5

ATMOSPHERIC DEPOSITION IN EUROPEAN FORESTS IN 2019

Aldo Marchetto, Peter Waldner, Arne Verstraeten, Daniel Žlindra

Summary

Studying the effects of atmospheric pollution on forest ecosystems requires an evaluation of air quality and of the amount of pollutants carried to the forests by atmospheric deposition. Pollutant flux towards ecosystems through deposition mainly follows two pathways: wet deposition of compounds dissolved in rain, snow, sleet or similar, and dry deposition of particulate matter through gravity or adsorption on forest canopy for example.

Pollutant deposition shows a relatively high local variability, related to the distribution of pollutant sources and the local topography, and *in-situ* measurement is needed to obtain accurate evaluations and to validate model estimates.

In 2019, the chemical composition of atmospheric deposition under the tree canopy was measured in 290 ICP Forests Level II permanent plots throughout Europe. In this report, we focus on acidifying, buffering, and eutrophying compounds in canopy throughfall deposition.

High values of nitrate deposition were mainly found in central Europe (Germany, Denmark, Belgium, Czechia, Switzerland, and eastern Austria), while for ammonium they were also found in northern Italy and Poland. While most of central Europe receives a moderate amount of sulphate deposition, high values are mainly found close to the largest point sources. In the southern part of Europe, sulphate deposition is also influenced by volcanic emission and by the episodic deposition of Saharan dust. The influence of marine aerosols was relevant at sites in coastal areas.

Calcium and magnesium deposition can buffer the acidifying effect of atmospheric deposition. High values of calcium deposition are reported in southern Europe, mainly related to the deposition of Saharan dust, and in eastern Europe. The correction for the marine contribution of calcium matters mainly for sites in central Europe and in Spain. In the case of magnesium, however, the distribution of the highest values is markedly reduced by the sea salt correction.

Introduction

The atmosphere contains a large number of substances of natural and anthropogenic origin. A large part of them can settle, or be adsorbed to receptor surfaces, or be included in rain and snow and finally reach land surface as wet and dry deposition.

In the last century, human activities led to a dramatic increase in the deposition of nitrogen and sulphur compounds.

Sulphur deposition almost exclusively occurs in the form of sulphate (SO_4^{2-}), derived from marine aerosol and from sulphuric acid formed in the atmosphere by the interaction of gaseous sulphur dioxide (SO_2) with water.

Sulphur dioxide emission derives from coal and fuel combustion, volcanoes, and forest fires and has increased since the 1850s, causing an increase in the deposition of sulphate and in deposition acidity, which can be partly buffered by the deposition of base cations, mainly calcium (Ca^{2+}) and magnesium (Mg^{2+}).

Natural sources of nitrogen (N) in the atmosphere are mainly restricted to the emission of N_2O and N_2 during denitrification and the decomposition of the nitrogen gas molecule in the air during lightning. However, human activities cause the emission of large amounts of nitrogen oxides (NO_x), released during combustions, and of ammonia (NH_3) deriving from agriculture and farming. They are found in atmospheric deposition in the form of nitrate (NO_3^-) and ammonium (NH_4^+).

Nitrogen compounds have two effects on the ecosystems: They are important plant nutrients that can produce ecosystem eutrophication, and both have strong effects on plant metabolism (e.g., Silva et al. 2015), forest ecosystem processes (e.g. Meunier et al. 2016) and biodiversity (e.g., Bobbink et al. 2010), but they can also act as acidifying compounds (Bobbink and Hettelingh 2011).

Emission and deposition of nitrate and ammonium are recently decreasing, but the trend is less evident than for sulphate (Waldner et al. 2014; EEA 2016).

Materials and methods

Atmospheric deposition is collected on the ICP Forests permanent plots under the tree canopy (throughfall samplers, Fig. 5-1a) and in a nearby clearance (open field samplers, Fig. 5-1b,c). Throughfall samples are used to estimate wet deposition, i.e. the amount of pollutants carried out by rain and snow, but they also include dry deposition from particulate matter collected by the canopy. The total deposition to a forest, however, also includes nitrogen taken up by leaves directly or organic nitrogen compounds. It can be estimated by applying canopy exchange models.

It is important to note the different behaviour of individual ions when they interact with the canopy: In the case of sulphate, calcium and magnesium, the interaction is almost negligible and it can be assumed that throughfall deposition includes the sum of wet and dry deposition.

Other ions, such as nitrate and ammonium, interact with the tree canopy and the associated microbial communities. For example, tree leaves can uptake ammonium ions and release potassium (K+) ions and organic compounds. Certain microorganisms of the phyllosphere can convert ammonium into nitrate through a process called canopy nitrification (Guerrieri et al. 2015). These canopy interactions strongly affect the composition of throughfall deposition.

Sampling, analysis and quality control procedures are harmonized on the basis of the ICP Forests Manual (Clarke et al. 2016). Quality control and assurance include laboratory ring-tests, use of control charts and performing conductivity and ion balance checks on all samples (König et al. 2010). In calculating ion balance, the charge of organic compounds was considered proportional to the dissolved organic carbon (DOC) content following Mosello et al. (2005, 2008).

In this report, we consider the 2019 yearly throughfall deposition, collected on 290 permanent plots and following the ICP Forests Manual.

Eight plots were excluded because the duration of sampling covered less than 90% (329 days) of the year, and 65 other plots were marked as "not validated" because the conductivity check was passed for less than 30% of the analysis of the year. Other plots were also marked as the laboratory did not participate in the mandatory Working Ring Test, or did not pass the minimum requirement. This applied to 2 plots for sulphate, 9 for nitrate, 16 for ammonium, 8 for calcium, and 5 for magnesium.

As the deposition of marine aerosol represents an important contribution to the total deposition of sulphate, calcium and magnesium, a sea-salt correction was applied, subtracting from the deposition fluxes the marine contribution, calculated as a fraction of the chloride (Cl^-) deposition according to the ICP Integrated Monitoring Manual (FEI 2013). Three more plots were marked for sea-salt corrected ions as the laboratory did not pass the test for chloride.

Results

The uneven distribution of emission sources and receptors and the complex orography of parts of Europe results in a marked spatial variability of atmospheric deposition. However, on a broader scale, regional patterns in deposition arise. In the case of nitrate, high and moderate throughfall deposition was mainly found in central Europe, including Germany, Czechia, Poland, Austria, Italy, Slovenia and Belgium, but single plots with high deposition values are also reported in other countries (Fig. 5-2).

