2012?tab=download.

Fares, S., Loreto, F., Kleist, E., Wildt, J., 2008. Stomatal uptake and stomatal deposition of ozone in isoprene and monoterpene emitting plants. Plant Biol (Stuttg) 10(1), 44-54.

Farquhar, G.D., von Caemmerer, S., Berry, J.A., 1980. A Biochemical Model of Photosynthetic CO 2 Assimilation in Leaves of C 3 Species. Planta 149, 78-90, Planta.

Freer-Smith, P.H., El-Khatib, A.A., Taylor, G., 2004. Capture of particulate pollution by trees: a comparison of species typical of semi-arid areas (Ficus nitida and Eucalyptus globulus) with European and North American species. Water Air Soil Poll. 155, 173-187.

Gobierno Vasco, 2017. Open Data Euskadi. Portal de acceso a los datos públicos del Gobierno Vasco. Available at: http://opendata.euskadi.eus/.

Gómez, M.C., Durana, N., Navazo, M., Alonso, L., García, J.A., Ilardia, J.L., 2004. Application of validation data tests from an on-line volatile organic compound analyser to the detection of air pollution episodes in urban areas. Anal. Chim. Acta 524, 41–49

Gromke, C., B. Ruck, 2007. Influence of trees on the dispersion of pollutants in an urban street canyon – Experimental investigation of the flow and concentration field. Atmos. Environ. 41, 3287–3302.

Guidolotti, G., Calfapietra, C., Pallozzi, E., De Simoni, G., Esposito, R., Mattioni, M., Nicolini, G., Matteucci, G., Brugnoli, E., 2017. Promoting the potential of flux-measuring stations in urban parks: An innovative case study in Naples, Italy. Agric. For. Meteorol. 233, 1–10.

Hansen, M.C., Potapov, P.V., Moore, R., Hancher, M., Turubanova, S.A., Tyukavina, A., Thau, D., Stehman, S.V., Goetz, S.J., Loveland, T.R., Kommareddy, A., Egorov, A., Chini, L., Justice, C.O., Townshend, J.R.G., 2013a. High-Resolution Global Maps of 21st-Century Forest Cover Change. Science 342, 850–53. Data available online from: http://earthenginepartners.appspot.com/science-2013-global-forest.

Hansen, M.C., Potapov, P.V., Moore, R., Hancher, M., Turubanova, S.A., Tyukavina, A., Thau, D., Stehman, S.V., Goetz, S.J., Loveland, T.R., Kommareddy, A., Egorov, A., Chini, L., Justice, C.O., Townshend, J.R.G., 2013b. Hansen/UMD/Google/USGS/NASA Tree Cover and Tree cover Loss and Gain, Country Profiles. University of Maryland, Google, USGS, and NASA. Accessed through Global Forest Watch on [date]. Available at: www.globalforestwatch.org.

Hao, Y., Peng, H., Temulun, T., Liu, L.-Q., Mao, J., Lu, Z.-N., Chen, H., 2018. How harmful is air pollution to economic development? New evidence from PM2.5 concentrations of Chinese cities. J. Clean. Prod. 172, 743-757.

Hicks B.D., Baldochi D.D., Hosker R.P., Hutchison B.A., Matt D.R., McMillen R.T., Satterfield L.C., 1985. On the use of monitored air concentrations to infer dry deposition. NOAA Technical Memorandum ERL ARL-141. Air Resosurces Laboratory, Silver Springs, MD.

Hicks, B.B., Baldocchi, D.D., Meyers, T.P., Hosker, R.P., Matt, D.R., 1987. A preliminary multiple resistance routine for deriving dry deposition velocities from measured quantities. Water, Air, and Soil Pollution 36, 311-330.

Hirabayashi, S., Kroll, C.N., Nowak, D.J., 2015. i-Tree Eco Dry Deposition Model Descriptions. i-Tree Eco model. Available at: https://www.itreetools.org/eco/resources/iTree_Eco_Dry_Deposition_Model_Descriptions.pdf.

Hosker Jr., R.P., and Lindberg, S.E., 1982. Review: Atmospheric deposition and plant assimilation of gases and particles. Atmos. Environ. 16-(5), 889-910.

Ibarra-Berastegi, G., Elias, A., Agirre, E., Uria, J., 2003. Traffic congestion and ozone precursor emissions in Bilbao (Spain). Environ. Sci. Pollut. R. 10, 361-367.

Ibarra-Berastegi, G., Elias, A., Barona, A., Saenz, J., Ezcurra, A., Diaz de Argandoña, J., 2008. From diagnosis to prognosis for forecasting air pollution using neural networks: Air pollution monitoring in Bilbao. Environ. Modell. Softw. 23, 622-637.

Ilan Levy, Y., Broday, D.M., 2017. Improving modeled air pollution concentration maps by residual interpolation. Sci. Total Environ. 598, 780–788.

Janhall, S., 2015. Review on urban vegetation and particle air pollution e Deposition and dispersion Atmos. Environ. 105, 130-137.

Killus, J.P., Meyer, J.P., Durran, G.E., Anderson, G.E., Jerskey, T.N., 1984. Continued research in mesoscale air pollution simulation modeling. Volume 5: refinements in numerical analysis, transport, chemistry, and pollutant removal. Publ. EPA/600/3.84/0954. Research Triangle Park, NC: United States Environmental Protection Agency.