The central European area of high and moderate ammonium throughfall deposition is larger than for nitrate, with higher throughfall deposition values particularly in Germany, Belgium, and northern Italy, western Slovakia and Poland (Fig. 5-3).

It is generally considered that negative effects of nitrogen deposition on forests become evident when inorganic nitrogen deposition (i.e. the sum of nitrate and ammonium deposition) is higher than a specific threshold, known as the critical load. Critical loads can be evaluated for each site by modeling, but more generic critical loads (empirical critical loads) are also being evaluated, ranging between 10 and 25 kg ha^{-1} y^{-1} (Bobbink and Hettelingh, 2011). In 2019, throughfall inorganic nitrogen deposition higher than 10 kg ha^{-1} y^{-1} were mainly measured in central Europe, including Germany, Belgium, northern Italy, Switzerland, Austria, and Czechia (Fig. 5.4). Total deposition of nitrogen is typically a factor 1 to 2 higher than (below canopy) throughfall deposition, due to nitrogen being taken up by tree leaves in the canopy.

The area with high and moderate throughfall deposition of sulphate is smaller than for the nitrogen compounds (Fig. 5-5): High values are mainly found close to the largest point sources. In the southern part of Europe, sulphate deposition is also influenced by volcanic emission and by the episodic deposition of Saharan dust. The area of moderate deposition extends to most of central Europe from Belgium to Bulgaria. The influence of marine aerosols was relevant at sites in coastal areas, where the correction for sea-salt contribution led to low throughfall deposition values, without relevant alterations in the pattern described above (Fig. 5.6).

Figure 5-1: Throughfall (a) and open-field (b, c) collectors for rain in summer (a) and for snow in winter (b) in the northern Alps, Switzerland (Image: WSL)

EVALUATIONS

Calcium and magnesium are also analysed in the ICP Forests deposition monitoring network, as their deposition can buffer the acidifying effect of atmospheric deposition, protecting soil from acidification. High values of calcium throughfall deposition are mostly reported in central and southern Europe (Fig. 5-7). The correction for the marine contribution was less relevant than in previous years. (Fig. 5-8): High sea-salt corrected calcium deposition is mainly found in southern Europe (Spain, Italy, Slovenia, Romania, and Greece) where the influence of wind-blown Saharan dust is remarkable.

On the contrary, in the case of magnesium, the distribution of the highest values, including a large portion of southern and central Europe (Fig. 5-9), is markedly reduced by the sea salt correction (Fig. 5-10).

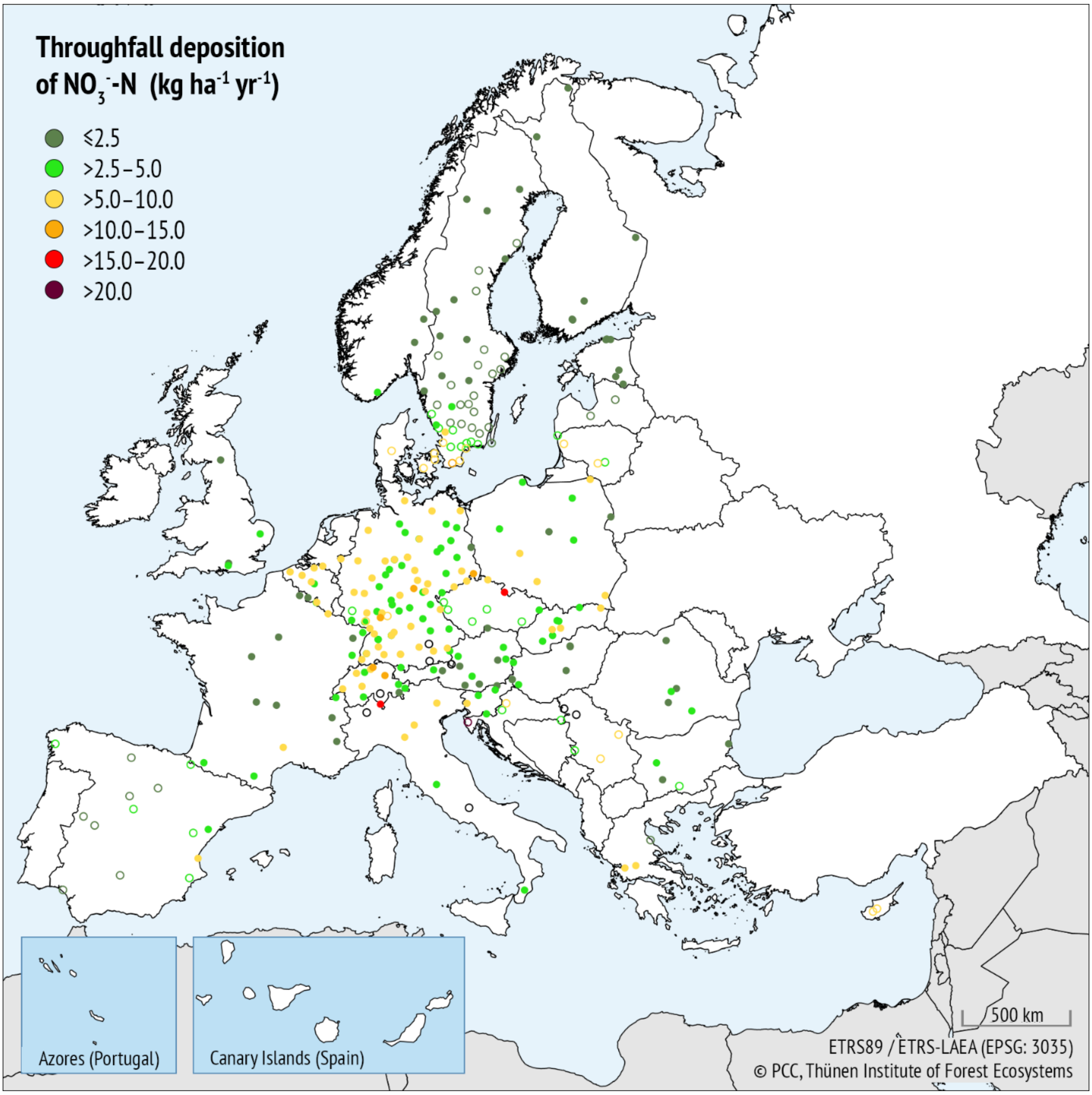

Figure 5-2: Throughfall deposition of nitrate-nitrogen (kg NO_3^--N ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days.

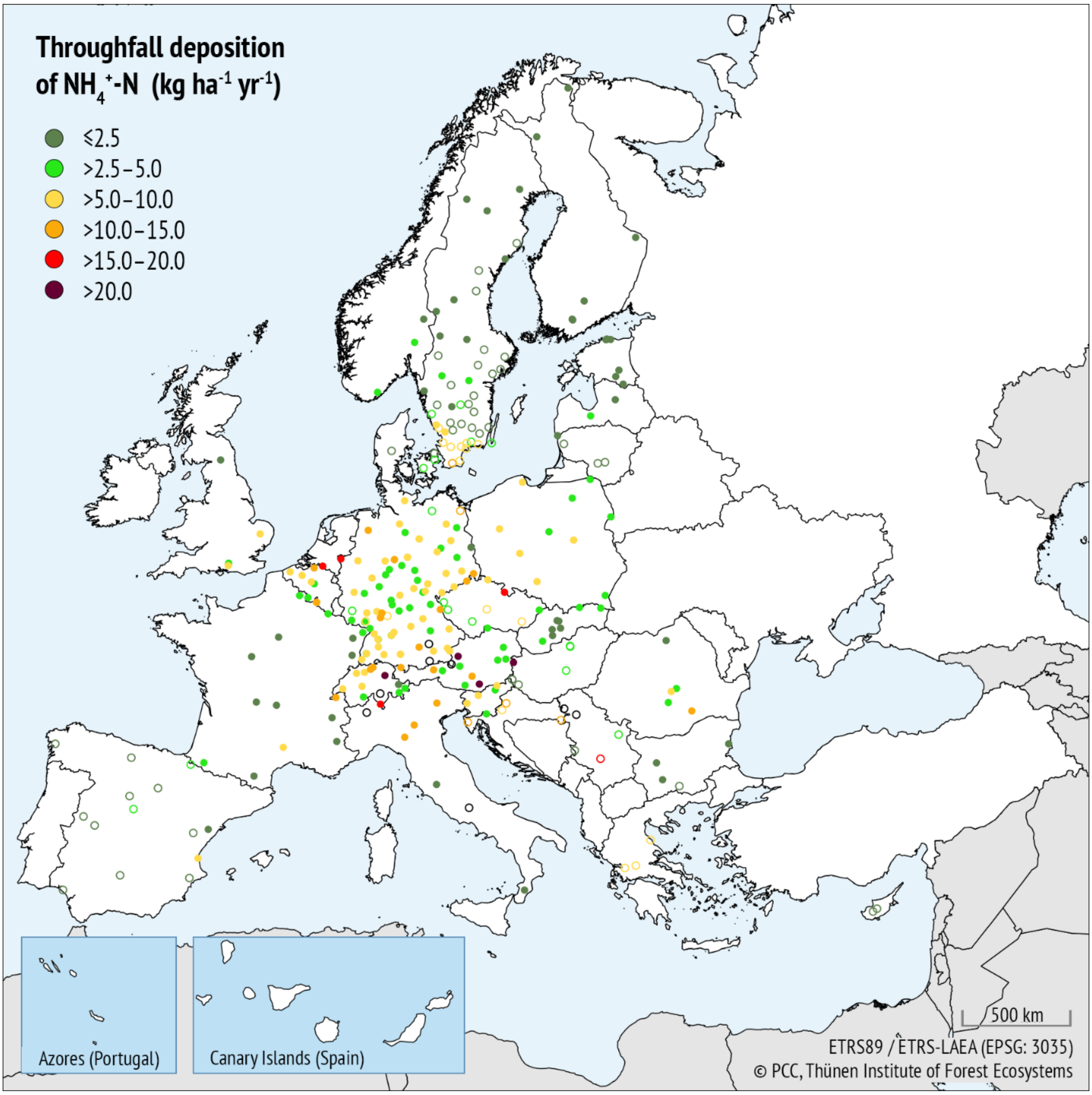

Figure 5-3: Throughfall deposition of ammonium-nitrogen (kg NH_4^+-N ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days.