Litschke, T., Kuttler, W., 2008. On the reduction of urban particle concentration by vegetation; a review. Meteorol. Z. 17, 229-240.

Lockwood, A. L., Filley, T. R., Rhodes, D. Shepson, P. B., 2008. Foliar uptake of atmospheric organic nitrates. Geophys. Res. Lett. 35, L15809.

Lovett, G.M., 1994. Atmospheric deposition of nutrients and pollutants in North America: an ecological perspective. Ecol. Appl. 4 (4), 629-650.

Mohan, S.M., 2016. An overview of particulate dry deposition: measuring methods, deposition velocity and controlling factors. Int. J. Environ. Sci. Te. 13(1), 387-402.

Murray, F.J., Marsh, L., Bradford, P.A., 1994. New York State energy plan, Vol. II: issue reports. Albany, NY: New York State Energy Office.

Myneni, R., Knyazikhin, Y., Park, T. (2015). MOD15A2H MODIS Leaf Area Index/FPAR 8-Day L4 Global 500m SIN Grid V006. NASA EOSDIS Land Processes DAAC. http://doi.org/10.5067/MODIS/MOD15A2H.006 (Terra).

Nowak, D.J., and Heisler, G.M., 2010. Air Quality Effects of Urban Trees and Parks. Research Series. National Recreation and Park Association. Available at: http://www.nrpa.org/uploadedFiles/nrpa.org/Publications_and_Research/Research/Papers/Nowak-Heisler-Summary.pdf.

Nowak, D.J., Crane, D.E., Stevens, J.C., 2006. Air pollution removal by urban trees and shrubs in the United States. Urban For. Urban Gree. 4, 115-123.

Nowak, D.J., Hirabayashi, S., Bodine, A., Greenfield, E., 2014. Tree and forest effects on air quality and human health in the United States. Environ. Pollut. 193, 119-129.

Nowak, D.J., Hirabayashi, S., Bodine, A., Hoehn, R., 2013. Modeled PM2.5 removal by trees in ten U.S. cities and associated health effects. Environ. Pollut. 178, 395-402.

OpenStreetMap, (2015). OpenStreetMap, OpenStreetMap Foundation (OSMF). Available at: https://www.openstreetmap.org/.

Pederson, J.R., Massman, W.J., Mahrt, L., Delany A., Oncley, S., Den Hartog, G., Neumann H.H., Mickle, R.E., Shaw R.H., Paw, K.T., Grantz, D.A., Macpherson, J.I., Desjardins, R., Schuepp, P.H., Pearson, R., Arcado, T.E., 1995. Cailifornia ozone deposition experiment: methods, results, and opportunities. Atmos. Environ. 29 (21), 3115-3132.

Petroff, A., Mailliat, A., Amielha, M., Anselmeta, F., 2008. Aerosol dry deposition on vegetative canopies. Part I: Review of present knowledge. Atmos. Environ. 42, 3625–3653.

Pope, C.A., Ezzati, M., Dockery, D.W., 2009. Fine-particulate air pollution and life expectancy in the United States. New Engl. J. Med. 360 (4), 376–386.

Prandtl, L., 1925. Bericht uber Untersuchungen zur ausgebildeten Turbulenz, Zs. angew. Math. Mech., 5, 136–139.

R Development Core Team, 2017. R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. Available at: http://www.R-project.org.

Ricardo-AEA, 2014. Update of the Handbook on. External Costs of Transport. Final Report. Report for the European Commission: DG MOVE. Ricardo-AEA/R/ ED57769. Issue Number 1. Available at: https://ec.europa.eu/transport/sites/transport/files/handbook_on_external_costs_of_transport_2014_0.pdf.

Ries, K., Eichhorn, J., 2001. Simulation of effects of vegetation on the dispersion of pollutants in street canyons. Meteorol. Z. 10 (4), 229–233.

RWDI, 2006. South Fraser Perimeter Road Regional Air Quality Assessment: Technical Volume 16 of the Environmental Assessment Application. BC Ministry of Transportation. Available at: www.gov.bc.ca/tran/.

Sanderson, K., 2008. Trees eat pollution products. Nature. doi:10.1038/news.2008.1046.

Schuck, A., Van Brusselen, J., Päivinen, R., Häme, T., Kennedy, P., Folving, S., 2002. Compilation of a Calibrated European Forest Map Derived from NOAA-AVHRR Data. Internal Report 13.

European Forest Institute, Joensuu, Finland, 44 p. + Annexes. Available at: http://www.efi.int/files/attachments/publications/ir_13_annexes.pdf.

Templer, P.H., Weathers, K.C., Ewing, H.A., Dawson, T.E., Mambelli, S., Lindsey, A.M., Webb, J., Boukili, V.K., Firestone, M.K. (2015). Fog as a source of nitrogen for redwood trees: evidence from fluxes and stable isotopes. J. Ecol. 103(6), 1397-1407.

Vailshery, L.S., Jaganmohan, M., Nagendra, H., 2013. Effect of street trees on microclimate and air pollution in a tropical city. Urban For. Urban Gree. 12, 408–415.

Wesely, M.L., 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. Atmos. Environ. 23 (6), 1293-1304.