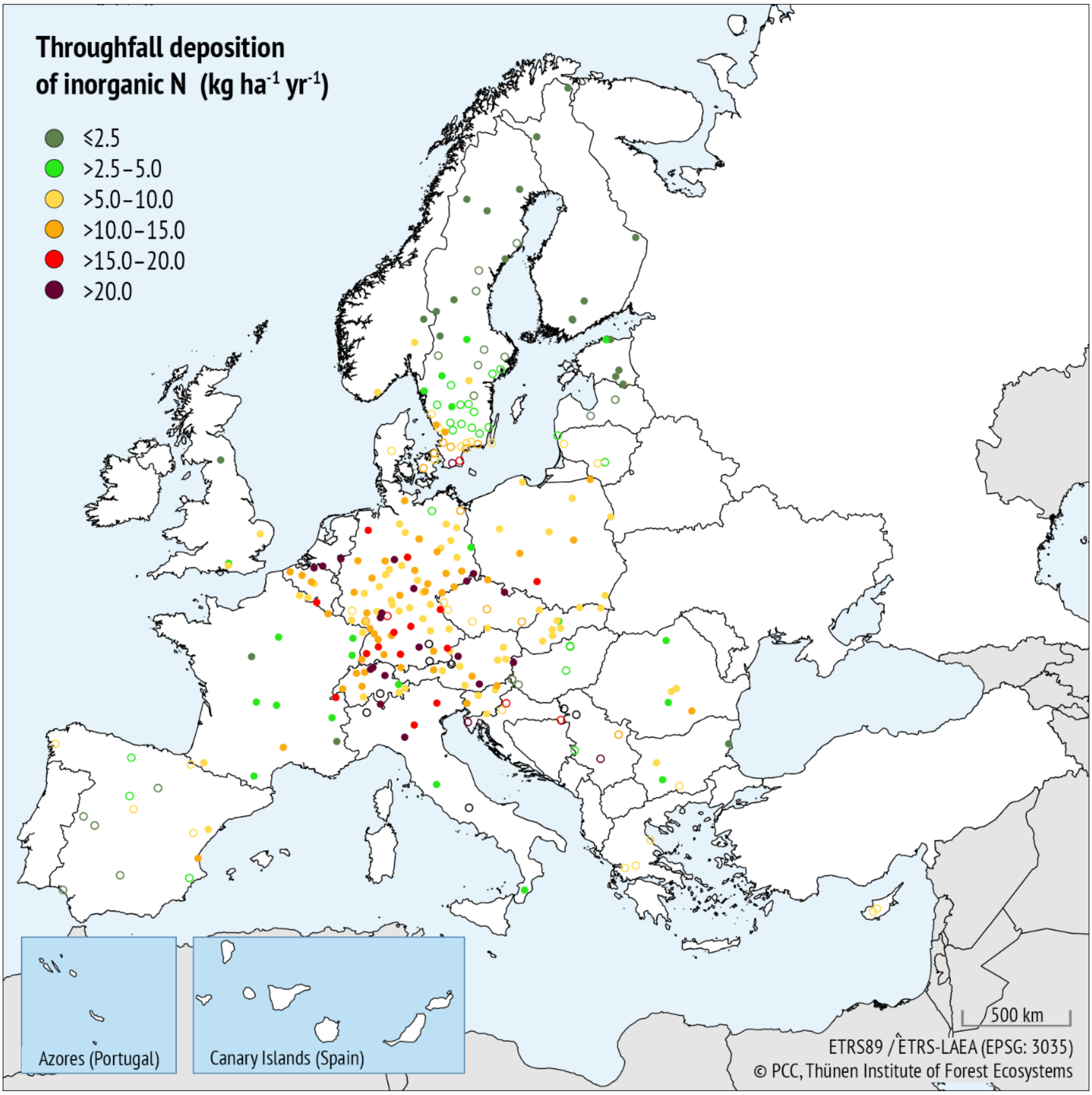

Figure 5-4: Throughfall deposition of inorganic nitrogen (NO_3^--N + NH_4^+-N) (kg N ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days.

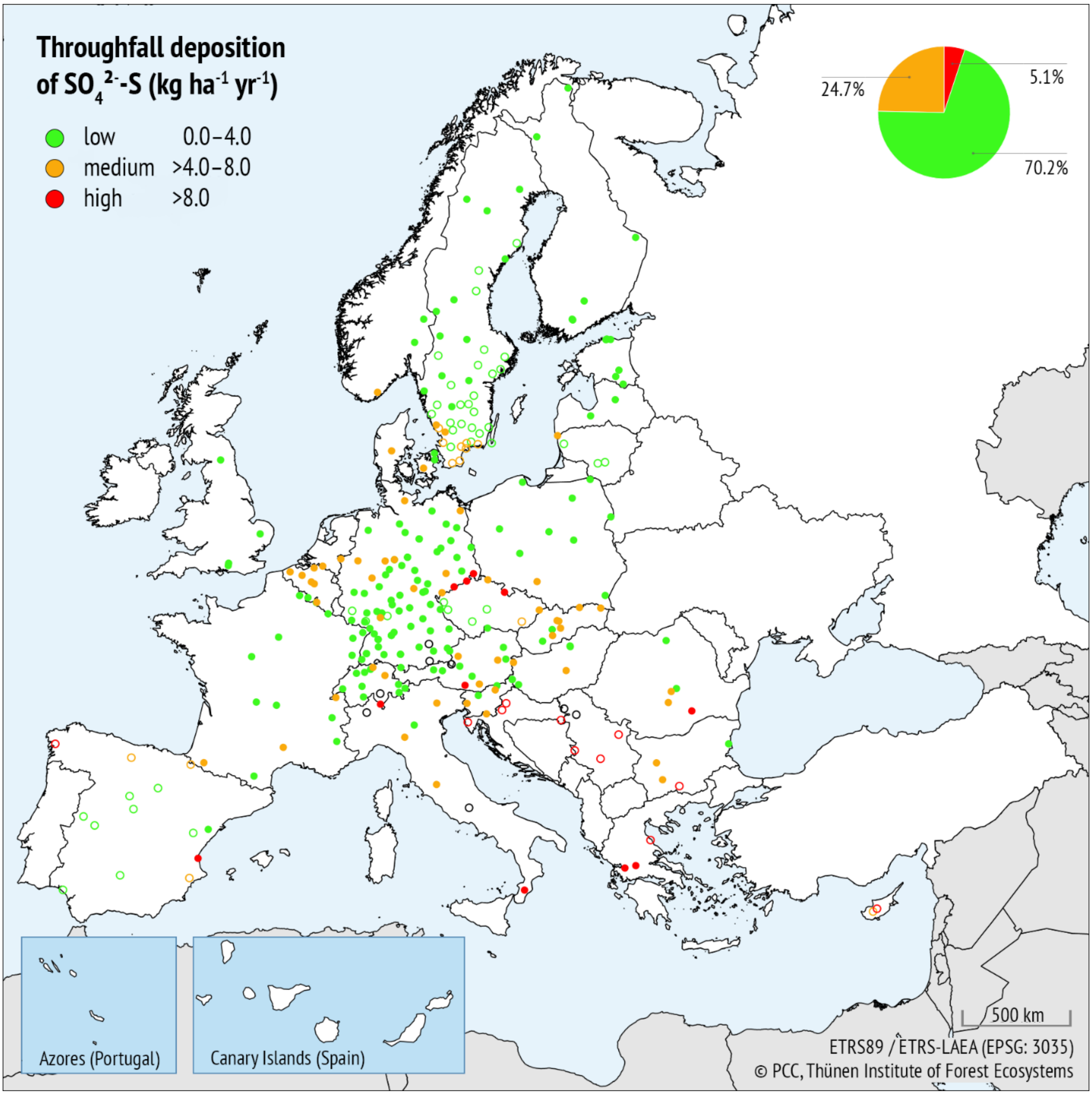

Figure 5-5: Throughfall deposition of sulphate-sulphur (kg SO_4^{2-}-S ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days. Legend: low (green, 0.0–4.0 kg SO_4^{2-}-S ha^{-1} yr^{-1}), medium (yellow, >4.0–8.0 kg SO_4^{2-}-S ha^{-1} yr^{-1}), high (red, >8.0 kg SO_4^{2-}-S ha^{-1} yr^{-1}).

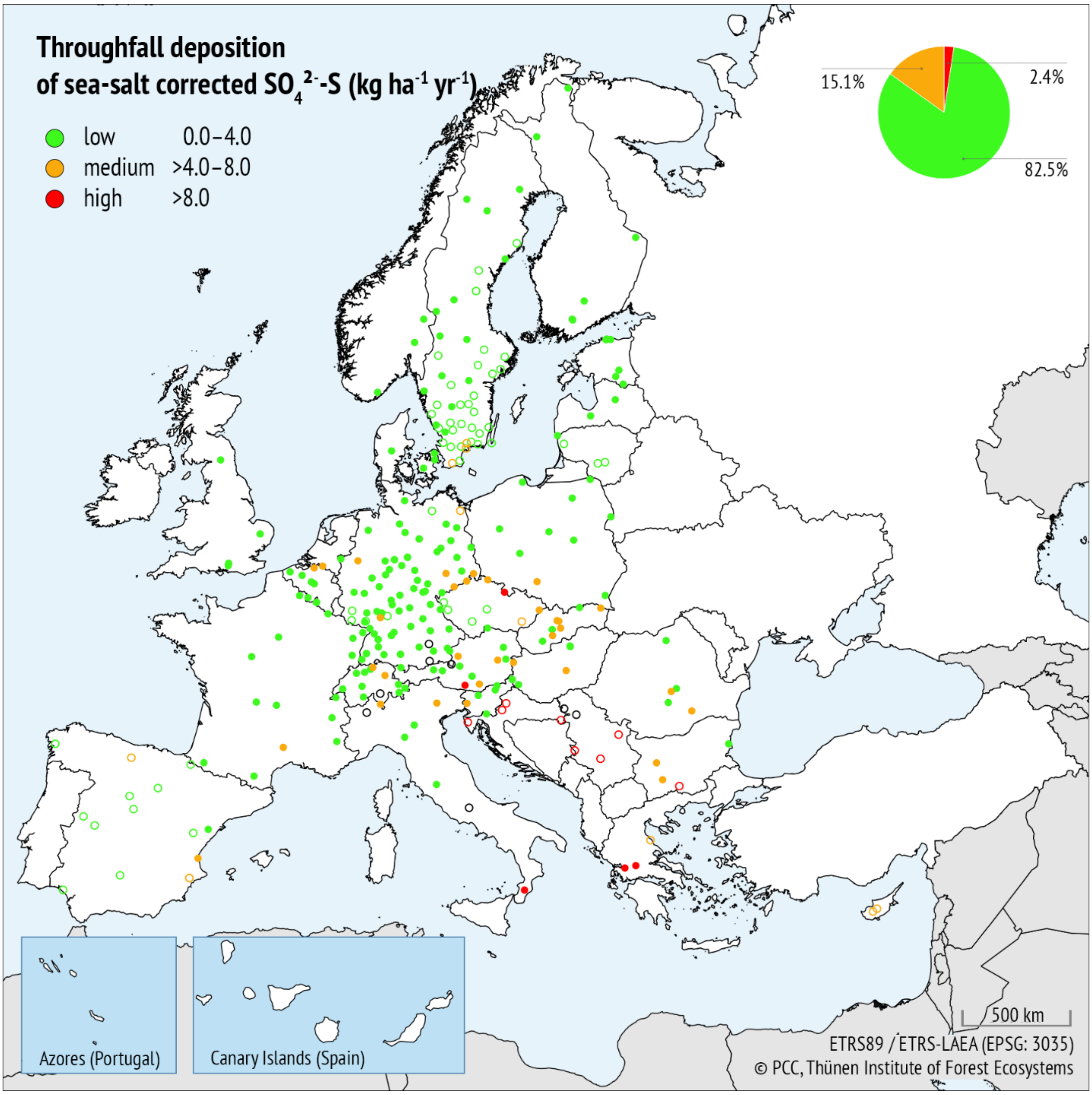

Figure 5-6: Throughfall deposition of sea-salt corrected sulphate-sulphur (kg SO_4^{2-}-S ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days. Legend: low (green, 0.0–4.0 kg SO_4^{2-}-S ha^{-1} yr^{-1}), medium (yellow, >4.0–8.0 kg SO_4^{2-}-S ha^{-1} yr^{-1}), high (red, >8.0 kg SO_4^{2-}-S ha^{-1} yr^{-1}).

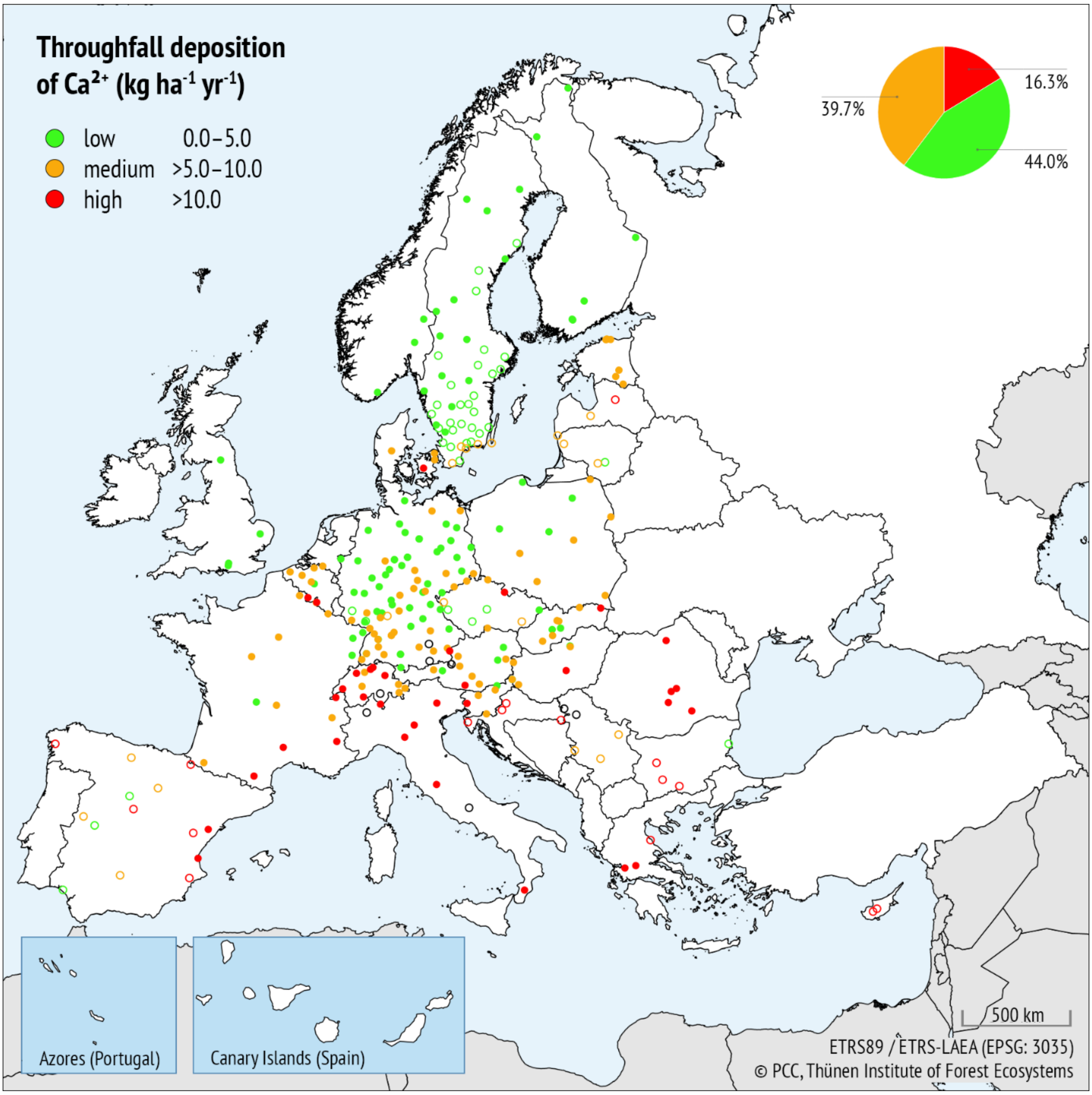

Figure 5-7: Throughfall deposition of calcium (kg Ca^{2+} ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days. Legend: low (green, 0.0–5.0 kg Ca^{2+} ha^{-1} yr^{-1}), medium (yellow, >5.0–10.0 kg Ca^{2+} ha^{-1} yr^{-1}), high (red, >10.0 kg Ca^{2+} ha^{-1} yr^{-1}).

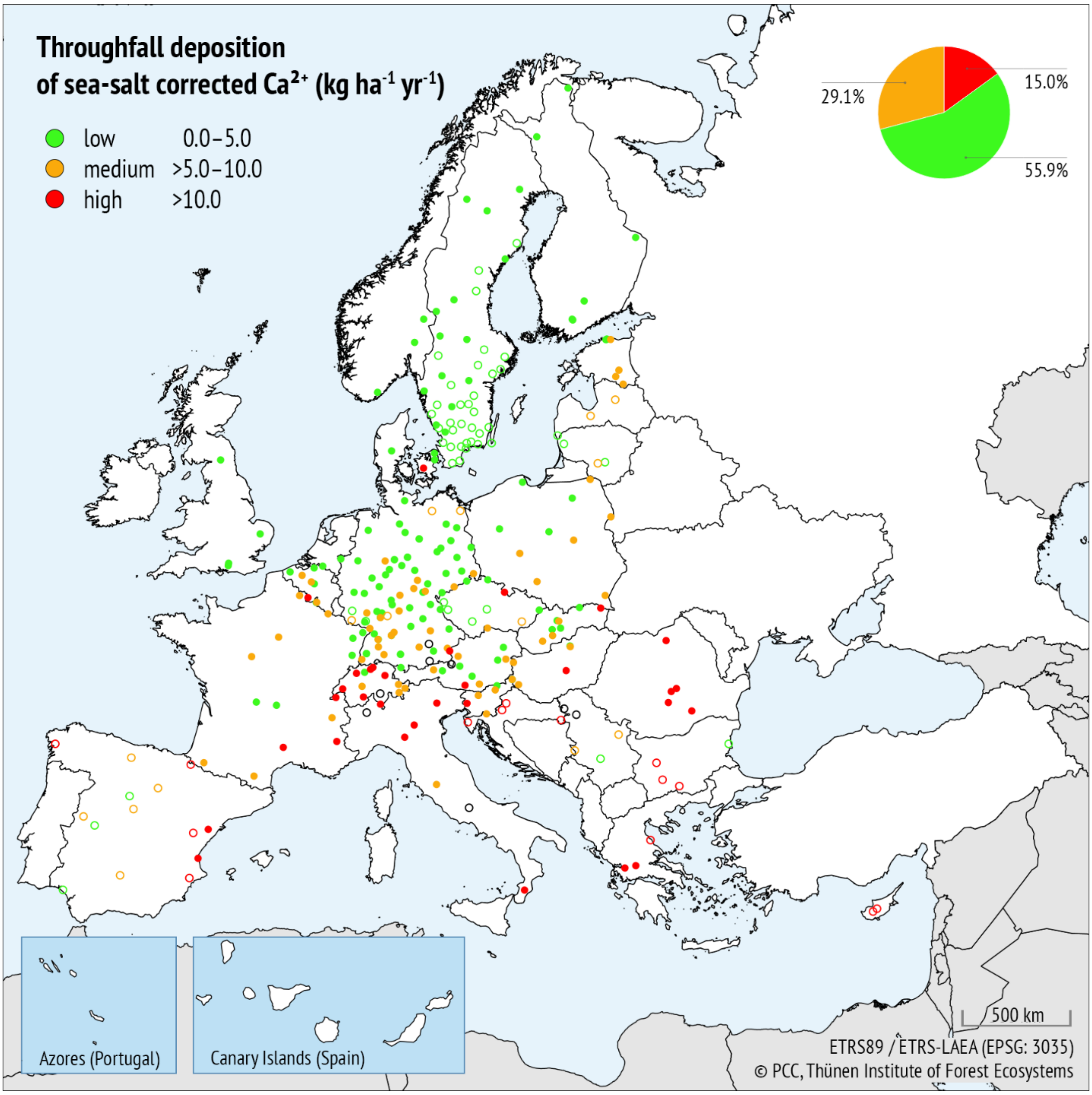

Figure 5-8: Throughfall deposition of sea-salt corrected calcium (kg Ca^{2+} ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days. Legend: low (green, 0.0–5.0 kg Ca^{2+} ha^{-1} yr^{-1}), medium (yellow, >5.0–10.0 kg Ca^{2+} ha^{-1} yr^{-1}), high (red, >10.0 kg Ca^{2+} ha^{-1} yr^{-1}).

EVALUATIONS

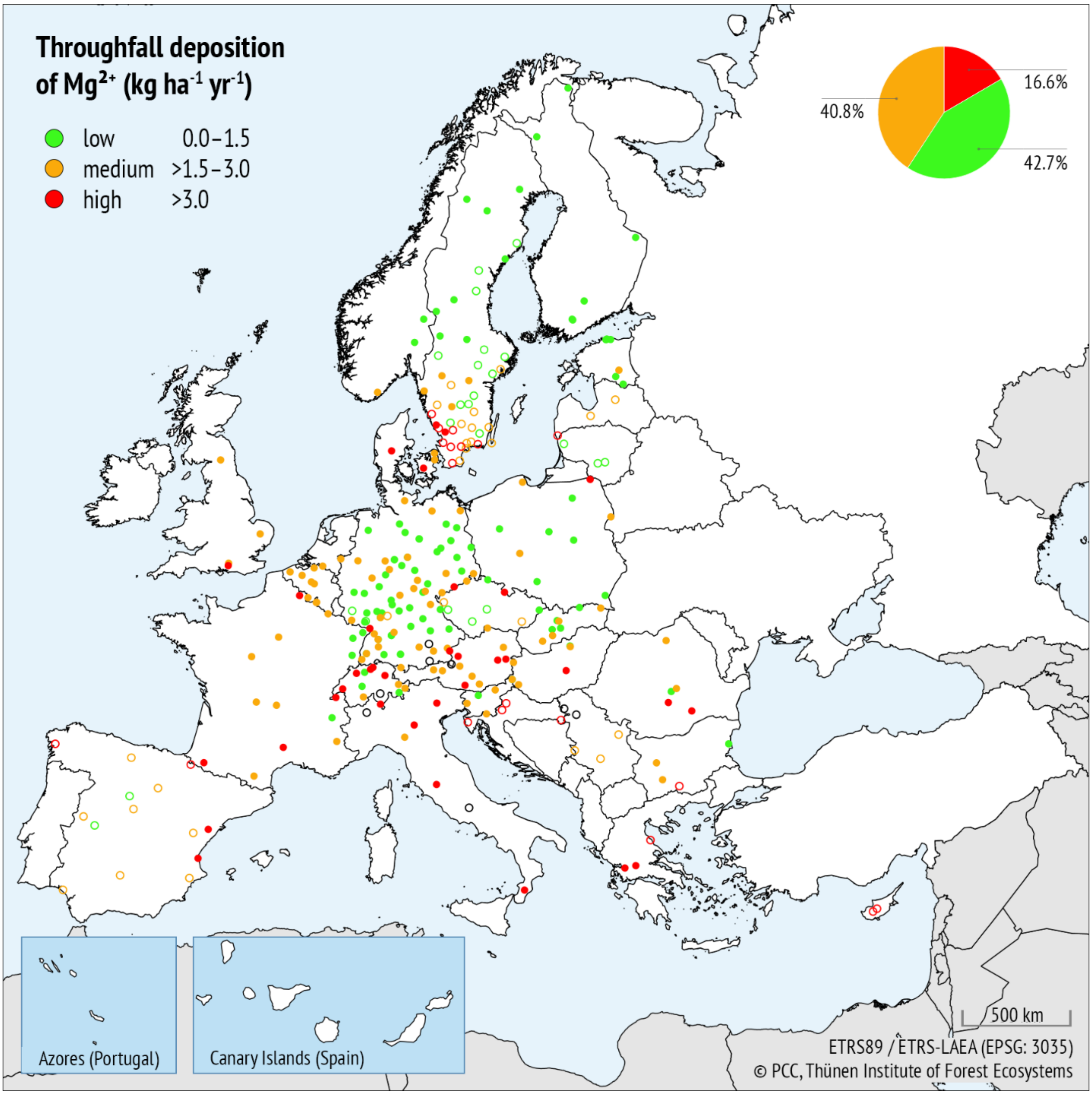

Figure 5-9: Throughfall deposition of magnesium (kg Mg^{2+} ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days. Legend: low (green, 0.0–1.5 kg Mg^{2+} ha^{-1} yr^{-1}), medium (yellow, >1.5–3.0 kg Mg^{2+} ha^{-1} yr^{-1}), high (red, >3.0 kg Mg^{2+} ha^{-1} yr^{-1}).

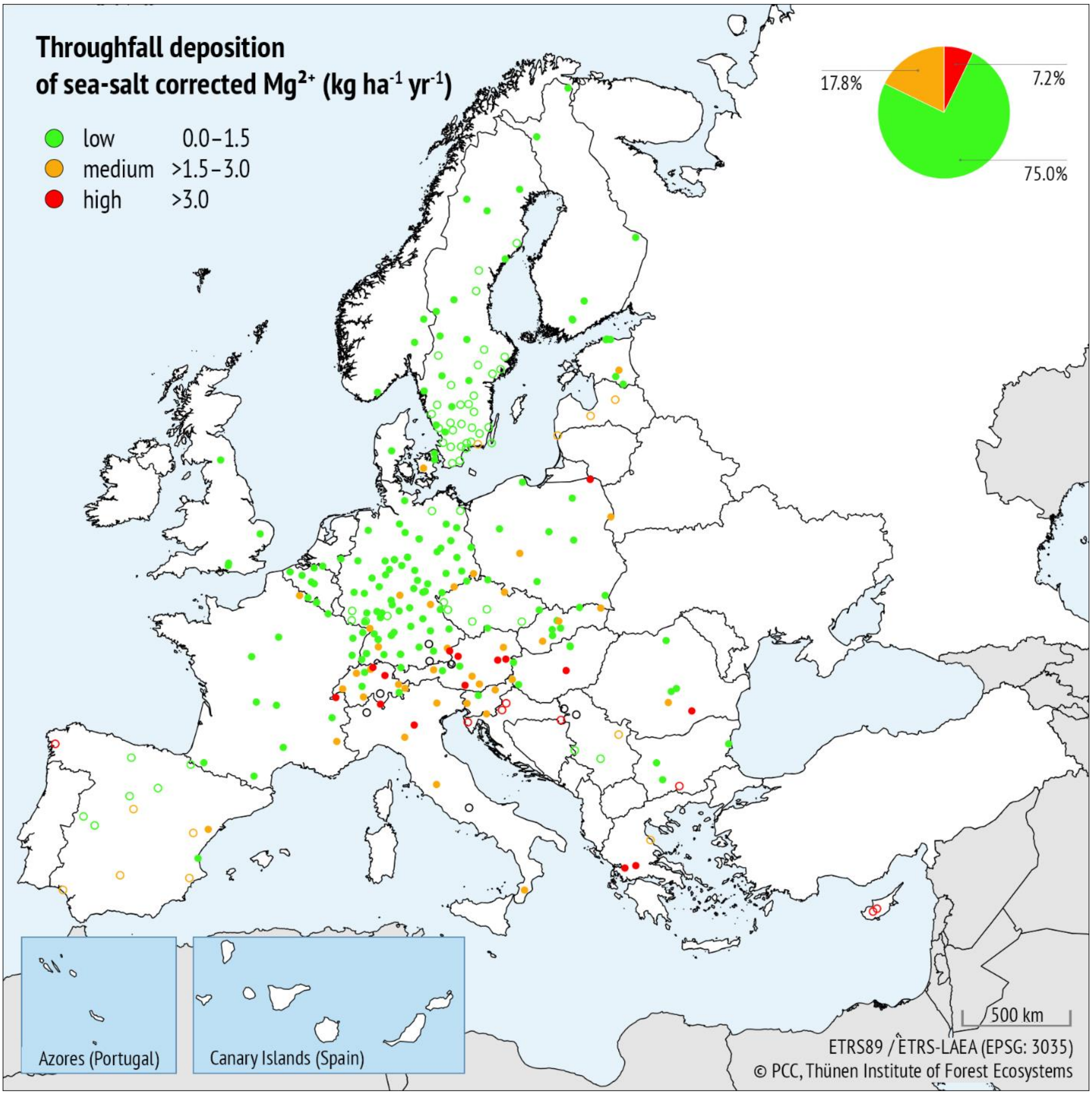

Figure 5-10: Throughfall deposition of sea-salt corrected magnesium (kg Mg^{2+} ha^{-1} yr^{-1}) measured in 2019 on the ICP Forests Level II plots and the Swedish Throughfall Monitoring Network. Colored dots: validated data. Colored circles: not validated data. Black circles: monitoring period shorter than 330 days. Legend: low (green 0.0–1.5 kg Mg^{2+} ha^{-1} yr^{-1}), medium (yellow, >1.5–3.0 kg Mg^{2+} ha^{-1} yr^{-1}), high (red, >3.0 kg Mg^{2+} ha^{-1} yr^{-1}).

References

Bobbink R, Hettelingh JP, eds (2011) **Review and revision of empirical critical loads and dose-response relationships.** Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM)

Bobbink R, Hicks K, Galloway J, Spranger T, Alkemade R, Ashmore M, Bustamante M, Cinderby S, Davidson E, Dentener F, Emmett B, Erisman JW, Fenn M, Gilliam F, Nordin A, Pardo L, De Vries W (2010) **Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis.** Ecol Appl 20:3059

Clarke N, Žlindra D, Ulrich E, Mosello R, Derome J, Derome K, König N, Lövblad G, Draaijers GPJ, Hansen K, Thimonier A, Waldner P (2016) **Part XIV: Sampling and Analysis of Deposition.** In: UNECE ICP Forests Programme Coordinating Centre (ed): Manual on methods and criteria for harmonized sampling, assessment, monitoring and analysis of the effects of air pollution on forests. Thünen Institute of Forest Ecosystems, Eberswalde, Germany, 32 p + Annex [http://www.icp-forests.org/manual.htm]

EEA (2016) **Emissions of the main air pollutants in Europe** – European Environment Agency, Copenhagen, Denmark. [https://www.eea.europa.eu/data-andmaps/indicators/main-anthropogenic-air-pollutantemissions/]

FEI (2013) **Data calculation** (Annex 7). In: Manual for Integrated Monitoring. http://www.syke.fi/nature/icpim > Manual for Integrated Monitoring. (Accessed 23.04.2020)

Guerrieri R, Vanguelova EI, Michalski G, Heaton THE, Mencuccini M (2015) **Isotopic evidence for the occurrence of biological nitrification and nitrogen deposition processing in forest canopies**. Glob Change Biol 21:4613–4626

König N, Kowalska A, Brunialti G, Ferretti M, Clarke N, Cools N, Derome J, Derome K, De Vos B, Fuerst A, Jakovljevič T, Marchetto A, Mosello R, O'Dea P, Tartari GA, Ulrich E (2016) **Part XVI: Quality Assurance and Control in Laboratories.** In: UNECE ICP Forests Programme Coordinating Centre (ed): **Manual on methods and criteria for harmonized sampling, assessment, monitoring and analysis of the effects of air pollution on forests.** Thünen Institute of Forest Ecosystems, Eberswalde, Germany, 46 p. + Annex [http://www.icp-forests.org/manual.htm]

Meunier CL, Gundale MJ, Sánchez IS, Liess A (2016) **Impact of nitrogen deposition on forest and lake food webs in nitrogen-limited environments**. Glob Change Biol 22:164–179

Mosello R, Amoriello M, Amoriello T, Arisci S, Carcano A, Clarke N, Derome J, Koenig N, Tartari G, Ulrich E (2005) **Validation of chemical analyses of atmospheric deposition in forested European sites.** J Limnol 64:93–102

Mosello R, Amoriello T, Benham S, Clarke N, Derome J, Derome K, Genouw G, Koenig N, Orrù A, Tartari G, Thimonier A, Ulrich E, Lindroos A (2008) **Validation of chemical analyses of atmospheric deposition on forested sites in Europe: 2. DOC concentration as an estimator of the organic ion charge.** J Limn 67:1–14

Silva LCR, Gómez-Guerrero A, Doane TA, Horwath WR (2015) **Isotopic and nutritional evidence for species- and sitespecific responses to N deposition and elevated CO_2 in temperate forests.** J Geophys Res Biogeosci 120:1110–1123

Waldner P, Marchetto A, Thimonier A, Schmitt M, Rogora M, Granke O, Mues V, Hansen K, Pihl Karlsson G, Žlindra D, Clarke N, Verstraeten A, Lazdins A, Schimming C, Iacoban, C, Lindroos AJ, Vanguelova E, Benham S, Meesenburg H, Nicolas M, Kowalska A, Apuhtin V, Napa U, Lachmanová Z,, Kristoefel F, Bleeker A, Ingerslev M, Vesterdal L, Molina J, Fischer U, Seidling W, Jonard M, O'Dea P, Johnson J, Fischer R, Lorenz M (2014) **Detection of temporal trends in atmospheric deposition of inorganic nitrogen and sulphate to forests in Europe.** Atmos Environ 95:363–374